SULFUR COATING OF UREA IN SHALLOW SPOUTED BEDS

by

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Abstract

Sulfur coated urea (SCU) is an effective and economical slow-release nitrogen fertilizer, and its production in a spouted bed was investigated. SCU was produced by batch and continuous operations. Higher quality products were typically produced by the batch process, but at significantly lower production rates than the continuous process. In order to understand such operations, mathematical models describing the coating process were developed and verified through experiments.

The production of SCU was studied in shallow spouted beds fitted with a pneumatic molten sulfur spray nozzle located at the cone inlet. Bed hydrodynamics, coating mechanism, particle coating distribution and product quality were examined under the following conditions: Bed diameter of cylindrical section - 0.24 and 0.45 m; bed height - 0.11 to 0.63 m; included cone angle - 60°; particle diameter - 2.1 to 2.8 mm; particle density 930 to 1490 kg/m³; main spouting air \leq 37 L(actual)/s; atomizing air \leq 0.87 L(actual)/s; urea feed rate - 7.6 to 20 g/s; sulfur injection rate - 2.1 to 6.1 g/s; orifice diameter - 21 to 35 mm; bed temperature - 18 to 70 °C; sulfur content \leq 60 %. The temperatures of atomizing air and molten sulfur were fixed for all runs at approximately 160 and 150 °C, respectively. The coating process was successfully modeled using mass and momentum balance equations, inertial sulfur droplet deposition as the dominant coating mechanism, and Monte Carlo simulations.

The hydrodynamic model was based on the one-dimensional mass and momentum balances suggested by Lefroy and Davidson (1969) for gas and particle motion in the spout, the axial pressure correlation given by Morgan and Littman (1980), and the vector form of the Ergun (1952) equation for gas motion in the annulus. The effect of atomizing air entering through the spray nozzle was successfully incorporated into the model by considering the total momentum flux into the bed. Conical beds were found to behave similar to conical-cylindrical beds having a column diameter of 80 % of the maximum conical bed diameter.

The dominant coating mechanism was deduced from the bed hydrodynamics and spray drop sizes produced by the pneumatic atomizing nozzle (type: internal mixing; Fluid Cap # 40100; Air Cap # 1401110; manufactured by Spraying System Co.). The drop sizes were found to range from approximately 6 to 50 μ m dia. The atomizing air flow rate did not affect the drop size distribution significantly under the conditions used in the present study. For the drop sizes produced and the hydrodynamic conditions prevailing in the spouted bed, inertial deposition was found to be the dominant mechanism for coating the bed particles.

On the basis of the bed hydrodynamics and the coating mechanism, the particle coating distributions were calculated utilizing the Monte Carlo method, and the quality of SCU particles was estimated from the coating distributions. The simulation results, which were in good agreement with the experimental data, imply that the product quality improved with increasing bed diameter, spouting and atomizing air flow rates, and that it decreased with increasing urea feed rates. Some improvement in product quality was also observed after changing the urea feed location and reducing the spray angle.

The model results also indicated that products with widely varying quality can be produced in a series of spouted beds at high production rates. This implies that the spouted bed is an effective and practical coating unit for producing SCU.

iii

Table of Contents

Abstract	ii
Table of Contents	iv
List of Tables	ix
List of Plates	x
List of Figures	xi
Acknowledgment	xv
Dedications	xvi
Chapter 1. Introduction	1
1.1. The UBC Spouted Bed Process	2
1.2. Objectives of Present Study	5
1.2.1. Bed Hydrodynamics	6
1.2.2. Coating Mechanisms	6
1.2.3. Overall Coating Performance	6
1.2.4. Benefits of the Study	7
Chapter 2. Literature Review	8
2.1. UBC Process	8
2.1.1. Product Quality	9
2.1.2. Effect of Bed Temperature on Product Quality	9
2.1.3. Effect of Sulfur Injection Rate on Product Quality	11
2.1.4. Effect of Atomizing Air on Product Quality	11
2.1.5. Effect of Bed Depth on Product Quality	12
2.1.6. Effect of Spouting Air Flow Rate on Product Quality	13
2.1.7. Effect of Chemical Additives on Product Quality	.13
2.2. Models of Spouted Bed Coating Process	13
2.3. Models and Correlations for Spouted Bed Hydrodynamics	16
2.3.1. Minimum Spouting Velocity	16

2.3.2. Solids Circulation and Bed Hydrodynamics	1
2.3.3. Spout Diameter	1
2.3.4. Pressure Profile in Annulus	1
2.4. Coating Mechanism	2
2.5. Monte Carlo Method	2
Chapter 3. Experimental Materials, Apparatus and Procedures	2
3.1. Experimental Materials	2
3.1.1. Urea	2
3.1.2. Sulfur	2
3.1.3. Particles Used in Hydrodynamics Study	2
3.2. Main Coating Apparatus	3
3.2.1. Spouted Bed	3
3.2.2. Sulfur Supply System	3
3.2.2.1. Sulfur Melter	3
3.2.2.2. Sulfur Filter	3
3.2.2.3. Sulfur Rotameter	3:
3.2.2.4. Sulfur Line	3
3.2.2.5. Nitrogen Supply	3'
3.2.3. Nozzle Assembly	3′
3.2.4. Urea Feeding Device	31
3.2.5. Product Withdrawal Device	39
3.2.6. Product Collector	39
3.2.7. Dust Collector	39
3.2.8. Air, Steam, and Water Supplies	40
3.3. Apparatus for Hydrodynamics Study	40
3.4. Apparatus for Spray Study	41
3.4.1. Spray Box	42
3.4.2. Spray Sampler	43
3.5. Coating Procedures	44
3.5.1. Start Up	44

3.5.2. Coating	45	
3.5.3. Shut Down and Clean-up	46	
3.6. Procedures for Hydrodynamics Study	46	
3.6.1. Minimum Spouting Velocity, U _{ms}	46	
3.6.2. Voidage of Loosely Packed Bed, ε_{mf}	47	
3.6.3. Mean Particle Diameter, d_p , and Sphericity, ϕ_s	47	
3.6.4. Diameter of Inlet Orifice, d _i	47	
3.6.5. Static Pressure in Annulus	48	
3.6.6. Air Velocity in the Spout, u_s	48	
3.6.7. Radial Velocity Profile at the Base of the Bed	49	
3.7. Procedures for Spray Studies	49	
3.7.1. Operating Limits of Spray Nozzle	49	
3.7.2. Spray Drop Size Measurements	50	
3.8. Product Quality Analysis	50	
3.8.1. Sulfur Content	50	
3.8.2. Particle Sulfur Content	51	
3.8.3. Seven Day Dissolution Test	52	
Chapter 4. Mathematical Models		
4.1. Simple Models	53	
4.1.1. Model I: Residence Time Model	53	
4.1.2. Model II: Simple Spray Zone Model	56	
4.1.3. Model III: Variable Concentration Spray Zone Model	61	
4.2. Model IV: Rigorous Model	62	
4.2.1. Calculation of Models for Solids Circulation Rate and Bed Hydrodynamics	63	
4.2.2. Determination of Coating Mechanism and Concentration Profile	66	
4.2.3. Calculation of Coating Distribution Using the Monte Carlo Method	68	
4.2.3.1. Limitation of Analytical Model	68	
4.2.3.2. Monte Carlo Procedure for Model III	69	

4.2.3.3. Monte Carlo Procedure for Model IV	70
4.2.3.3.1. Continuous Operation	70
4.2.3.3.2. Batch Operation	72
Chapter 5. Results and Discussions	74
5.1. Bed Hydrodynamics	74
5.1.1. Minimum Spouting Velocity, U _{ms}	75
5.1.2. Pressure Profile in Annulus	85
5.1.3. Velocity Profile in Annulus	89
5.1.4. Velocity Profile in Spout	89
5.1.5. Solids Movement	94
5.2. Spray Studies	94
5.2.1. Operating Limits	94
5.2.2. Spray Drop Size Distribution and Average Drop Size	96
5.2.3. Coating Mechanism and Sulfur Spray Concentration	102
5.3. Coating Distribution and Product Quality	105
5.3.1. Coating Distribution	107
5.3.2. Product Quality	110
5.3.3. Effect of Operating and Model Variables on Coating Distri- bution	112
5.3.3.1. Effect of Operating Time	112
5.3.3.2. Effect of Sample Size	115
5.3.3.2.1. Numerical Sampling	115
5.3.3.2.2. Manual Sampling	116
5.3.3.3. Effect of Spray Angle	118
5.3.3.4. Effect of Feed Location	120
5.3.3.5. Effect of Beds-in-Series	121
5.3.3.6. Effect of Model Variables, x_e and x_c	123
5.3.4. Sensitivity Analysis Using Model IV	125
5.4. Commercial Implications	130
Chapter 6. Conclusions and Recommendations	131
6.1. Conclusions	31

6.1.1. Bed Hydrodynamics	131
6.1.2. Spray Studies	132
6.1.3. Coating Distribution and Product Quality	132
6.2. Recommendations for Further Work	133
Nomenclature	134
References	141
Appendix I: Supporting Derivations and Methodology	149
I-1. Determination of Shutter Area	150
I-2. Model II Derivation for Forced Urea Feed	152
I-3. Calculation Method for Vector Ergun Equation	154
I-4. Sulfur Sampling Devices	156
I-5. Batch Coating Model	161
I-6. Minimum Spouting Velocity Predictions by Wan-Fyong et al. Equation	163
Appendix II: Experimental Data and Calculated Results	164
Appendix III: Computer Program Listings	179
Appendix IV: Calibration Results	218

•

List of Tables

Table	<u>page</u>
1.1 Major agronomic benefits associated with SCU usage (Tisdale et al., 1985)	1
2.1 Range of operating conditions used in previous studies on the UBC process	9
3.1 Selected physical properties of urea (Perry et al., 1984)	27
3.2 Selected chemical and physical properties of sulfur (Stauffer Chemical Co.)	28
3.3 Properties of common sulfur allotropes (Donahue and Meyer, 1965; Dale and Ludwig, 1965)	28
3.4 Physical properties of bed particles	30
3.5 Types and capacities of Brooks rotameters used in this work	36
4.1 Assumptions used in simple models	58
4.2 Selected infinite series definitions	59
5.1 Operating ranges applicable to this work	75
5.2 Operating conditions under which the air velocities in the spout and the pres- sure profiles in the annulus were determined	76
5.3 Axial pressure profile near the spout-annulus interface	77
5.4 Air velocity profile in the spout	78
5.5 Fitted constants for the Wu et al. (1987) equation	80
5.6 Operating ranges of the spray studies	95
5.7 Results of drop size studies at various operating conditions	100
5.8 Sauter mean diameters relative to the operating limits	101
5.9 Mechanical collection efficiency (using correlations suggested by Clift et al., 1981) at selected operating conditions in the spout	102
5.10 Operating range applicable to coating distribution and product quality studies 1	106
5.11 Operating conditions investigated for the coating study	106
5.12 Errors associated with sample sizes based on the Model II and III results	16

List of Tables Continued

Table	page
5.13 Effect of feed location (using Model II with $x_c = 0.5$, $\overline{X}_s = 0.25$)	121
5.14 Range of model variables examined for sensitivity analysis	127
5.15 Results of sensitivity analysis using Model IV ($\overline{X}_s = 0.25$, $T = 60$ °C)	128
I-1 Direct sulfur sampling	158
I-2 Hot tip: packed bed $S \Rightarrow SO_2$ converter	158
I-3 Hot tip: plate $S \Rightarrow SO_2$ converter	159
I-4 Hot tip: laser beam $S \Rightarrow SO_2$ converter	159
I-5 Hot tip: three tube hot wire $S \Rightarrow SO_2$ converter	160
I-6 Hot tip: two tube hot wire $S \Rightarrow SO_2$ converter	160
II-1 Minimum spouting velocity data	165
II-2 Spray drop size distribution	174
II-3 Measured sulfur content of individual particles for selected continuous runs	176
II-4 Predicted sulfur content of individual particles for selected continuous runs	177
II-5 Measured sulfur content of individual particles for batch runs shown in Figures 4.3 and 5.25	178
IV-1 Calibration equations for flowmeters and refractometer	219

List of Plates

Plate	<u>page</u>
3.1 Urea	24
3.2 Sulfur coated urea produced by batch process	24
3.3 Sulfur coated urea produced by continuous process	25
3.4 Polyformaldehyde	25
3.5 Polyethylene	26
3.6 Polystyrene	26

List of Figures

Figure	page
1.1 Schematic diagram of UBC spouted bed coating unit for producing sulfur coated urea (heavy arrows indicate direction of solids flow)	3
1.2 Sulfur content of CIL and UBC products (Tsai, 1986)	5
2.1 Effect of bed temperature on product quality. (The scatter results from the effects of other variables)	10
2.2 Effect of atomizing air flow rate on dissolution rate normalized for the sulfur flow rate (Weiss, 1981)	12
3.1 Viscosity of sulfur at low temperature range (Freeport Sulfur Co., 1954)	29
3.2 Simplified flowsheet of UBC spouted bed facility	31
3.3 Sectional view of spouted bed column	32
3.4 Shutter assembly (dimensions are given in mm) designed by Mathur, Meisen and Lim (1978)	33
3.5 Sectional view of 18.9 L sulfur reservoir	34
3.6 Sectional view of "slip-on" sulfur line connector	36
3.7 Sectional view of nozzle, perforated plate and steam chamber (all dimensions in mm; designed by Meisen, Lee and Le, 1986)	38
3.8 Modifications to the top of the spouted bed (i.e., see Figure 3.3) for hydrody- namics study	41
3.9 Schematic diagram of static pressure probe	42
3.10 Schematic diagram of S-type pitot tube (1/8" tubes and 3/8" tube are held in place with silver solder)	42
3.11 Spray box assembly	43
3.12 Simplified drawing of the rotating sulfur droplet sampler	44
3.13 Sectional view of atomizing nozzle and inlet air assembly (not drawn to scale)	48
4.1 Model I – perfectly mixed vessel	54

List of Figures Continued

Figure	<u>page</u>
4.2 Prediction of Model I using different numbers of continuously stirred tanks in	
series	55
4.3 Prediction of Model I	57
4.4 System described by Model II	58
4.5 Sectional view of the lower spout	66
4.6 Concentration balance on coating material in the spout	67
5.1 Comparison between experimental and predicted U_{ms} values	79
5.2 Radial velocity profile 10 mm above the shutter in an empty bed ($Q_s = 27.9$ L/s, $d_i = 24.7$ mm and $T = 65$ °C)	80
5.3 Effect of atomizing air on minimum spouting velocity (minimum spouting ve- locity is based only on the main spouting air flow rate; $D = 0.45$ m, $H = 0.31$ m, $d_i = 35$ mm)	82
5.4 Momentum flow of air into the spouted bed (dashed and solid lines represent 0.24 m and 0.45 m dia. beds, respectively)	82
5.5 Effect of bed height on minimum spouting velocity, Ums. (Cone-cylinder junc- tions are denoted by dashed lines; solid lines represent indicated relation- ships fitted to experimental data)	83
5.6 Comparison between experimental data and predictions from correlation based on the optimum diameter D'	83
5.7 Pressure profile in the annulus (conical bed, Run H22)	86
5.8 Axial pressure profile in the annulus near the spout-annulus interface (conical- cylindrical bed, Run H1)	86
5.9 Axial pressure profile in the annulus (conical bed, Run H22, column diameter used in calculation)	87
5.10 Axial pressure profile in the annulus (conical bed, Run H22, D' used in calcu- lation)	87
5.11 Axial pressure profile in the annulus ($H = 0.53$, Run H17)	88

List of Figures Continued

Figure	page
5.12 Comparison between measured and predicted pressure profile in the annulus	88
5.13 Effect of atomizing air on axial air velocity profile in the spout (Q_s is fixed at approximately 32 L/s in all runs)	9 0
5.14 Air velocities in the spout determined experimentally and from Model IV hydrodynamics (Run H1)	91
5.15 Air velocities in the spout determined experimentally and from Model IV hydrodynamics and Equation (5.5) (Run H22)	91
5.16 Operating limits for atomization of sulfur	96
5.17 Number distribution and predictions using log-normal equation (Run S1a)	97
5.18 Log-normal representation of drop size distribution (Run S1a)	98
5.19 Nukiyama-Tanasawa representation of drop size distribution (Run S1a)	98
5.20 Predictions of Sauter mean diameter	01
5.21 Sulfur concentration profile for various spray angles	.04
5.22 Sulfur concentration profile for various coating runs ($\phi = 20^{\circ}$)	04
5.23 Comparison between measured and predicted coating distributions for Run C17	08
5.24 Comparison between measured and predicted coating distributions for Run C38	08
5.25 Coating distributions of batch products	09
5.26 Relationship between 7-day dissolution and sulfur content for batch products 1	11
5.27 Comparison between measured and predicted product quality	12
5.28 Effect of operating period on coating distribution1	14
5.29 Effect of sample size on coating distribution (Models II and III)1	17
5.30 Effect of sample size on coating distribution (Model IV)	17
5.31 Effect of sample size (manual sampling)1	18
5.32 Effect of spray angle on coating distribution (results from Model IV)	19

List of Figures Continued

Figure	page
5.33 Effect of the spray concentration on coating distribution (results were ob- tained from Model III using Equation (4.19))	. 119
5.34 Effect of feed location	122
5.35 Product coating distributions for beds-in-series (from Model IV)	122
5.36 Sensitivity analysis of x_e on coating distribution using Model II ($x_c = 0.6$)	124
5.37 Sensitivity analysis of x_c on coating distribution using Model II ($x_e = 0.2$)	124
5.38 Model predictions of product quality	126
5.39 Effect of a large x_c on coating distribution ($x_c = 0.05$)	129
I-1 Shape of 1/5th of the shutter	150
I-2 Shape of a section of fully open shutter with the same base length (t) shown in Figure I-1	151
I-3 Simplified flow sheet of a forced feed system	152
I-4 Minimum spouting velocity predictions using Wan-Fyong et al. (1969) equation	163
IV-1 Calibration curve for the lower capacity spouting air rotameter	220
IV-2 Calibration curve for the higher capacity spouting air rotameter	221
IV-3 Calibration curve for the atomizing air rotameter	222
IV-4 Calibration curve for the sulfur rotameter	223
IV-5 Calibration curve for the urea feeder	224
IV-6 Predictions using the calibrated value of C_o for the static-pitot tube	225
IV-7 Predictions using the calibrated value of C_o for the S-type pitot tube	226
IV-8 Calibration curve for the Abbey refractometer	227

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and my daughter Ashleigh...

Chapter 1. Introduction

Sulfur coated urea (SCU) has been proven as an effective and economical slow release nitrogen fertilizer. SCU is produced by applying a light coating of water resistant sulfur on urea granules. In soil, the sulfur is slowly degraded by microorganisms and the urea is thereby exposed. For this reason SCU is classified as a slow release nitrogen (SRN) fertilizer. Previous studies (Davis, 1973; Waddington and Duich, 1976; Allen et al., 1971) showed that SCU is at least as effective as other SRN fertilizers or repeated applications of uncoated nitrogen fertilizers. Moreover, SCU is the least expensive SRN fertilizer currently on the market, and has the highest nitrogen content. Other agronomic benefits of using SCU are summarized in Table 1.1.

The major disadvantages of SCU, are that the sulfur increases the soil acidity and lowers the nitrogen content of the fertilizer. Although lime application can mitigate the acidifying effect and greater fertilizer dosage can make up the necessary nitrogen requirement, both remedies add to the total cost. Therefore, it is important to reduce the sulfur content of SCU without significantly lowering the quality of SCU as a slow release nitrogen fertilizer.

Benefits as a slow-release nitrogen (SRN) fertilizer:	Benefits of sulfur [*] (for sulfur deficient soils):		
• Increases efficiency of nitrogen use by the crop	• Increases intensity of grass		
 Reduces toxicity to the crop 	color		
Reduces application cost	• Enhances crop growth when		
• Decreases fertilizer run-off which pollutes local	applied with nitrogen		
waters			
Increases fertilizer storage life			
* Sulfur and nitrogen are macronutrients for crops.			

Table 1.1: Major agronomic benefits associated with SCU usage (Tisdale et al., 1985).

In order to produce high quality SCU an understanding of the production process is important. Two processes have been studied for manufacturing SCU: the Tennessee Valley Authority (TVA) rotary drum process and the UBC spouted bed process. The TVA process and its development were summarized by Tsai (1986) and will not be repeated here. The UBC process is the objective of this study and its description and developments are summarized in the following section.

1.1. The UBC Spouted Bed Process

Development of the spouted bed sulfur coating process started in 1975 by Meisen and Mathur. The equipment consisted mainly of a cylindrical vessel with a conical base filled with urea granules as shown in Figure 1.1. Air injected at the base of the apparatus forms a jet (spout) carrying particles entrained from the dense surrounding region (annulus). The particles are carried upwards until they reach the top of the bed (fountain) whence they fall back into the annulus. A cyclic pattern of particle movement is thereby established.

Coating is accomplished by spraying molten sulfur into the bottom of the bed coaxially with the spouting air. Each time a urea granule passes through the spray zone, it acquires a layer of sulfur which solidifies (if the bed is properly operated) by the time the particle reaches the top of the bed. Repeated passages through the spray zone build up the coat and reduce coat imperfections.

The SCU quality, expressed in terms of the 7-day dissolution $(D_{25})^1$ value, was found to depend principally on bed temperature and sulfur flow rate. Initial experiments and dissolution tests demonstrated poor reproducibility. Operational problems including nozzle

 $^{^{1}}D_{25}$ denotes the percentage of urea which dissolves when 50 g of sample containing 25 wt% sulfur are placed in 250 mL of water at 37.8°C for 7 days.



Figure 1.1: Schematic diagram of UBC spouted bed coating unit for producing sulfur coated urea (heavy arrows indicate direction of solids flow).

plugging and sulfur handling difficulties led to further studies by Meisen and co-workers (Zee, 1977 and Lim, 1978).

Successful batch-wise coating was achieved by Weiss and Meisen (1981, 1983). The product quality (D_{25}) was comparable to that of the CIL product made by the TVA process, and was found to depend on the sulfur droplet size, the spray distribution and the nature of the urea surface.

Some operational aspects of the continuous spouted bed process were examined by Meisen and Tsai (1986). In their initial study, they found the product from the UBC process gave higher D_{25} values (i.e., had lower quality) than those of the CIL product. They suspected that the higher D_{25} values of the UBC product resulted from a significant fraction of uncoated particles in the product. This is supported by Figure 1.2 which is a plot of the percentage of particles which contain less than a certain sulfur content (denoted by Probability %) as a function of sulfur content. The cumulative percentage is shown on a "probability scale". The latter implies that the plot would be a straight line if the sulfur content is normally distributed around the mean value. Figure 1.2 shows that 27% of the particles contained less than 10 % sulfur while only 5 % of the particles from the CIL product contained less than 10 % sulfur. Meisen and Tsai suspected that these uncoated particles resulted from fresh urea particles bypassing the spray zone and leaving the spouted bed prematurely. By changing the feed location and operating variables such as the bed height and the flow rate of the spouting air, a product comparable to the CIL product in terms of D_{25} values was obtained.

Many experiments are generally required to determine the effects of operating variables on the product quality. Since the cost of the experiments is high, an alternative method is sought. One way to reduce the number of experiments is to develop a mathematical model describing the coating process.



Figure 1.2: Sulfur content of CIL and UBC products (Tsai, 1986).

1.2. Objectives of Present Study

The main objective of the present study is to develop mathematical models for the spouted bed coating process for the production of sulfur coated urea. The models use probabilistic approaches and involve empirical and theoretical sub-models for the fluid and particle hydrodynamics and for the coating mechanism. Particular emphasis is placed on shallow beds (i.e., conical beds and beds extending just above the conical section) since they have low pressure drops and are therefore of greatest commercial interest. Other objectives of this study include the development and verification of the sub-models. The latter are presented in the following subsections.

1.2.1. Bed Hydrodynamics

Although many hydrodynamic models have been reported for conventional spouted beds, the hydrodynamics of spouted beds configured for coating has not been addressed in the literature. In particular, the special geometry at the air inlet, atomizing air and coating agent make the coater behave differently from conventional beds. The objectives of the hydrodynamic study are to:

- determine the effects of atomizing air and bed geometry on the air velocities in the bed, including the minimum spouting velocity;
- develop mathematical models (or modify existing models) to describe the bed hydrodynamics in a spouted bed coater;
- verify the models by experimentally examining the fluid velocities in the spout and the pressure distribution in the annulus.

1.2.2. Coating Mechanisms

The coating mechanism governs the rate at which the sulfur droplets deposit onto urea particles and, ultimately, the spray concentration profile in the bed. The objectives of this part of the study are to determine the coating mechanism by experimentally analyzing the spray droplet size distribution.

1.2.3. Overall Coating Performance

An important objective of this work is the verification of the model predictions on the coating performance of the bed. Once the model is developed, the following verifications are conducted:

- the coating distribution of particles is determined for selected experiments;
- the coating distribution is correlated with the product quality expressed in terms of D₂₅ values.

1.2.4. Benefits of the Study

Use of a mathematical model may be an inexpensive and fast alternative to conducting experiments to determine the optimum operating conditions. The model may also be useful in designing commercial plants because the product quality information is readily predictable for any size and number of beds the plant may require. The commercial implications based on model predictions are presented in Chapter 5.

A better understanding of the following areas is also achieved as a result of this study:

- the effectiveness of coating models of various complexities is identified;
- the bed hydrodynamics of a spouted bed configured for coating are elucidated;
- the sulfur atomization and urea-sulfur contact mechanisms are characterized.

It should be noted that the physical bonding of sulfur on urea, the related effect of temperature and the influence of chemical additives are not examined in this work. The effect of bed temperature on product quality is investigated only through its impact on bed hydrodynamics.

Chapter 2. Literature Review

2.1. UBC Process

Although the spouted bed coating process has been studied in the past (Singiser et al., 1966; Umaki and Mathur, 1976), the effects of individual operating variables on product quality were not investigated until Meisen and Mathur commenced their research in 1975. The results obtained by Meisen and co-workers since 1975 are reviewed in this section.

The batch-wise² production of sulfur coated urea was studied by Meisen and co-workers – Mathur (1978), Zee (1977), Lim (1978), and Weiss (1981, 1983), and the continuous³ production was studied by Meisen and Tsai (1986). Most of the earlier work was devoted to improving the operational aspects of the process. (Detailed description of equipment modifications are provided by Weiss (1981) and by Tsai (1986)). With an improved coating facility, Lim was able to quantitatively explain the effects of the principal operating variables on the product quality for the batch process. Weiss followed up with more equipment modifications and more extensive studies of the batch process. Tsai extended the investigation to continuous operation. The operating conditions studied are summarized in Table 2.1.

The principal operating variables that were found to affect the product quality were bed temperature (T_b) , sulfur injection rate (W_s) , atomizing air flow rate (Q_a) , bed depth (H),

² The "batch process" refers to a process where a batch of urea is placed in the spouted bed, coated and discharged.

³ The "continuous process" refers to the process where urea is continuously fed to the bed and coated product (SCU) is continuously discharged from the bed.

Authors	<i>H</i> , m	<i>Т_b</i> , °С	Q_{s} , m ³ /min	Q_{a} , m ³ /h	W _s , g/min	<i>d_i</i> , mm
Lim, 1978	~ 0.41	37 - 80	0.38 - 1.28	0.39 - 0.83	54 - 86	16.1
Weiss, 1981	0.27 - 0.54	48 - 86	0.89 - 1.44	0.4 - 0.79	34 - 260	n/a
Tsai, 1986	0.15 & 0.24	50 - 87	0.6 - 1.1	0.35 - 0.65	22 - 76	n/a

Table 2.1: Range of operating conditions used in previous studies on the UBC process. (Fixed variables: Column diameter = 0.15 m, cone angle = 60°, nozzle type = "internal-mixing" (supplied by Spraying Systems Co., fluid cap #2050, air cap #67147)).

spouting air flow rate (Q_s) and chemical additives. A summary and review of the findings and explanations by Lim, Weiss, and Tsai are provided in the following sub-sections.

2.1.1. Product Quality

The quality of sulfur coated urea may be evaluated by laboratory or field tests. The seven day dissolution test was developed by TVA to measure the product quality in the laboratory and the dissolution value (D_{25}) obtained from this test has been adopted as the standard measure of the product quality by Meisen and co-workers.

Tsai found that products could have the same D_{25} value even though their instantaneous dissolution rates were very different. In particular, high initial dissolution rates were typically observed with products from the continuous process.

2.1.2. Effect of Bed Temperature on Product Quality

Previous researchers of the TVA process (Shirley and Meline, 1975) and the UBC process found that the bed temperature was the most important operating parameter affecting the product quality. For both processes, the optimum coating temperature was near 80 °C; however, the results (see Figure 2.1) for the UBC process show considerable scatter. The scatter was explained by the effects of other operating variables on D_{25} (Meisen and Mathur, 1978; Weiss and Meisen, 1983).



Figure 2.1: Effect of bed temperature on product quality. (The scatter results from the effects of other variables).

Upon examination of the coat surface under high magnification, Weiss was able to provide some explanations for the optimum coating temperature. For bed temperatures below 80 °C, premature freezing of sulfur droplets before impingement onto the bed particles prevented the sulfur from spreading evenly on the urea surface. As a result, the surface appeared lumpy with gaps between the lumps, and the presence of these gaps enhanced the passage of water to the urea core thereby increasing the urea dissolution rate. Urea coated significantly above 80 °C showed cracks in the coats. The cracks were thought to be caused by the contraction of sulfur as it changed from the monoclinic allotrope (S_{β}) of lower density to the orthorhombic form (S_{α}) of higher density upon cooling. For bed temperatures below 80 °C, S_{β} was assumed to be absent from the coats and therefore major thermal contraction did not take place.

2.1.3. Effect of Sulfur Injection Rate on Product Quality

The experimental results obtained by Lim and Weiss suggest that the sulfur feed rate had a strong effect on the product quality. To determine the relationship between the sulfur injection rate and D_{25} , Weiss conducted several experiments by varying only the sulfur feed rate at a given bed temperature. The results showed the logarithm of D_{25} to be inversely proportional to the sulfur flow rate. According to the Nukiyama-Tanasawa equation (1939), the spray droplet size (d_s) increases with liquid flow rate (Q_l) : i.e.,

$$d_{s} = \frac{585}{u_{r}} \left(\frac{\sigma_{l}}{\rho_{l}}\right)^{0.5} + 597 \left(\frac{\mu_{l}}{\sqrt{\sigma_{l}\rho_{l}}}\right)^{0.45} \left(\frac{1000Q_{l}}{Q_{a}}\right)^{1.5}$$
(2.1)

Weiss therefore concluded that the product quality improves with sulfur droplet size.

2.1.4. Effect of Atomizing Air on Product Quality

Lim (1978) found that the product quality decreased as the atomizing air flow rate was increased. Weiss found this relationship to be linear when his results were normalized to take into account the effect of sulfur flow rate on product quality (see Figure 2.2). Using the relationship given by Nukiyama and Tanasawa (Equation (2.1)), Weiss confirmed that smaller spray droplet sizes gave rise to a lower product quality (or higher D_{25} values).

The present hydrodynamic study (see Chapter 5) showed that the atomizing air flow rate could significantly influence the solids circulation and the size of the spray zone, in addition to the spary droplet size. Moreover, the spray angle was observed to vary with atomizing air flow rate. These effects of atomizing air flow rate could ultimately alter the product quality; however, such effects were not addressed in the previous studies.



Figure 2.2: Effect of atomizing air flow rate on dissolution rate adjusted to a reference sulfur flow rate (Weiss, 1981).

2.1.5. Effect of Bed Depth on Product Quality

The batch studies conducted by Lim (1978) suggested that reducing the bed height improves the product quality. Shorter particle cycle times associated with shallower beds were thought to increase the chance of particles receiving uniform coats and hence improve the product quality.

However, Weiss' experimental results showed that the bed height had little effect on product quality. In his investigation on the effect of bed depth, the spouting air flow rate had to be increased with bed depth to maintain spouting (other variables were fixed). Increasing the spouting air flow rate, according to Mathur and Epstein (1974), increases the urea circulation rate and consequently lowers the sulfur deposition on the urea particles per pass. owever, Weiss found that the changes resulting from the variation in bed depth and spouting air flow rate did not alter the product quality significantly.

Tsai found the bed depth to have a more pronounced influence on the product quality in the continuous process. In oder to explain his results, Tsai introduced the concept of the "spray zone". The spray zone was assumed to cover the lower portion of the spout and consequently the particles that enter the spout above the spray zone did not receive any additional coating. As the bed height increases, more particles by-pass the spray zone, resulting in more inadequately coated particles and a lower product quality.

The concept of the "spray zone" is explored in the development of the mathematical models in Chapter 4.

2.1.6. Effect of Spouting Air Flow Rate on Product Quality

Lim (1978) found that the spouting air flow rate had little effect on the product quality in the batch coating process. Weiss and Tsai did not investigate the effect of the spouting air flow rate.

2.1.7. Effect of Chemical Additives on Product Quality

Silicone (Dow Corning 200) was found to improve the product quality according to Weiss (1981). Other chemical additives including CO_2 , NH_3 , N_2 and liquid dicyclopentadiene resulted in no major improvements in product quality in the study conducted by Lim (1978).

2.2. Models of Spouted Bed Coating Process

Basically three approaches for modeling spray coating in spouted beds have been reported in the past. The earliest paper was published by Umaki and Mathur in 1976. The model of a continuous granulation process was based on mass and number balances which took into account particle growth by solute deposition, particle breakage and dust formation due to particle abrasion and undeposited spray droplets. The model correlated the experimental particle growth rate data reasonably well. Unfortunately, as pointed out by Mann (1978), the assumptions of constant bed weight, and constant ratio of the formation rate of fresh nuclei to the number of particles in the bed implied that the particle growth resulted in a reduction of the formation of fresh nuclei, which would ultimately lead to just one or two large particles in the bed. This model was also limited to predicting the mean bed particle size. The particle size distribution, which is an important feature of slow release fertilizers, could not be predicted.

In 1983, Mann developed a model which predicted the coating distribution of the solids produced in a spout-fluid bed equipped with a draft tube and operating in batch mode. Mann believed that the coating distribution is mainly affected by the number distribution of passages through the spray zone and the distribution of the coating mass deposited on the particles per passage. Based on the findings of Cox (1967) and Mann (1974, 1975, 1979 and 1981) that the latter two distributions asymptotically approached normal distributions with time, Mann developed a 5-parameter model. The parameters were operating time, mean and variance of cycle time, and mean and variance of coating amount per cycle. Mann suggested 90 short experimental runs to relate the latter four variables to bed diameter, annulus width, height of draft tube from the base, atomizer type, air flow rate, and coating solution flow rate.

For a coating process in which the coat itself does not significantly add to the size of the bed particles and for a fixed bed geometry, this approach appears to be valid. However, if the coating material significantly increases the size of the bed particles, short experiments may not give proper indication of the effect of the latter four variables. The increase in the

particle size results in particles receiving more coating material per cycle; this adds to the bed height which may, in turn, decrease the cycle time of the bed particles. Moreover, as the model is modified for continuous operation, additional operating variables may need to be considered. In such cases, the number of experiments required to determine the relationship between the operating variables and the model variables will increase significantly. Such an increase in the number of experiments may render this approach ineffective for practical purposes.

In 1984, Berruti et al. developed a mathematical model for predicting the size distribution of solids formed in a continuous spouted bed coating process. They assumed perfect particle mixing, constant feed composition (i.e., feed rates of coating material and bed particles), no particle segregation at the product discharge, constant hold up and negligible particle breakage and fines formation. On the basis of a particle population balance, which was developed by Randolph and Larson (1971) for describing a crystallization process, the model predicted the size distribution of the bed particles under transient conditions. Prior knowledge of the feed rate, mean feed particle size and growth rate were required to determine the product size distribution. The authors kept the growth rate and the mean feed particle size constant while altering the feed rate. Their results showed that the size distribution of the product particles approached a log-normal distribution.

Two assumptions, namely perfect particle mixing and a well-dispersed homogeneous gas phase containing the coating agent, seem, however, unrealistic for the spouted bed processes.

None of the three aforementioned models considered, in detail, the coating mechanisms and particle circulation patterns inside the spouted bed. Umaki and Mathur (1976), and Berruti et al. (1984) treated the bed as a perfectly stirred vessel and assumed that particles in all regions of the bed received the same amount of coating. Mann (1983) assumed a spray zone, but his model variables were empirical. These variables could not explain the particle circulation in the bed and the coating mechanism.

2.3. Models and Correlations for Spouted Bed Hydrodynamics

Since only a few hydrodynamic models and correlations for shallow spouted beds are available in the literature, those that are widely used for standard spouted beds are also considered here. It should be noted that detailed and critical review of most hydrody-namic models and correlations considered here can be found elsewhere (Mathur and Epstein, 1974; Epstein and Grace, 1984; Krzywanski, 1992).

2.3.1. Minimum Spouting Velocity

For cylindrical vessels up to about 0.6 m in diameter with conical base, the most reliable (within \pm 10 %) correlation for the minimum spouting velocity is, according to Epstein and Grace (1984), the Mathur-Gishler (1955) equation:

$$U_{\rm ms} = \frac{d_p}{D} \left(\frac{d_i}{D}\right)^{1/3} \sqrt{2gH \frac{\rho_p - \rho}{\rho}}$$
(2.2)

Wu et al. (1987) modified the Mathur-Gishler equation by separating the density and bed height terms in the following way:

$$U_{ms} = k\sqrt{2gH} \left(\frac{d_p}{D}\right)^{\alpha} \left(\frac{d_i}{D}\right)^{\beta} \left(\frac{H}{D}\right)^{\gamma} \left(\frac{\rho_p - \rho}{\rho}\right)^{\delta}$$
(2.3)

The constants k, α , β , γ , and δ were calculated using a standard least squares technique to fit 112 data points. The values of the constants are 10.6, 1.05, 0.266, -0.095 and 0.256, respectively. The modified equation improved the U_{ms} predictions significantly, especially at temperatures well above ambient.

2.3.2. Solids Circulation and Bed Hydrodynamics

Basically three approaches for predicting the particle circulation rates have been reported in the literature. Two approaches, according to Morgan et al. (1985), involve using onedimensional particle force, and mass and momentum balances in the spout. A more recent and rigorous approach is based on the theory of plasticity for the solids motion in the annulus (Krzywanski et al., 1992; Amirshahidi, 1984; Khoe, 1980). The former two approaches also predict fluid velocity and voidage profiles in the spout, while the latter approach does not. However, Krzywanski et al. (1992) combined the theory of plasticity with the vector Ergun (1952) equation in the annulus and the two-phase momentum equations in the spout to solve for the bed hydrodynamics.

The most recent force balance model developed by Lim and Mathur (1978) had problems with the stiffness of the model equations at the bed inlet, although its predictions using experimental values away from the inlet as the initial conditions were reasonable. Khoe (1980) and Amirshahidi (1984) determined the solids flow in the annulus based on the theory of plasticity. The model developed by Khoe was strongly dependent on experimental results – the magnitudes and locations of sources and sinks were found experimentally. Amirshahidi encountered difficulties in the computation of the stress field in the conical region which is required to calculate the velocity field of solids. The theory of plasticity was applied to solve solids flow in the annulus; the solids flow in the rest of the bed would require additional equations. The Krzywanski et al. model required basic information such as wall and internal friction angles as well as the fluid velocity profile at the fluid inlet, which are not easily calculated nor readily available.

Although the model developed by Krzywanski et al. is the most comprehensive model available, applying it to the current study requires extraordinary computational resources. Furthermore, the model must be corrected for the unusual geometry at the bed inlet due to the presence of the nozzle in the coating unit (see Chapter 5). Accounting for the nozzle is a complex task because the flow rates of atomizing air and spouting air, the location and type of the spray nozzle all affect the boundary conditions of the model. Moreover, the bed hydrodynamics were found to be very sensitive to the friction angles which probably vary with the amount of sulfur on the particles.

The mass and momentum balances model, on the other hand, is much simpler, and yet, it was generally found to provide good approximations of solids circulation and bed hydrodynamics with little or no modification (Lefroy and Davidson, 1969; Morgan et al., 1985; Stocker, 1987). This model, however, required accurate estimates of spout diameter (D_s) , pressure distribution in the spout (P_s) , particle-fluid interaction (β_p) and air flow into the annulus (U_r) . The correlations for the first two of these variables are reviewed in the next two sections.

2.3.3. Spout Diameter

Although several spout shapes have been observed (Mathur and Epstein, 1974), a constant spout diameter has been commonly assumed (Epstein and Grace, 1984). The most recent correlation of the average spout diameter was developed by Wu et al. (1987) which is a modification of the equation given by Bridgewater and Mathur (1972). The modified equation resulted from a new set of fitted constants determined by applying a least squares fit to their data (which were obtained with D = 0.154 m; $\rho_p = 2600$ kg/m³; $0.945 < d_p < 1.665$ mm; $12.7 < d_i < 26.6$ mm; 0.168 < H < 1.38 m; $0.168 < \rho < 1.259$ kg/m³; $10.9 < \mu < 32.0 \times 10^{-6}$ kg/m·s), i.e.

$$D_{s} = 5.61 G^{0.433} D^{0.583} \mu^{0.133} / (\rho_{b} \rho g)^{0.283}$$
(2.4)

where $G = \rho U$ (2.5)

and $\rho_b = \rho_p (1 - \varepsilon_{mf}).$ (2.6)

This equation was found to give better predictions of their data at elevated temperatures than the McNab (1972) equation.

2.3.4. Pressure Profile in Annulus

Several pressure distribution correlations have been reported in the literature. Lefroy and Davidson (1969) used an empirical correlation based on the pressure measurements at the spout-annulus interface. The only model with a strong theoretical basis was developed by Epstein and Levine (1978); it was derived from the Ergun equation (1952) and the Mamuro and Hattori flow correlation (1968), and is given by

$$\frac{P_a - P_H}{-\Delta P_f} = \frac{1}{h(2\alpha_p - 1)} \Big[2(\alpha_p - 1)[1.5(h^2 - x^2) - (h^3 - x^3) + 0.25(h^4 - x^4)] + 3[3(h^3 - x^3) - 4.5(h^4 - x^4) + 3(h^5 - x^5) - (h^6 - x^6) + 0.143(h^7 - x^7)] \Big]$$
(2.7)

where
$$\alpha_p = 2 + \frac{129\mu(1 - \varepsilon_{mf})}{\rho d_p U_{mf}}$$
, (2.8)

$$h = H/H_m, \tag{2.9}$$

and $x = z/H_m$. (2.10)

This equation describes the pressure distribution in the annulus but, since there is a relatively small pressure drop between the spout and annulus (Rovero et al., 1985), it can also be used to estimate the pressure distribution in the spout.

Morgan and Littman (1980) developed the following general pressure drop correlation based on experimental pressure measurements reported in the literature:

$$\Delta P_{ms} / \Delta P_{mf} = 1 - Y \tag{2.11}$$

where

$$Y^{2} + [2(X - 0.2) - 1.8 + (3.24/\varphi)]Y + [(X - 2)(X - 0.2) - (3.24X/\varphi)] = 0, (2.12)$$
$$X = 1/[(H/D) + 1], (2.13)$$

and
$$\varphi = 7.18 \left(\frac{\rho U_{mf} U_i}{(\rho_p - \rho)g d_i} \phi_s (5\phi_s^3 - 7.57\phi_s^2 + 4.09\phi_s - 0.516) - \frac{d_i}{D} \right) + 1.07$$
 (2.14)

Fluid flow models for the annulus can also be modified to predict the pressure distribution with the aid of expressions such as the Ergun (1952) equation. Mamuro and Hattori (1968) derived a fluid flow model for the annulus using Darcy's law and Rovero et al. (1983) modified the Mamuro-Hattori equation for beds having a conical base by substituting

$$A_a = \pi \left(D^2 - D_s^2 \right) / 4 = A_0 \text{, for } z > H_c$$
(2.15)

and
$$A_a = \pi \left[(2z \tan(\theta/2) + D_b)^2 - D_s^2 \right] / 4$$
, for $z \le H_c$ (2.16)

into
$$\frac{dQ_a}{dz} \left(\frac{d^2 Q_a}{dz^2} \right) = \frac{BQ_a}{A_a} - BU_{aH_a}$$
(2.17)

where
$$B = 18U_{aH_m} A_a^2 / H_m^3$$
 (2.18)

and
$$Q_a = A_a U_a$$
 (2.19)

The boundary conditions are:

$$Q_a = Q_{a0}$$
 at $z = 0$ (2.20)

$$Q_a = U_{aH_a} A_a \text{ at } z = H_m. \tag{2.21}$$

2.4. Coating Mechanism

Four mechanical collection processes⁴ have been reported in the literature (Clift et al., 1981; Lunde and Lapple, 1957): diffusional deposition, inertial deposition, direct interception and gravitational settling. These processes are often aided by electrophoretic, thermophoretic and diffusiophoretic effects (Meisen et al., 1971). However, only the inertial deposition mechanism is considered because it was inferred that it was the dominating

⁴ collection process, in this study, corresponds to the mechanisms by which the atomized sulfur droplets deposit onto the bed particles.

mechanism under the experimental conditions prevailing in this study. Thus, the review in this section covers more recent inertial deposition correlations reported in the literature.

According to Clift et al. (1981), the correlation given by Thambimuthu (1980) is the most reliable equation for predicting the inertial impaction efficiency of a single spherical collector:

$$\eta = (S_t / (S_t + 0.062\varepsilon))^3, \text{ for } 0.002 < S_t < 0.02$$
(2.22)

where
$$S_t = (u - v) d_c^2 \rho_c / 18 \mu d_p$$
 (2.23)

The range of Stokes numbers (S_{ν}) , however, is rather small and is not generally applicable to the conditions of this study.

Earlier work by Behie et al. (1972) led to equations which are valid for a wider range of Stokes numbers:

$$\eta = 0, \text{ for } S_t \le 0.083$$
 (2.24)

$$\eta = 0.0036 - 0.2323S_t + 2.422S_t^2 - 2.033S_t^3, \text{ for } 0.083 < S_t \le 0.6$$
 (2.25)

$$\eta = S_t^2 / (S_t + 0.5)^2$$
, for $S_t > 0.6$ (2.26)

These equations for the single particle collection efficiency were obtained based on the assumption that the aerosols were rigid and spherical.

2.5. Monte Carlo Method

The term "Monte Carlo" was introduced by von Neumann and Ulam during World War II, as a code word for the secret work at Los Alamos; it was suggested by the gambling casinos at the city of Monte Carlo in Monaco (Rubinstein, 1981). The Monte Carlo method has been often confused with "stochastic simulation", and Ripley (1987) suggested that the term "Monte Carlo method" should have the more specialized meaning of "doing something clever and stochastic with simulation". Rubinstein (1981) defines stochastic simulation as statistical sampling experiments with a model over *time* which involves the use of a random number; Monte Carlo simulation as a technique using random or pseudorandom numbers for solving model equations. The latter definition is used to describe the model developed in this work.

Although this type of simulation is often viewed as a "method of last resort" to be employed when everything else has failed, recent advances in simulation methodologies, availability of software, and technical developments have made Monte Carlo simulation one of the most widely used and accepted tools in system analysis and operations research (Rubinstein, 1981). The reasons for using the Monte Carlo method, in the past, included

- (i) The data are impossible or very expensive to obtain (e.g. the performance of largescale rocket engines and the effect of proposed tax cuts on the economy);
- (ii) The system cannot be described in terms of a set of mathematical equations for which analytic solutions are obtainable;
- (iii) The solution to a mathematical model cannot be obtained by straight-forward analytic techniques;
- (iv) The experimental verification of the mathematical models describing the system is impossible or very costly to perform.

In this work, the Monte Carlo method was used because of reason (iii).

Chapter 3.

Experimental Materials, Apparatus and Procedures

The coating apparatus developed by Meisen et al. (1986) was used for all experiments conducted in this study. Minor modifications to the spouted beds were necessary for the study of bed hydrodynamics and a spray box was built to determine size distribution of the spray droplets. The experimental procedures used for the coating studies were similar to those described by previous workers (Weiss, 1981 and Tsai, 1986). Since the apparatus used in this work was largely the same as that used by previous workers, more emphasis is placed, in this chapter, on describing the modifications to the equipment.

3.1. Experimental Materials

Urea and sulfur are the only materials used to produce sulfur coated urea in this study; wax, silicone and other chemical additives were not needed. Urea and sulfur coated urea particles are shown in Plates 3.1 - 3.3. The chemical and physical properties of sulfur and urea are important in determining various operating limits of the coating process, and are discussed in the following subsections.

Initially, only urea and sulfur coated urea (SCU) were considered for the hydrodynamics study; however, considerable attrition was encountered with these particles. Therefore, polyformaldehyde, polyethylene and polystyrene particles in addition to the urea and SCU particles, were also used for this purpose. They are shown in Plates 3.4, 3.5 and 3.6, respectively, and discussed in Section 3.1.3.



Plate 3.1: Urea.



Plate 3.2: Sulfur coated urea produced by batch process.



Plate 3.3: Sulfur coated urea produced by continuous process.



Plate 3.4: Polyformaldehyde.



Plate 3.5: Polyethylene.



Plate 3.6: Polystyrene.

3.1.1. Urea

Urea was supplied by Sherritt-Gordon Ltd. and produced by the NSM fluidized bed granulation process. Selected properties of urea are listed in Table 3.1.

The operating bed temperature was kept above 60 °C. Heavy attrition was observed at room temperatures, i.e., dust build up on the Plexiglas column was observed almost immediately after spouting started. The attrition rate gradually decreased with increasing temperature; however, no data were collected to determine the effect of temperature on the attrition rate. The lower operating limit of 60°C was adopted based on visual inspection of dust build up on the column walls.

Other factors that appeared to influence the attrition rate were spouting air flow rate, spouting air orifice diameter, and bed diameter. Higher spouting air flow rates, small orifices, and smaller bed diameter increased the attrition rate.

Table 3.1: Selected physical properties of urea.

Melting point (Perry et al., 1984)133°CSphericity1.0Particle density (Perry et al., 1984)1335 kg/m³

Mesh (CDN)	Aperture Size (mm)	Wt. fraction (%)
+ 6	3.36	2.8
-6+7	2.83	24.5
- 7 + 8	2.38	33.5
- 8 + 10	2.0	30.1
- 10 + 16	1.19	9.1
- 16		trace
mean size = $\left[\sum_{i} x_{i} / d_{pi}\right]^{-1}$	2.16	

Size distribution (Sherritt-Gordon Ltd.)

3.1.2. Sulfur

PETROSUL International Ltd. provided sulfur for this work. The sulfur purity exceeded 99.5 wt % due to the presence of only minor traces of ash and carbon (total of 0.10 % on average).

Selected properties of solid and liquid sulfur are given in Tables 3.2 and 3.3, and Figure

Physical state (@ 21°C, 1 atm)	Solid
Bulk density, kg/m ³	1200 - 1394 Lumps, 560 - 960 Powder
Boiling point	444°C
Melting point	119°C (approximate)
Odor	None
Flash point	188°C, COC
Auto ignition temperature (dust in air)	190°C
Vapor pressure @ 20°C	< 0.0001 mm Hg
Explosive limits (dust in air)	between 35 and 1400 g/m ³

Table 3.2: Selected chemical and physical properties of sulfur (Stauffer Chemical Co.).

Table 3.3: Properties of common sulfur allotropes (Donahue and Meyer, 1965; Dale and Ludwig, 1965).

Property	S _α	S _β	
Common name	Orthorhombic sulfur	Monoclinic sulfur	
Recommended name	Orthorhombic (α) sulfur	Monoclinic (β) sulfur	
Molecular formula	S ₁₂₈	S ₄₈	
Crystalline form	Orthorhombic	Monoclinic	
Unit cell	16 molecules of $S_{\lambda}(S_8)$	6 molecules of $S_{\lambda}(S_8)$	
Stability region	< 95.5°C	95.5°C to 119°C	
Color	Opaque yellow at 24°C	Between yellow and orange	
Density, kg/m ³	2070	1960	
Shore B-2 hardness	90	1.96	
Tensile strength, kPa	330	410	

3.1. The minimum sulfur viscosity occurs at 159°C; higher temperatures result in the formation of polymeric sulfur which is very viscous as shown in Figure 3.1. The explosive limits and auto ignition temperatures were considered in designing the sulfur spray box. The sulfur allotropes listed in Table 3.3 were important in defining particle properties in the simulation of the coating process, and assessing the quality of sulfur coated urea.

3.1.3. Particles Used in Hydrodynamics Study

Considerable attrition was encountered with urea and sulfur coated urea (SCU) particles and, as a result, they were restricted to determining the minimum spouting velocity in the



Figure 3.1: Viscosity of sulfur at low temperature range (Freeport Sulfur Co., 1954).

Material	<i>d_n</i> (mm)	ρ_p^* (kg/m ³)	\mathcal{E}_{mf}	<i>\$</i>
Urea	2.16	1335	0.42	1
SCU, 19 % Sulfur	2.27	1427	0.42	1
SCU, 28 % Sulfur	2.33	1471	0.42	1
SCU, 31 % Sulfur	2.36	1490	0.42	1
Polyethylene	2.80	927	0.40	1
Polystyrene	2.30	1045	0.44	0.85
Polyformaldehyde	2.70	1385	0.43	1

Table 3.4: Physical properties of bed particles.

* The density was measured in the standard way by using a pycnometer.

0.24 m bed. Other materials were therefore used as well including polystyrene, polyethylene, and polyformaldehyde particles (see Plates 3.4 - 3.6) with similar size and density as urea. Attrition was also encountered with polystyrene, and its study was limited to determining the hydrodynamics in the 0.24 m bed. The physical properties of these particles are given in Table 3.4.

3.2. Main Coating Apparatus

As shown by Figure 3.2, the main components of the experimental apparatus were the spouted bed, sulfur supply system, nozzle assembly, urea feeding and product withdrawal devices, and the steam, air and water supply systems. The coating facility was rebuilt by Meisen et al. (unpublished report, 1986) incorporating some of the equipment used earlier by Tsai. The major changes included larger beds (0.24 and 0.45 m diameters), different nozzle assembly, larger sulfur melting pot, and urea feeding and product withdrawal devices.



Figure 3.2: Simplified flowsheet of UBC spouted bed facility.

3.2.1. Spouted Bed

The coating operation took place in a spouted bed which consisted of a Plexiglas column, a stainless steel cone with a shutter assembly, and a stainless steel cap. Columns 0.24 and 0.45 m in diameter were used in this study. The dimensions of the 0.24 m column and the cone extension for the 0.45 m column are given in Figure 3.3.

The 0.24 m column had four 25.4 mm dia. product discharge holes drilled at heights 0.28, 0.36, 0.44 and 0.56 m from the base of the cone, while the larger column had only one discharge hole (32 mm dia.) at the top of the cone (0.36 m from the base of the cone). Urea



Figure 3.3: Sectional view of spouted bed column

was fed through a Plexiglas tube opposite the product discharge locations and just above the bed. The Plexiglas column could operate continuously at temperatures up to 110°C.

The shutter mechanism controlled the size of the orifice opening at the base of the bed. As shown in Figure 3.4, the shutter consisted of five S-shaped, overlapping stainless steel leaves arranged in a circle. The range of opening was 3.2 to 38 mm dia. Since the shutter



Figure 3.4: Shutter assembly (dimensions are given in mm) designed by Mathur, Meisen and Lim (1978).

shape changed from a circular to an irregular shape as the shutter was closed, calculating its open area required a special procedure which is given in Appendix I.

3.2.2. Sulfur Supply System

Only minor modifications were made to the system designed by Meisen et al. (1986). The sulfur and atomizing air lines to the upper plate had to be reinforced to allow a "slip-on" type of sulfur line connector (see Section 3.2.2.4). This type of connector was necessary for a completely steam traced sulfur line. Major components of the sulfur supply system were the sulfur melter, filter, rotameter, flow control valve, sulfur line and nitrogen supply.

3.2.2.1. Sulfur Melter

The sulfur melter used in this study was originally designed as the sulfur reservoir connected to a steam jacketed sulfur melter. Unfortunately, the steam supply was insufficient to maintain the reservoir and the melter at the desired temperature. In this work, only the sulfur reservoir was used because of its large capacity (18.9 L). A schematic diagram of the reservoir is shown in Figure 3.5.

The reservoir was a modified Bink 83-5404 pressure tank (0.3 m dia. OD and 0.48 m high), insulated with fiberglass. Holes were drilled through the cap to facilitate sulfur feeding, molten sulfur withdrawl, and pressurizing. The solid sulfur fed into the reservoir was melted by contact with a stainles steel steam coil (9.5 mm dia. tube wound to 0.178 m dia. nine times). A full sulfur charge of 30 kg melted in approximately six hours. A pressure relief valve was also added on top of the reservoir to prevent excessive pressure build-up.



Figure 3.5: Sectional view of 18.9 L sulfur reservoir.

3.2.2.2. Sulfur Filter

The stainless steel 316 in-line cartridge filter (Rigimesh, manufactured by Pall Canada Ltd.) was located at the mouth of the molten sulfur outlet. The screen size of the filter was 149 μ m (screen size #100).

3.2.2.3. Sulfur Rotameter

A standard rotameter tube (Brooks, Model R-6M-25-A) inside a steam heated brass block was used as the sulfur rotameter (designed by Weiss and Meisen, 1983). Two stainless steel pieces on the top and bottom of the brass casing were used to hold the rotameter tube in place and Viton O-rings were used to seal the ends. Two polycarbonate windows with heat resistant gaskets were placed in front and back of the brass block to allow a clear view of the rotameter tube. A set of glass and stainless steel floats was used. The specifications of all rotameters are given in Table 3.5, and their calibration curves are presented in Appendix IV.

3.2.2.4. Sulfur Line

The main sulfur line was a 6.4 mm dia. 316 SS tube enclosed by 13 mm dia. O.D. TFE tubing overbraided with 304 SS and insulated with fiberglass. Complete steam tracing was not possible at the points where the line joined the melter, rotameter and base of the bed and frequent plugging was observed at the connection to the base of the bed. A fully steam traced connection required a "slip-on" connector as shown in Figure 3.6. This connector was enclosed in a 19 mm O.D. SS tube for the passage of steam. No sulfur plug-

Stream	Maximum Capacity	Rotameter
Atomizing air	0.44 L/s	Tube: R-7M-25-1 Float: Glass
Spouting air	23.9 L/s	Tube: R-12M-25-4 Float: 12-RS-221
	42.3 L/s	Tube: R-12M-127-3 Float: 12-RS-221
Sulfur	1.7 g/s	Tube: R-6M-25-1 Float: glass and stainless steel

Table 3.5: Types and capacities of Brooks rotameters used in this work.



Figure 3.6: Sectional view of "slip-on" sulfur line connector.

ging of the sulfur lines was experienced with this modification.

3.2.2.5. Nitrogen Supply

Industrial grade pressurized nitrogen (typically up to 20 psi) was used to force sulfur out of the melter. The sulfur flow rate was controlled by the N_2 pressure using the regulator on the gas cylinder. The rotameter valve was also used to control the flow rate of the sulfur.

3.2.3. Nozzle Assembly

The nozzle assembly consisted of a perforated plate, "steam chamber", and spray nozzle as shown in Figure 3.7. The perforated plate served as a flow straightener and air distributor for the spouting air. The steam chamber kept the sulfur and atomizing air lines at a constant temperature. The atomizing air and sulfur lines were connected to the upper flange and sealed with Viton O-rings.

An "internal-mixing" type pneumatic nozzle was used for spraying sulfur (see Figure 3.7). It consisted of fluid cap # 40100, air cap # 1401110 and retainer ring manufactured by Spraying Systems Co. Molten sulfur flowed through the fluid cap that narrowed into a fine tip, before entering the air cap with the atomizing air. Atomizing air entered through three equally spaced holes into the gap between the air cap and the fluid cap. The air and sulfur streams converged just above the nozzle tip thus forming sulfur droplets.

3.2.4. Urea Feeding Device

Urea pellets were stored in a 170 L steel drum with a conical base. The drum was placed 0.75 m above the spouted bed. From the storage bin, urea pellets fell into a vibrating magnetic feeder (Model F-T01A, manufactured by FMC Corp.) mounted directly under



Figure 3.7: Sectional view of nozzle, perforated plate and steam chamber (all dimensions in mm; designed by Meisen et al., 1986).

neath the bin. The urea feed rate was controlled with an electric controller (Model CSCR 1B, FMC Corp.). The urea was introduced into the bed through a 25 mm ID Plexiglas tube just above the annulus near the wall and opposite the product withdrawal port. Urea could not be fed below the surface of the annulus due to slow moving bed particles near the wall.

3.2.5. Product Withdrawal Device

The product discharged through a column slot and a 25.4 mm ID Plexiglas tube. The tube was connected to a PVC flexible plastic hose which directed the SCU into the product storage bin. The discharge mechanism depended on the gravity and air flow into the storage bin from the bed resulting from the pressure difference between the two.

3.2.6. Product Collector

The product leaving the spouted bed was collected in a large wooden box $(1.21 \times 0.85 \times 0.64 \text{ m high})$. The box was air sealed with silicone and gaskets to contain any dust. Three holes were drilled through the top board: a 19 mm dia. hole for the incoming product, a 54 mm dia. hole for air exhaust, and a 0.3 m dia. hole for cleaning the box. The 0.3 m hole was covered with a 13 mm thick Plexiglas lid held in place by attaching the cover to a 0.05 x 0.33 m Plexiglas board inside the box. A 25 mm dia. hole was also drilled through the bottom board to empty the box.

3.2.7. Dust Collector

Urea and sulfur fines elutriated from the top of the spouted bed passed through a flexible exhaust hose into a water scrubber (see Figure 3.2). The treated air was then vented directly into the laboratory exhaust system. A nylon mosquito mesh (approximately 1 mm mesh size) was placed on top of the bed in the exhaust air line to prevent the bed particles from leaving the bed.

3.2.8. Air, Steam, and Water Supplies

All air lines were connected to the laboratory compressed air supply (maximum pressure of 510 kPa). Instrument air (maximum pressure of 650 kPa) was used for sulfur atomization to provide cleaner air. Rotameters were used for all flow measurements. The type and capacities of the rotameters are given in Table 3.5.

The spouting air was heated by a steam heater and a 3 kW electric heater. The atomizing air was heated by a flexible electric heating tape (Type silicone rubber, 312 Watts; manufactured by Thermolyne Corp.) wrapped around the air line. The electric heater was controlled by a proportional-integral controller supplied by Omega Engineering Inc. (Model No. 49J, range of 0 to 200°C). The temperatures were monitored with iron-constantan thermocouples.

Steam was generated by a 30 kW, three phase, "Electro-Steam" boiler (Type F-10; manufactured by Fulton Ltd.) capable of steam flows up to 48 kg/h at 720 kPa. The operating pressure was set at 650 kPa for all runs in this study. The pressure downstream from the boiler typically fell into the range of 580 to 620 kPa. All steam traps discharged into a common atmospheric header that drained into the main sewer system.

Cold tap water was used in the water scrubber.

3.3. Apparatus for Hydrodynamics Study

The modifications to the spouted beds for the hydrodynamics study included replacing the stainless steel caps with 1/2" (13 mm) plywood caps. The new caps were slotted to allow a sampling tube to enter from the top of the bed, and the tube could be positioned at any radial position. The modified cap is shown in Figure 3.8.



Figure 3.8: Modifications to the top of the spouted bed (i.e., see Figure 3.3) for hydrodynamics study.

A static pressure probe (see Figure 3.9) and an S-type pitot tube (see Figure 3.10) were used to measure the static pressure in the annulus and the fluid velocity in the spout, respectively. The tubes were secured to the top of the bed by a 64×230 mm Plexiglas sampling tube support and Swagelok fittings.

3.4. Apparatus for Spray Study

Sulfur spray droplets produced by the atomizing nozzle described in Section 3.2.3 were captured in an air-sealed spray box. The box was designed to be placed immediately above the nozzle assembly (see Section 3.2.3). The nozzle assembly was not modified for this study. The two main components of the spray box are the box and the spray sampler.



Figure 3.9: Schematic diagram of static pressure probe.



To water manometer

Figure 3.10: Schematic diagram of S-type pitot tube (1/8" tubes and 3/8" tube are held in place with silver solder).

3.4.1. Spray Box

The design of the 0.61 x 0.61 x 1.22 m spray box used to sample sulfur droplets is shown in Figure 3.11. The box was constructed of a rigid 1/2" (13 mm) plywood base, back and side walls with 6.4 mm Plexiglas viewing panels and a 3.2 mm aluminum cap. Side walls were slotted (57 mm) for the insertion of the sampling tube 0.15 to 0.3 m from the base of



Figure 3.11: Spray box assembly.

the nozzle, and the slots were sealed once the sampling tube was placed inside the box. A light source (100 W lamp) was placed behind the box to improve the visibility of the spray. An aluminum plate was placed over the base of the bed to force auxiliary air (normally used as the spouting air in spouted bed operations) along the walls to reduce spray depositions on the walls and to improve the spray's visibility. The spent air was cleaned using an air filter (1 μ m pore size; type: Blue #100 SYN; manufactured by B.C. Air Filter Ltd.) before being vented into the laboratory exhaust system.

3.4.2. Spray Sampler

The spray sampler consisted of an outer aluminum shell and a glass slide support made of a 51 mm O.D. aluminum rod as shown in Figure 3.12. The rod was machined to hold

1





229 x 22.2 mm Slot with 1.6mm grooves

Figure 3.12: Simplified drawing of the rotating sulfur droplet sampler.

three sample cells (25.4 x 76.2 mm glass slides). The sampling unit could be withdrawn through the side of the box to replace the glass slides. The outer shell was secured by rings fitted to the sampler tube supports, and could be rotated using a rotating handle outside the box.

3.5. Coating Procedures

The operating procedures for the production of sulfur coated urea in a spouted bed consists of three steps: "start up", "coating", and "shut down".

3.5.1. Start Up

Due to the lengthy melting time of sulfur, the sulfur was typically melted overnight. This step required the following procedure: the boiler (steam generator) was turned on with all valves connected to the coating facility closed, the melter was filled with solid sulfur, the water flow to the scrubber was turned on (the water from the scrubber served to reduce loud crackling noises created by the water traps), and the main power supply and the temperature readout were switched on to monitor the temperatures at various points of the equipment. Once the pressure in the boiler reached 80 psig (650 kPa), the steam was introduced to the melter by opening the appropriate valves.

In the meantime, the urea hopper was filled and the spray nozzle was placed in position. The nozzle was checked to ensure there were no plugs.

3.5.2. Coating

The coating operation required the following steps: (i) the spouted bed was placed in the "coating" position with the shutter open; (ii) atomizing air, spouting air, and their respective heaters were turned on; (iii) the urea feeder was turned on after the air temperatures reached 80 and 160 °C for the spouting and atomizing air, respectively; (iv) the bed was left spouting with the withdrawal outlet open until steady state temperatures were observed; (v) the nitrogen supply (set at approximately 5 psig) was opened to the sulfur melter, and the valve attached to the rotameter was opened (an electric heating gun was frequently used to unblock the valve); (vi) the nitrogen pressure regulator and the rotameter valve were adjusted to set the sulfur flow rate to the desired values; (vii) steady state (indicated by a constant bed temperature and fountain height) was usually reached after ten minutes of coating for a given set of operating conditions, and product samples were collected; (viii) when the experiment was complete, the nitrogen supply valve and the urea feeder were turned off; (ix) the pressure was relieved from the melter by opening the pressure relief valve and the sulfur flow valve was shut off after the sulfur had flowed back into the melter; (x) the air heaters were turned off, the atomizing air was reduced to minimum, and the shutter and the spouting air valve were closed simultaneously, (xi) the spouted bed was detached from its base, and the bed particles were unloaded.

The operating bed temperature (T_b) was taken as the steady state bed temperature after the sulfur was injected into the bed. This temperature was typically 5 to 10°C higher than the temperature before the sulfur injection. To take a sample of the product, the flexible hose in the product withdrawal line was disconnected from the product storage bin and attached to a small container. A sudden surge of particles resulted from the pressure difference between the bed and atmosphere, and approximately a minute was allowed for the bed to return to steady state conditions before the products were sampled. The sampling time was also recorded to determine the production rate.

All samples at different operating conditions were collected by repeating the above procedure for consistency, i.e. the bed was emptied before starting a new run.

3.5.3. Shut Down and Clean-up

The procedure for shutting down was as follows: the spray nozzle was removed, inspected for plugging, and soaked in 50 % NaOH solution, all sources of steam, power, water and air were turned off, and the column was cleaned.

3.6. Procedures for Hydrodynamics Study

3.6.1. Minimum Spouting Velocity, U_{me}

The minimum spouting velocity was found by spouting the particles and then gradually reducing the air flow rate until spouting ceased. U_{ms} was calculated from the total volumetric air flow rate, Q_{ms} , just before cessation of spouting, i.e.,

$$U_{\rm ms} = 4Q_{\rm ms} / \pi D^2 \tag{3.1}$$

where D denotes the diameter of the cylindrical portion of the bed. It should be noted that the total air flow rate (Q_t) is the sum of the spouting (Q_s) and atomizing (Q_a) air flow rates. The errors in the measurements, calculated from the rotameter scale readings and from repeat measurements, were typically less than ± 5 %. •

3.6.2. Voidage of Loosely Packed Bed, ε_{mf}

The voidage (ε_{mf}) was determined by quickly inverting and re-inverting a glass cylinder (0.1 m dia. by 0.6 m high) filled approximately one-third with particles. The voidage was calculated from the following equation:

$$\varepsilon_{mf} = \frac{V_b - m_p / \rho_p}{V_b} \tag{3.2}$$

3.6.3. Mean Particle Diameter, d_p , and Sphericity, ϕ_s

The mean diameter was found by sieving the particles using #5 to #14 Tyler mesh screens and the equation

$$d_{p} = 1 / \sum_{i} x_{i} / d_{pi}$$
(3.3)

where x_i denotes the mass fraction of particles with diameter d_{pi} .

With the exception of polystyrene, all particles were nearly spherical (see Plates 3.1 - 3.5). The sphericity of the elliptical-cylindrical polystyrene particles (see Plate 3.6) was determined by measuring the principal dimensions (~ 2.2 x 3.1 x 3.3 mm) of a representative sample containing over 80 particles. Other particles were assumed to be perfect spheres, i.e., $\phi_s = 1$.

3.6.4. Diameter of Inlet Orifice, d_i

The orifice is partly obstructed by the presence of the atomizing air nozzle (see Figure 3.13). The effective orifice diameter (d_i) is therefore calculated from

$$d_i^2 = d_o^2 - d_a^2 \tag{3.4}$$

and
$$d_o^2 = 4A_o/\pi$$
 (3.5)

where A_o , d_o , and d_a denote the area of the orifice opening, the area equivalent diameter of the orifice opening and the diameter of atomizing air nozzle, respectively.



Figure 3.13: Sectional view of atomizing nozzle and inlet air assembly (not drawn to scale).

3.6.5. Static Pressure in Annulus

The static pressure was determined by using the probe shown in Figure 3.9. The probe was inserted into the top of the bed and moved horizontally and vertically to the desired location. The pressure readings were taken with a water manometer, one leg of which was open to the atmosphere.

3.6.6. Air Velocity in the Spout, u,

The air velocity in the spout was determined by using the S-type Pitot tube shown in Figure 3.10 and connected to a water manometer. The tube was inserted into the top of the bed and lowered to the desired position. It was then moved radially until the maximum pressure difference was noted. This corresponded to the axis of the spout and was used to calculate u_s . (Therefore, the measured u_s corresponds to the maximum velocity in the spout.)

The tube was calibrated against an ASME (1971) standard static pitot tube to 40 m/s in a wind tunnel (0.30 x 0.41 m by 3.7 m long). The standard pitot tube coefficient (C_o) was then determined from the following equation:

$$u = C_o \sqrt{2\Delta P / \rho} \tag{3.6}$$

The calibration curve is shown in Appendix IV.

3.6.7. Radial Velocity Profile at the Base of the Bed

In the absence of the bed particles, the radial air velocity distribution 10 mm above the shutter was measured with a static pitot tube connected to a water manometer. The tube consisted of a 1/16" (1.6 mm) stainless steel inner tube and a 1/8" (3.2 mm) stainless steel outer tube. The calibration curve and coefficient (C_o) for the static pitot tube are provided in Appendix IV.

3.7. Procedures for Spray Studies

3.7.1. Operating Limits of Spray Nozzle

The operating envelope of the spray nozzle used in this work was investigated by determining the upper and lower limits of the atomization velocity. The lower limit or the minimum atomization velocity (u_{ma}) was determined by gradually reducing the atomizing air flow rate from a fully atomized state until no atomization was observed. The velocity was calculated from the superficial air flow rate referenced to the nozzle orifice (d_{at}) . The upper limit or the minimum pulsating velocity (u_{mp}) was determined by gradually increasing the atomizing air flow rate until inconsistent atomization (indicated by flickering of the spray) was observed.

3.7.2. Spray Drop Size Measurements

All samples of sulfur droplets were collected 0.2 m above the nozzle. At this distance, the sprays were fully developed and the spray density was low enough for easy sampling. The sampling time and the slot size on the protective tube were also varied to obtain low spray density (effects of sample time and the slot size are discussed in Chapter 5).

The procedure for collecting the spray samples was as follows: (i) the glass slides were placed on the slide holder (see Figure 3.12); (ii) the holder was placed in the spray box; (iii) the spray was turned on at desired sulfur and atomizing air flow rates; (iv) the outer shell was rotated using the rotating handle to expose the glass slides to the spray; (v) the glass slides were removed and the procedure was repeated for a new operating condition.

The sulfur drops on the glass slides were analyzed under a microscope attached to a LI-ETZ TAS Image Analyzer available in the Department of Metals and Materials Engineering at UBC. At least ten frames of size 750 by 750 μ m were analyzed to obtain approximately 100 droplets for each run. Depending on the operating condition, most drop samples contained more than 100 drops in ten frames. Non-spherical particles were not included in the analysis.

3.8. Product Quality Analysis

3.8.1. Sulfur Content

The total sulfur deposited on the urea particles was determined by two methods. One method involved mass balance calculations based on the urea and sulfur feed rates; negligible loss of sulfur and urea fines was assumed. The other method was more direct and is referred to as the crush test. In the latter method, either the sulfur or urea could be analyzed to determine the sulfur content. The former was used in this work to obtain the mean sulfur content of the product.

The procedure for the crush test was as follows: a weighted sample (approximately 20 g) of sulfur coated urea was ground in a crucible with 10 mL of water to obtain a fine slurry (the exposed urea readily dissolved). The slurry was washed into a filter (1 μ m pores) with excess water. The filtered sulfur was left over night to dry and the dried sulfur was weighed the following day. The results could be reproduced to approximately \pm 1 % for batch products, and \pm 3 % for continuous products.

3.8.2. Particle Sulfur Content

Up to one hundred individual SCU particles, randomly selected, were analyzed to determine the sulfur content distribution for each product. The analysis of sulfur (as described in Section 3.8.1) was difficult due to the relatively small mass of sulfur on each particle. Consequently, the urea content was analyzed to determine the sulfur content of each particle. Each particle was weighed, placed in a 0.5 mL vial and crushed (using a paper clip) in 0.5 mL of distilled water. The solution was capped and left overnight to ensure the complete dissolution of urea. The refractive index of the solution was measured with an Abbe refractometer (Model JB7150, Bausch and Lomb Optical Co.). The urea concentration was determined from the refractometer calibration curve (see Appendix IV). The urea content in the solution could be calculated once its concentration was known. The sulfur content was determined by the difference between the urea and the total particle weights.

The error due to the particle weight measurement (typically 5 to 30 mg) was as large as \pm 20 % while the error due to the urea content measurement using the refractometer was even higher (up to \pm 25 %) for 5 mg particle. However, the errors for particles bigger than 5 mg were typically less than \pm 10 and 12 %, respectively.

2

3.8.3. Seven Day Dissolution Test

The standard seven day test developed by TVA was used, i.e. 50 g of sulfur coated urea and 250 mL of distilled water were placed in a capped jar for seven days at 37.8 °C in an incubator. The solution was carefully poured out and stirred. A sample of the solution was analyzed under the refractometer for the urea concentration as mentioned in the previous section (Section 3.8.2). The seven day dissolution rate (U_{Dm}) was determined as

$$U_{Dm} = \frac{\text{weight of urea dissolved}}{\text{weight of urea in sample}} \times 100\%$$
(3.7)

The results could be reproduced to approximately ± 2 % for batch products, and to less than ± 5 % for continuous products.

Chapter 4. Mathematical Models

Mathematical models describing the spouted bed coating process for the production of sulfur coated urea (SCU) are developed in this chapter. The first section deals with simple models based on assumptions used for previously published coating models (see Section 2.2). The simple models are then improved by progressively relaxing various assumptions to determine how they limit these models. Subsequent sections deal with the development of a "rigorous" model and programming strategy.

4.1. Simple Models

The objectives of developing simple models are to test some of the assumptions used in the previously mentioned models in the literature, to determine the limitations of the models, and to establish the need to develop a more comprehensive model. The simple models can provide quick estimates of the coating performances of a spouted bed, which may be useful in preliminary plant design.

4.1.1. Model I: Residence Time Model

In developing the first model, two principal assumptions are made: perfect mixing of the bed particles and uniform sulfur spray concentration throughout the bed. In addition, negligible coating (sulfur spray) is assumed lost. A schematic diagram of Model I is shown in Figure 4.1. The tracer tests conducted by previous investigators (Becker and Sallans, 1961; Kugo et al., 1965; Barton et al., 1968) on spouted beds indicated that the residence time distribution of the bed particles approximated that of a perfectly mixed vessel. The residence time distribution, $F{t}$, for bed particles leaving a perfectly mixed vessel is given by:

$$F\{t\} = 1 - e^{-t/t} \tag{4.1}$$



Figure 4.1: Model I – perfectly mixed system.

 $F{t}$ may be regarded as the probability that a particle entering the bed at time zero leaves after time $\leq t$.

In Model I, the amount of coating (or sulfur), m_s , on a particle leaving the bed is assumed to be directly proportional to its residence time in the bed, i.e.,

$$m_s \propto t$$
 (4.2)

This assumption is consistent with assuming that a uniform spray concentration exists throughout the bed. It is convenient to express the particle coat distribution in terms of the sulfur content of the particle (X_s) rather than m_s , i.e.,

$$X_s = \frac{m_s}{m_s + m_u} \tag{4.3}$$

where m_u denotes the mass of urea in a coated particle. Assuming m_u is the same for all particles, Equations (4.2) and (4.3) may be combined to give

$$t/\bar{t} = \frac{X_s}{\overline{X_s}} \frac{1-\overline{X_s}}{1-X_s}$$
(4.4)

Equation (4.1) may now be rewritten in terms of X_s as well:

$$G\{X_s\} = 1 - \exp\left(-\frac{X_s}{\overline{X_s}}\frac{1 - \overline{X_s}}{1 - X_s}\right)$$
(4.5)

where $G\{X_s\}$ denotes the probability that a particle has a sulfur content less than or equal to X_s . Only the average sulfur content of the product $(\overline{X_s})$ must be known to determine the sulfur content distribution of the product. Typical results from this equation are plotted in Figure 4.2 (n = 1).

The correlation given by Barton et al. (1968) showed that the particle residence time distribution in the spouted bed deviated from that in the perfectly mixed bed, i.e.,

$$F\{t\} = 1.026 - 0.92 \exp(-1.087(t/t - 0.1))$$
(4.6)

The results were explained by using mixed-flow models where 8-10% of the total bed volume in the spouted bed was in plug flow (Quinlan and Ratcliffe, 1970). Chatterjee (1970)



Figure 4.2: Prediction of Model I using different numbers of continuously stirred tanks in series.
interpreted the residence time distributions in terms of two continuously stirred tanks (CST). The results based on these models are also shown in Figure 4.2. The results of CST's in series were calculated from the following expression (Hill, 1977):

$$F\{t\} = 1 - e^{-nt/\bar{t}} \left[1 + \frac{nt}{\bar{t}} + \frac{1}{2!} \left(\frac{nt}{\bar{t}}\right)^2 + \dots + \frac{1}{(n-1)!} \left(\frac{nt}{\bar{t}}\right)^{n-1} \right]$$
(4.7)

Note that, to obtain Figure 4.2, t/\bar{t} in all cases was replaced with the relationship given by Equation (4.4). Batch results for a coating time \bar{t} are also given in Figure 4.2 for comparison.

Tsai's coating distribution data are shown in Figure 4.2 and are fairly well represented in the mid-range of sulfur concentrations by Equation (4.5), except that most of the experimental data are slightly under predicted. In the same range, some improvement was observed with the model given by Barton et al. (1968). Models based on several CST's deviate further from the experimental data.

The presence of significant fractions of uncoated urea particles produced in the continuous process and the particle sulfur content distribution found in the batch product (see Figure 4.3) cannot be predicted by Model I. This leads to the conclusion that the assumption of a uniform spray concentration in the bed is not valid for spouted bed coaters.

4.1.2. Model II: Simple Spray Zone Model

The aforementioned deficiencies of Model I suggest the presence of a limited spray zone which some particles do not enter before leaving the spouted bed. In order to incorporate such a spray zone, Model II was created on the premise that the bed could be divided into three regions: spout, annulus and fountain. The spray zone is located in the lower region of the spout, and the amount of coat a particle receives per pass through the spray zone is fixed, i.e., the same amount of coat is applied on all particles regardless of the positions at



Figure 4.3: Prediction of Model I (Equation (4.7), $n = \infty$) and batch product from the current work ($\overline{X}_s = 0.25$, $W_s = 4.1$ g/s, $d_i = 28.2$ mm, H = 0.25 m, D = 0.24 m, $Q_a = 0.57$ L/s and $T_b = 75$ °C).

which they enter the spray zone. It is presumed that urea is introduced directly into the annulus and that SCU is withdrawn from the fountain. Other assumptions are listed in Table 4.1. The system described by Model II is illustrated in Figure 4.4.

The mathematical derivation of the model equations is accomplished by examining the probability of particles receiving certain amounts of coating by the time they leave the bed. For example, the total probability of particles receiving no coat before exiting the bed (P_{0c}) is the sum of the probability of the particles by-passing the spray zone (x_b) combined with the probability of leaving the bed (x_e) , i.e.,

$$P_{0c} = x_{e}x_{b} + x_{e}x_{r}x_{b}^{2} + x_{e}x_{r}^{2}x_{b}^{3} + \dots + x_{e}x_{r}^{n-1}x_{b}^{n} + \dots$$

$$= \sum_{n=1}^{\infty} x_{e}x_{r}^{n-1}x_{b}^{n}$$
(4.8)

		Perfect mix	ing	Spray zone					
	Spout	Annulus	Fountain	Location	Sulfur drop- let concen- tration				
Model I	Yes	Yes	Yes	All	Uniform				
Model II	No	No	Yes	Lower spout	Fixed				
Model III	No	No	Yes	Lower spout	Variable				

Table 4.1: Assumptions used in simple models.



b). Simplified Spouted Bed

Figure 4.4: System described by Model II.

where $x_r = 1 - x_e$ denotes the probability of a particle not exiting from the bed when it leaves the fountain. The terms on the right hand side of Equation (4.8) represent the probabilities of particles not receiving any coat after one to an infinite number of cycles. Inspection of the infinite series formulas given in Table 4.2 reveals that Equation (4.8) reduces to:

$$P_{0c} = x_e x_b / (1 - x_r x_b)$$

Similarly, the probability of particles receiving one or more coats (P_{kc}) is given by

$$P_{lc} = {\binom{1}{1}} x_e x_c + {\binom{2}{1}} x_e x_r x_c x_b + {\binom{3}{1}} x_e x_r^2 x_c x_b^2 + \dots + {\binom{n}{1}} x_e x_r^{n-1} x_c x_b^{n-1} + \dots$$

Table 4.2: Selected infinite series definitions.

$$f\{x\} = \frac{1}{1-x} = \sum_{n=0}^{\infty} x^n = 1 + x + x^2 + x^3 + \dots$$

$$f'\{x\} = \frac{1}{(1-x)^2} = \sum_{n=1}^{\infty} nx^{n-1} = 1 + 2x + 3x^2 + 4x^3 + \dots$$

$$f''\{x\} = \frac{2}{(1-x)^3} = \sum_{n=2}^{\infty} n(n-1)x^{n-2} = 2 + 6x + 12x^2 + 20x^3 + \dots$$

$$f'''\{x\} = \frac{6}{(1-x)^4} = \sum_{n=3}^{\infty} n(n-1)(n-2)x^{n-3} = 6 + 24x + 60x^2 + \dots$$

$$\vdots$$

$$f^{*}\{x\} = \frac{k!}{(1-x)^{k+1}} = \sum_{n=k}^{\infty} \frac{n!}{(n-k)!} x^{n-k}$$

$$= \sum_{n=1}^{\infty} nx_{e} x_{r}^{n-1} x_{c} x_{b}^{n-1}$$

$$= x_{e} x_{c} / (1 - x_{r} x_{b})^{2}$$

$$P_{2c} = {\binom{2}{2}} x_{e} x_{r} x_{c}^{2} + {\binom{3}{2}} x_{e} x_{r}^{2} x_{c}^{2} x_{b} + {\binom{4}{2}} x_{e} x_{r}^{3} x_{c}^{2} x_{b}^{2} + \dots + {\binom{n}{2}} x_{e} x_{r}^{n-1} x_{c} x_{b}^{n-1} + \dots$$

$$= \sum_{n=2}^{\infty} {\binom{n}{2}} x_{e} x_{r}^{n-1} x_{c}^{2} x_{b}^{n-2}$$

$$= x_{e} x_{c}^{2} x_{r} / (1 - x_{r} x_{b})^{3}$$

$$\vdots$$

$$P_{kc} = x_{e} x_{c}^{k} x_{r}^{k-1} / (1 - x_{r} x_{b})^{k+1}$$
(4.9)

where $x_c = 1 - x_b$ denotes the probability of a particle going into the spray zone.

The amount of coat (m_{sl}) each particle receives per pass through the spray zone can be related to the average coat on the particles $(\overline{m_s})$,

$$\overline{m_s} = \sum_{k=0}^{\infty} k \cdot m_{sl} \cdot P_{kc} \bigg/ \sum_{k=0}^{\infty} P_{kc} = \sum_{k=0}^{\infty} k \cdot m_{sl} \cdot P_{kc}$$
(4.10)

since the denominator in the first equality of Equation (4.10) is equal to one. Substituting P_{kc} from Equation (4.9) into Equation (4.10) and applying infinite series definitions, $\overline{m_{k}}$ reduces to

$$m_s = m_{s1} \cdot x_c / x_e \tag{4.11}$$

Assuming all urea particles have the same mass, the sulfur content distribution can be calculated from Equation (4.9). Furthermore, it follows from Equations (4.3) and (4.11) that

$$X_{s}\{k\} = \frac{k \cdot m_{s1}}{k \cdot m_{s1} + m_{u}}, \text{ for } k = 0, 1, 2, ..., \infty$$
$$= \frac{k \cdot x_{e} \cdot \overline{X_{s}}}{k \cdot x_{e} \cdot \overline{X_{s}} + (1 - \overline{X_{s}})x_{e}}, \text{ for } k = 0, 1, 2, ..., \infty$$
(4.12)

The variables x_e , x_c and $\overline{X_s}$ must be specified or known in order to use this model.

The average sulfur content $(\overline{X_s})$ can be determined experimentally by a crush test of the SCU or it may be calculated from the sulfur and urea flow rates. The probability of particles exiting from the fountain, x_e , may be determined if the circulation rate (W_c) is known;

$$x_e = W_p / W_c \tag{4.13}$$

The production rate of SCU (W_p) can be calculated from the urea feed rate (W_u) ,

$$W_{p} = W_{\mu} / (1 - \overline{X_{s}}) \tag{4.14}$$

assuming negligible loss of sulfur and urea fines.

The probability of the particles entering the spray zone (x_c) is more difficult to determine. It may be estimated by using the average mass of coating material deposited on the particles per pass; however, such an estimate requires information about the deposition rate of the coat onto the bed particles and the density of particles in the spray zone. This information is not readily available. One method of determining this information is to correlate x_c and x_e to the operating variables. The value of x_c may be determined by optimizing x_c to fit experimental coat distribution data.

This model can also be modified for operations where the urea feed is forced into the spray zone by moving the feed location near the wall of the column. (Tsai (1986) claimed that such a change in feed location improved SCU quality.) The assumptions used in this model are that all urea feed particles pass through the spray zone, and that the volume of the particles does not change significantly as a result of coating. (See Appendix I for the details of the derivation.) The resulting model equations are

$$P_{kc} = \frac{x_{s} x_{c}^{k-1} x_{r}^{k-1}}{(1 - x_{b} x_{r})^{k}}, \text{ for } k = 1, 2, \dots, \infty$$
(4.15)

and
$$\overline{X_s}\{k\} = \frac{k \cdot x_s \overline{X_s}}{k \cdot x_r \overline{X_s} + (x_b x_s + x_c)(1 - \overline{X_s})}$$
 for $k = 1, 2, ..., \infty$ (4.16)

The assumption that all particles receive equal amounts of coat as they pass through the spray zone is questionable since the spray concentration decreases with height. Particles which enter the spray zone further away from the nozzle will be exposed to a lower sulfur droplet concentration and spend less time in the spray zone. Such particles receive less coating material than the particles that enter the spray zone closer to the nozzle. In the following section, a model which addresses such a spray zone is developed.

4.1.3. Model III: Variable Concentration Spray Zone Model

The purpose of developing Model III is to see the effects of varying the sulfur concentration in the spray zone on sulfur content distribution. The sulfur droplet concentration (C_c) in the spray zone is assumed to vary with the distance from the nozzle, i.e.,

$$C_c \propto (z_c - z)^a \tag{4.17}$$

where z_c = height of the spray zone, z = distance from the nozzle, and a = index. The amount of coat a particle receives is assumed to be directly proportional to the sulfur droplet concentration in the spout, i.e.,

$$m_{\rm sl} \propto C_c \tag{4.18}$$

Therefore, it follows that

$$m_{s1} = k_c (z_c - z)^a \tag{4.19}$$

The probability of a particle entering at any location in the spray zone is the same; therefore, the probability density $(\Phi\{z\})$ may be expressed as

$$\Phi\{z\} = k_p \tag{4.20}$$

Since $\int_0^{z_c} \Phi\{z\} dz = x_c$, then $k_p = x_c/z_c$. The average amount of coat a particle receives $(\overline{m_s})$ is given by

$$\overline{m_{s1}} = \int_0^{z_c} m_{s1} \Phi\{z\} dz / \int_0^{z_c} \Phi\{z\} dz$$
(4.21)

which, after solving for k_c and substituting into Equation (4.19), gives

$$m_{s1} = (a+1)m_{s1}(z_c - z)^a / z_c^a$$
(4.22)

All other assumptions are the same as for Model II.

Due to the complexity of deriving an analytical expression for Model III, the Monte Carlo method was applied to find the coating distribution. See Section 4.2.3 for more details.

4.2. Model IV: Rigorous Model

The ultimate goal of the present study is to model the sulfur coating of urea in a spouted bed by developing sub-models for particle circulation and coating mechanism, and then, based on the histories of single particles, determine the coating distribution. This model should require only the operating and design specifications as inputs and should not depend on other information to determine the coating distribution. The approach taken to develop the mathematical model for predicting the concentrations of the sulfur droplets in the gas phase (i.e. spray zone) in the spouted bed is similar to that taken by Meisen and Mathur (1974) for a spouted bed aerosol collector. The bed is divided into three regions as shown in Figure 1.1. Each region is considered separate and distinct from each other although interactions exist along the boundaries. The hydrodynamics and coating in the fountain are expected to be unimportant and, therefore, they are not considered further in this work.

Several key assumptions are required for Model IV. The particles in the fountain are assumed to be perfectly mixed, while those in the spout and annulus are assumed to be in plug flow. In addition, particle segregation due to changing size and density is assumed to be absent. The change in sulfur properties resulting from solidification and allotropic transformations is neglected as far as their effect on bed hydrodynamics, bed temperature and particle circulation is concerned. Consequently the effects such as the influence of bed temperature on the quality of coating (Tsai, 1986; Weiss, 1981) cannot be determined from Model IV.

The Model IV equations and methods used to calculate the bed hydrodynamics, sulfur droplet concentration and coating distribution are outlined in the following sections. The computer program listings for Model IV are given in Appendix III.

4.2.1. Calculation of Solids Circulation Rate and Bed Hydrodynamics

The hydrodynamics model used in this work is based on the one-dimensional gas and solids mass and momentum balances for the spout developed by Lefroy and Davidson (1969) and the vector form of the Ergun (1952) equation for gas flow in the annulus. The spout diameter is assumed constant and estimated using Equation (2.4). The pressure distribution at the spout-annulus interface (equal to the radially averaged pressure in the spout) is determined by Equation (2.11) and

$$P_a\{z\} = \Delta P_{pss}\{H\} - \Delta P_{pss}\{z\} \tag{4.23}$$

Equation (4.23) is based on the findings of Grbavcic et al. (1976) that the axial pressure gradient at any elevation above the spout inlet is independent of the height of the bed. By specifying the pressure distribution at the interface, the annulus gas flow problem can be uncoupled from the hydrodynamics in the spout.

The vector Ergun equation, as suggested by Stanek and Szekely (1974) and modified for the annulus by Rovero et al. (1983), is given by

$$-\nabla P_a = \mathbf{U}(f_1 + f_2 |\mathbf{U}|) \tag{4.24}$$

where
$$f_1 = 150 \ \mu (1 - \varepsilon_a)^2 / d_p^2 \varepsilon_a^3$$
 (4.25)

and
$$f_2 = 1.75 \ \rho(1 - \varepsilon_a) / d_p \varepsilon_a^3$$
 (4.26)

The equation is solved subject to atmospheric pressure at the top of the annulus, the extended Morgan-Littman distribution at the spout-annulus interface and zero normal pressure gradient at the cylindrical and conical walls of the column. Because the method for solving Equation (4.24) is lengthy and is very similar to the method outlined by Rovero et al., it is given in Appendix I. It should be noted, however, that second order finite difference equations are used to solve for the fluid stream functions (see Appendix I), from which the gas velocity components and pressure can be calculated. Additional assumptions are that $\varepsilon_a = \varepsilon_{mf}$ and that $|v_a| \ll |u_a|$. The solution of the equation governing fluid in the annulus also provides the leakage flow (U_r) of gas from the spout. The latter is required by the gas mass balance in the spout.

The mass and momentum balances of Lefroy and Davidson (1969) for gas and solids motion in the spout are given by

$$A_s \frac{d(\varepsilon_s u_s)}{dz} + \pi D_s U_r = 0$$
(4.27)

$$A_s \frac{d((1-\varepsilon_s)v_s)}{dz} + \pi D_s V_r = 0$$
(4.28)

$$\rho A_s \frac{d(\varepsilon_s u_s^2)}{dz} = -\varepsilon_s A_s \frac{dP_s}{dz} - \beta_p (u_s - v_s) |u_s - v_s| A_s$$
(4.29)

$$\rho_p A_s \frac{d((1-\varepsilon_s)v_s^2)}{dz} = -(1-\varepsilon_s)A_s \frac{dP_s}{dz} + \beta_p (u_s - v_s)|u_s - v_s|A_s -(1-\varepsilon_s)(\rho_p - \rho)A_s g$$

$$(4.30)$$

Equations (4.27) and (4.28) represent the gas and solids mass balances while Equations (4.29) and (4.30) are the gas and solids momentum balances, respectively. The terms on the right hand sides of the momentum equations account for the normal stress, drag between the two phases and, in case of solids, the gravity force. The particle-fluid interaction parameter (β_p) is approximated by the Richardson and Zaki (1954) equation as applied by Lefroy and Davidson (1969):

$$\beta_p = 0.33(1 - \varepsilon_s)\rho / d_p \varepsilon_s^{178}$$
(4.31)

The initial conditions for Equations (4.27), (4.29) and (4.30) are:

$$\varepsilon_s = 1, u_s = u_0, \text{ and } v_s = 0 \text{ at } z = 0.$$
 (4.32)

This set of equations is solved numerically using UBC ODEPACK (Moore, 1989) running under the UBC MTSG (Runnals, 1987) main frame operating system. Once ε_s , u_s and v_s have been obtained, the particle entrainment flux (V_r) at the spout-annulus interface can be calculated from Equation (4.28).

Finally, the radially averaged particle velocity in the annulus can be determined by equating the solids up-flow in the spout with the solids down-flow in the annulus at any height (z), i.e.,

$$v_a = v_s A_s (1 - \varepsilon_s) / A_a (1 - \varepsilon_a) \tag{4.33}$$

As well as radial uniformity, Equation (4.33) assumes that there is no particle segregation in the annulus and negligible mass change due to coating in the spout.

4.2.2. Determination of Coating Mechanism and Concentration Profile

In determining the coating mechanism and the concentration of sulfur droplets in the spouted bed, it is assumed that the solidification of sulfur droplets, as mentioned earlier, does not occur in the gas phase. Furthermore, it is established from the spray studies (see Section 5.2.3) that the primary mechanism for sulfur droplet deposition on the bed particles is inertial impaction. The latter phenomenon occurs when the gas carrying small sulfur droplets approaches the bed particles; the gas deflects around the particles, while the sulfur droplets, by virtue of their greater inertia, deposit on the bed particles. The validity of the assumption that inertial deposition is the dominant collection mechanism is based on the measured droplet size distribution and information about aerosol collection provided by Clift et al. (1981).

The inertial impaction coating deposition rate (N_i) on the spherical bed particles per unit volume of the bed is calculated from

$$N_i = C_s A_p \eta |u - v| \tag{4.34}$$

where the projected area of particles per unit bed volume (A_p) is

$$A_p = 1.5(1 - \varepsilon) / d_p \tag{4.35}$$

The expressions given by Behie et al. (1972), i.e., Equations (2.23) - (2.26), were used to determine the inertial impaction efficiency (η).

Two additional assumptions are used to calculate the sulfur spray concentration. First, the sulfur spray is presumed not to penetrate into the annulus (i.e., coating only occurs in the spout), since the spray occupies a very small volume of the spout (see Figure 4.5). Second, coating does not occur until the spray reaches the spout-annulus interface (at which point the spray is assumed to be fully developed). The second assumption was based on visual observations of the bed particles in a half column spouted bed and of the sulfur spray in the spray box. The concentration of the bed particles is low in the lower section



Figure 4.5: Sectional view of the lower spout.

of the spout and the particles are located primarily near the wall of the spout. The spray only occupies a small area just above the nozzle.

With these assumptions, a mass balance for coating material in the spout gives (see Figure 4.6):

$$\frac{\mathrm{d}(C_s u_s \varepsilon_s A_s)}{\mathrm{d}z} = 0, \text{ for } z < h_{fds}$$
(4.36)

$$\frac{\mathrm{d}(C_s u_s \varepsilon_s A_s)}{\mathrm{d}z} + N_i A_s = 0, \text{ for } z \ge h_{fds}$$

$$(4.37)$$

The initial condition is

$$C_s = C_{s0} \text{ at } z = 0$$
 (4.38)

Note that Equation (4.38) assumes that the sulfur droplet concentration below $z = h_{fds}$ can be smeared out radially, in order that a one-dimensional approximation may be applied. Equations (4.36) and (4.37) are solved simultaneously with Equations (4.27) to (4.30) using UBC ODEPACK. The spray angle of 20° (provided by the manufacturer of the spray nozzle for spraying water into still air) is used in the calculations to determine h_{fds} .



Figure 4.6: Mass balance on coating material in the spout.

4.2.3. Calculation of Coating Distribution Using the Monte Carlo Method

4.2.3.1. Limitation of Analytical Model

The equations governing Models I and II can be solved analytically. Such models are called deterministic models. However, Model III and Model IV are not amenable to analytical solutions. For example Models II - IV may be represented by

$$\Pr\{m_{s} \leq m_{s}^{*}\} = \sum_{i=1}^{\infty} \Pr\{N=i\} \Pr\{\sum_{j=1}^{i} m_{s1} \leq m_{s}^{*} | N=i\}$$
(4.39)

where $\Pr\{m_s \leq m_s^*\}$ is the probability that no more than m_s^* is deposited on a particle during its residence in the bed. $Pr\{N = i\}$ denotes the probability that a particle stays in the

bed for *i* cycles, and $\Pr\left\{\sum_{j=1}^{i} m_{s1} \le m_{s}^{*} | N = i\right\}$ is the conditional probability that the total

coat a particle receives is less than m_i^* given that the particle goes through *i* cycles before exiting the bed. From the analysis of the exit probabilities, it can be shown that

$$\Pr\{N=i\} = x_e x_b^{i-1} \tag{4.40}$$

Mann (1983) showed that the remainder of the term in Equation (4.39) can be reduced to

$$\Pr\left\{\sum_{j=1}^{i} m_{s1} \le m_{s}^{*} | N = i\right\} = \Pr^{*i}\left\{m_{s1} \le m_{s}^{*} | N = i\right\}$$
(4.41)

where $\Pr^{*i} \{m_{s1} \le m_s^* | N = i\}$ denotes the *i*th order convolution of $\Pr\{m_{s1} \le m_s^* | N = i\}$. Simple analytical expressions can be obtained if an exact solution of the convolution in Equation (4.41) exists and if an infinite series representation exists for Equation (4.39). If both conditions are satisfied, Equation (4.39) can be reduced to a simple analytical expression. The equation can usually be solved numerically if the conditions are not satisfied. However, the derivations of analytical expressions or numerical solutions can be difficult for complicated models such as Models III and IV, and are susceptible to errors from dealing with large arrays of coating probabilities or from numerical procedures. Moreover, both processes must be repeated if model specifications change (e.g., change in feed location for Model II). The errors associated with these methods can be avoided, and the repeat derivations are unnecessary if the Monte Carlo method is used.

4.2.3.2. Monte Carlo Procedure for Model III

The path of a particle and the sulfur deposited on the particle are simulated from given values of x_e , x_c and a, and random parameters N_{RI} and N_{R2} . As each particle enters the bed, its path through the spout is determined by N_{RI} , from which m_{sI} can be calculated

$$m_{s1} = 0, \text{ for } N_{R1} > x_c$$
 (4.42)

$$m_{s1} = (a+1)\overline{m_{s1}}(z_c - z)^a / z_c^a, \text{ for } N_{R1} \le x_c$$
(4.43)

where $z = N_{RI} \cdot H$, $0 < N_{RI} < 1$ and z_c is the distance indicating the upper limit of the spray zone. Once the particle leaves the spout and enters the fountain, its path from the fountain is determined by N_{R2} , i.e.,

if
$$N_{R2} < x_{e}$$
, the particle leaves the bed (4.44)

if
$$N_{R2} \ge x_e$$
, the particle stays in the bed (4.45)

where $0 < N_{R2} < 1$. If the particle stays in the bed, the procedure is repeated. If the particle leaves the bed, another particle is introduced until a sufficient number of particles is simulated to determine the coating distribution. The value of $\overline{m_{s1}}$ is determined from the following definition of the mean sulfur weight in the product $(\overline{m_s})$:

$$\frac{1}{m_s} \approx \frac{\sum \text{coating on the particles in units of } \overline{m_{s1}}}{\text{number of particles sampled}}$$
(4.46)

and the sulfur content is found by applying Equation (4.3). The random numbers N_{RI} and N_{R2} are generated using the UBC RANDOM (Nicol, 1986) package available in UBC MTSG (Runnals, 1987). A pseudo-random number generator was used, i.e. the same 'seed' value produces the same sequence of random number.

4.2.3.3. Monte Carlo Procedure for Model IV

The Monte Carlo method (see Section 2.5) was used to determine the distribution of sulfur on the particles leaving the spouted bed. The sulfur deposit on a particle is determined by following the path of the particle in the bed and calculating the amount of coating material the particle receives until it leaves the bed. The procedure is then repeated for another feed particle until a sufficient number of particles has been considered to give a good representation of the coating distribution. The detailed procedure for determining the coat on an individual particle during its residence in a spouted bed is given in the following subsections:

4.2.3.3.1. Continuous Operation

1. When a feed particle enters the unit or a cycling particle returns via the fountain to the top of the bed, its radial position at the top of the annulus is randomly assigned. The feed urea particle, as it enters the bed, is assumed to land within $(\Delta D_f =)$ 0.03 m of the column

wall at the surface of the annulus. This assumption is based on the observations that majority of particles fall within 0.03 m of the column. The radial position of the feed particle on the annulus surface is assigned using a random number generated by the UBC RAN-DOM (Nicol, 1986) package available in the MTSG (Runnals, 1987) main frame operating system at UBC. The generated number (N_R) falls between 0 and 1, and it is used to assign the radial position (D_r) of the particle according to

$$N_R = \frac{D_r^2 - D_f^2}{D^2 - D_f^2} \tag{4.47}$$

where
$$D_f = D - \Delta D_f$$
 (4.48)

 ΔD_f denotes twice the width (adjacent to the bed wall) of the bed surface area occupied by the feed and D denotes the column diameter. Similarly, the radial position of the returning particle (via the fountain) on the annulus is calculated from

$$N_R = \frac{D_r^2 - D_s^2}{(D_f^2 - D_s^2) + (1 - x_f)(D^2 - D_f^2)}, \text{ for } D_r \ge D_f$$
(4.49)

$$N_R = \frac{(1 - x_f)(D_r^2 - D_f^2) + (D_f^2 - D_s^2)}{(D_f^2 - D_s^2) + (1 - x_f)(D^2 - D_f^2)}, \text{ for } D_r < D_f$$
(4.50)

where
$$x_f = \frac{W_u / \rho_u}{v_a (D^2 - D_f^2)(1 - \varepsilon_a)\pi / 4|_H}$$
 (4.51)

The denominator in Equations (4.49) and (4.50) represents the total area occupied by the recycling particles, and x_f represents the fraction of area the feed particles occupy in the area defined by $\pi (D^2 - D_f^2)/4$.

2. The particle is followed around the bed, i.e. entry into the annulus, transport in the annulus, entry into the spout, transport in the spout, entry into the fountain, transport in the fountain, entry into the discharge tube or re-entry into the annulus. As the particle spends significantly more time in the annulus than the spout or fountain, the total cycle time (t_c) is estimated using the residence time of the particle in the annulus:

$$t_{c} = \int_{\hat{H}}^{z_{c}} dz \, / \, v_{a}\{z\} \tag{4.52}$$

 z_s can be determined by equating the particle flow rate at the surface of the annulus between D and D_r to that in the spout at z_s , i.e.,

$$v_a(1-\varepsilon_a)(D_c^2-D_r^2)\Big|_H = v_s(1-\varepsilon_s)D_s^2\Big|_{z_s}$$
(4.53)

where
$$v_a|_H = v_s \frac{1 - \varepsilon_s}{1 - \varepsilon_a} \frac{D_s^2}{D^2 - D_s^2}\Big|_H$$
 (4.54)

The total residence time (t_r) in the bed can be found from the sum of the cycle times

$$t_r = \sum_{i=1}^{N} t_c \tag{4.55}$$

where N denotes the total number of cycles a particular particle makes before leaving the bed. The coating amount per cycle (m_{sl}) can be calculated from

$$m_{s1} = \int_{z_s}^{H} A_p^* C_s |u_s - v_s| \eta / v_s \, dz, \text{ for } z_s > h_{fds}$$
(4.56)

$$m_{s1} = \int_{h_{fds}}^{H} A_{p}^{*} C_{s} |u_{s} - v_{s}| \eta / v_{s} \, dz, \text{ for } z_{s} \le h_{fds}$$
(4.57)

where A_p^* is the projected area of a single urea particle and h_{fds} is the height at which the spray becomes fully developed and, hence, coating occurs beyond this point (see Figure 4.6). The spray angle (ϕ) was assumed to be 20° and h_{fds} is calculated from

$$h_{fds} = D_s \tan(\phi/2)/2$$
 (4.58)

Since coating is assumed not to occur in the annulus and fountain, m_{sI} in Equations (4.56) and (4.57) represent the total amount of sulfur deposited on the particle in one cycle.

3. The position of the particle is reassigned on the annulus surface. If the particle lands in the exit stream, the simulation ends for this particular particle and begins for another. Otherwise, the next cycle begins as before. The sulfur content $(X_{sp,i})$ of the *i*th particle leaving the bed is given by

$$X_{sp,i} = \sum_{j=1}^{N} m_{s1} / (\sum_{j=1}^{N} m_{s1} + m_{\mu})$$
(4.59)

where m_u is the mass of urea in a single bed particle. Because the average sulfur content of simulated particles (\overline{X}_{sp}) does not always agree with the sulfur content calculated from the sulfur and urea feed rates (\overline{X}_s) , especially when a small number of particles are simulated, $X_{sp,i}$ is normalized as follows:

$$X_{sp,i} = X_{sp,i} \overline{X}_{sp} / \overline{X}_{s}$$
(4.60)

In total, 1000 particles were typically simulated to determine the coating distribution for each set of operating condition. The values of \overline{X}_s and \overline{X}_{sp} typically differed by 2 % before normalizing according to Equation (4.60). The effect of the sample size is discussed in Chapter 5.

4.2.3.3.2. Batch Operation

The simulation procedure for the batch coating operation is the same as that for the continuous operation except that the start and the termination of the particle simulation are different. The simulation of batch coating terminates when the particle's residence time is equal to the total coating time of the batch product. The cycle and residence times are calculated from Equations (4.52) and (4.55). The detailed derivation of the batch model is given in Appendix I.

Chapter 5. Results and Discussions

The results of the studies conducted on the bed hydrodynamics and sulfur spray are discussed in the first two sections of this chapter. The third section deals with the coating experiments including the coating distribution and product quality. The commercial implications of the present work are then discussed.

5.1. Bed Hydrodynamics

The minimum spouting velocity, pressure in the annulus, and fluid velocity in the spout were measured in the absence of sulfur injection. The validity of expressions presented in Chapter 2 were compared with the experimental data obtained from the present coating unit. Modifications were made in the expressions to improve their predictive ability where necessary and possible. The modifications resulted from the presence of the pneumatic atomizing nozzle in the spouting air inlet, the presence of atomizing air, and operating with shallow conical beds.

The ranges of variables examined in this study are summarized in Table 5.1. A total of 327 runs were performed to determine the minimum spouting velocity and the spout velocity and annulus pressure profiles were measured in 32 runs. The operating conditions of the experiments performed to determine the pressure profiles and spout velocities are given in Table 5.2. Pressure and velocity data are given in Tables 5.3 and 5.4, respectively. The minimum spouting velocity data are provided in Appendix II.

Parameter	Operating Range	Parameter	Operating Range
Н	0.11 - 0.63 m	Q _a	0 - 0.87 L/s
D	0.24 and 0.45 m	Т	18 - 70 °C
d_i	21 - 35 mm	ρ	$1 - 1.2 \text{ kg/m}^3$
Q _s	0 - 37 L/s	-	

Table 5.1: Operating ranges applicable to this work.

5.1.1. Minimum Spouting Velocity, U_{ms}

Besides providing the lower limit of operation, the minimum spouting velocity (U_{ms}) information may be used to study the effects on bed hydrodynamics of atomizing air, presence of the nozzle, and operating with shallow conical beds.

Figure 5.1 provides comparison between the measured U_{ms} and the values calculated by the standard equations due to Mathur and Gishler (1955) and Wu et al. (1987) (given by Equations (2.2) and (2.3), respectively). The values of the constants in Equation (2.3) are repeated in Table 5.5.

As shown by Figure 5.1, poor agreement was obtained between the experimental measurements and the aforementioned standard equations. The discrepancy was thought to result from determining the experimental U_{ms} values based on the total air flow rate (i.e., the sum of Q_a and Q_s) without due regard to the momentum of the main and atomizing air streams. The atomizing air, which enters at high velocity (up to 131 m/s) in the centre of the spout, should enhance the effectiveness of the main spouting air stream (typical velocities: 22 to 78 m/s) and therefore reduce the minimum spouting velocity. This is confirmed by the radial velocity profile measured at the bottom of the cone (just above the shutter) in the absence of bed particles (see Figure 5.2).

Run	Particle Type	Q_s	Q_a	H	D	d_i	T	ρ
#		L/s	L/s	m	m	m	°C	kg/m ³
H1	Polyformaldehyde	33.77	0.53	0.29	0.24	0.0308	60	1.06
H2	Polyformaldehyde	32.96	0.53	0.29	0.24	0.0308	66	1.04
H3	Polyformaldehyde	28.50	0.53	0.29	0.24	0.0308	65	1.05
H4	Polyformaldehyde	29.65	0.53	0.29	0.24	0.0308	61	1.06
H5	Polyformaldehyde	31.97	0.87	0.29	0.24	0.0308	67	1.04
H6	Polyformaldehyde	31.89	0.70	0.29	0.24	0.0308	68	1.04
H7	Polyformaldehyde	31.53	0.53	0.29	0.24	0.0308	66	1.04
H8	Polyformaldehyde	31.19	0.00	0.29	0.24	0.0308	68	1.04
H9	Polyformaldehyde	33.15	0.53	0.29	0.24	0.0351	68	1.04
H10	Polyformaldehyde	31.82	0.53	0.29	0.24	0.0282	70	1.03
H11	Polyformaldehyde	30.47	0.53	0.29	0.24	0.0247	71	1.03
H12	Polyformaldehyde	27.78	0.53	0.29	0.24	0.0212	66	1.04
H13	Polyformaldehyde	28.65	0.53	0.29	0.24	0.0308	21	1.20
H14	Polyformaldehyde	33.77	0.53	0.34	0.24	0.0308	66	1.06
H15	Polyformaldehyde	33.97	0.53	0.37	0.24	0.0308	62	1.05
H16	Polyformaldehyde	36.02	0.53	0.37	0.24	0.0308	66	1.04
H17	Polystyrene	34.17	0.53	0.53	0.24	0.0308	64	1.05
H18	Polystyrene	28.50	0.53	0.34	0.24	0.0308	65	1.05
H19	Polyethylene	31.81	0.53	0.36	0.24	0.0308	69	1.03
H20	Polyethylene	27.54	0.53	0.29	0.24	0.0308	62	1.05
H21	Polyethylene	29.48	0.53	0.30	0.45	0.0308	60	1.06
H22	Polyformaldehyde	31.17	0.53	0.25	0.45	0.0308	62	1.05
H23	Polyformaldehyde	35.39	0.53	0.31	0.45	0.0308	62	1.05
H24	Polyformaldehyde	32.05	0.00	0.29	0.45	0.0308	62	1.05
H25	Polyformaldehyde	32.91	0.87	0.29	0.45	0.0308	62	1.05
H26	Polyformaldehyde	31.17	0.53	0.29	0.45	0.0308	62	1.05
H27	Polyformaldehyde	33.97	0.53	0.29	0.45	0.0308	62	1.05
H28	Polyformaldehyde	32.57	0.53	0.29	0.45	0.0308	62	1.05
H29	Polyformaldehyde	32.57	0.53	0.29	0.45	0.0282	62	1.05
H30	Polyformaldehyde	30.46	0.53	0.29	0.45	0.0247	62	1.05
H31	Polyformaldehyde	35.39	0.53	0.29	0.45	0.0351	62	1.05
H32	Polyformaldehyde	32.08	0.53	0.29	0.45	0.0308	24	1.19

Table 5.2:Operating conditions under which the air velocities in the spout and the
pressure profiles in the annulus were determined.

- Table 5.3: Axial pressure profile near the spout-annulus interface. (R Run number as shown in Table 5.2; z height from the base of the bed, mm; pressures are expressed in mm water).
- a) Small Bed (D = 0.24 m)

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H32

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	Run\z	15	35	55	75	95	115	135	155	175	195	215	235	255	275	295	315	335	355	385	435	485	535
	H1	96	92	86	82	78	68	56	48	40	29	22	14	5	0	-	-	-	-	-	-	-	•
	H2.	86	88	83	78	76	66	57	46	36	32	22	16	8	4	-	-	-	•	-	-	-	-
	H3	-	98	92	87	79	73	58	48	39	32	26	18	9	6	-	-	-	-	-	-	-	-
	H4	97	98	91	84	78	70	61	50	42	31	20	12	6	2	-	-	-	•	-	-	-	-
	H5	94	96	92	85	80	70	62	49	40	32	25	17	8	2	-	•	-	-	-	•	-	-
	H6	91	92	90	81	76	65	56	46	36	30	21	15	7	0	-	•	•	•	-	-	•	•
	H7	89	93	87	82	75	68	60	49	38	28	25	17	6	1	-	-	-	-	-	•	-	•
	H8	97	100	94	89	82	70	65	56	47	39	28	22	13	6	•	-	-	•	-	•	•	-
	H9	91	105	94	88	83	73	62	51	41	31	23	14	7	0	-	-	-	-	-	-	-	-
	H10	-	86	81	79	75	68	61	51	43	36	28	19	10	4	-	-	-	•	-	-	· •	-
	H11	-	76	76	71	67	64	60	50	40	32	28	21	10	5	• •	-	-	-	-	-	-	-
	H12	-	67	75	71	72	63	59	51	47	40	32	22	8	0	-	-	-	•	-	-	-	-
	H13	-	82	80	75	68	63	59	49	39	29	19	10	2	0	•	-	-	-	-	-	-	-
	H14	102	114	111	108	100	92	86	72	62	54	44	34	28	20	12	1	2	-	-	• ·	•	-
	H15	-	-	-	-	107	103	90	85	81	67	62	52	-	35	-	21	1	-	-	-	-	-
	H16	63	105	104	99	96	83	73	70	•	65	-	55	-	38	-	22	•	1	•	-	•	-
	H17	154	148	149	140	134	128	128	127	-	119	-	99	-	-	86	-	78	-	52	36	21	1
	H18	67	62	59	-	55	49	47	•	37	-	27	-	20	-	7	-	0	•	•	•	•	-
	H19	99	95	87	83	82	74	70	64	-	49	-	35	-	25	•	19	•	2	•	•	•	-
	H20	76	73	66	60	59	52	46	35	26	25	•	12	-	2	-	-	-	-	-	-	-	-
b) La	rge Bed ((D =	0.45	m)																			
	Run\z	20	4	10	60	80	1	00	120	14	0	160	180	20	00	220	240	26	50	270	280) 3	00
	H21	60	e	50	56	53		48	44			34	-	2	3	•	11		•	-	0		•
	H22	74	-	72	67	62		55	49	-		34	-	1	8	-	1		-	-	-		-
	H23	91	9)6	86	80		73	-	6	3	-	49		•	33	-	2	5	-	-		0
	H24	80	8	30	76	70		64	-	5	1	-	35		-	17	-		-	0	-		-
	H25	80	-	17	74	69		62	-	5	0	-	34		-	19	•		-	0	-		-
	H26	81	8	81	76	70		64	-	5	1	-	35		•	18	-		-	0	-		-
	H27	80	8	30	76	72		64	-	5	1	-	35		-	18	-		-	0	-		-
	H28	81	8	35	76	74		63	-	5	3	-	36		•	22	-		-	2	-		-
	H29	76	م ا	73	70	65		63	-	4	9	-	36		•	19	-		-	0	-		-
	H30	49	(50	60	58		56	-	4	7	-	34		-	18	-		-	0	-		•
	H31	97	9	92	84	80)	74	-	5	6	-	40		-	22	-		-	1	•		-

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- Table 5.4: Air velocity profile in the spout. (R Run number as shown in Table 5.2; z height from the base of the bed, mm; velocities are expressed in m/s).
- a) Small Bed (D = 0.24 m)

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Run\z	105	125	145	165	185	205	225	245	265	285	305	325	335	355	405	455	505
H1	37.6	30.5	27.5	24.6	21.9	19.4	18.1	15.9	-	•	-	•	-	•	-	-	•
H2	37.6	33.2	26.8	23.2	21.2	19.0	17.5	15.2	-	-	-	-	-	-	-	-	-
H3	34.7	28.2	22.6	20.2	17.5	16.8	15.2	13.9	-	-	-	-	-	-	-	-	-
H4	35.6	29.3	24.1	20.7	18.8	17.4	15.9	14.2	-	-	-	-	-	-	-	-	•
H5	39.9	32.5	26.4	22.7	21.2	19.0	17.2	15.6	-	-	-	-	-	-	-	-	-
H6	39.0	30.9	25.9	23.3	20.3	18.7	16.9	14.8	-	-	-	-	-	-	-	-	-
H7	37.9	30.0	25.3	22.7	19.6	17.9	16.8	15.2	-	-	•	-	-	-	-	-	-
H8	38.0	30.1	25.9	22.1	19.0	18.0	16.5	14.8	-	-	-	-	-	-	-	-	-
H9	33.3	28.3	24.4	20.9	18.7	16.9	15.7	14.4	-	•	-	-	•	•	-	•	-
H10	35.7	30.1	25.5	22.8	19.7	18.0	15.3	13.0	-	-	-	-	-	-	-	-	•
H11	31.5	27.0	23.4	20,4	18.4	16.9	14.9	13.0	•	-	-	· •	-	-	-	-	•
H13	-	13.8	17.0	19.5	16.3	14.9	13.8	12.5	8.8	-	-	-	-	-	-	-	-
H14	40.8	32.5	27.5	24.1	20.7	18.8	15.9	15.9	15.1	14.2	12.3	7.1	-	-	-	-	-
H16	-	25.3	24.3	17.5	16.8	14.3	14.3	14.3	14.3	13.4	12.4	11.9	11.9	-	-	•	-
H17	-	-	34.8	28.8	24.0	-	18.2	-	14.3	-	12.9	-	-	10.1	10.1	10.1	8.0
H18	38.4	31.8	26.5	21.2	17.9	-	14.3	-	12.9	-	8.8	-	-	-	-	•	-
H19	30.3	23.9	20.4	17.6	13.0	11.9	-	10.8	-	10.8	-	10.8	•	-	-	-	-
H20	25.9	21.1	17.1	14.2	11.3	10.1	9.4	8.0	5.0	-	-	-	-	•	-	-	-

b) Large Bed (D = 0.45 m)

Run\z	120	140	160	180	200	220	240	260	280	300
H21	33.7	27.5	22.7	19.4	16.7	15.1	14.2	12.8	10.7	•
H22	36.5	30.4	25.2	22.0	19.5	17.4	15.1	•	-	-
H23	41.4	34.5	30.0	26.2	22.0	19.5	17.4	16.3	14.7	11.8
H24	37.5	30.6	25.9	22.2	19.2	17.1	15.5	13.8	-	•
H25	42.0	34.0	28.3	22.2	20.5	19.2	17.1	14.7	-	-
H26	37.0	30.0	25.9	22.2	18.8	17.1	15.5	13.3	-	-
H27	41.4	34.0	30.2	24.2	21.7	19.2	17.4	15.1	-	•
H28	38.2	31.7	27.4	23.4	19.8	17.8	17.1	14.7	-	-
H29	36.5	30.8	25.9	22.2	20.5	18.5	15.5	12.8	-	-
H30	32.8	27.8	24.2	21.1	19.5	17.8	15.9	13.8	-	-
H31	30.0	27.4	23.9	20.5	18.5	17.1	14.7	-	-	-
H32	29.8	24.9	21.2	18.4	16.8	15.0	12.1	-	-	•



Figure 5.1: Comparison between experimental and predicted U_{ms} values.

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Authors	k	α	β	γ	δ	S _d (m/s)						
Wu et al. (1987)	10.6	1.05	0.266	-0.095	0.256	0.171						
Mathur-Gishler (1955)	1	1	1/3	0	0.5 0.092							
Present Work:												
All beds ^a	6.10	1.27	0.551	-0.004	0.520	0.033						
Conical-Cylindrical beds only ^a	18.5	1.19	0.373	-0.193	0.263	0.030						
Conical beds only ^a	0.147	0.610	0.243	0.508	0.477	0.0095						
All beds ^b	4.47	1.22	0.492	-0.007	0.518	0.034						
All beds ^c	13.5	1.17	0.372	-0.148	0.289	0.033						
^a The diameter of the cylindr ^c The diameter is given by <i>D</i> cylindrical beds.	^a The diameter of the cylindrical bed is used; ^b The maximum annulus diameter (D_m) is used; ^c The diameter is given by $D' = 0.8 D_m$ for the conical beds and $D' = D$ for the conical- cylindrical beds											

Table 5.5: Fitted constants for U_{ms} correlations.



Distance from Centre, mm

Figure 5.2: Radial velocity profile 10 mm above the shutter in an empty bed ($Q_s = 27.9 \text{ L/s}, d_i = 24.7 \text{ mm} \text{ and } T = 65 ^{\circ}\text{C}$).

To examine the effect of Q_a on U_{ms} , two series of experiments were performed with polystyrene particles under the following conditions: D = 0.24 m and H = 0.35 m; D = 0.45 m and H = 0.31 m. In both cases, the inlet orifice diameter (d_i) was 35 mm. The results obtained with the larger bed are shown in Figure 5.3 and it is apparent that, as Q_a was increased, the main air flow rate (Q_s) decreased more rapidly in order to achieve minimum spouting. As a result, U_{ms} decreased with Q_a . The total momentum flow into the beds, which results from Q_a and Q_s , was therefore calculated as

$$M_{t} = \frac{4\rho Q_{s}^{2}}{\pi d_{o}^{2}} + \frac{4\rho Q_{a}^{2}}{\pi d_{at}^{2}}$$
(5.1)

where d_{at} denotes the diameter of the nozzle tip through which the atomizing air is discharged (see Figures 3.7 and 3.13). As can be seen from Figure 5.4, the total momentum flow at minimum spouting is approximately constant. Since Equations (2.2) and (2.3) do not account for the momentum introduced into the bed by the atomizing air, Q_a was incorporated into the equations by calculating a normalized inlet diameter (d_i) given by

$$d_i'^2 = 4\rho(Q_s + Q_a)^2 / \pi M_t$$
(5.2)

The total momentum flux (M_t) is found from Equation (5.1).

Since Equations (2.2) and (2.3) were developed primarily for conical-cylindrical beds, experiments were undertaken to examine U_{ms} as a function of bed height for shallow beds where the particles are restricted to a portion of the conical section only. The experiments were conducted with polystyrene particles under the following conditions: $d_i = 25 \text{ mm}$, $Q_a = 0.53 \text{ L/s}$, $T = 60 \,^\circ\text{C}$. The results are shown in Figure 5.5 and indicate that when the bed is confined to the conical section, U_{ms} is not proportional to $H^{0.5}$ as suggested by the Mathur-Gishler equation, but it is approximately proportional to H. More specifically, Table 5.5 indicates that $U_{ms} \propto H^{1.08}$ for conical beds. The $H^{0.5}$ relationship suggested by the Mathur-Gishler equation predicts higher U_{ms} values than are found experimentally for shallow conical beds. The reason is that, in reality, more air flows through the spout than



Figure 5.3: Effect of atomizing air on minimum spouting velocity (minimum spouting velocity is based only on the main spouting air flow rate; D = 0.45 m, H = 0.31 m, $d_i = 35$ mm).



Figure 5.4: Momentum flow of air into the spouted bed (dashed and solid lines represent the 0.24 m and 0.45 m dia. beds, respectively).

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Figure 5.5: Effect of bed height on minimum spouting velocity, Um. (Cone-cylinder junctions are denoted by dashed lines; solid lines represent indicated relationships fitted to experimental data).



Figure 5.6: Comparison between experimental data and predictions from correlation based on the optimum diameter D'.

implied by this equation because the inlet air is unable to spread into the annulus as effectively since the annulus is smaller and the wall is closer to the centre of the bed.

Since the constants obtained for the Wu et al. equation are different in the case of conical and conical-cylindrical beds, an attempt was made to unify the results by using the maximum bed diameter (D_m) covered by the particles. When the bed particles extend into the cylindrical region of the spouted bed, this diameter corresponds to the cylinder diameter. The coefficients resulting from the least squares fit are presented in Table 5.5.

Upon observing the solids movement in the conical beds, D_m was suspected to be an inappropriate variable representing U_{ms} . The bed particles near the wall on the surface of the annulus were observed to move significantly slower than the particles near the centre of the bed. These differences in particle velocities were much more pronounced in conical beds than in conical-cylindrical beds. Consequently, the conical beds were suspected to behave as conical-cylindrical beds with $D < D_m$.

To determine the "effective" bed diameter (D') representing U_{ms} for shallow, conical beds the following relationship was considered:

$$D' = c D_m \tag{5.3}$$

The proportionality constant (c) was determined by obtaining the best fit between the experimental and predicted results. The optimum value of c was found to be 0.8 and the corresponding coefficients in the Wu et al. equation are also given in Table 5.5. As shown by Figures 5.1 and 5.6, the agreement between predicted and experimental results is improved. The optimal, modified Wu et al. equation thus becomes:

$$U'_{ms} = 13.5\sqrt{2gH} \left(\frac{d_p}{D'}\right)^{1.17} \left(\frac{d'_i}{D'}\right)^{0.372} \left(\frac{H}{D'}\right)^{-0.148} \left(\frac{\rho_p - \rho}{\rho}\right)^{0.289}$$
(5.4)

It is interesting to note that Equation (5.4) implies that $U'_{ms} \propto H^{0.352}$ for deep beds. The corresponding exponents of H for the equations of Wu et al. (1987) and Mathur and

Gishler (1955) are 0.4 and 0.5, respectively. This result also supports the contention made by Wu et al. that the H and ρ terms should not be combined as suggested by Mathur and Gishler. For shallow conical beds where $D' \propto H$, Equation (5.4) reduces to Q_{ms} (= $\pi/4$ $\cdot U'_{ms}(D')^2$) $\propto H^{0.96}$, i.e., the relationship is virtually linear. This agrees well with the experimental data shown in Figure 5.5 (note that U_{ms} is based on the column diameter in this figure; hence, $Q_{ms} \propto U_{ms}$). The conical bed data also could be well represented by the Wan-Fyong et al. (1969) equation, corrected for atomizing air (see Appendix I).

5.1.2. Axial Pressure Profile in Annulus

The axial pressure profiles near the spout-annulus interface are given in Table 5.3. The temperature, atomizing air and spouting air flow rates did not have a significant effect on the pressure profiles. Furthermore, the variations in the radial pressure profiles were small for conical-cylindrical and conical beds (see, for example, Figure 5.7).

The axial pressure profiles for two beds are shown in Figures 5.8 and 5.9 together with predictions based on the equations provided by Epstein-Levine (1980), Rovero et al. (1983) (which is the Epstein-Levine equation modified for beds with a conical base), and Morgan and Littman (1980). $U_{m\beta}$ U_p , U_{aHm} and H_m in these equations were calculated from the correlations given by Grace (1982), Clift et al. (1978), Epstein et al. (1978) and McNab and Bridgwater (1977), respectively. The first two expressions under-predicted the axial pressure profile whereas the Morgan-Littman equation gave fairly good agreement in the case of conical-cylindrical beds (see Figure 5.8). By contrast, all three equations performed poorly for conical beds as shown by Figure 5.9. However, when the equations are modified by using D' (see Equation (5.3)), the agreement is significantly improved, especially in the case of the Morgan-Littman equation (see Figure 5.10).

Further examination of the Morgan-Littman correlation showed that for the deeper bed (0.53 m) and changes in d_i , the agreement was not as good. In general, the correlation



Figure 5.7: Pressure profile in the annulus (conical bed, Run H22).



Figure 5.8: Axial pressure profile in the annulus near the spout-annulus interface (conical-cylindrical bed, Run H1).

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Figure 5.9: Axial pressure profile in the annulus (conical bed, Run H22, column diameter used in calculation).



Figure 5.10: Axial pressure profile in the annulus (conical bed, Run H22, D' used in calculation).

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Figure 5.11: Axial pressure profile in the annulus (H = 0.53, Run H17).



Figure 5.12: Comparison between measured and predicted pressure profile in the annulus.

over-estimated the pressure drop in the 0.53 m bed (see Figure 5.11), and under-estimated the effect of d_i . In an attempt to improve the predictions, the optimal constants in Equation (2.14) for the 0.24 m bed data were determined by a least squares technique. Although the optimal constants improved the agreement for deeper beds, the predictions for the shallower beds were somewhat impaired.

5.1.3. Velocity Profile in Annulus

The radial pressure variation in the annulus was found to be small (see Figure 5.7). However, even small pressure gradients can lead to significant changes in the air flow rate in the annulus. If no radial variation of pressure is assumed and the Morgan-Littman correlation is used, significant errors in the air flow rate result. This result is due to the larger flow area away from the centre of the bed. For this reason, the vector Ergun (1952) equation is solved to determine the fluid flow in the annulus.

The solution of the vector Ergun equation for Run H22 is shown in Figure 5.12 and the method of solution is given in Appendix I. The model predictions, in general, were in good agreement with the measured values. Moreover, significant improvement to the spout velocity predictions was observed when the fluid flow in the annulus is calculated using the vector Ergun equation rather than the one-dimensional equation.

5.1.4. Axial Velocity Profile in Spout

The measured spout velocity profiles are given in Table 5.4. The effect of atomizing air flow on u_s is significant and is particularly noticeable in the lower section of the bed as shown by Figure 5.13. The total spouting air flow exhibits a similar effect on u_s . Changes in bed height did not greatly influence the velocity profile in the spout.

The spout velocities predicted by the mass and momentum balance equations (Lefroy and Davidson, 1969) introduced in Chapter 4 represented the measured velocities well (e.g.,

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Figure 5.13: Effect of atomizing air on axial air velocity profile in the spout (Q, is fixed at approximately 32 L/s in all runs).



Figure 5.14: Air velocities in the spout determined experimentally and from Model IV hydrodynamics (Run H1).



Figure 5.15: Air velocities in the spout determined experimentally, from Model IV hydrodynamics and from Equation (5.5)(Run H22).
see Figures 5.14 and 5.15) if the aforementioned modified equations were used. The correlations and assumptions used in the mass and momentum balance equations included:

- The spout diameter is constant and is given by the Wu et al. correlation (1987);
- The pressure profile is given by the Morgan-Littman (1980) equation modified for conical beds and effective bed diameter D';
- The air flow rate in the annulus is given by the vector Ergun (1952) equation;
- The fluid-particle interaction term proposed by Richardson and Zaki (1954) is valid;
- The spouting air flow rate is calculated from the total momentum flux into the bed rather than the total flow rate,
- The air density is based on the initial spouting air temperature rather than the average bed temperature.

A sensitivity analysis for these variables showed that the strongest factor influencing the agreement between the velocity predictions and measurements was the correlation used to predict the pressure profile; good agreement with the velocity measurements was observed when the difference between the predicted and measured pressures was small.

Figures 5.14 and 5.15 show more pronounced deviations between the measured and predicted velocities near the base of the bed. Regardless of the presence of the atomizing air, the measured velocity was expected to be higher than the predicted (i.e., average) velocity since the measurements were based on the highest pressure difference indicated by the pitot tube (which corresponds to the highest velocity). The large discrepancy near the inlet may be the result of a large radial velocity gradient in the spout (Krzywanski, 1992; Abramovich, 1963).

Moreover, the velocity profiles shown in Figures 5.2 and 5.13 suggest considerable influence of the atomizing air on the radial velocity profiles, especially near the base of the bed. In an attempt to model the atomizing air velocity, the following equations were developed, based on the assumptions that no mixing between the atomizing jet and spouting air occurred and that the solids voidage remained the same as the average voidage in the spout:

$$d(\varepsilon_s u_{as} A_{as})/dz = 0 \tag{5.5}$$

where
$$A_{as} = \pi D_{as}^2 / 4$$
 (5.6)

and
$$dD_{as}/dz = k_{\theta} |u_{as} - u_s|/(|u_{as}| + |u_s|)$$
 (5.7)

Equation (5.7) is given in Abramovich (1963). D_{as} represents the spread of the jet boundary layer and k_{θ} represents the fully developed jet angle in still air. Using the spray angle (k_{θ}) value of 20°, Equation (5.5) was solved together with Equations (4.27) to (4.30). The results are presented in Figure 5.15 and show that the atomizing air velocity changes rapidly to a value slightly higher than the spouting air velocity a short distance away from the base of the bed. Above this height, the atomizing air velocity changes at the same rate as the spouting air velocity. This result is most likely due to the assumption that no mixing occurs across the jet boundary layer. In reality, mixing across the atomizing air jet boundary should occur (Rushton and Oldshue, 1953; Abramovich, 1963) especially with the entrainment of solids into the spout, and consequently, the boundary should quickly disappear. To model the atomizing air velocity in the spout appropriately, a means of estimating the mixing length should be devised; however, this task was beyond the scope of this thesis and is pursued no further.

Other explanations for the discrepancy in the measured and predicted velocities near the base of the bed include the constant spout diameter assumption and the inability to predict the pressure peak by the Morgan-Littman correlation. Most spout shapes quickly diverge to a constant spout diameter (Lim, 1978); therefore, the gas velocities in the lower spout region will be higher than if a constant spout diameter is assumed. In most of the pressure measurements (see Table 5.3), a pressure peak occurred near the base of the bed and none of the correlations given in the literature predicts such behavior. It is very likely that the

inability to predict the pressure peak by the correlation used in this work resulted in a lower predicted spout velocity near the base of the bed.

5.1.5. Solids Movement

The claims that particle segregation occurs in the annulus were made by Kutlouglu et al. (1983), Cook and Bridgewater (1978), Robinson and Waldie (1978), and Piccinini et al. (1977). The claims were visually investigated by using sulfur coated urea particles. Using a batch of screened particles of size less than 1.17 mm and greater than 3.35 mm (using US # 16 and # 6 sieves) dyed blue and red, respectively, the movement of the tagged particles in a larger batch of sulfur coated urea particles was observed in a six inch half-column. The urea used for this experiment was a low grade urea, and contained 10 % by mass (after coating) of particles with $d_p > 3.35$ mm and 25 % with $d_p < 1.17$ mm. Inspection of the these particles spouting at various bed heights and air flow rates using a video camera showed no observable particle segregation in the annulus.

Observations of the solids also indicated that the solids near the spout-annulus interface moved faster than the solids near the wall; this is consistent with the findings by Rovero et al. (1985).

5.2. Spray Studies

The operating ranges examined are summarized in Table 5.6. The drop size data detected by the image analyzer are extensive and are included in Appendix II.

5.2.1. Operating Limits

As stated in Chapter 3, the operating range of the atomizing nozzle used in this study was limited by no atomization and inconsistent atomization at low and high atomizing air flow

Fixed Operating Conditions						
Atomizing Air Temperature 160 °C						
Auxiliary (Spouting Air) Temperature	60 °C					
Steam Pressure	75 - 80 psi					
Molten Sulfur Temperature	~ 150 °C					
Varied Operating Conditions						
Atomizing Air Flow Rate	0.23 to 0.70 L/s					
Sulfur Flow Rate	2.1 to 6.1 g/s					

 Table 5.6:
 Operating ranges of the spray studies.

rates, respectively. Both limits can be seen in Figure 5.16. The lower limit of the atomization velocity (U_{ma}) could be well represented by

$$U_{ma} = 13.8 + 1.61 W_s \tag{5.8}$$

where W_s and U_{ma} have units of kg/h and m/s, respectively. The upper limit of consistent atomization or the minimum pulsating velocity (U_{mp} , in m/s) obeyed the following linear relationship for $W_s > 8$ kg/h:

$$U_{mn} = 3.88 + 4.59W_s \tag{5.9}$$

 U_{ma} corresponds to the minimum energy required to atomize sulfur and Equation (5.8) should be valid for most fluid atomizers. U_{mp} corresponds to the maximum atomization energy, i.e. additional air is wasted. This upper limit is probably caused by the pressure feed system used in this work and may not apply if other feed systems such as a sulfur pump were used.

The atomizing air velocity used in the coating runs was kept approximately mid-way between the operating limits to ensure that the spray was properly developed.



Figure 5.16: Operating limits for atomization of sulfur.

5.2.2. Spray Drop Size Distribution and Average Drop Size

A typical sulfur droplet size distribution detected by the image analyzer is shown in Figure 5.17. The majority of droplets fell into the size range of 6 to 50 μ m and appeared to follow a log-normal distribution (Orr, 1966), i.e.,

$$N = \frac{\Sigma N}{\ln \sigma_g \sqrt{2\pi}} \exp\left(\frac{-(\ln d_s - \ln d_{gc})^2}{2\ln^2 \sigma_g}\right)$$
(5.10)

where N = frequency of observation of the spray droplet diameter d_s

$$\ln d_{gc} = \Sigma (N \ln d_s) / \Sigma N \tag{5.11}$$

$$\ln \sigma_g = \sqrt{\Sigma \left[N \left(\ln d_s - \ln d_{gc} \right)^2 \right] / \Sigma N}$$
(5.12)



Figure 5.17: Experimental drop size distribution and predictions using lognormal equation (Run S1a).

A log-normal plot is shown in Figure 5.18. The Nukiyama-Tanasawa distribution (Lewis et al., 1948) gave a good representation of the larger drop sizes (see Figure 5.19), but failed for the range 6 to 50 μ m where the highest frequencies of drops were recorded. The Nukiyama-Tanasawa equation is given by

$$\ln\left(\frac{N}{d_s^2 \Delta d_s}\right) = \ln k_1 - k_2 d_s^{k_3} \tag{5.13}$$

where Δd_s denotes the diameter range of the N droplets. The log-normal distribution was therefore used to represent the drop size data, and the mean drop size and the deviation were calculated from Equations (5.11) to (5.12), respectively.



Figure 5.18: Log-normal representation of drop size distribution (Run S1a).



Figure 5.19: Nukiyama-Tanasawa representation of drop size distribution (index of 1/3 appeared to give optimum fit; Run S1a).

The frequency of the droplets outside the 6 to 50 μ m size range was much lower (see Figure 5.17) and therefore only the droplets in the size range 6 to 50 μ m were used in the calculations. Two methods were employed to determine the drop size distribution: the first method employed Equations (5.11) and (5.12) to determine the coefficients for Equation (5.10); the second method used an optimization procedure to determine the coefficients directly, i.e.,

Minimize
$$\sum \left(N_{counted} - \int_{cut} \frac{\Sigma N}{\ln \sigma_g \sqrt{2\pi}} \exp \left(\frac{-(\ln d_s - \ln d_{gc})^2}{2\ln^2 \sigma_g} \right) d(\ln d_s) \right)^2$$
 (5.14)

where 'cut' represents the droplets in a given size range and ΣN denotes the frequency of droplets in each cut. The values of σ_g , d_s , and ΣN that best represent the 6 to 50 μ m data according to Equation (5.10) were determined using this least squares technique. Both results can be seen in Figure 5.17. Although both methods appear to give good representation to this set of data (Run S1a), the second method resulted in large errors for runs with cuts that deviated from the log-normal shape. To avoid this error, only the first method was used for all subsequent calculations including the results given in Table 5.7.

The Sauter mean diameter (d_{sv}) , also known as the surface-volume diameter, was then calculated from

$$\ln d_{sv} = \ln d_{gc} + 2.5 \ln^2 \sigma_g \tag{5.15}$$

using the calculated values of d_{gc} and σ_{g} . The results are presented in Tables 5.7 and 5.8. The Sauter diameter is commonly used for mass transfer operations (Mugele and Evans, 1951) including coating operations. For liquids atomized in small converging nozzles with compressed air, Nukiyama and Tanasawa (1939) correlated d_{sv} in the following way:

$$d_{sv} = k_1 / u_r + k_2 (1000 Q_1 / Q_a)^{15}$$
(5.16)

where d_{sv} and u_r (= $u_a - u_l$) are expressed in μ m and m/s, respectively. The optimum values of the constants k_l and k_2 were -560 and 3.45, respectively, using the data given in Table 5.7. The drop size data obtained above the minimum pulsating velocity did not ap-

Run #	N	$d_{gc,}\mu \mathrm{m}$	σ _{g,} μm	$d_{sv_{sv_{s}}}\mu m$	$Q_{a,}$ L/s	W _s , g/s
S1a	254	20.5	1.47	29.8	0.357	3.12
S1b	101	20.3	1.40	27.0	0.357	3.12
S1c	123	18.4	1.61	32.5	0.357	3.12
Slabc	478	19.9	1.50	30.1	0.357	3.12
S2a	223	16.8	1.54	26.8	0.314	3.12
S2b	140	18.8	1.54	30.0	0.314	3.12
S2ab	363	17.5	1.55	28.2	0.314	3.12
S 3	251	15.8	1.55	25.4	0.442	3.12
S4	229	17.9	1.80	42.6	0.336	3.12
S 5	87	15.0	1.56	24.7	0.314	2.14
S 6	73	13.7	1.57	22.7	0.272	2.14
S 7	58	15.7	1.53	24.2	0.229	2.14
S 8	287	27.0	1.57	44.7	0.698	6.06
S9	184	16.9	1.88	45.9	0.570	6.06
S10	111	14.4	1.75	31.7	0.442	6.06
S 11	356	20.3	1.68	44.3	0.527	5.08
S12	184	17.9	1.83	44.5	0.442	5.08
S 13	430	18.1	1.76	40.3	0.612	5.08
S14	236	14.6	1.66	27.7	0.442	4.10
S14a	696	13.5	1.58	22.8	0.442	4.10
S14b	80	18.6	1.67	36.3	0.442	4.10
S14c	744	14.3	1.65	26.9	0.442	4.10
S 15	140	16.4	1.62	29.4	0.527	4.10
S16	128	17.2	1.70	34.4	0.357	4.10

Table 5.7: Results of drop size studies at various operating conditions.

pear to follow any clear trend (see Table 5.8) and were therefore not used in the optimization calculations. The results are displayed in Figure 5.20 and show poor agreement with the measured data. The following power-law equation, determined by a least squares method, gives a better representation of the data:

$$d_{sv} = 261(Q_a / Q_{ma})^{1.7}(Q_l / Q_a)^{1.9}$$
(5.17)

The units of d_{sv} are mm. This equation suggests that Q_a and Q_l should not be grouped together without adding a separate term to correct for their differing effects on d_{sv} . Equation (5.17) clearly shows (from the exponents of the Q_l and Q_a which are 1.9 and 0.2, respectively) that the effect of Q_l on d_{sv} is much greater than that of Q_a .

	Near middle of operating limits	Near pulsating point	Above pulsat- ing point	W _s , g/s
Q_{α} , L/s	0.229	0.272	0.315	2.14
<i>d_{sv},</i> μm	24.7	22.7	24.7	
Q_{α} , L/s	0.315	0.357	0.442	3.12
<i>d_{sy},</i> μm	30.0	29.8	25.4	
Q_{α} , L/s	0.360	0.442	0.527	4.10
<i>d_w</i> , μm	34.4	27.7	29.4	
Q_{α} , L/s	0.442	0.527	0.612	5.08
$d_{sv} \mu m$	44.5	44.3	40.3	-
Q_{σ} L/s	0.442	0.57		6.06
<i>d_{sv},</i> μm	45.9	44.7		

Table 5.8: Sauter mean diameters relative to the operating limits.



Figure 5.20: Predictions of Sauter mean diameter.

The main errors in d_{sv} were suspected to arise from uncertainties associated with the sample size, sample location, and sampling procedure. The errors associated with the sample size and the sample locations were investigated in runs S1a to S2b (see Table 5.7), and the largest error was ± 10 %. The maximum error associated with the rotation speed of the protective tube was even higher at ± 31 % (runs S14 and S14b).

In the model coating calculations which follow, it is assumed that all sulfur droplets have the same size equal to the Sauter mean diameter calculated from Equation (5.17).

5.2.3. Coating Mechanism and Sulfur Spray Concentration

The assumption that inertial deposition is the dominating mechanism for the deposition of sulfur droplets on the bed particles is verified by using the collection efficiency equations suggested by Clift et al. (1981). Table 5.9 shows typical values of the collection efficiency due to diffusion (E_D) , inertial deposition (E_I) , gravitational settling (E_G) and direct inter-

Table 5.9: Mechanical collection efficiency (using correlations suggested by Clift et al., 1981 and Behie et al., 1972) at selected operating conditions in the spout (ρ_c = 1773 kg/m³, d_p = 2.16 mm, μ = 2.08 x 10⁻⁵ kg/m·s).

<i>U</i> , m/s		5	5	0
<i>d_s,</i> μm	6	50	6	50
S,	0.40	27	4.0	275
$E_D = (4.36 / \varepsilon) (\wp / Ud_s)^{2/3}$	3.3 x 10 ⁻⁶	7.9 x 10 ⁻⁷	7.1 x 10-7	1.7 x 10 ⁻⁷
$E_{I} = \frac{0.0036 - 0.2323S_{t} + 2.422S_{t}^{2}}{-2.033S_{t}^{3}, 0.083 < S_{t} \le 0.6}$	0.165	_	-	
$E_I = S_t^2 / (S_t + 0.5)^2, \ S_t > 0.6$	-	0.965	0.788	0.996
$E_G = 0.075 \sqrt{N_G}$	0.00074	0.0061	0.00023	0.0019
$E_{DI} = 6.3 N_{DI}^2 \varepsilon^{-2.4}$	0.00011	0.0077	0.00011	0.0077

ception (E_{DI}) at selected operating conditions. The values of the air velocity (U) and the droplet size (d_s) given in Table 5.9 are close to the operating conditions in the spout. The results clearly indicate that inertial deposition is the dominating collection mechanism under the present coating conditions.

The collection efficiency alone, however, is not sufficient to calculate the sulfur droplet deposition rate onto bed particles; information on the spray distribution above the nozzle is required also. Consequently, experiments were attempted to determine the spray distribution; however, a suitable device for detecting the sulfur concentration in the bed could not be developed. A sampler without a heated tip could not deal with the molten sulfur spray, which plugged up the tip of the sampler. Consequently, efforts were made to heat the tip electrically (to oxidize the sulfur) with NiCr wires wrapped around the tip. Unfortunately, the wires only lasted a few seconds in the bed because they could not withstand collisions with the fast moving bed particles and the presence of urea dust which left carbon residue on wires. Internal heating of the tip was not possible due to space limitations of the sampler (i.e., the sampler had to be small enough to be placed in the spout without significantly upsetting the upstream flow). The details of the samplers which were tested in this work are provided in Appendix I.

Since the spray distribution could not be measured directly, the assumptions concerning the spray angle (see Chapter 4) and the height (h_{fds}) at which the sulfur droplets start to deposit onto the bed particles could not be verified. However, the angle and height could be deduced from the particle coating distribution, which is the topic of the next section. The predicted sulfur concentration profiles for various spray angles for Run C17 are shown in Figure 5.21; corresponding values for various coating runs using a spray angle of 20° are shown in Figure 5.22. The operating conditions for the runs shown in Figures 5.21 and 5.22 are given in Table 5.11. Decreasing spout velocity causes the initial increase in the sulfur concentration until $z = h_{fds}$. Note that in Figure 5.21, the spray angle



Figure 5.21: Predicted sulfur concentration profile for various spray angles.



Figure 5.22: Predicted sulfur concentration profile for various coating runs ($\phi = 20^{\circ}$).

of 180° corresponds to completely dispersed spray droplets in the main air stream or $h_{fds} =$ 0; in reality, such a spray angle does not exist.

5.3. Coating Distribution and Product Quality

The coating distribution and quality of products were measured according to the methods outlined in Chapter 3. The coating distribution was also calculated using the models presented in Chapter 4 and the product quality was calculated using the methods outlined in this section. The operating ranges applicable to this study are listed in Table 5.10, and the experimental and model results are given in Table 5.11.

One deviation from the methods described in Chapter 3 relates to the measurement of the urea feed rate. The urea feed rates often did not match the calibrated rates (see Appendix IV); as a result, the urea feed rate (W_{μ}) was determined from the product discharge rate (W_{p}) and the mean sulfur content found by the crush test (\overline{X}_{sm}) : i.e.,

$$W_{u} = W_{p} (1 - \overline{X}_{sm}) \tag{5.18}$$

The mean sulfur content (\overline{X}_s) based on the feed rates of urea and sulfur (also given in Table 5.11) was never in complete agreement with and was usually somewhat larger than \overline{X}_{sm} . One reason for the discrepancy is that the products may have been sampled before steady state conditions were achieved. Consequently, the predicted product quality, U_{D} , (see Section 5.3.2) given in Table 5.10 is based on \overline{X}_s rather than \overline{X}_{sm} and represents the seven day urea dissolution value for the products obtained under steady state operations.

It should be noted that comparisons between the coating distribution predictions of Models II and III with Models I and IV are not possible as the relationships between the variables in Models II and III to the operating variables were not determined in this work (see Chapter 4). Furthermore, the predictions of Models II and III could not be compared directly with the measured results for the same reason. Consequently, the discussions regar-

Operating Variable	Operating Range
Spouting air flow rate, Q_s	20.1 - 29.2 L/s
Atomizing air flow rate, Q_a	0.23 - 0.78 L/s
Bed Height, H	0.28 - 0.36 m
Bed Diameter, D	0.24 and 0.45 m
Spouting air orifice diameter, d_i	24.7 - 35.1 mm
Urea feed rate, W_{μ}	7.6 - 19.9 g/s
Sulfur injection rate, W_s	3.12 - 6.06 g/s
Bed Temperature, T_b	57 - 64 °C

Table 5.10: Operating range applicable to coating distribution and product quality studies.

Table 5.11: Operating conditions investigated for the coating study.

Run	D _i	Q,	Qa	W _s	W _u	T _b	D	H	\overline{X}_{sm}	$U_{\rm Dm}$	\overline{X}_{s}	U_{D}
#	(mm)	(L/s)	(L/s)	(g/s)	(g/s)	(°C)	(m)	(m)	%	%	%	%
C17	24.7	21.1	0.44	4.1	8.6	57	0.24	0.28	25.9	50.7	32.3	42.2
C19	24.7	21.1	0.44	4.1	5.65	57	0.24	0.28	32.6	37.5	42.1	32.8
C20	24.7	21.1	0.44	4.1	14.6	57	0.24	0.28	23.0	64.2	21.9	59.4
C21	24.7	21.1	0.44	4.1	19.9	57	0.24	0.28	17.1	69.2	17.1	70.0
C22	24.7	22.8	0.23	5.08	10.1	63	0.24	0.28	30.0	52.3	33.3	41.9
C23	24.7	24.5	0.44	5.08	8.75	64	0.24	0.28	32.7	44.1	36.8	38.5
C24	24.7	22.8	0.57	5.08	10.1	64	0.24	0.28	28.0	40.6	33.4	41.6
C25	24.7	20.1	0.44	4.1	9.91	64	0.24	0.28	26.1	48.2	29.3	46.8
C26	24.7	22.8	0.44	4.1	9.73	64	0.24	0.28	23.1	46.3	29.7	46.6
C29	28.2	24.5	0.44	4.1	9.92	64	0.24	0.28	22.5	51.0	29.3	47.7
C30	35.1	29.2	0.44	4.1	10.5	64	0.24	0.28	21.8	74.6	28.2	n/s
C31	24.7	21.1	0.23	3.12	9.43	60	0.24	0.28	21.1	64.7	24.9	54.0
C33	24.7	21.1	0.53	6.06	11.3	60	0.24	0.28	24.5	38.0	34.9	39.2
C38	24.7	22.8	0.44	4.1	11.7	60	0.24	0.36	23.2	51.5	25.9	52.3
C39	24.7	27.6	0.44	4.1	7.6	64	0.45	0.355	17.6	49.9	35.1	n/s
C40	24.7	27.6	0.78	6.06	11.1	64	0.45	0.355	15.3	72.4	35.3	n/s

ding Models II and III results are mostly confined to a sensitivity analysis of the model variables.

5.3.1. Coating Distribution

Forty sulfur coated urea particles were randomly sampled and the sulfur content of the individual particles were determined using the particle crush test described in Chapter 3. The sulfur content $(X_{sp,i})$ was then normalized according to

$$(X_{sp,i})_{new} = X_{sp,i} \overline{X}_{sp} / \overline{X}_{sm}$$
(5.19)

where \overline{X}_{sp} is the average sulfur content of the 40 particles sampled. The results can be seen in Figures 5.23 and 5.24 for Runs C17 and C38, respectively; the results for other selected runs are provided in Appendix II.

As shown by Figure 5.24, Model IV with the spray angle (ϕ) of 20° and Model I appear to give good predictions of the measured sulfur content distributions. Both models, however, under-predicted the amount of the inadequately coated particles (i.e., $X_s < 10$ %). This result was expected for Model I as discussed already in Chapter 4. In case of Model IV, the size of the actual spray zone was suspected to be lower than that predicted, which meant that the actual spray angle was larger or that coating occurred below the assumed h_{fds} . It is unlikely that the spray angle is larger than the angle provided by the manufacturer ($\phi = 20^\circ$) as the angle should decrease in the presence of spouting air (see Equation (5.5)). Therefore, h_{fds} was suspected to be incorrect.

Consequently, the effect of h_{fds} was investigated by examining the coating distribution of batch products. The batch products were used because the effects of the feed location and exit probability are absent, and the influence of h_{fds} can be isolated. Figure 5.25 shows the coating distributions for the batch products coated for 210 and 480 s under the following conditions: $W_s \approx 3.5$ g/s, $H \approx 0.3$ m, D = 0.24 m, $T \approx 60$ °C and $d_i = 28.2$ mm.



Figure 5.23: Comparison between measured and predicted coating distributions for Run C17.



Figure 5.24: Comparison between measured and predicted coating distributions for Run C38.



Figure 5.25: Coating distributions of batch products.

Again, the model under-predicted the amount of inadequately coated particles but, when $(h_{fds})_{new} = 0.5(h_{fds})_{\phi=20^{\circ}}$ was used, the predictions improved considerably. The physical explanation for this result (assuming the spray angle is fixed at 20°) is that the bed particles start to be coated at a height which is approximately half of the distance at which the spray reaches the spout-annulus interface. Significant improvements were also noted for the continuous data in both Figure 5.24 when these new h_{fds} values were used in the calculations.

The discrepancy between the measured and predicted distributions may be attributed to uncertainties in the measurements of the sulfur flow rate, urea feed rate, and individual particle sulfur content. The error in the sulfur flow rate is typically ± 1 scale reading on the sulfur rotameter which amounts to less than ± 5 % error in the flow rate. The error in

the urea feed rate using Equation (5.18) is less than ± 9 %. The largest error contributing to the coating distribution is probably the sulfur content measurements of individual particles (see Chapter 3); the error was as large as ± 25 % in determining the urea content of individual particles. The errors in the measurements of \overline{X}_{sm} and U_{Dm} amounted to less than 3 %.

The discrepancies between the measured and predicted distributions shown in Figures 5.23 to 5.25 fall within the maximum measurement errors; however, consistently lower values of \overline{X}_{sm} compared to \overline{X}_s in Table 5.11 suggest that other errors may be involved. Other errors were suspected to result from the small sample sizes used to measure the coating distribution and from sampling beds operating under unsteady state conditions. These matters are further investigated in Section 5.3.3.

5.3.2. Product Quality

The measured and predicted values of the product quality (expressed in terms of the seven day dissolution test) are given in Table 5.11. In determining the predicted values, the quality of an individual particle was assumed to be the same as that of batch products having the same sulfur content.

The quality of the batch product was measured and correlated using a hyperbolic function which was found to give a good representation of the data: i.e.,

$$U_{D,i} = \frac{1}{2} \left[\tanh\left(\frac{c_1 - X_{sp,i}}{X_{sp,i}(c_2 - X_{sp,i})}\right) + 1 \right]$$
(5.20)

where $U_{D,i}$ and $X_{sp,i}$ represent the seven day urea dissolution rate and sulfur content, respectively. Twenty-three batch runs were conducted to find the optimum values of c_1 and c_2 which were 0.198 and 0.715, respectively, using a non-linear least squares optimization method. The results are plotted as Figure 5.26. All runs were conducted under the fol-



Figure 5.26: Relationship between 7-day dissolution and sulfur content for batch products.

lowing initial operating conditions: T = 60 °C, H = 0.25 m, D = 0.24 m, $d_i = 24.7$ mm, $Q_s = 25$ L/s, $Q_a = 0.44$ L/s and $W_s = 4.1$ g/s. Adjustment in the spouting air flow rate (Q_s) was required to keep the bed from collapsing, and the bed temperature (T) varied up to 15 °C due to the steady injection of molten sulfur (introduced at ~150 °C). Other than these operating variables, only the operating time was varied to control the sulfur content of the products.

The predicted dissolution values $(U_{D,i})$ were calculated from Equation (5.20) and averaged to determine the predicted product quality (U_D) given in Figure 5.27 and Table 5.11. All model predictions are based on steady state operations. Of all runs listed in Table 5.11, only four runs were approximately under steady state when product sampling occurred



Figure 5.27: Comparison between measured and predicted product quality.

(see next section), and they are noted in Figure 5.27. Other runs had probably not reached steady state when sampling occurred and, as a result, the measured urea dissolutions were higher than the predicted dissolutions.

5.3.3. Effect of Operating and Model Variables on Coating Distribution

5.3.3.1. Effect of Operating Time

All product samples were collected after (at least) ten minutes of coating operation at which time steady state was assumed to exist (see Chapter 3). For a typical coating operation (e.g., $W_p = 14$ g/s, $\rho_p = 1400$ kg/m³, $\varepsilon = 0.42$, H = 0.28 m and D = 0.24 m), the average residence time (\bar{t}) of the bed particles is ~400 s, and ten minutes (t = 600 s) of operation corresponds to $t/\bar{t} = 1.5$ and $F(t) \approx 0.8$ for a perfectly mixed bed of particles

(see Equation (4.1)). (F(t) = 1 indicates the true steady state condition, and occurs when $t \rightarrow \infty$.) For most coating runs, the average coating time was approximately 20 minutes which corresponds to $t/\bar{t} = 3$ and $F(t) \approx 0.95$.

The large values of F(t) indicate that most of the runs were close to steady state conditions; however, the coating distribution and sulfur content results indicated that some runs might not have reached steady state at the time of sampling. Consequently, an experiment was conducted to investigate this effect. The effect of the operating time was investigated in a deep bed (0.36 m) with a very low urea feed rate (approximately 4 g/s) to accentuate the effect. The average residence time of the particles under this condition was ~20 minutes. The results can be seen in Figure 5.28. As indicated by the changing shapes of the coating distribution curve, steady state was not achieved after ten minutes ($t/\bar{t} = 0.5$) of coating operation. Even after operating for two hours ($t/\bar{t} = 6$ and F(t) = 0.998; i.e., very close to steady state for a perfectly mixed bed), the sulfur content of the heavily coated fraction was still changing. Similar trends were also found in Tsai's (1986) investigation with a 0.15 m bed after three hours of continuous operation.

An explanation for this result is that the spouted bed does not behave as a perfectly mixed bed. The spouted bed may be seen as a partly well-mixed and partly plug-flow vessel in which the bed particles spend most of their time in the plug flow zone (annulus). Consequently, the spouted bed approaches steady state more slowly than does a perfectly mixed vessel.

The steady state condition is also indicated by the values of sulfur content. The measured sulfur content values (\overline{X}_{sm}) for the three samples collected are given in Figure 5.28 and the sulfur content based on the feed rates of urea and sulfur (\overline{X}_s) was 42.3 %. When steady state is achieved, \overline{X}_{sm} should approach \overline{X}_s . (The fact that the \overline{X}_{sm} values for the one and two hour runs are slightly higher than the \overline{X}_s value is probably due to uncertainty in pro-



Figure 5.28: Effect of operating period on measured coating distribution.

duction rate measurement). Consequently, the sulfur content can be used as a measure to determine whether steady state was achieved. In this work, steady state was assumed to exist if the following condition was met:

$$X_s - X_{sm} \le 3\%$$
 (5.23)

The value of 3 % was chosen based on the uncertainty of the crush test (see Chapter 3). The steady state results based on Equation (5.23) are noted in Figure 5.27.

The results from this investigation provide an explanation for the discrepancy between the predicted and measured results for the heavily coated fraction in Run 17 (see Figure 5.23). The product was probably sampled from the bed while it was still under a transient operation (e.g., $\overline{X}_s - \overline{X}_{sm} > 6$ %; see Table 5.11) and, as a result, its heavily coated fraction is shown to deviate from the model steady-state predictions.

5.3.3.2. Effect of Sample Size

Ideally, the sample size should be as large as possible to obtain a small sampling error; however, the sample size is limited by the resources available for the analysis. The sample sizes used to determine the coating distributions in this work were limited to 1000 and 40 particles for numerical (Monte Carlo) and manual sampling, respectively. These sizes were chosen on the basis of the costs associated with computer time and the time required to analyze the coated urea products.

5.3.3.2.1. Numerical Sampling

The errors associated with sample sizes and the effect of sample size on coating distribution were investigated for the various mathematical models. The errors associated with the sample sizes were determined by comparing the results of Models II and III. Models II and III were chosen for this purpose because both models should yield identical results when a = 0 for Model III (see Equation (4.19)), but the methods of solution are different - i.e., the Model II solutions are exact whereas those of Model III are based on the numerically approximate Monte Carlo method. The differences between the model predictions of the average coating amount were calculated from

$$\sigma = \sqrt{\sum_{i=1}^{n} [(m_{s1})_i - (m_{s1})_{\infty}]^2 / n_{sim}},$$

where $(m_{s1})_i$ represents the Model III results given by Equation (4.22), $(m_{s1})_{\infty}$ represents the Model II results given by Equation (4.11), and n_{sim} represents the number of simulations performed. The results are summarized in Table 5.12 and include three simulations per sample size. For each simulation, a different seed or initialization was used to generate random numbers. Note that the m_{s1} values are directly related to the average sulfur content (\overline{X}_{sm}) values of the sampled particles; therefore, the σ values apply to \overline{X}_{sm} as well. The results are also plotted in Figure 5.29. The step-wise nature of the curves in

N	seed $= 1$	seed $= 2$	seed = 5	σ, %
100	6.75446 x 10 ⁻⁷	6.17060 x 10 ⁻⁷	5.83438 x 10 ⁻⁷	7.0
500	6.42544 x 10 ⁻⁷	5.58211 x 10 ⁻⁷	6.06776 x 10 ⁻⁷	5.7
1000	6.46193 x 10 ⁻⁷	5.66462 x 10 ⁻⁷	6.10643 x 10 ⁻⁷	5.4
5000	6.11628 x 10 ⁻⁷	5.86313 x 10 ⁻⁷	6.03037 x 10 ⁻⁷	2.0
10000	6.09172 x 10 ⁻⁷	6.07758 x 10 ⁻⁷	6.07282 x 10 ⁻⁷	1.1
8	6.06776 x 10 ⁻⁷	6.06776 x 10 ⁻⁷	6.06776 x 10 ⁻⁷	0.0

Table 5.12: Values of m_{s1} for various sample sizes based on the Model II and III results.

Figure 5.29 are due to the spray zone assumption used in Model II, and are discussed further in Section 5.3.3.3.

Besides showing considerable scatter in the average and individual sulfur content values, the probabilities of the particles with the lower sulfur content are higher for smaller sample sizes. This trend is probably due to the small probabilities associated with particles having a high sulfur content which could easily be missed when the sample size is small. This effect appears to be more pronounced in Figure 5.29 for N = 100.

The deviation between the measured results (N = 40) and the predicted results (N = 1000)for Run C38 (see Figure 5.24) appears similar to the deviation between the N = 50 and N = 1000 results shown in Figure 5.30. Since Run C38 had probably reached steady state at the time of sampling (as indicated by $\overline{X}_s \cong \overline{X}_{sm}$), the deviation between the predicted and measured results probably arose from the small sample size used to determine the measured coating distribution.

5.3.3.2.2. Manual Sampling

A similar investigation was conducted for the manually sampled results. Two samples of 40 particles from Run C17 were analyzed for their sulfur content, and the sum of the two



Figure 5.29: Effect of sample size on predicted coating distribution (Models II and III).



Figure 5.30: Effect of sample size on predicted coating distribution (Model IV).

samples was used as the third sample. The results are plotted in Figure 5.31. The scatter provides some measure of the magnitude of the errors between the two sample sizes.

5.3.3.3. Effect of Spray Angle

The spray angle (ϕ) is a term used to define the lower height at which the coating occurs (h_{fds}) in Model IV. It should not be confused with the actual spray angle of the sulfur spray as the spray density of an actual spray varies radially while the density within ϕ does not. Furthermore, the effect of increasing ϕ may be seen as that of decreasing h_{fds} .

As shown by Figure 5.32, decreasing the spray angle reduces the fraction of uncoated particles. This result is similar to the Model III results for decreasing the value of a, as



Figure 5.31: Effect of sample size (manual sampling).



Figure 5.32: Effect of spray angle on coating distribution (results from Model IV).



Figure 5.33: Effect of the spray concentration on coating distribution (results were obtained from Model III using Equation (4.19)).

shown by Figure 5.33. A large value of *a* corresponds to a rapid decrease in the spray concentration in the spray zone. As ϕ or *a* increases, the size of spray zone is reduced and more particles leave the bed uncoated. The fact that a 40° spray angle (i.e., equivalent to $(h_{fds})_{new} = 0.5(h_{fds})_{\phi=20^\circ}$) describes the experimental data better as shown in Figure 5.24 and 5.25, is probably due to the fact that the actual spray zone was lower than assumed. As mentioned in Section 5.3.1, this improvement was probably achieved by allowing coating to occur below the assumed h_{fds} and was not caused by an actual increase in ϕ .

The step increases associated with the a = 0 curve in Figure 5.33 and the curves representing smaller spray angles in Figure 5.32 resulted because a significant portion of bed particles have the same probability of receiving the same amount of coat in a discrete number of passes through the spray zone before leaving the bed. The first step represents the fraction of particles receiving one coat before leaving the bed, and subsequent steps represent the fractions of particles receiving more than one coat before leaving the bed. For $a \neq$ 0, few bed particles have identical probabilities of passing through the same fraction of the spray zone; therefore, the steps disappear from the figures. These steps, however, can be seen in all Model II results (e.g., $N = \infty$ case in Figure 5.29).

5.3.3.4. Effect of Feed Location

Tsai (1986) claimed that the product quality improved as a result of introducing the urea feed near the wall of the bed. Particles which descend in the annulus near the wall, enter the lower portion of the spout where the spray zone is located. If all particles are forced into the spray zone, uncoated particles are avoided. However, Model II predictions in Table 5.13 indicate that the change in the coating distribution is small for $x_e (= W_p/W_c) =$ 0.05. The explanation for this is that the forced feed ensures at least one pass through the spray zone, but since a small x_e value indicates the amount of coat per cycle is very small and the total number of cycles is large, one additional cycle through the spray zone will not significantly add to the total coat on the particles. Under the operating conditions ex-

	$x_e = 0.05$				$x_e = 0.2$			
Arbitra loca	Arbitrary feed location		Feed location at the wall		Arbitrary feed location		Feed location at the wall	
P [*] _{kc} , %	$\overline{X_s}$, %	$P_{kc}^{*}, \%$	$\overline{X_s}$,%	$P_{kc}^{*}, \%$	$\overline{X_s}$, %	$P_{kc}^{*}, \%$	$\overline{X_s}$,%	
5	0	0	0	17	0	0	0	
14	3	10	3	44	12	40	12	
22	6	19	6	63	21	64	21	
29	9	27	9	75	29	78	29	
36	12	34	12	84	35	87	35	
48	17	47	17	89	41	92	41	
57	21	57	21	93	45	95	45	
65	25	65	25	95	49	97	49	
71	29	72	29	97	52	98	52	
77	32	77	32	98	55	99	55	
81	35	81	35	99	58	99	58	
84	38	85	38	99	60	100	60	
87	41	88	41	99	62	100	62	
90	44	91	44	100	64	100	64	
	P_{kc}^{\bullet} = cumulative probability							

Table 5.13: Effect of feed location (using Model II with $x_c = 0.5$, $\overline{X}_s = 0.25$).

amined in this study, x_e was less than 0.05 and only small changes were observed in the coating distribution for various feed locations. The effect of the feed location on the coating distribution for the conditions of Run C17 can be seen in Figure 5.34.

5.3.3.5. Effect of Beds-in-Series

The greatest improvement in the coating distribution can be achieved by operating several beds-in-series, as shown by Figures 4.2 and 5.35. Models I and IV both predict drastic reduction in uncoated and heavily coated fractions for large numbers of beds-in-series; the distributions approach those of the batch products (see Figure 4.3). These improvements in the product are of commercial importance and are discussed further in Section 5.4. (In Model IV, one bed size was used regardless of number of beds-in-series.)



Figure 5.34: Effect of feed location (results from Model IV).



Figure 5.35: Product coating distributions for beds-in-series (from Model IV).

5.3.3.6. Effect of Model Variables, x_e and x_c

The variables x_c and x_e in Models II and III characterize two main factors influencing the coating distribution. Their relationships to the operating variables were not determined in this work as a large number of experiments are required (see Chapter 4); however, the effect of operating variables on these model variables can be explained qualitatively. The size of the spray zone is characterized by x_c , and increasing x_c is equivalent to increasing the spray zone or reducing the by-pass zone. The number of times the particles pass through the spray zone is characterized by $x_e (= W_p/W_c)$, and increasing x_e is equivalent to increasing the particle discharge rate, decreasing the circulation rate, or both.

The operating variables that affect x_c are Q_s , Q_a , W_s and ϕ . Q_s determines the impaction velocity of the droplets onto the bed particles and influences both η and N_i in Equations (2.26) and (4.34). Q_a and W_s determine the droplet size and influence η in Equation (2.26). ϕ influences the spray zone directly. The value of x_c increases with Q_a and decreases with Q_s , W_s and ϕ .

The operating variables that affect x_e are W_p and W_c . W_p is directly affected by W_u and W_c is influenced by Q_s , H, D and d_i . W_c increases with Q_s , H, D and d_i . The value of x_e increases with W_p and decreases with W_c .

The effects of x_c and x_e on coating distribution are shown in Figures 5.36 and 5.37. The results presented in Figure 5.36 indicate that for $x_e = 0.05$ and $x_c = 0.6$, over 30 % of particles receive three coats or less. Three coats on a particle are likely not enough to completely cover the particle. To increase the number of coats on the particles, x_e must be decreased and x_c increased (see Figure 5.37).

The variables x_e and x_c can be varied to simulate various types of beds. These variables are also useful in predicting the presence of uncoated urea particles in the product stream.



Figure 5.36: Sensitivity analysis of x_e on coating distribution using Model II ($x_c = 0.6$).



Figure 5.37: Sensitivity analysis of x_c on coating distribution using Model II ($x_e = 0.2$).

Both plug flow and CST residence time models can be represented by manipulating the x_e and x_c values. Plug flow is simulated when $x_e = 1$ which is equivalent to a spouted bed equipped with a draft tube which also serves as a discharge tube. Continuous stirred tank behavior is simulated when $x_e = 0$. Model I predictions can be duplicated by Model II when $x_c = 1$. However, a finite fraction of uncoated urea particles in the product stream is only predicted when $x_c < 1$. Note also that Model III becomes Model II when a = 0.

5.3.4. Sensitivity Analysis Using Model IV

Certain operating variables $(Q_s, Q_a, d_i, H, D \text{ and } W_p)$ had little effect on the coating distribution and were not discussed in the previous section. In this section, the effects of all the principal variables are examined. Model IV provided good predictions of the measured data (see Figures 5.23 - 5.25 and 5.27); therefore, its results are used to identify the main variables influencing the product quality.

The principal factor affecting the product quality is the sulfur content. The amount of sulfur deposited on the urea product indicates the thickness of coating; the quality of the coat, in turn, determines the rate of urea dissolution. The model and experimental results in Figure 5.38 clearly indicate that the sulfur content is the major factor influencing the product quality.

Since the sulfur content is the dominant factor influencing the product quality, and because Model I is based only on \overline{X}_s , Model I should also provide a good estimate of the results. As shown by Figure 5.38, Model I does give a good estimate of Model IV results, and both models agree well with the experimental results regardless of the condition (steady state or transient) under which the samples were collected. This result is not surprising since the value of x_e is very small for the operating conditions considered in this study. The discrepancy between the predicted and measured results are likely due to the presence of significant amounts of uncoated urea in the bed under transient conditions, as all coat-



Figure 5.38: Model predictions of product quality.

ing operations started with uncoated urea and insufficient time was allowed for the bed to reach steady state.

The differences between the Model I and Model IV predictions are attributed to the effects of other operating variables on the product quality. To determine the effects of the principal operating variables, a sensitivity analysis was conducted. The sulfur content and the bed temperature were fixed at 25 % and 60 °C, respectively, and only one variable was changed at a time. The range of model variables examined are given in Table 5.14 and the results are shown in Table 5.15. The same base condition was not used for all runs as a change in one variable often resulted in a change in other variables (e.g., an increase in the urea feed rate required an increase in the sulfur feed rate to produce 25 % sulfur coated urea). U/U_{ms} values of either 1.1 or 1.2 were used in all runs, unless otherwise specified.

Variable	Operating Range
Spouting air velocity, U/U_{ms}	1.05 - 1.4
Atomizing air flow rate, Q_a	0.44 - 0.78 L/s
Bed Height, H	0.25 - 0.55 m
Bed Diameter, D	0.25 - 0.45 m
Spouting air orifice diameter, d_i	21.2 - 28.2 mm
Urea feed rate, W_{μ}	6.43 - 22.9 g/s
Spray angle, ϕ	12 - 180°
Bed particle size, d_p	2.1 - 4.3 mm
Feed location, ΔD_f	0.02 m - Arbitrary
Sample size, N	50 - 5000 particles
Number of beds in series, N_{beds}	1 - 8 beds

Table 5.14: Range of model variables examined for sensitivity analysis.

The range of model variables studied actually exceeded the range that was verified for the hydrodynamics study and, as a result, the coating model predictions may not provide accurate results for certain experimental conditions (e.g., for deep beds and large orifice sizes).

The results given in Table 5.15 show that the maximum change in U_D is less than 4 % for a single bed. Such a small change in the product quality may be explained by examining the effect of each operating variable on coating distribution; however, the effect of most operating variables may be explained more easily by considering the effects of Model II variables x_e and x_c (see Section 5.3.3.6). In this manner, the effects of many operating variables on the coating distribution can be simplified to the effect of just the two model variables.

For the operating conditions examined, $x_e < 0.1$. For such small values of x_e , the effect of x_c on the coating distribution is noticeable (see Figure 5.39) only for the inadequately coated ($\overline{X}_s < 15$ %) and heavily coated ($\overline{X}_s > 30$ %) particles, i.e., little change in the
Bed Diameter		Atomiz	zing Air	Urea Fe	ed Rate	Orifice 1	Diameter
<i>D</i> , m	U _D ,%	Q_{α} , L/s	U _D ,%	W _w , g/s	U _D , %	<i>d_i</i> , mm	U _D ,%
0.25	54.2	0.44	52.5	6.43	53.9	21.2	53.4
0.35	53.6	0.61	52.2	12.3	54.2	_24.7	54.2
0.45	53.3	0.78	52.0	22.9	54.3	28.2	53.9
Bed Height		Spouti	ng Air	Bed Par	ticle Id.	Spray	Angle
<i>H</i> , m	U _D , %	U/U _{ms}	<i>U</i> _{<i>D</i>} ,%	<i>d_n</i> , mm	U _D , %	φ	U _D ,%
0.25	53.9	1.05	54.7	2.1	54.5	180°	56.0
0.35	53.9	1.1	54.2	3.2	54.4	36°	52.9
0.45	54.2	1.2	54.4	4.3	54.6	18°	52.5
0.55	54.2	1.4	53.7			12°	52.7
Feed Location							
ΔD_f , m	0.02	0.05	0.1	0.15	$D - D_s$		
U _D ,%	53.4	53.9	54.2	54.3	54.3		
Sample Size							
N	50	100	500	1000	5000		
$U_{_{D}}, \%$	58.4	53.3	54.9	54.0	54.2		
Beds in Series							
N _{beds}	1	2	4	8			
U _D ,%	54.0	46.4	40.8	35.9			
Model I Prediction: $U_D = 54.2 \%$							

Table 5.15: Results of sensitivity analysis using Model IV ($\overline{X}_s = 0.25$, T = 60 °C).

range 15 % $< \overline{X}_s < 30$ % is observed for $x_e = 0.05$, even though the x_c value is quite different. This change in the coating distribution does not change the product quality significantly because the product quality is determined primarily by the fraction of particles having 15 % $< \overline{X}_s < 30$ % (see Figure 5.26 or Equation 5.20).

The most significant improvement in product quality occurs when urea is coated in a series of beds. This result is consistent with the above explanation that the fraction in the range



Figure 5.39: Effect of a large change in x_c on coating distribution ($x_e = 0.05$).

15 % < \overline{X}_s < 30 % determines the product quality. Figure 5.35 clearly shows that this fraction becomes larger as the number of beds increases.

Since Model I provides an accurate prediction of the fraction in the range 15 % $< \overline{X}_s < 30$ % (see Figure 5.24), it also provides an accurate prediction of the product quality (see Table 5.15). In addition, the Model I solution can be obtained easily; hence, it may be useful in obtaining quick and accurate estimates of the product quality. However, it should be noted that the inadequately coated and heavily coated fractions are also important (e.g., to the appearance of the product and design specifications which require information on the maximum bed particle size) and the Model I predictions of these fractions may be poor.

The effect of sample size can also be seen in Table 5.15. The purpose of including these values was to check the error associated with the sample size. The results in terms of the deviations from the value obtained for 5000 particles are similar to those of the standard deviations calculated for the coating distribution (see Table 5.12).

5.4. Commercial Implications

The findings of this study are applicable to the design of commercial coating plants. Model I provides a quick estimate of the product quality suitable for preliminary plant designs, while Model IV may be used for more detailed studies. Models II and III can aid with conceptualizing spouted bed coating processes.

Commercial plants will normally have considerably higher capacities than the current apparatus. Higher capacities may be obtained in two ways: increasing the bed size and increasing the number of beds in parallel. The latter option should be considered only if the former option does not provide the desired capacity, because an increase in the number of beds can pose operational difficulties. These include controlling the distributions of air and liquid sulfur to different beds. Larger beds, on the other hand, may increase the production rate without sacrificing the product quality (presuming the hydrodynamics and x_e stay constant).

It is unlikely that the product quality can be controlled by varying the production rate alone. As discussed already, the product quality is determined primarily by the number of beds in series. The product quality increases with the number of beds-in-series, but because operational difficulties also increase, the minimum number of beds that meets the design quality specification should be used. Note that the increase in the product quality is large when the number of beds increases from one to two beds, but the increase in the product quality becomes smaller for each additional bed.

Chapter 6. Conclusions and Recommendations

6.1. Conclusions

The spouted bed coating process for the batch-wise and continuous production of sulfur coated urea has been successfully modeled using mass and momentum balance equations and inertial sulfur droplet deposition as the dominating coating mechanism.

Specific conclusions that may be drawn from the results given in Chapter 5 are listed in the following sub-sections.

6.1.1. Bed Hydrodynamics

- The minimum spouting velocity (U_{ms}) varies linearly with the bed height in conical beds.
- U_{ms} varies proportionally with the total momentum due to atomizing air and spouting air flow into the bed.
- U_{ms} , axial pressure profile in the annulus and air velocity in the spout in conical beds are similar to the corresponding quantities in conical-cylindrical beds with the column diameter equal to 80 % of the maximum diameter of conical beds.
- U_{ms} for shallow beds could be well represented by the correlation given by Equation (5.4).
- The Morgan-Littman (1980) correlation provides accurate axial pressure profiles in the annulus for shallow spouted beds if the diameter modification for conical beds is used.

• The mass and momentum balances suggested by Lefroy and Davidson (1969) give accurate predictions of air velocity in the spout, provided that the vector form of the Ergun (1952) equation for gas flow in the annulus and the modifications listed above are used.

6.1.2. Spray Studies

- Consistent sulfur atomization by the nozzle used in this work occurs in the range defined by Equations (5.8) and (5.9).
- The size of the sulfur droplets typically ranged from 6 to 50 μm dia. and is well represented by a log-normal distribution.
- The average droplet size is affected primarily by the sulfur feed rate, and is well represented by the power-law correlation given by Equation (5.17).
- The dominant mechanism by which the sulfur droplets deposit onto the bed particles is inertial impaction.

6.1.3. Coating Distribution and Product Quality

- The spray angle, urea feed location and the number of beds-in-series significantly affect the amount of lightly coated and heavily coated particles in the sulfur coated urea product.
- The product quality is primarily affected by the average sulfur content of the product and the number of beds-in-series. Spray angle, feed location, atomizing and spouting air flow rates, urea and sulfur feed rates, and bed diameter also affect the product quality, but to a much lesser extent.
- The Monte Carlo method provides good representation of coating distribution if a sufficient number of particles are simulated.
- The spouted bed coating unit is capable of producing sulfur coated urea of widely varying quality at a high production rate.

6.2. Recommendations for Further Work

Certain difficulties and assumptions in modeling the coating process resulted from the limitations of the one-dimensional models used in this work. A significant number of assumptions could be reduced if a fully two-dimensional (axi-symmetric) model is developed. For example, h_{fds} would not have to be assumed because the particle concentration and the spray distribution in the spout could be determined in a two-dimensional model. Some experimental work may be necessary to develop a model of the radial spray density in the spray region. The model should also be a three-phase model as the effects of sulfur spray on the bed hydrodynamics cannot be properly determined with the two-phase model developed in this work. The two-dimensional, two-phase model developed by Krzywanski (1992) would be a good starting point for such a model.

Before commercialization of the spouted bed process, a pilot plant consisting of several beds-in-series should be evaluated. Amongst the foreseeable problems are control of the distribution of air and sulfur to different beds, transportation of large particles from bed to bed, and separation of large particles from the product. These problems cannot be addressed until a pilot plant is actually available. A continuous sulfur supply system should also be incorporated into the pilot plant to study the effects of operating time.

The hydrodynamics of larger diameter beds should also be examined because many correlations used in this work may not apply to larger beds (Lim and Grace, 1987).

Finally, the sulfur droplet distribution in the spouted bed could not be directly measured using the devices examined in this work. A further study on such a device could be valuable and it would advance aerosol detection in complex environments. The devices studied here could be a good starting point for such an effort.

Nomenclature

A _{as}	Local spray area, m ²
A _o	Area of shutter opening, m ²
A _p	Projected area of spout particles per unit volume of spout, m ⁻¹
A_p^*	Projected area (cross-sectional area) of a particle, m ²
A _s	Spout cross-sectional area, m ²
a	Exponent in Equation (4.17), dimensionless
С	Concentration of sulfur droplets in the gas phase, kg/m ³
C _c	Spray concentration defined for Model III, kg/m ³
Co	Pitot tube coefficient, dimensionless
с	Fitted coefficient of D in Equation (5.3)
P	Diffusion Coefficient, m ² /s
D	Column diameter, m
$D\{z\}$	Local bed diameter, m
D'	"Effective" bed diameter $(D' = 0.8 D_m \text{ in conical section and } D' = D \text{ in conical-cylindrical section}), m$
D ₂₅	Seven day urea dissolution for products with 25 % sulfur content, fractional
D _{as}	Local diameter of the spray, m
D_{f}	$D - \Delta D_{f} \mathbf{m}$
D _{ini}	Location of feed particle on the surface of the bed during its first cycle, m
D _m	Maximum annulus diameter for the conical bed, m
D,	Location of bed particles on the surface of the bed, m
D,	Spout diameter, m
d_a	Diameter of atomizing air nozzle cap at top, m

d_{at}	Diameter of nozzle orifice, m
d_c	Diameter of aerosol particle, m
d_{gc}	Geometric counted mean of droplet size, m
d_i	Orifice diameter based on the flow area of Q_s , m
d_i'	Corrected orifice diameter in Equation (5.2), m
d_o	Shutter diameter based on the flow area, m
d_p	Average diameter of bed particle, m
d_{pi}	Mean diameter of adjacent Tyler screen apertures, m
d _s	Diameter of sulfur spray droplets, m
d _{sv}	Sauter mean (surface-volume) diameter, m
$F{t}$	Distribution function of residence time, dimensionless
f_{1}, f_{2}	Coefficients of Ergun equation, kg/m ³ s and kg/m ⁴
$E_{D}, E_{D},$	Collection efficiency due to diffusion, direct interception, gravitation and
E_G, E_I	inertial impaction, dimensionless
G	ho U
$G\{X_s\}$	Exit sulfur content distribution, dimensionless
g	Acceleration due to gravity, m/s ²
H	Loosely packed static bed height, m
H_{c}	Height of cone base, m
H_m	Maximum spoutable bed depth, m
h	H/H _m
h _{fds}	Height at which the spray coating begins, m
k	Coefficient in Equation (2.3); number of passes through the spray zone
k _c	Proportionality constant in Equation (4.19)
k_p	Probability constant in Equation (4.20), m ⁻¹
k _e	Angle of fully developed spray jet, °
М.	Momentum flow of spouting and atomizing air into the bed, $kg \cdot m/s^2$

m_p	Total weight of the bed particles, kg
m _s	Weight of sulfur in sulfur coated urea, kg
\overline{m}_s	Mean weight of sulfur in sulfur coated urea, kg
m_s^*	Conditional weight of sulfur deposited on urea, kg
m _{sl}	Amount of sulfur deposited on a bed particle per pass through the spray zone, kg
\overline{m}_{sl}	Mean weight of sulfur picked up per pass through spray zone, kg
m _u	Weight of urea in sulfur coated urea, kg
<i>m</i> _u	Mean weight of urea in sulfur coated urea, kg
Ν	Sample size, particles; number of cycles, cycles
N_a^*	Deposition rate of sulfur droplets in the annulus between streamlines, kg/s
N _{DI}	Interception parameter, dimensionless
N _G	Gravitational settling parameter, dimensionless
N _i	Deposition rate of sulfur droplets on spout particles per unit volume of spout, kg/s·m ³
N _R	Numerically generated random number between 0 and 1
n	Number of beds in series in Equation (4.7)
ñ	Direction normal to streamlines, m
Р	Pressure, Pa
P_f	Pressure in fluidized bed, Pa
$P_a\{z\}$	Pressure at z, Pa
$\Pr\{z\}$	Probability of particle entering at z into the spray zone, fractional
P _{kc}	Probability of particle entering the spray zone k times, fractional
Q _a	Volumetric atomizing air flow rate, m ³ /s
Q_l	Flow rate of spray liquid, m ³ /s
Q _{ma}	Atomizing air flow rate at minimum atomization, m ³ /s
Q_{ms}	Volumetric flow rate at minimum spouting, m ³ /s

Q,	Volumetric spouting air flow rate, m ³ /s		
Qt	Total volumetric flow rate = $Q_a + Q_s$, m ³ /s		
r	Radial distance from the centre of the bed, m		
S	Distance along the stream line, m		
S _d	Standard deviation of U_{ms} predictions, m/s		
S _t	Stoke's number, dimensionless		
T, T _b	Bed temperature, °C		
t	Time, s		
ī	Mean residence time of solids, s		
t _c	Cycle time, s		
t _r	Residence time, s		
U	Superficial air velocity, m/s		
U	Vector form of air velocity, m/s		
U_D	Predicted seven day urea dissolution rate, fractional		
U_{Dm}	Measured seven day urea dissolution rate, fractional		
U _{ms}	Minimum spouting velocity based on column diameter (D), m/s		
U' _{ms}	Minimum spouting velocity based on D' , m/s		
U,	Volumetric rate of air cross-flow per unit area of spout-annulus interface, m/s		
U _t	Terminal settling velocity, m/s		
U	Interstitial air velocity, m/s		
u _r	Relative velocity between gas and liquid at the orifice, m/s		
V _b	Total bed volume, m ³		
V,	Entrainment rate of bed particles into the spout per unit area of spout- annulus interface, m/s		
v	Local bed particle velocity, m/s		
W _c	Circulation rate of bed particles, kg/s		

W_p	Production rate, kg/s
W _s	Sulfur injection rate, kg/s
W _u	Urea feed rate, kg/s
X	Defined in Equation (2.13), dimensionless
X,	Sulfur content, dimensionless
\overline{X}_{s}	Mean sulfur content of sulfur coated urea based on urea and sulfur feed rates, fractional
\overline{X}_{sm}	Mean sulfur content of sulfur coated urea based on a crush test, fractional
\overline{X}_{sp}	Mean sulfur content of simulated (SCU) particles, fractional
$X_{sp,i}$	Sulfur content of individual particle, fractional
x	z/H _m
x _b	Probability of particle by-passing the spray zone, fractional
x _c	Probability of particle entering the spray zone, fractional
x _e	Probability of particle exitting the bed after each cycle, fractional
x _f	Fraction of area occupied by the urea feed in the area defined by ΔD_{fw} , fractional
x _r	1 - x_e , fractional
Y	Defined in Equation (2.11)
Z	Axial destance from base of the bed, m
z _c	Length of spray zone from base of the bed, m
<i>Z</i> _s	Location of particle's entry into the spout for urea feed particles, m

Greek Letters

α_p	Defined in Equation (2.8)
$\alpha, \beta, \varepsilon, \gamma$	Exponent in Equation (2.3)
β_p	Particle-fluid interaction term, dimensionless

Ζ	1	Difference
Z	MD_f	Twice the width of annulus covered by feed adjacent to the wall of the spouted bed at the top of the bed, m
ε	:	Voidage, fractional
đ	$\{z\}$	Probability density, m ⁻¹
¢)	Spray angle, °
¢) ₅	Particle shape factor or sphericity, fractional
r	n	Collection efficiency, fractional
q	0	Defined in equation (2.14), dimensionless
μ	l	Fluid viscosity, kg·m/s
μ	4	Viscosity of liquid spray, kg·m/s
0)	Included spouted bed cone angle, °
ρ)	Fluid density, kg/m ³
ρ	a	Atomizing air density, kg/m ³
ρ	Ь	Bulk density of loose-packed solids, kg/m ³
ρ	c	Aerosol density, kg/m ³
ρ	1	Density of liquid spray, kg/m ³
ρ	p	Particle density, kg/m ³
ρ	22	Density urea particles, kg/m ³
Σ	;	Sum
σ		Standard deviation (Table 5.12), fractional
σ	g	Log-standard geometric deviation, m
σ	1	Surface tension of liquid spray, dyn/cm
ψ	r	Stream function, dimensionless
\overline{V}	,	Directional gradient

Subscripts

0	Condition at $z = 0$
1	per cycle
a	Annulus; atomizing air
as	Atomizing air in spout
Н	Condition at $z = H$
i	Pertaining to an individual particle or 'cut'
1	Liquid spray
ma	At minimum atomization
mf	At minimum fluidization
mp	At minimum pulsation
ms	At minimum spouting
P	Bed particle
5	Spout; sulfur
u	Urea

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Appendix I:

Supporting Derivations and Methodology

In this section, selected experimental and numerical methods used to determine various model and operating variables are provided. All symbols used in this section are consistent with the symbols used in the main body of this thesis, unless specified otherwise. The following is a list of supporting materials given in this section

I-1. Determination of Shutter Area

I-2. Model II Derivation for Forced Urea Feed

I-3. Calculation Method for Vector Ergun Eqaution

I-4. Sulfur Sampling Devices

I-5. Batch Coating Model

I-6. Minimum Spouting Velocity Predictions by Wan-Fyong et al. Equation

I-1. Determination of Shutter Area

As shown by Figure 3.4, a shutter was used to control the area of spouting air flow into the bed. The shutter consisted of five leaves which formed a circle when the shutter was fully open and formed a pentagon when fully closed. The shape of the shutter opening was an irregular shape when the shutter was not fully closed or fully opened. In this section, the area of the shutter was determined by examining the shape of the shutter for all positions.

When the shutter was fully open, the shape of the opening was close to a perfect circle. this shape was assumed to be a circle and the radius of the circle is denoted by R. The shape of the shutter was not a circle nor a



pentagon when the shutter was not fully open, and the area of the shutter representing one-fifth of the area is shown in Figure I-1. The measured width of the shutter opening, D_e , is given by

$$D_e = a + b + t \tag{I-1}$$

where a, b and t are given in Figure I-1. The angle between lines a and b is equal to 36°; therefore,

$$b = a\cos(36^{\circ}) \tag{I-2}$$

and
$$c = \sqrt{a^2 - b^2}$$
 (I-3)

The length of t can be determined by considering a section of the fully open shutter shown in Figure I-2. The length of t is given by

$$t = R - L \tag{I-4}$$

where $L = \sqrt{R^2 - c^2}$ (I-5)

The angle, ϕ , between R and L is then



$$\phi = \cos^{-1}(L/R) \tag{I-6}$$

and the area outside of the triangle is given by

$$C = \phi R^2 - c \cdot L \tag{I-7}$$

By substituting Equations (I-2) and (I-4) into Equation (I-1), the value of a can be determined:

$$a = \frac{-\beta \pm \sqrt{\beta^2 - 4\alpha\gamma}}{2\alpha} \tag{I-8}$$

where
$$\alpha = 2(1 + \cos 36^\circ)$$
 (I-9)

$$\beta = 2(R - D_e)(1 + \cos 36^\circ)$$
 (I-10)

$$\gamma = D_e^2 - 2D_e R \tag{I-11}$$

Substituting *a* into Equations (I-2) and (I-3) gives *b* and *c*. The values of *t* and ϕ can be determined from Equations (I-4) to (I-6). It follows that the area of the shutter (A_o) shown in Figure I-1 is given by

$$A_o = 5(b \cdot c + C) \tag{I-12}$$

and the orifice diameter, d_i , based on the flow area is given by

$$d_i = \sqrt{A_o / (\pi/4)} \tag{I-13}$$

I-2. Model II Derivation for Forced Urea Feed

Model II derivation for the spouted bed coating unit that forces the feed into the spray zone is given in this section. The assumptions are that all feed enters the spray zone in the first cycle and that the volume of the bed particles does not change significantly. The simplified diagram of this system is shown below.



Figure I-3: Simplified flow sheet of a forced feed system.

Since the volume of the particles is assumed to be constant, the percentage of the feed that enters the spray zone is equal to that leaving the bed; therefore, the spray zone will be reduced by the same amount for the returning particles. It follows that

$$x_{c} = (1 - x_{e} - x_{b}^{*}) / (1 - x_{e})$$
(I-14)

and
$$x_b = x_b^* / (1 - x_e)$$
 (I-15)

where x_c and x_b are probabilities of returning particles entering and by-passing the spray zone. x_b^* is the absolute probability of particles by-passing the spray zone. The probability of particles not receiving any coat is zero since all particles are forced through the spray zone as it is introduced into the bed. The probability of particles receiving one or more coat is given by

$$P_{1c} = x_{e} + x_{e}x_{b}x_{r} + x_{e}x_{b}^{2}x_{r}^{2} + x_{e}x_{b}^{3}x_{r}^{3} + \cdots$$

$$P_{2c} = x_{e}x_{c}x_{r} + 2x_{e}x_{c}x_{b}^{2}x_{r} + 3x_{e}x_{c}x_{b}^{2}x_{r}^{2} + 4x_{e}x_{b}^{3}x_{r}^{3} + \cdots$$

$$P_{3c} = x_{e}x_{c}^{2}x_{r}^{2} + 3x_{e}x_{c}^{2}x_{b}^{2}x_{r} + 6x_{e}x_{c}^{2}x_{b}^{2}x_{r}^{2} + 10x_{e}x_{c}^{2}x_{b}^{3}x_{r}^{3} + \cdots$$

$$\vdots$$

$$P_{kc} = \binom{k}{k}x_{e}x_{c}^{k-1}x_{r}^{k-1} + \binom{k+1}{k}x_{e}x_{c}x_{b}^{2}x_{r} + \binom{k+2}{k}x_{e}x_{c}x_{b}^{2}x_{r}^{2} + \cdots$$
(I-16)

Using the infinite series representation in Table 4.2, this equation can be reduced to

$$P_{kc} = \frac{x_{e} x_{c}^{k-1} x_{r}^{k-1}}{(1 - x_{b} x_{r})^{k}}$$
(I-17)

and \overline{m}_{s} using Equation (4.10) becomes

$$m_{s} = k \cdot m_{s} (x_{b} + x_{c} / x_{s}) \tag{I-18}$$

the sulfur content distribution equation is obtained by substituting above expression into

Equation (4.3), which is gives

$$\overline{X_s}(k) = \frac{k \cdot x_e \overline{X_s}}{k \cdot x_r \overline{X_s} + (x_b x_e + x_c)(1 - \overline{X_s})} \text{ for } k = 1, \infty$$
(I-19)

1-3. Calculation Method for Vector Ergun Equation

The basic derivation is identical to that given in Rovero et al. (1983). The derivation is shown here only for verification purposes.

The vector Ergun equation is given by

$$-\nabla P_a = \mathbf{U}(f_1 + f_2|\mathbf{U}|) \tag{I-20}$$

Applying the curl operation to this equation gives

$$-\nabla \times \mathbf{U} = \mathbf{U} \times \nabla \ln(f_1 + f_2 |\mathbf{U}|) \tag{I-21}$$

Introducing stream functions defined for axisymmetric motion about the axis of the flow,

$$u_z = -\frac{1}{r}\frac{\partial\psi}{\partial r}$$
 and $u_r = \frac{1}{r}\frac{\partial\psi}{\partial z}$ (I-22)

Equation (I-21) can be rewritten as

$$\begin{cases} \frac{f_1r}{f_2} \left[\left(\frac{\partial \psi}{\partial z} \right)^2 + \left(\frac{\partial \psi}{\partial r} \right)^2 \right]^{1/2} + \left[2 \left(\frac{\partial \psi}{\partial r} \right)^2 + \left(\frac{\partial \psi}{\partial z} \right)^2 \right] \right] \frac{\partial^2 \psi}{\partial r^2} \\ + \left\{ \frac{f_1r}{f_2} \left[\left(\frac{\partial \psi}{\partial z} \right)^2 + \left(\frac{\partial \psi}{\partial r} \right)^2 \right]^{1/2} + \left[2 \left(\frac{\partial \psi}{\partial z} \right)^2 + \left(\frac{\partial \psi}{\partial r} \right)^2 \right] \right\} \frac{\partial^2 \psi}{\partial z^2} \\ - \left\{ \frac{f_1}{f_2} \left[\left(\frac{\partial \psi}{\partial z} \right)^2 + \left(\frac{\partial \psi}{\partial r} \right)^2 \right]^{1/2} + \frac{2}{r} \left[\left(\frac{\partial \psi}{\partial z} \right)^2 + \left(\frac{\partial \psi}{\partial r} \right)^2 \right] \right\} \frac{\partial \psi}{\partial r} \\ + 2 \left[\frac{\partial \psi}{\partial z} \frac{\partial \psi}{\partial r} \frac{\partial^2 \psi}{\partial r \partial z} \right] = 0 \end{cases}$$

(I-23)

Equation (I-10) was solved with the following boundary conditions:

at the wall,

 $\psi = 0 \tag{I-24}$

at the bed surface,

$$\partial \psi / \partial z = 0$$
 (I-25)

and at the spout-annulus interface,

$$\frac{\mathrm{d}P_a}{\mathrm{d}z} = -\frac{1}{r}\frac{\partial\psi}{\partial r}\left\{f_1 + \frac{f_2}{r}\left[\left(\frac{\partial\psi}{\partial z}\right)^2 + \left(\frac{\partial\psi}{\partial r}\right)^2\right]^{1/2}\right\}$$
(I-26)

 dP_{a}/dz was determined from the Morgan-Littman (1980) equation. Equations (I-23) to (I-26) were expressed in second order finite differences forms and solved iteratively using a successive under-relaxation factor of 0.7.

Using the calculated velocity field, the pressure profile was then determined by taking divergence of Equation (I-20) and imposing the continuity condition for an incompressible fluid: i.e.,

$$\nabla^2 P_a = -f_2(\mathbf{U} \cdot \nabla \mathbf{U}) \tag{I-27}$$

Expansion yields

$$\frac{1}{r}\frac{\partial}{\partial r}\left[r\frac{\partial P_a}{\partial r}\right] + \frac{\partial^2 P_a}{\partial z^2} = -\frac{f_2}{\left(u_r^2 + u_z^2\right)^{1/2}} \left\{u_r^2\frac{\partial u_r}{\partial r} + u_r u_z\left(\frac{\partial u_z}{\partial r} + \frac{\partial u_r}{\partial z}\right) + u_z^2\frac{\partial u_z}{\partial z}\right\} (I-28)$$

which is solved with the following boundary conditions:

at the wall

$$\partial P_a / \partial \bar{n} = 0 \tag{I-29}$$

at the bed surface

$$P_a = P_H \tag{I-30}$$

and at the spout-annulus interface, the pressure distribution given by the Morgan-Littman is applied. Second order finite difference approximations were used for all equations except at the cone wall boundary where a first order approximation was used.

1-4. Sulfur Sampling Devices

Various sulfur sampling devices were tested for use in the spouted bed and none were found to be satisfactory. In this section, the problems associated with the devices considered for measuring the sulfur concentration in the spouted bed are summarized to aid those wishing to continue this work. Brief descriptions of each device as well as measurement procedures and problems associated with each device are provided in Tables I-1 to I-6.

All devices considered here include a tip with 1 mm opening. The direct sampling of sulfur (without heating the sampling tube) was not possible as molten sulfur smothered the tip of the sampler and immediately plugged the tip. Heated samplers were therefore considered. Series of tests were conducted with the packed bed heating element; however, electrical contact between the iron particles could not be established when the iron particles were in a loose state. Sintering of iron particles resulted in a very low electrical resistance across the packed bed and partial sintering resulted in a very fragile bed. Plate design and laser beam methods were also considered, but they were regarded difficult to construct and costly.

Finally, the quartz tube designs using NiCr wires to heat the tip were built and tested. The 3/8" three-tube was found too large to place in the spout. A smaller 1/4" two-tube was found to be a satisfactory size for sampling in the spout. However, this design also failed. The problems inherent to this design were that the quartz tube eventually broke as the tube was moved around the bed (i.e., slight force against this tube broke the tube), the exposed NiCr wires could not withstand the presence of urea dust and the impact of SCU particles, and wire sizes that can be used were limited.

The main problem associated with hot tip samplers was that the samplers must stay red hot in order to convert S to SO_2 upon impaction of S. None of the samplers could accomodate enough room for insulating material. As a result, the heating tip was cooled by the air, bed materials, and sulfur spray. Unless a considerable amount of wattage can be forced into the heating elements, the element cannot stay red hot. The temperature gradient in the sampler tube caused urea to melt over the heating wires which prevented the wires from getting red hot. Due to space limitations on the sampler, the maximum wire size that can be used to heat the tip was limited to 1/30", and this wire size limited the maximum heating capacity of the wires to approximately 120 W.

In summary, an appropriate device for measuring the sulfur concentration in the spouted bed had to meet the following criteria:

- the device has to be slim enough to enter the spout without upsetting the upstream conditions,
- the device must withstand the presence of excess sulfur and urea dust at the tip,
- the tube must be strong enough to withstand the collisions to fast moving bed particles and lateral movements through the annulus,
- and the device must be large enough to accomodate electrical heating element (the heating element should not be exposed to urea or sulfur dust).

None of the devices considered in this work met all these conditions.

Description of Device	1/8" SS tube connected to an in-line filter and a vacuum pump.
Method of Sampling	Vacuum suction; isokinetic sampling; length of sample timed using a stop-watch; calibrate sulfur collected as a result of in- trodution and withdrawl of sampler in and out of the spout
Collection Mechanism	Direct sulfur sampling
Method of Analysis	Gravitational method using CS_2 after urea is washed using H_2O ; filter and parts leading to the filter are analyzed for the sulfur content
Likely Problems	Plugging of the sampler tip due to solidification of sulfur at the tip of the sampler, and due to excess amount of sulfur near the spray nozzle; difficulty of moving the sampler into the spout using short tube (which must be inserted through the side of the wall) – longer tubes will be difficult for analysis using the gravitational method; hazards of working with CS_2

Table I-1: Direct sulfur sampling.

Table I-2: Hot tip: packed bed $S \Rightarrow SO_2$ converter.

Description of Device	3/8" SS outer tube with alumina inner sleeve and high resistant (electrically) pellets sintered into a packed bed; the packed bed is heated to a red hot state by passing electricity through it; the tip end of the sampler serves as an electric lead and the other lead is inserted through the side of the sampler using non-elec- tric conducting fittings
Method of Sampling	Vacuum suction; isokinetic sampling; by electrically heating the bed, S is converted into SO_2
Collection Mechanism	Heating and reacting spray deposits into SO ₂
Method of Analysis	Amount of SO_2 determined via SO_2 analyzer, and amount of S determined from volumetric flow rates and stoichiometry
Problems Observed	Plugging of the tip due to difficulties in heating the tip electri- cally; difficulties of sintering the bed to give a certain electrical resistance – usually the resistance became too low (small voi- dage) or too high (little sintering, resulting in no electric con- tact); localized heating

Table I-3: Hot tip: plate $S \Rightarrow SO_2$ converter.

Description of Device	3/8" SS outer tube with alumina inner sleeve and high resistant (electrically) plate; the plate is heated to a red hot state by
	the side of the sampler using non-electric conducting fittings
Method of Sampling	Vacuum suction; isokinetic sampling; by electrically heating the plate, S is converted into SO_2
Collection Mechanism	Heating the spray (deposited via inertial impaction onto the plate) and converting S to SO_2
Method of Analysis	Amount of SO_2 determined via SO_2 analyzer, and amount of S determined from volumetric flow rates and stoichiometry
Likely Problems	Plugging of the tip due to difficulties in heating the tip electri- cally; difficulties of manufacturing the plate with the electrical leads, and variable density plate

Table I-4: Hot tip: laser beam $S \Rightarrow SO_2$ converter.

Description of Device	3/8" mm quartz or SS tube with a beam sink (spherical ball bearing) located at the end of the sampler
Method of Sampling	Vacuum suction; isokinetic sampling; heat the ball with the la- ser beam
Collection Mechanism	Zapping the S spray (deposited via inertial impaction onto the ball) into SO_2
Method of Analysis	Amount of SO_2 determined via SO_2 analyzer, and amount of S determined from volumetric flow rates and stoichiometry
Likely Problems	Inaccurate trajectory of the laser beam due to vibrations of the sampler in the bed contributed by the air and particles hitting the sampler; plugging of the tip due to difficulties in heating the tip electrically; incorporation of the laser beam to the exist- ing apparatus due to lengthy nature of laser beam source; cost and availability of laser beam

Description of Device	3/8" quartz outer tube, 1/4" quartz inner shell and 1/8" quartz heating tube to hold the heating element; the tip and the inner shell are heated using NiCr wires; two electrical leads are separated by the with the quart tubes for the inner shell and outer wires are exposed to the bed
Method of Sampling	Vacuum suction; isokinetic sampling; by electrically heating the wire, S is converted into SO_2
Collection Mechanism	Spray deposits on to the tip of the inner shell via inertial impaction onto the plate, and converts to SO_2 upon impaction
Method of Analysis	Amount of SO_2 determined via SO_2 analyzer, and amount of S determined from volumetric flow rates and stoichiometry
Problems Observed	Too large to be placed in the spout

Table I-5: Hot tip: three tube hot wire $S \Rightarrow SO_2$ converter.

Table I-6: Hot tip: two tube hot wire $S \Rightarrow SO_2$ converter.

Description of Device	1/4" quartz outer shell and 1/8" quartz inner tube to hold the heating element; the tip and the inner shell are heated using NiCr wires; two electrical leads are separated by the with the quart tubes for the inner shell and outer wires separated physi- cally by the strength of the wires; all wires are exposed to the material
Method of Sampling	Vacuum suction; isokinetic sampling; by electrically heating the wire and therefore the inner shell, S is converted into SO_2
Collection Mechanism	Spray deposits on to the tip of the inner shell via inertial impaction onto the inner shell, and converts to SO_2 upon impaction
Method of Analysis	Amount of SO_2 determined via SO_2 analyzer, and amount of S determined from volumetric flow rates and stoichiometry
Problems Observed	NiCr wires cannot withstand the presence of urea, urea vapour and impaction of bed particles; melting and reaction of urea at the sides of the sampler, and eventually making its way to the heating elements and greatly lowering the temperature (the heating element could not sustain the red color) of the heating element; presence of large urea particles in the bed as result of urea agglomeration due to melted urea

I-5. Batch Coating Model

Model IV may be manipulated to predict coating distributions of batch products. In the continuous coating operation, the simulation of a particle in the bed begins as the particle enters the bed and the simulation ends as the particle leaves the bed. The probability associated with the urea feed location determines the path of the particle during its first cycle through the bed, and the probability associated with the returning particles from the fountain determines the subsequent paths. The exit probability of the particle in the fountain determines the termination of the simulation for each particle. In the batch operation, all particles have the same residence time in the bed, and hence, the probabilities associated with the feed location and with the particles exiting no longer apply. Instead, the total residence time of the particle indicates the termination of the simulation. The coating amount a particle receives in each cycle can be calculated in the same way as a particle in the continuous operation; therefore, the only additional information required to determine the coating distribution for the batch product is the cycle time.

The cycle time for the particle is calculated from Equation (4.52) and the total residence time from Equation (4.55). The simulation ends when the total cycle time is equal to the total operating time.

A correction to the above equation is necessary for the first cycle. In the first cycle or at the start of a coating operation, bed particles could be located anywhere in the bed. To determine the location of each particle, the Monte Carlo method is applied. First, the vertical position of the particle in the annulus is determined using a random number (N_p) :

$$N_R = (V_{tc0} - A_s h_i) / V_{tot}, \text{ for conical section}$$
(I-31)

$$N_R = (V_c + (h_i - h_c)A - A_s h_i) / V_{tot}, \text{ for cylindrical section}$$
(I-32)

where V_{tc0} = bed volume in the conical section; function of h_i

 A_s = area of the spout

 V_{tot} = total bed volume

 V_c = total bed volume in conical section

A = bed surface area in the cylindrical section

 $h_c = \text{cone height}$

The radial position (D_i) at h_i is determined using another random number,

$$N_R = (D_i^2 - D_s^2) / (D^2 \{h_i\} - D_s^2)$$
(I-33)

where $D\{z\}$ = bed diameter at h_i

 $D_s =$ spout diameter

The location where the particle enters the spout (h_s) can be calculated from the following particle mass balance which assumes no radial velocity gradient in the annulus:

$$v_a(1-\varepsilon_a)A_a\big|_{h_i} = v_s(1-\varepsilon_s)A_s\big|_{z_s}$$
(I-34)

where v_a = particle velocity in the annulus at h_i

 A_a = annulus area at h_i

 ε_{a} , ε_{s} = voidages in the annulus and the spout, respectively

 v_s = particle velocity in the spout at z_s

The value of z_s can be determined from a root finding method. The values v_a and A_a are given by

$$v_a = (1 - \varepsilon_s) v_s A_s \big|_{h_i} \big/ (1 - \varepsilon_a) A_a \big|_{h_i}$$
(I-35)

and $A_a = \frac{\pi}{4} (D_b + 2h_i \tan 30^\circ)$, for conical section (I-36)

$$A_{a} = \frac{\pi}{4}D^{2}$$
, for cylindrical section (I-37)

where $D_b =$ bed diameter at the base of the bed

D = bed diameter of the cylindrical section of the bed

The cycle time can then be calculated from Equation (4.52) with h_i and z_s as the limits. For subsequent cycles, the cycle time can be calculated using the same equation with the limits of H and z_s .

I-6. Minimum Spouting Velocity Predictions by Wan-Fyong et al. Equation

The Wan-Fyong et al. (1969) equation was correlated using their conical (plus short cylinder) bed data obtained under the following operating conditions: cone angle = 10 - 70°; d_i = 26 - 76 mm; H = 70 - 300 mm; $d_p = 0.35 - 4$ mm; $\rho_s = 450 - 1390$ kg/m³; $\rho_b = 200 - 790$ kg/m³. For 60° conical bed, their equation becomes

$$(U_i)_{ms} = 0.748 U_t (H/d_i)^{0.82}$$
 (I-38)

where $U_i = 4Q_{ms} / \pi d_i^2$ and U_t is the average terminal velocity of bed particles. The agreement between this equation and the experimental data for the conical bed was good using the corrected d_i' (see Figure I-4); however, it suggests $H^{0.82}$ and $d_i^{1.18}$ relationship with Q_{ms} (in this work $H^{0.96}$ and $d_i^{0.37}$ relationship was found with Q_{ms}).



Figure I-4: Minimum spouting velocity predictions using Wan-Fyong et al. (1969) equation.
Appendix II:

Experimental Data and Calculated Results

This section includes measured data and calculated results that are too lengthy to be in the main body of this thesis. All data are converted to SI units and tabulated in the following order:

Table II-1: Minimum spouting velocity data.

Table II-2: Spray drop size distribution.

Table II-3: Measured sulfur content of individual particles for selected continuous runs.

Table II-4: Predicted sulfur content of individual particles for selected continuous runs.

Table II-5: Measured sulfur content of individual particles for batch runs shown in

Figures 4.3 and 5.25.

<i>d_p</i> , mm	$ ho_p$, kg/m ³	<i>d_i</i> , mm	<i>Q</i> _{<i>a</i>} , L/s	<i>Q_s</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , °C
2.33	1471	25.1	0.000	20.8	0.255	0.24	18
2.33	1471	25.1	0.447	20.5	0.255	0.24	18
2.33	1471	25.1	0.878	20.1	0.255	0.24	18
2.33	1471	29.9	0.000	22.5	0.255	0.24	18
2.33	1471	29.9	0.447	22.1	0.255	0.24	18
2.33	1471	29.9	0.878	21.6	0.255	0.24	18
2.33	1471	29.9	0.447	21.3	0.255	0.24	55
2.33	1471	29.9	0.878	20.3	0.255	0.24	55
2.33	1471	29.9	0.000	22.0	0.255	0.24	55
2.33	1471	25.1	0.447	19.7	0.255	0.24	58
2.33	1471	25.1	0.878	18.9	0.255	0.24	58
2.33	1471	25.1	0.000	20.2	0.255	0.24	58
2.33	1471	29.9	0.447	21.3	0.255	0.24	71
2.33	1471	29.9	0.878	20.3	0.255	0.24	71
2.33	1471	29.9	0.000	21.6	0.255	0.24	71
2.33	1471	29.9	0.447	24.7	0.360	0.24	55
2.33	1471	29.9	0.878	24.1	0.360	0.24	55
2.33	1471	29.9	0.000	25.2	0.360	0.24	55
2.33	1471	25.1	0.447	22.9	0.360	0.24	55
2.33	1471	25.1	0.878	22.1	0.360	0.24	55
2.33	1471	25.1	0.000	23.2	0.360	0.24	55
2.36	1490	29.9	0.447	21.7	0.255	0.24	62
2.36	1490	29.9	0.878	20.6	0.255	0.24	62
2.36	1490	29.9	0.000	22.3	0.255	0.24	62
2.36	1490	25.1	0.447	20.3	0.255	0.24	62
2.36	1490	25.1	0.878	19.5	0.255	0.24	62
2.36	1490	25.1	0.000	20.8	0.255	0.24	62
2.27	1427	29.9	0.447	20.9	0.260	0.24	61
2.27	1427	29.9	0.878	19.9	0.260	0.24	61
2.27	1427	29.9	0.000	21.2	0.260	0.24	61
2.27	1427	25.1	0.447	19.1	0.260	0.24	61
2.27	1427	25.1	0.878	18.3	0.260	0.24	61
2.27	1427	25.1	0.000	19.4	0.260	0.24	61
2.16	1335	29.9	0.000	20.1	0.270	0.24	20
2.16	1335	29.9	0.447	19.5	0.270	0.24	20
2.16	1335	29.9	0.878	18.7	0.270	0.24	20
2.16	1335	25.1	0.000	18.6	0.270	0.24	20
2.16	1335	25.1	0.447	18.3	0.270	0.24	20
2.16	1335	25.1	0.878	17.7	0.270	0.24	20

 Table II-1: Minimum spouting velocity data.

d _p , mm	ρ_p , kg/m ³	<i>d</i> _{<i>i</i>} , mm	<i>Qa</i> , L/s	<i>Qs</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , °C
2.16	1335	29.9	0.000	23.9	0.405	0.24	
2.16	1335	29.9	0.447	23.6	0.405	0.24	20
2.16	1335	29.9	0.878	23.3	0.405	0.24	20
2.16	1335	25.1	0.000	22.0	0.405	0.24	20
2.16	1335	25.1	0.447	21.6	0.405	0.24	20
2.16	1335	25.1	0.878	21.2	0.405	0.24	20
2.16	1335	29.9	0.000	18.7	0.105	0.24	20 60
2.16	1335	29.9	0.447	18.3	0 270	0.24	60
2.16	1335	29.9	0.878	17.6	0 270	0.24	60
2.16	1335	25.1	0.000	17.5	0.270	0.24	60
2.16	1335	25.1	0.447	16.8	0.270	0.21	60
2.16	1335	25.1	0.878	16.0	0.270	0.24	60
2.16	1335	29.9	0.000	22.6	0.405	0.24	60
2.16	1335	29.9	0.447	22.3	0.405	0.24	60
2.16	1335	29.9	0.878	21.6	0.405	0.24	60
2.16	1335	25.1	0.000	20.2	0.405	0.24	60
2.16	1335	25.1	0.447	19.8	0.405	0.24	60
2.16	1335	25.1	0.878	19.2	0.405	0.24	60
2.8	927	24.7	0.000	24.2	0.297	0.45	24
2.8	927	28.2	0.000	25.1	0.297	0.45	24
2.8	927	30.8	0.000	25.3	0.297	0.45	24
2.8	927	35.1	0.000	26.9	0.297	0.45	24
2.8	927	24.7	0.528	23.7	0.297	0.45	24
2.8	927	28.2	0.528	24.5	0.297	0.45	24
2.8	927	30.8	0.528	25.0	0.297	0.45	24
2.8	927	35.1	0.528	26.5	0.297	0.45	24
2.8	927	24.7	0.868	23.4	0.297	0.45	24
2.8	927	28.2	0.868	24.0	0.297	0.45	24
2.8	927	30.8	0.868	24.3	0.297	0.45	24
2.8	927	35.1	0.868	26.0	0.297	0.45	24
2.8	927	21.2	0.000	20.2	0.249	0.45	24
2.8	927	24.7	0.000	21.3	0.249	0.45	24
2.8	927	28.2	0.000	22.1	0.249	0.45	24
2.8	927	30.8	0.000	22.7	0.249	0.45	24
2.8	927	35.1	0.000	24.0	0.249	0.45	24
2.8	927	21.2	0.528	19.6	0.249	0.45	24
2.8	927	24.7	0.528	20.8	0.249	0.45	24
2.8	927	28.2	0.528	21.8	0.249	0.45	24
2.8	927	30.8	0.528	22.1	0.249	0.45	24

Table II-1 continued.

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<i>dp</i> , mm	$ ho_p$, kg/m ³	<i>d</i> _{<i>i</i>} , mm	<i>Qa</i> , L/s	<i>Q_S</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , °C
2.8	927	35.1	0.528	23.5	0.249	0.45	24
2.8	927	21.2	0.868	19.4	0.249	0.45	24
2.8	927	24.7	0.868	20.3	0.249	0.45	24
2.8	927	28.2	0.868	21.2	0.249	0.45	24
2.8	927	30.8	0.868	21.5	0.249	0.45	24
2.8	927	35.1	0.868	22.5	0.249	0.45	24
2.8	927	21.2	0.000	23.6	0.304	0.45	60
2.8	927	24.7	0.000	24.9	0.304	0.45	60
2.8	927	28.2	0.000	25.6	0.304	0.45	60
2.8	927	30.8	0.000	26.0	0.304	0.45	60
2.8	927	35.1	0.000	27.6	0.304	0.45	60
2.8	927	21.2	0.528	23.5	0.304	0.45	60
2.8	927	24.7	0.528	24.5	0.304	0.45	60
2.8	927	28.2	0.528	25.2	0.304	0.45	60
2.8	927	30.8	0.528	25.8	0.304	0.45	60
2.8	927	35.1	0.528	26.9	0.304	0.45	60
2.8	927	21.2	0.868	23.1	0.304	0.45	60
2.8	927	24.7	0.868	24.2	0.304	0.45	60
2.8	927	28.2	0.868	24.8	0.304	0.45	60
2.8	927	30.8	0.868	25.2	0.304	0.45	60
2.8	927	35.1	0.868	26.0	0.304	0.45	60
2.8	927	21.2	0.000	19.6	0.241	0.45	60
2.8	927	24.7	0.000	20.5	0.241	0.45	60
2.8	927	28.2	0.000	21.1	0.241	0.45	60
2.8	927	30.8	0.000	21.7	0.241	0.45	60
2.8	927	35.1	0.000	22.6	0.241	0.45	60
2.8	927	21.2	0.528	19.3	0.241	0.45	60
2.8	927	24.7	0.528	20.2	0.241	0.45	60
2.8	927	28.2	0.528	20.6	0.241	0.45	60
2.8	927	30.8	0.528	21.2	0.241	0.45	60
2.8	927	35.1	0.528	22.2	0.241	0.45	60
2.8	927	21.2	0.868	18.7	0.241	0.45	60
2.8	927	24.7	0.868	19.5	0.241	0.45	60
2.8	927	28.2	0.868	19.8	0.241	0.45	60
2.8	927	30.8	0.868	20.2	0.241	0.45	60
2.8	927	35.1	0.868	21.5	0.241	0.45	60
2.7	1385	21.2	0.000	23.0	0.245	0.45	22
2.7	1385	24.7	0.000	24.9	0.245	0.45	22
2.7	1385	28.2	0.000	26.3	0.245	0.45	22

<i>d</i> _{<i>p</i>} , mm	ρ_p , kg/m ³	d _i , mm	<i>Qa</i> , L/s	<i>Q_S</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , °C
2.7	1385	30.8	0.000	26.6	0 245	0.45	
2.7	1385	35.1	0.000	28.0	0.245	0.45	22
2.7	1385	21.2	0.528	20.1	0.245	0.45	22
2.7	1385	24.7	0.528	24.4	0.245	0.45	22
2.7	1385	28.2	0.528	25.8	0.245	0.45	22
2.7	1385	30.8	0.528	26.3	0.245	0.45	22
2.7	1385	35.1	0.528	27.8	0.245	0.45	22
2.7	1385	21.2	0.868	22.2	0.245	0.45	22
2.7	1385	24.7	0.868	23.8	0.245	0.45	22
2.7	1385	28.2	0.868	24.9	0.245	0.45	22
2.7	1385	30.8	0.868	25.5	0.245	0.45	22
2.7	1385	35.1	0.868	26.6	0.245	0.45	22
2.7	1385	21.2	0.000	27.7	0.289	0.45	22
2.7	1385	24.7	0.000	28.6	0.289	0.45	22
2.7	1385	28.2	0.000	29.5	0.289	0.45	22
2.7	1385	30.8	0.000	30.2	0.289	0.45	22
2.7	1385	35.1	0.000	31.6	0.289	0.45	22
2.7	1385	21.2	0.528	27.4	0.289	0.45	22
2.7	1385	24.7	0.528	28.3	0.289	0.45	22
2.7	1385	28.2	0.528	29.1	0.289	0.45	22
2.7	1385	30.8	0.528	29.8	0.289	0.45	22
2.7	1385	35.1	0.528	31.2	0.289	0.45	22
2.7	1385	21.2	0.868	27.0	0.289	0.45	22
2.7	1385	24.7	0.868	28.0	0.289	0.45	22
2.7	1385	28.2	0.868	28.7	0.289	0.45	22
2.7	1385	30.8	0.868	29.3	0.289	0.45	22
2.7	1385	35.1	0.868	30.8	0.289	0.45	22
2.7	1385	21.2	0.000	24.2	0.243	0.45	65
2.7	1385	24.7	0.000	25.4	0.243	0.45	65
2.7	1385	28.2	0.000	26.0	0.243	0.45	65
2.7	1385	30.8	0.000	26.3	0.243	0.45	65
2.7	1385	35.1	0.000	28.5	0.243	0.45	65
2.7	1385	21.2	0.528	23.8	0.243	0.45	65
2.7	1385	24.7	0.528	24.8	0.243	0.45	65
2.1	1385	28.2	0.528	25.7	0.243	0.45	65
2.1	1385	30.8	0.528	26.0	0.243	0.45	65
2.1	1385	35.1	0.528	27.9	0.243	0.45	65
2.1	1385	21.2	0.868	23.2	0.243	0.45	65
2.1	1385	24.7	0.868	24.2	0.243	0.45	65

<i>d_p</i> , mm	$ ho_p$, kg/m ³	<i>d_i</i> , mm	<i>Qa</i> , L/s	<i>Q_s</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , ℃
2.7	1385	28.2	0.868	25.4	0 243	0.45	65
2.7	1385	30.8	0.868	25.4	0.243	0.45	65
2.7	1385	35.1	0.868	26.9	0.243	0.45	65
2.7	1385	21.2	0.000	27.7	0.289	0.45	65
2.7	1385	24.7	0.000	28.5	0.289	0.45	65
2.7	1385	28.2	0.000	29.3	0.289	0.45	65
2.7	1385	30.8	0.000	29.8	0.289	0.45	65
2.7	1385	35.1	0.000	31.8	0.289	0.45	65
2.7	1385	21.2	0.528	27.3	0.289	0.45	65
2.7	1385	24.7	0.528	28.3	0.289	0.45	65
2.7	1385	28.2	0.528	28.8	0.289	0.45	65
2.7	1385	30.8	0.528	29.3	0.289	0.45	65
2.7	1385	35.1	0.528	31.2	0.289	0.45	65
2.7	1385	21.2	0.868	27.0	0.289	0.45	65
2.7	1385	24.7	0.868	27.8	0.289	0.45	65
2.7	1385	28.2	0.868	28.4	0.289	0.45	65
2.7	1385	30.8	0.868	28.8	0.289	0.45	65
2.7	1385	35.1	0.868	30.4	0.289	0.45	65
2.7	1385	24.7	0.000	33.7	0.332	0.45	65
2.7	1385	28.2	0.000	34.9	0.332	0.45	65
2.7	1385	24.7	0.528	33.5	0.332	0.45	65
2.7	1385	28.2	0.528	34.5	0.332	0.45	65
2.7	1385	30.8	0.528	36.7	0.332	0.45	65
2.7	1385	24.7	0.868	33.2	0.332	0.45	65
2.7	1385	28.2	0.868	34.2	0.332	0.45	65
2.7	1385	30.8	0.868	35.6	0.332	0.45	65
2.3	1045	24.7	0.000	22.0	0.293	0.45	65
2.3	1045	28.2	0.000	23.2	0.293	0.45	65
2.3	1045	30.8	0.000	23.8	0.293	0.45	65
2.3	1045	35.1	0.000	26.0	0.293	0.45	65
2.3	1045	24.7	0.528	21.2	0.293	0.45	65
2.3	1045	28.2	0.528	22.6	0.293	0.45	65
2.3	1045	30.8	0.528	23.5	0.293	0.45	65
2.3	1045	35.1	0.528	25.4	0.293	0.45	65
2.3	1045	24.7	0.868	20.6	0.293	0.45	65
2.3	1045	28.2	0.868	21.7	0.293	0.45	65
2.3	1045	30.8	0.868	22.3	0.293	0.45	65
2.3	1045	35.1	0.868	23.8	0.293	0.45	65
2.3	1045	24.7	0.000	26.4	0.341	0.45	65

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d _p , mm	$ ho_p$, kg/m ³	<i>d_i</i> , mm	<i>Qa</i> , L/s	<i>Q_s</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , °C
2.3	1045	28.2	0.000	27.0	0.341	0.45	65
2.3	1045	30.8	0.000	28.0	0.341	0.45	65
2.3	1045	35.1	0.000	30.4	0.341	0.45	65
2.3	1045	24.7	0.528	25.6	0.341	0.45	65
2.3	1045	28.2	0.528	26.7	0.341	0.45	65
2.3	1045	30.8	0.528	27.5	0.341	0.45	65
2.3	1045	35.1	0.528	29.8	0.341	0.45	65
2.3	1045	24.7	0.868	25.0	0.341	0.45	65
2.3	1045	28.2	0.868	26.2	0.341	0.45	65
2.3	1045	30.8	0.868	27.0	0.341	0.45	65
2.3	1045	35.1	0.868	29.0	0.341	0.45	65
2.3	1045	24.7	0.000	30.8	0.344	0.45	65
2.3	1045	28.2	0.000	31.6	0.344	0.45	65
2.3	1045	30.8	0.000	33.0	0.344	0.45	65
2.3	1045	35.1	0.000	35.8	0.344	0.45	65
2.3	1045	24.7	0.528	30.3	0.344	0.45	65
2.3	1045	28.2	0.528	32.1	0.344	0.45	65
2.3	1045	30.8	0.528	32.5	0.344	0.45	65
2.3	1045	35.1	0.528	35.3	0.344	0.45	65
2.3	1045	24.7	0.868	29.9	0.344	0.45	65
2.3	1045	28.2	0.868	31.6	0.344	0.45	65
2.3	1045	30.8	0.868	31.9	0.344	0.45	65
2.3	1045	35.1	0.868	34.5	0.344	0.45	65
2.7	1385	21.2	0.000	23.7	0.245	0.24	18
2.7	1385	24.7	0.000	25.2	0.245	0.24	18
2.7	1385	28.2	0.000	26.0	0.245	0.24	18
2.7	1385	30.8	0.000	27.1	0.245	0.24	18
2.7	1385	35.1	0.000	28.1	0.245	0.24	18
2.7	1385	21.2	0.357	23.3	0.245	0.24	18
2.7	1385	24.7	0.357	24.9	0.245	0.24	18
2.7	1385	28.2	0.357	25.8	0.245	0.24	18
2.7	1385	30.8	0.357	27.0	0.245	0.24	18
2.7	1385	35.1	0.357	28.1	0.245	0.24	18
2.7	1385	21.2	0.528	23.2	0.245	0.24	18
2.7	1385	24.7	0.528	24.8	0.245	0.24	18
2.7	1385	28.2	0.528	25.7	0.245	0.24	18
2.7	1385	30.8	0.528	26.7	0.245	0.24	18
2.7	1385	35.1	0.528	28.0	0.245	0.24	18
2.7	1385	21.2	0.698	23.0	0.245	0.24	18

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T٤	able	II-1	continued.

<i>d</i> _{<i>p</i>} , mm	$ ho_{p}$, kg/m ³	<i>d</i> _{<i>i</i>} , mm	<i>Qa</i> , L/s	<i>Q_S</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , °C
2.7	1385	24.7	0.698	24.6	0.245	0.24	18
2.7	1385	28.2	0.698	25.4	0.245	0.24	18
2.7	1385	30.8	0.698	26.5	0.245	0.24	18
2.7	1385	35.1	0.698	27.7	0.245	0.24	18
2.7	1385	21.2	0.868	22.8	0.245	0.24	18
2.7	1385	24.7	0.868	24.5	0.245	0.24	18
2.7	1385	28.2	0.868	25.3	0.245	0.24	18
2.7	1385	30.8	0.868	26.3	0.245	0.24	18
2.7	1385	35.1	0.868	27.5	0.245	0.24	18
2.7	1385	21.2	0.000	23.6	0.245	0.24	65
2.7	1385	24.7	0.000	25.3	0.245	0.24	65
2.7	1385	28.2	0.000	26.2	0.245	0.24	65
2.7	1385	30.8	0.000	27.1	0.245	0.24	65
2.7	1385	35.1	0.000	28.8	0.245	0.24	65
2.7	1385	21.2	0.357	23.5	0.245	0.24	65
2.7	1385	24.7	0.357	25.0	0.245	0.24	65
2.7	1385	28.2	0.357	25.9	0.245	0.24	65
2.7	1385	30.8	0.357	26.8	0.245	0.24	65
2.7	1385	35.1	0.357	28.5	0.245	0.24	65
2.7	1385	21.2	0.528	23.3	0.245	0.24	65
2.7	1385	24.7	0.528	24.9	0.245	0.24	65
2.7	1385	28.2	0.528	25.8	0.245	0.24	65
2.7	1385	30.8	0.528	26.7	0.245	0.24	65
2.7	1385	35.1	0.528	28.4	0.245	0.24	65
2.7	1385	21.2	0.698	23.1	0.245	0.24	65
2.7	1385	24.7	0.698	24.7	0.245	0.24	65
2.7	1385	28.2	0.698	25.6	0.245	0.24	65
2.7	1385	30.8	0.698	26.2	0.245	0.24	65
2.7	1385	35.1	0.698	28.0	0.245	0.24	65
2.7	1385	21.2	0.868	22.8	0.245	0.24	65
2.7	1385	24.7	0.868	24.4	0.245	0.24	65
2.7	1385	28.2	0.868	25.3	0.245	0.24	65
2.7	1385	30.8	0.868	25.8	0.245	0.24	65
2.7	1385	35.1	0.868	27.7	0.245	0.24	65
2.7	1385	30.8	0.000	28.7	0.280	0.24	65
2.7	1385	30.8	0.528	28.3	0.280	0.24	65
2.7	1385	30.8	0.528	30.9	0.330	0.24	65
2.7	1385	30.8	0.528	32.1	0.370	0.24	65
2.7	1385	30.8	0.528	33.7	0.430	0.24	65

<i>d</i> _{<i>p</i>} , mm	$ ho_p$, kg/m ³	<i>d</i> _i , mm	<i>Qa</i> , L/s	<i>Q_s</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , °C
2.8	927	21.2	0.000	21.0	0 265	0.24	65
2.8	927	24.7	0.000	22.7	0.265	0.24	65
2.8	927	28.2	0.000	23.3	0 265	0.24	65
2.8	927	30.8	0.000	24.4	0.265	0.24	65
2.8	927	35.1	0.000	26.1	0.265	0.24	65
2.8	927	21.2	0.357	20.8	0.265	0.24	65
2.8	927	24.7	0.357	22.4	0.265	0.24	65
2.8	927	28.2	0.357	23.0	0.265	0.24	65
2.8	927	30.8	0.357	24.1	0.265	0.24	65
2.8	927	35.1	0.357	25.8	0.265	0.24	65
2.8	927	21.2	0.528	20.6	0.265	0.24	65
2.8	927	24.7	0.528	22.0	0.265	0.24	65
2.8	927	28.2	0.528	22.8	0.265	0.24	65
2.8	927	30.8	0.528	23.8	0.265	0.24	65
2.8	927	35.1	0.528	25.6	0.265	0.24	65
2.8	927	21.2	0.698	20.6	0.265	0.24	65
2.8	927	24.7	0.698	21.7	0.265	0.24	65
2.8	927	28.2	0.698	22.5	0.265	0.24	65
2.8	927	30.8	0.698	23.3	0.265	0.24	65
2.8	927	35.1	0.698	25.5	0.265	0.24	65
2.8	927	21.2	0.868	20.3	0.265	0.24	65
2.8	927	24.7	0.868	21.4	0.265	0.24	65
2.8	927	28.2	0.868	22.2	0.265	0.24	65
2.8	927	30.8	0.868	22.6	0.265	0.24	65
2.8	927	35.1	0.868	25.0	0.265	0.24	65
2.3	1045	21.2	0.000	20.8	0.275	0.24	65
2.3	1045	24.7	0.000	21.3	0.275	0.24	65
2.3	1045	28.2	0.000	21.8	0.275	0.24	65
2.3	1045	30.8	0.000	22.5	0.275	0.24	65
2.3	1045	35.1	0.000	24.3	0.275	0.24	65
2.3	1045	21.2	0.357	20.4	0.275	0.24	65
2.3	1045	24.7	0.357	21.1	0.275	0.24	65
2.3	1045	28.2	0.357	21.5	0.275	0.24	65
2.3	1045	30.8	0.357	· 22.0	0.275	0.24	65
2.3	1045	35.1	0.357	23.7	0.275	0.24	65
2.3	1045	21.2	0.528	20.4	0.275	0.24	65
2.3	1045	24.7	0.528	21.0	0.275	0.24	65
2.3	1045	28.2	0.528	21.3	0.275	0.24	65
2.3	1045	30.8	0.528	21.7	0.275	0.24	65

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<i>d_p</i> , mm	$ ho_{p}$, kg/m ³	<i>d_i</i> , mm	<i>Qa</i> , L/s	<i>Q_s</i> , L/s	<i>H</i> , m	<i>D</i> , m	<i>T</i> , °C
2.3	1045	35.1	0.528	23.3	0.275	0.24	65
2.3	1045	21.2	0.698	20.1	0.275	0.24	65
2.3	1045	24.7	0.698	20.7	0.275	0.24	65
2.3	1045	28.2	0.698	20.8	0.275	0.24	65
2.3	1045	30.8	0.698	21.4	0.275	0.24	65
2.3	1045	35.1	0.698	22.8	0.275	0.24	65
2.3	1045	21.2	0.868	19.6	0.275	0.24	65
2.3	1045	24.7	0.868	20.2	0.275	0.24	65
2.3	1045	28.2	0.868	20.4	0.275	0.24	65
2.3	1045	30.8	0.868	21.0	0.275	0.24	65
2.3	1045	35.1	0.868	22.4	0.275	0.24	65
2.3	1045	21.2	0.528	21.6	0.275	0.24	65
2.3	1045	24.7	0.528	22.4	0.275	0.24	65
2.3	1045	28.2	0.528	22.5	0.275	0.24	65
2.3	1045	30.8	0.528	23.8	0.275	0.24	65
2.3	1045	35.1	0.528	25.2	0.275	0.24	65

Table II-1 continued.

.

<i>d_s</i> \Run #	1a	1b	lc	2a	2b	3	4	5	6	7	8
2.93	7	2	0	7	0	5	0	7	0	1	0
8.79	15	3	16	41	16	57	62	21	29	12	8
14.65	60	20	41	69	40	76	40	20	23	20	40
20.51	58	32	17	50	37	46	33	12	7	10	22
26.37	48	10	9	21	11	23	19	6	4	3	41
32.23	18	6	7	12	7	11	11	5	3	2	34
38.09	13	3	11	5	11	4	13	1	3	3	29
43.95	6	4	5	5	4	4	14	5	2	0	30
49.8	4	4	2	1	3	5	11	4	0	2	16
55.66	3	5	5	2	3	2	3	1	0	1	17
61.52	3	2	2	0	4	4	4	1	0	0	12
67.38	2	2	2	2	1	1	8	1	0	1	7
73.24	2	1	1	1	0	2	0	0	0	1	5
79.1	0	1	0	0	0	2	3	1	0	0	7
84.96	3	1	0	1	0	3	1	0	0	1	2
90.82	2	0	1	0	0	2	2	0	0	0	3
96.68	2	2	1	0	1	0	0	0	0	0	2
102.54	2	0	0	1	0	1	1	0	0	0	1
108.4	1	0	0	1	0	2	1	0	0	1	1
114.26	1	1	0	1	1	0	1	0	1	0	0
120.12	0	0	0	0	0	0	2	0	0	0	0
125.98	1	1	0	1	0	0	0	0	0	0	3
131.84	0	0	0	0	0	0	0	0	0	0	1
137.7	0	0	0	0	0	0	0	1	0	0	0
143.55	0	0	1	0	0	0	0	0	0	0	2
149.41	0	0	0	0	0	1	0	1	0	0	0
>150	3	1	2	2	1	0	0	0	1	0	4
Total (N)	254	101	123	223	140	251	229	87	73	58	287

Table II-2: Spray drop size distribution.

<i>ds</i> \Run #	9	10	11	12	13	14	14a	14b	14c	15	16
2.93	0	0	0	0	0	23	0	0	0	0	2
8.79	57	47	48	53	103	71	165	17	306	28	49
14.65	37	18	88	33	91	40	216	23	179	29	34
20.51	14	10	49	20	57	24	108	6	92	20	12
26.37	5	6	42	12	42	17	57	8	44	18	6
32.23	8	7	33	15	22	8	41	3	21	17	8
38.09	9	3	22	13	29	5	27	5	18	8	4
43.95	13	3	21	11	19	6	27	5	9	6	4
49.8	12	3	14	8	19	6	22	1	15	0	1
55.66	4	0	12	7	11	4	6	4	12	5	1
61.52	1	2	2	3	10	6	6	0	10	2	1
67.38	3	0	8	4	6	1	10	2	2	0	0
73.24	1	2	6	1	5	3	1	0	3	0	1
79.1	4	0	3	0	2	1	4	0	4	1	0
84.96	1	1	0	1	2	2	1	2	4	1	1
90.82	5	1	2	0	3	2	0	1	4	1	0
96.68	0	0	3	0	1	2	2	0	1	0	1
102.54	4	0	0	1	1	3	2	0	4	0	0
108.4	1	1	0	3	3	3	0	0	4	1	0
114.26	1	0	2	2	0	2	1	0	0	0	0
120.12	1	2	0	0	2	0	0	0	0	0	0
125.98	0	1	0	0	0	1	0	0	0	0	0
131.84	1	0	0	0	0	0	0	1	1	0	0
137.7	0	1	0	1	0	1	0	0	3	0	0
143.55	0	1	0	0	0	1	0	0	0	0	0
149.41	0	0	0	1	0	0	0	0	3	0	0
>150	2	2	1	5	2	4	0	2	5	3	3
Total (N)	184	111	356	194	430	236	696	80	744	140	128

Table II-2 continued.

Run: 17a	17b	19	24	23	28	29	38	39
0.0	0.0	0.0	0.0	0.8	0.0	0.0	0.0	0.0
0.0	0.0	0.0	0.0	1.4	0.0	0.0	0.0	0.0
0.2	3.5	0.0	0.0	1.5	0.0	0.0	0.3	0.0
1.3	3.5	0.0	2.2	3.0	0.9	0.6	1.0	0.1
1.7	4.9	6.2	4.7	6.4	2.5	3.7	1.0	0.9
2.2	5.0	10.6	5.7	6.8	10.0	4.9	3.2	2.2
2.5	5.5	12.1	8.5	9.4	12.5	5.6	4.2	3.4
6.0	7.0	13.9	15.3	11.2	13.5	6.0	4.2	4.4
7.3	8.9	14.5	16.3	12.2	14.6	9.0	8.3	6.0
7.7	11.1	16.5	16.7	13.8	14.7	10.9	9.1	7.7
11.9	11.4	17.5	17.2	14.0	15.2	12.0	9.6	8.5
15.2	12.6	23.2	18.9	17.3	15.4	13.3	11.2	8.7
16.5	12.7	25.3	19.4	20.6	17.5	13.5	11.6	10.0
19.4	14.4	32.3	19.6	23.1	17.7	13.6	11.7	10.6
19.7	15.3	32.6	19.7	23.3	17.9	13.9	12.6	10.8
20.5	15.8	33.9	21.9	23.9	18.4	16.1	13.0	11.5
20.6	19.4	34.6	23.6	25.7	18.7	16.6	13.4	13.4
20.6	20.1	· 34.8	23.7	27.6	19.3	17.5	16.5	14.0
21.5	21.0	36.5	25.1	29.7	19.4	19.2	17.1	14.1
22.0	22.7	36.7	25.7	30.1	20.6	19.8	17.2	17.1
24.0	24.2	37.8	26.0	32.0	21.1	19.9	19.2	17.4
25.0	26.7	38.3	26.2	34.3	21.7	21.5	19.4	17.7
26.3	28.8	38.7	28.9	35.1	22.1	23.3	20.1	18.0
28.6	29.4	39.0	30.9	37.6	22.6	23.5	20.9	19.0
29.5	29.5	40.2	32.1	38.8	22.9	23.9	21.9	19.1
29.8	30.5	40.8	32.2	38.8	23.0	26.6	22.2	19.7
30.8	32.0	41.2	34.7	40.0	23.7	29.1	24.7	22.3
32.4	32.9	41.4	35.2	42.8	26.0	29.5	26.6	24.2
33.3	33.3	42.7	35.5	48.3	26.1	32.3	26.8	24.3
35.3	34.9	42.7	37.5	49.0	26.8	32.4	27.1	24.8
36.0	35.7	43.7	37.8	49.9	27.4	33.1	31.3	26.9
39.0	36.1	43.8	37.9	50.6	27.5	33.5	31.6	27.0
40.1	36.8	45.2	39.4	52.5	27.9	33.9	31.8	28.0
41.4	37.0	46.9	40.3	55.8	28.5	33.9	34.5	28.3
42.5	38.6	47.7	40.6	58.6	29.2	34.2	35.6	31.0
43.4	38.9	48.2	41.3	65.4	30.6	35.1	35.9	34.0
44.2	39.3	48.7	44.8	65.5	32.9	35.7	40.0	34.6
46.4	47.8	53.3	45.2	66.6	35.0	38.0	42.6	36.5
46.9	54.6	56.5	46.4	68.2	40.0	38.1	53.2	41.5
49.6	58.3	58.6	57.4	68.5	50.3	40.7	55.1	47.1

 Table II-3:
 Measured sulfur content of individual particles for selected runs (sulfur content values given in percent).

Run: 17	19	24	23	28	29	38
0.1	0.5	1.1	3.4	0.7	0.2	0.0
3.2	3.7	2.8	4.8	5.0	2.8	0.5
3.6	4.3	4.7	4.8	5.0	2.8	1.9
3.7	7.2	4.7	5.1	5.5	3.0	3.5
4.2	7.7	4.9	5.7	7.1	4.6	3.6
6.8	9.1	5.4	7.0	9.6	5.4	4.4
7.0	11.3	6.8	9.0	10.7	6.0	5.5
8.3	12.5	8.6	9.3	13.2	7.6	6.7
10.1	13.7	9.1	10.2	14.0	8.1	6.9
10.5	15.1	9.6	11.3	15.0	8.7	7.4
11.1	16.3	10.8	13.1	16.7	10.1	8.4
12.5	17.5	12.8	13.5	17.6	10.6	9.7
13.4	18.8	13.2	14.4	18.9	11.4	10.2
14.1	20.0	13.9	16.0	20.0	12.3	11.2
15.2	21.0	15.1	17.1	20.9	13.1	12.0
16.3	22.4	16.6	18.0	21.9	14.1	12.6
17.4	24.0	17.5	19.2	23.5	15.0	13.2
18.6	25.1	18.4	20.5	24.9	15.7	13.8
19.6	26.4	19.6	21.8	26.5	16.4	14.5
20.4	28.1	20.6	22.7	27.7	17.2	15.3
21.4	29.2	21.7	23.8	28.7	18.0	16.4
22.2	30.2	22.7	25.0	30.1	19.2	17.2
23.2	31.6	23.6	26.2	31.5	20.2	17.9
24.3	32.5	24.9	27.3	32.4	21.1	18.5
25.0	33.8	26.2	28.6	33.5	21.9	19.5
26.1	35.1	27.5	29.9	34.6	22.8	20.1
27.3	36.4	28.8	31.4	35.7	23.9	21.3
28.3	37.6	30.2	32.7	36.8	24.7	22.4
29.3	38.5	31.0	34.2	37.7	25.2	23.3
30.4	39.7	32.4	35.3	39.4	26.7	24.0
31.0	41.1	33.3	36.4	40.6	28.5	24.9
32.3	42.2	34.3	37.4	41.6	29.7	26.6
34.3	43.3	35.7	38.9	43.0	30.8	27.8
35.4	44.2	37.0	39.9	44.6	31.9	28.6
36.5	45.7	38.0	41.7	45.7	32.9	29.7
37.5	46.8	39.6	43.0	47.0	34.5	31.1
38.7	48.2	40.3	44.3	48.0	35.3	32.1
39.9	50.1	41.8	45.6	49.7	36.6	33.0
41.4	51.1	43.2	47.0	51.2	37.9	33.6
42.5	52.7	44.1	48.0	52.4	39.3	34.9
43.3	54.4	45.4	49.0	54.2	40.8	36.8

 Table II-4:
 Predicted sulfur content of individual particles for selected continuous runs (sulfur content values given in percent).

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45.2	55.2	46.8	50.7	55.6	43.2	38.3
46.6	56.9	49.2	53.0	57.3	44.9	40.2
49.6	59.5	51.0	54.7	59.0	46.9	41.5
51.7	61.0	52.9	57.4	60.7	48.3	43.0
54.0	63.2	55.1	58.8	63.5	50.7	45.0
56.0	65.2	57.9	60.9	65.8	53.3	47.3
58.7	69.3	61.0	64.3	68.2	56.6	52.0
61.8	72.6	65.8	69.3	71.3	61.7	55.9
75.5	83.9	85.4	87.9	83.5	72.3	66.0

Table II-5:Measured sulfur content of individual particles for batch runs shown in
Figures 4.3 and 5.25 (sulfur content values given in percent).

I	Data given	in Figure 4	.3	Data given in Figure 5.25			
				<u>t =</u>	<u>210 s</u>	<u>t =</u>	<u>480 s</u>
13	20.6	247	10 1	0.7	10.2	3.2	1 2 1
4.5	20.0	24.7	20.2	1.2	10.5	3.2	23.1
11 /	20.7	25.0	20.5	1.2	11.9	3.0	24.0
11.4	20.7	25.1	20.0	1.5	11.9	5.0	24.9
12.1	20.9	25.1	20.0	20	12.5	5.0	25.9
13.1	21.0	25.2	20.0	2.0	12.0	5.7	20.1
15.0	21.0	25.5	20.9	2.2	13.2	71	20.7
16.6	22.0	25.7	29.4	2.0	14.5	87	20.1
17.0	22.0	25.7	29. 4 20.5	3.1	14.5	87	20.5
18.3	22.2	25.0	20.3	3.7	15.0	80	29.0
18.7	22.5	25.9	30.5	30	16.8	10.2	29.2
18.7	22.5	20.0	30.5	10	16.0	12.2	29.0
10.7	22.0	20.0	30.7	54	10.9 17 A	13.5	31.0
10.1	22.0	20.2	31.3	54	17.7	15.4	31.0
10.3	22.0	27.2	31.5	54	17.7	15.0	21.7
10.5	22.0	27.5	31.5	60	17.0	16.6	32.0
19.5	23.0	27.5	32.0		17.0	10.0	32.0
10.5	23.5	27.5	32.0	9 1	17.9	19.5	22.9
19.5	23.5 73.4	27.0	32.2	84	10.2	18.5	310
20.0	23.4	27.0	32.5	0. 4 8.5	19.5	21.2	35.6
20.0	23. 4 24.7	28.0	343	85	21.2	21.2	37.6
20.0	24.7	20.0	34.5	9.5	21.2	22.0	38.5
20.0	24.7	28.0	34.8	9.4 9.4	26.0	22.1	30.5
20.0	24.7	28.1	37.6	10.0	20.5	22.2	30.8
20.5	24.7	28.2	37.6	10.0	33.8	22.9	40.2

Appendix III: Computer Program Listings

The source code listings (written in FORTRAN) and a description of the main computer program used to determine the coating distribution and product quality are given in this section. The numerical procedure, solution methods and definitions of selected variables in the COM-MON statements used in the program are described first. The computer program represents Model IV and includes hydrodynamics, spray distribution and Monte Carlo models.

The procedure for solving Model IV including the solution methods for solving the mass and momentum equations in the spout and the fluid flow in the annulus are given below:

Numerical Procedure

- (i) Read in the experimental data, convert equipment units to metric units using calibration equations, and determine properties of air using equations of state.
- (ii) Calculate bed parameters such as U_{mf} , U_{ms} , v_t and D_s ;
- (iii) Determine the axial pressure distribution $(P_a\{z\})$ using Equations (2.11) and (4.23), and calculate the values of SPLINE parameters (cubic interpolation parameters);
- (iv) Solve for the steam function (ψ) using Equation (I-23), velocities $(u_a\{z,r\})$ using Equation (I-22) and pressures $(P_a\{z,r\})$ in the annulus using Equation (I-28); determine the fluid flow rate across the spout-annulus interface and calculate the values of SPLINE parameters for the exchange flux U_r ;
- (v) Solve Equations (4.27), (4.29), (4.30), (4.36) and (4.37) to determine ε_s , v_s , u_s and C_s , and determine the corresponding values of SPLINE parameters;
- (vi) Calculate m_{sl} at various positions on the surface of the bed (D_r) and the values of SPLINE parameters;

(vii) Determine the particle sulfur content distribution with the Monte Carlo methods outlined in Section 4.2.3.3.

In all steps, the calculated information is stored as SPLINE parameters which provide this information whenever it is required through a simple calculation.

Solution Methods for Steps (iv) and (v)

In step (iv), the vector Ergun (1952) equation was written in terms of the stream function (i.e., Equation (I-23)) which was then expressed in second order finite differences forms and solved using an successive under-relaxation factor of 0.7. The grid size was set in such a way that nodes were located on the conical wall. Twenty-one grids (between D and D_s) were used in the radial direction, and the number of grids in the axial direction was calculated from the number of grids in the radial direction exceeded that in the radial direction. The convergence criterion of of 10⁻⁹ for the maximum change in the stream function value from iteration to iteration was used. (Decreasing this value to 10⁻¹⁰ did not significantly improve the results, but the number of iterations required to meet the criterion nearly doubled). For most runs, convergence was made to find the optimum relaxation factor; however, the rate of convergence appeared to be slower with smaller values, and larger values resulted in no convergence.

The same method was applied for solving the pressure profile in the annulus (i.e., Equation (I-28)), except that linear or first order approximations were used to predict the normal pressure derivative (Equation (I-29)) at the conical wall.

In step (v), the set of non-linear, coupled ordinary differential equations were solved using LSODE (in UBC ODEPACK, Moore, 1989) which utilizes the Gear method (Gerald and Wheatley, 1984). The solution could be obtained in most cases provided the equations were rewritten (and solved) in terms of u_s , $u_s \varepsilon_s$ and $v_s^2(1-\varepsilon_s)$. Absolute and relative tolerance values of 10^{-12} and 10^{-5} , respectively, were used.

Variables in the COMMON statements

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'H.COM1'	contains operating variables, bed constants and properties of bed particles:
AA0	Cross-sectional area of the annulus in the cylindrical section
AAB	Cross-sectional area of the cylindrical portion of the bed above the shutter and below the cone
AR	Archimedes number
AS	Cross-sectional area of the spout
CK1,CK2	Coefficients of the Ergun (1952) equation
DB	Diameter of the cylindrical portion of the bed above the shutter and below the cone
DCOL	D_c
DI	d_i
DIST	d_i
DNI	Diameter of the atomizing air orifice for the air cap of the nozzle
DP	d_p
DPF	ΔP_f ; Epstein and Grace (1984)
DPU	Diameter of urea particles
DS	D_s
ETAC	η
GRAV	g
н	H
HB	Length of the cylindrical portion of the bed above the shutter and below the cone
HCOAT	h _{fds}
ннм	H/H _m
НМ	H_m ; McNab and Bridgwater (1977)
LFEED	Feed type code - forced or arbitrary feed of urea
NDS	Dimension of the arrays used for all SPLINE routines
NPRINT	Print code
PH	P_{H}
PI, PI4	π, π/4
PSI	Sphericity of bed particles
QA	Q_a
QS	\mathcal{Q}_s
QT	Q _t
RHO	ρ

.

RHOAT	Density of atomizing air
RHOP	$ ho_p$
RHOSF	Density of liquid sulfur @ 150 °C
RHOSM	Density of monoclinic sulfur
RHOSR	Density of rhombic sulfur
RHOU	Density of solid urea
Т	T _b
ТА	Temperature of atomizing air
тν	Terminal velocity based on the average bed particle size; Grace (1986)
TAN30	tan(30°)
THETA	Included cone angle
UAHM	$U_{aHm} = 0.9 U_{mf}$, Epstein et al. (1978); Superficial gas velocity at maximum spoutable bed
	depth
UCOL	Superficial air velocity based on the column diameter
UMF	U _{mf} ; Grace (1982)
UMSMG	U_{ms} based on the Mathur-Gishler equation
UNIVC	Universal gas constant
VISC	μ .
VOID	ϵ_{mf}
WMA	Molecular weight of air
WS	W _s
WU	W _u
WTOT	W_{t} Production rate of sulfur coated urea
XSM	\overline{X}_{sm}
xs	\overline{X}_{s}
XUD	U_D ; Seven day dissolution of urea in the product

'H.COM4' includes the following arrays that are used in SPLINE routines:

CS	C _s
cz	Z
ES	E _s
NSP	Dimension of R and Z arrays
PAN	P _a
QAN	Q_{a}

R	Radial distance from the centre of the bed
SMT	Amount of sulfur on a bed particle calculated by the Monte Carlo method
VS	vs
z	Vertical distance from the base of the bed
ZMT	Vertical distance corresponding to SMT

Appendix III: Computer Program Listings

IMPLICIT REAL*8(A-H,O-Z) IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' REAL AAR(6), BB(20,6), AAZ(20), PLX(21, 101) READ(5,*,END=5)NRUN,IDI,R1,RA,RS,FU,TB,IDCOL,IH,XSM,XUD,IC,WTOT COMMON /GRIM/ PLX, AAZ, AAR, BB, NA, NS, IR0, IZ0, NR WRITE(8,*)NRUN, IDI, R1, RA, RS, FU, TB, IDCOL, IH, XSM, XUD, IC, WTOT GOTO 20 WRITE(8,*)' VOID = ', VOID **5 CONTINUE** C CALL PLOTAL WRITE(8,*) ' ** That"s All Folks! **' CALL PRINTO STOP 20 CONTINUE WS = CAL(4,RS,RS)WU = CAL(5,FU,FU)CALL DIPRTY(IH, IDCOL, IDI, IC) WRITE(8,*)'RUN#,H,D,DI,WU,WS,WTOT,XS,XSM,DP=',NRUN,H,DCOL + ,DI,WU,WS,WTOT,XS,XSM,DP C IF(KK.GE.2) CALL PLOTA2 IF(IDCOL.EQ.2) DCOL = DCOL*0.81T = TB + 273. TA = 160. + 273. CALL EOS(T,RHO,VISC) CALL EOS(TA.RHOAT, VISCAT) QA = CAL(3, RA, RHOAT)QS = CAL(1,R1,RHO)QT = QA + QSXUD = XUD/100.PH = 0.WRITE(8,*)'T,TA,RHO,VISC= ',T,TA,RHO,VISC UCOL = QT / PI4/DCOL**2 IF (DI.GT.25.*DP) WRITE(7,*)'DI/DP > 25, UNSTABLE BED' AR = DP**3 * (RHOP-RHO) * GRAV * RHO / VISC**2 HM = DCOL**2 / DP * (DCOL/DI)**(2./3.) * 700. / AR * + ((1.+35.9E-6*AR)**.5-1.)**2 RHOB = (1.-VOID)*RHOP GU = RHO*UCOL WRITE(8,*)'1111,RHOB,VOID,RHOP=',RHOB,VOID,RHOP DS = 5.61 * GU**0.433 * DCOL**0.583 * VISC**0.133 / (RHOB*RHO*GRAV)**0.283 + WRITE(8,*)'2222, VISC, AR, RHOB, QS= ', VISC, AR, RHOB, QS

CANGLE = 20.

HCOAT = DS/2./DATAN(CANGLE/180.*PI/2.)

SUBROUTINE DATINS

PROGRAM SPCOAT

INCLUDE 'H.COM1'

IF(KK.EQ.20) THEN

NPRINT = NPRINT + 1

CALL PRINTO

CALL DATINS

CALL STREAM

CALL PLOTS1

CALL PLOTMT

GOTO 10

END

CALL INITIA

10 KK = KK + 1

STOP

ENDIF

KK = 0 NPRINT = 0

184

AS = DS**2*PI4 AAB = PI4*(DB*DB-DS*DS)AA0 = PI4*(DCOL**2-DS*DS)PMAX= H * (RHOP-RHO) * (1.-VOID) * GRAV X11 = QA/(.1*.0254)**2 * DI**2*QS CALL SUBUMS(DIST,UMS) CALL SPRAY(QAMIN,QAMAX) C DI = DIST ETAC = DSP**2*RHOSF/18./VISC/DP DPST = DP*(RHO*GRAV*(RHOP-RHO)/VISC**2)**(1./3.) UMFST = DSQRT(27.2**2+.0408*AR)-27.2 ST0 = (RHO**2/VISC/GRAV/(RHOP-RHO))**(1./3.)UMF = UMFST/ST0/DPST WDP = DLOG10(DPST)VTST = 10**(-1.64758 + 2.47864*WDP - 1.09703*WDP**2 + + 0.17129*WDP**3) TV = VTST/ST0UAHM = 0.9 * UMF HHM = H/HMDPPSI=DP*PSI CK1 = 150.*VISC*(1.-VOID)**2/DPPSI**2/VOID**3 CK2 = 1.75*RHO*(1.-VOID)/DPPSI/VOID**3 DPF = -H*(CK1+CK2*UMF)*UMF WRITE(7,65)NRUN, DP, RHOP, RHO, QT, QA, H, DCOL, DI, VOID, PSI C WRITE(11,66)RUNNO(1:8),PRTYPE(1:12),QT,QA,H,DCOL,DI,RHO WRITE(7,67)WS,WU,DSP,QAMIN,QAMAX,DIST,XS,XSM,XUD,UMS WRITE(7,70)DS,UCOL,UMF,UAHM,TV,HM,DPF,PMAX,CK1,CK2 65 FORMAT('1'/' ** Run # ',12/' Dp = ',F6.4,' Rhop = ',F5.0, + ' Rho = ',F4.2,' Qs = ',F7.5, + ' Qa = ',F7.5/' H = ',F4.2,' D = ',F4.2,' Di = ',F6.4, + ' Void = ',F5.3,' Psi (Sphericity) = ',F4.2) 66 FORMAT(A9,A14,2X,F7.5,F9.5,F6.2,F6.2,F8.4,F6.2) 67 FORMAT(' Ws = ',F7.5,' Wu = ',F7.5,' DSP(Spray) = ',F9.7/ + 'Qamin = ',F7.5,'Qamax = ',F7.5,'Di" = '.F6.4/ + 'Xs = ',F4.3,' Xsm = ',F4.3,' Xud = ',F4.3,' Ums = ',F7.5) 70 FORMAT(' Ds = ',F6.4,' Ucol = ',F5.3,' Umf = ',F5.3, + ' UaHm = ',F5.3/' TV (Term Vel.) = ',F4.2,' Hm = ', + F4.2,' Dpf (dPmf) = ',F7.1,' Pmax = ',F6.1/ + 'CK1 (Ergun) = ',F6.1,' CK2 (Ergun) = ',F6.1,//) RETURN

WRITE(8,*) 'HCOAT, CANG, DS = ', HCOAT, CANGLE, DS

END

SUBROUTINE DIPRTY(IH, IDCOL, IDI, IC) IMPLICIT REAL*8(A-H,O-Z) COMMON /INLETD/ DIX INCLUDE 'H.COM1' IF(IH.EQ.1)THEN H = 0.28ELSEIF(IH.EQ.2) THEN H = 0.36ELSEIF(IH.EQ.3) THEN H = 0.355ELSE WRITE(8,*) ' *** Wrong IH Spec ***' ENDIF IF(IDCOL.EQ.1) THEN DCOL = 0.24ELSEIF(IDCOL.EQ.2) THEN DCOL = 0.45ELSE WRITE(8,*) ' *** Wrong IDCOL Spec ***' ENDIF IF(IC.EQ.1)THEN XS = WS/(WS+WU)ELSEIF(IC.EQ.0) THEN XS = XSMWU = (1.-XS)*WS/XS ELSEIF(IC.EQ.2) THEN WU = WTOT XS = WS/(WS+WU)ELSE WRITE(8,*)'**** Wrong Sulfur Feed Code ****', IC ENDIF WTOT = WS + WU RHOP = 1./((1.-XS)/RHOU+XS/RHOSR)UWT = RHOU*PI/6.*DPU**3 SWT = UWT/(1.-XS)*XSDP = ((UWT+SWT)/RHOP*6./Pi)**(1./3.)IF(IDI.EQ.1) THEN DIX = (15./16.)*2.54/100. ELSEIF(IDI.EQ.2) THEN DIX = (1. + 1./16.)*2.54/100.

ELSEIF(IDI.EQ.3) THEN

Appendix III: Computer Program Listings

DIX = (1. + 3./16.)*2.54/100.ELSEIF(IDI.EQ.4) THEN DIX = (1. + 9./32.)*2.54/100.ELSEIF(IDI.EQ.5) THEN DIX = (1. + 7./16.)*2.54/100.ELSE WRITE(8,*)'***** Wrong DI Spec *****' STOP ENDIF DI = CAL(8,DIX,DIX) END

SUBROUTINE SUBUMS(DIPR, UMS) IMPLICIT REAL*8(A-H,O-Z) EXTERNAL FUMS INCLUDE 'H.COM1' **DIMENSION WORK(80)** X = DI NF = 1SCALE = 1.D10 EPSL = 1.D-10 IC = 1 MI = 500LOG =6 CALL DPOWEL(FM,X,NF,SCALE,EPSL,IC,MI,LOG,WORK,IE,FUMS,*32) GOTO 33 32 CONTINUE WRITE(7,*)'error in POWEL, IE = ',IE **33 CONTINUE** DIPR = X GH = DSQRT(2.*GRAV*H) UMS =13.50*(DP/DCOL)**1.165 *(DIPR/DCOL)**0.3715 + *(DCOL/H)**0.1475*((RHOP-RHO)/RHO)**0.2894*GH WRITE(8,*)'values Of FM,X are ',FM,X END

DOUBLE PRECISION FUNCTION FUMS(X,ND)

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' $VA = QA/PI4/DAT^{*2}$ $AI = PI4^{+}DI^{++}2$ GH = DSQRT(2.*GRAV*H)XX = XIF(X.LE.0.) THEN FUMS = -X*10000.+1000. RETURN ENDIF UMS =13.50*(DP/DCOL)**1.165 *(XX/DCOL)**0.3715 + *(DCOL/H)**0.1475*((RHOP-RHO)/RHO)**0.2894*GH QMS = UMS*PI4*DCOL**2 QTX = QMS+QA VTX = QTX/XX**2/PI4QMS2 = QTX*VTX*AI-(WS*VA+RHOAT*QA*VA)*AI/RHO IF(QMS2.LE.0.)QMS2 = -QMS2*1000.QMS1 = DSQRT(QMS2)FUMS = ((QMS-QMS1)*1000.)**2 RETURN END

SUBROUTINE SPRAY(QAMIN,QAMAX)

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' UAMIN = 13.7893 + 1.6086*WS*3600. UAMAX = 3.881397 + 4.592215*WS*3600. QAMIN = UAMIN*DAT**2*PI4 QAMAX = UAMAX*DAT**2*PI4 DSP = 0.261012*(QA/QAMIN)**1.7015*(WS/RHOSF/QA)**1.8759 RETURN END

```
SUBROUTINE HSPOUT
  IMPLICIT REAL*8(A-H,O-Z)
   EXTERNAL FUNC
  INCLUDE 'H.COM1'
  INCLUDE 'H.COM4'
   DIMENSION Y(4), RTOL(4), ATOL(4), RWORK(80), IWORK(25), IPAK(100)
   DATA RTOL/4*1.D-5/ATOL/4*1.D-12/ITOL.ITASK.ISTATE, IOPT/1.1.1.0/
   DATA LRW, LIW, MF/80, 25, 22/MUA/4/
* Solve the Lefroy Davidson Equations
   U0 = QT/PI4/DI^{*2}
   VS(1) = 0.
   US(1) = U0
   ES(1) = 1.
   CS(1) = WS/PI4/DS**2/U0
   ISTATE = 1
   WRITE(7,102)HB,1.0,U0,0.,CS(1)
 102 FORMAT(//4X,'H',10X,'VOID.',7X,'AIR V.',6X,'PART. V.',3X,
  - 'SULF, C'//5G12.4)
 103 FORMAT(7G12.4)
   X1 = CZ(1)
   N = NSP
  DCOL = 0.445
С
   Y(1) = U0
   Y(2) = U0
   Y(3) = 0.D0
   Y(4) = CS(1)
   DO 200 I=2,N
     X_2 = CZ(I)
     CALL LSODE(FUNC, MUA, Y, X1, X2, ITOL, RTOL, ATOL, ITASK, ISTATE,
             IOPT.RWORK,LRW,IWORK,LIW,JAC,MF)
  1
     IF(ISTATE.NE.2) WRITE(8,*)'****** ISTATE = ',ISTATE
     V8 = 0.00
     VOIDM1 = 1.- Y(1)/Y(2)
     IF(Y(3).GT.0.)V8 = DSQRT(Y(3)/(1.D0-VOIDM1))
     IF(I/2*2.EQ.I)WRITE(7,103)X2,Y(1)/Y(2),Y(2),V8,Y(4)
     IF(X2.GT.H)THEN
       WRITE(8,*)'Z>H, Z = ',X2
       GOTO 200
     ENDIF
     VS(I) = V8
```

```
US(I) = Y(2)
   ES(I) = Y(1)/Y(2)
   CS(1) = Y(4)
   PRINT(1, NPRINT, I) = Y(4)
200 CONTINUE
  PRINT(1, NPRINT, 1) = CS(1)
  WSL = CS(N)*US(N)*ES(N)*AS
  WCIRC = VS(N)*(1.-ES(N))*AS*RHOP
  WPROD = WU/(1.-XS)
  WRITE(7,203)WSL,WSL/WS*100,WCIRC,WPROD
203 FORMAT(/' Sulfur Lost = ',G12.4,' KG/S = ',G12.4,'%'/
 - 'Part Circ Rate = ',G12.4,' KG/S, Prod Rate = ',G12.4)
  CALL MYSPLN(CZ.CS.CQ.CR.CT.N.NDS)
  CALL MYSPLN(CZ,VS,VQ,VR,VT,N,NDS)
  CALL MYSPLN(CZ.ES.EQ.ER.ET.N.NDS)
  CALL MYSPLN(CZ,US,UQ,UR,UT,N,NDS)
   WRITE(8,*)' HSPOUT OK'
  RETURN
  END
```

SUBROUTINE FUNC(M,X,Y,Z)

- * This subroutine provides RHS of the differential equations.
- * Parameters are

ENDIF RETURN END

- * Z = array containing RHS values of differential equations.
- * X = independent variabLE, Z/H
- * Y = array containing dependent variables
- * M = number of differential equations

```
IMPLICIT REAL*8(A-H,O-Z)
   REAL*8 Z(M), Y(M)
   INCLUDE 'H.COM1'
   E = Y(1)/Y(2)
C WRITE(8,1) X,Y(1),Y(2),Y(3),E
C 1 FORMAT('X, U*E, U, V2*E, E = ',5G11.4)
   EM = 1.-E
   VS = 0.
   IF(EM.GT.0)THEN
     VS2 = Y(3)/EM
     VS = 0.
     IF(VS2.GT.0.)VS = DSQRT(Y(3)/EM)
   ENDIF
   IF(E, LE, 0.)E = 0.000001
   DUV = Y(2) - VS
   UV = DABS(DUV)^*DUV
   B = FSI(E, 1)
   BB = B^{\dagger}UV
   DPZ = QAINT(X, 10)
   DQZ = QAINT(X,8)
   Z(1) = -DQZ/AS
   Z(2) = -DPZ/Y(2)/RHO - BB/RHO/Y(1) - Z(1)/E
   Z(3) = BB/RHOP - DPZ/RHOP*EM - EM*(RHOP-RHO)*GRAV/RHOP
   IF(X.GE.HCOAT) THEN
     Z(4) = -(1.5 \times EM \times ETA(DABS(DUV)) \times DABS(DUV)/DP + Z(1))
   +
      Y(4)/Y(1)
     Z(4) = - (4./DS*DQZ+1.5*EM*ETA(DABS(DUV))*DABS(DUV)/DP+Z(1))*
С
C + Y(4)/Y(1)
   ELSE
     Z(4) = -Z(1)*Y(4)/Y(1)
```

DOUBLE PRECISION FUNCTION ETA(X)

* Calculates impaction efficiency

```
IMPLICIT REAL*8(A-H,O-Z)
INCLUDE 'H.COM1'
SS = ETAC*X
IF(SS.LE.0.083D0)THEN
ETA = 0.D0
ELSEIF(SS.GT.0.6D0)THEN
ETA = SS*SS/(SS+0.5D0)**2
ELSE
ETA = 0.036-0.2323*SS+2.422*SS*SS-2.033*SS**3
ENDIF
END
```

DOUBLE PRECISION FUNCTION FSI(X,I)

* FLUID-SOLID interaction term estimation.

- * 1. RICHARDSON-ZAKI EQUATION
- * 2. ERGUN'S EQUATION
- * 3. MODIFIED (BY 4) R-Z EQUATION
- * 4. MORGAN & LITTMAN CORRELATION

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' IF(I.EQ.1)THEN FSI = 0.33*(1.-X)*RHO/DP/X**1.78 ELSEIF(I.EQ.2)THEN FSI = 1.75*(1.-X)*RHO/DP ELSEIF(I.EQ.3)THEN FSI = 0.109*(1.-X)*RHO/DP/X**1.64 ELSEIF(I.EQ.4)THEN FSI = RHO/DP/X*(78.44-633.94*X + 2124.52*X*X - 3772.5*X**3 + 3741.3*X**4 - 1964.27*X**5 + 426.45*X**6) + ELSE WRITE(7,*)'ERROR IN FSI CODE' ENDIF RETURN END

DOUBLE PRECISION FUNCTION CAREA(X)

* This function finds the area of the annulus (cone-base).

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' IF(X.LT.HB)THEN CAREA = AAB ELSE D = DB + 2.*(X-HB)*TAN30 IF(D.LT.DCOL) THEN CAREA = PI4*(D*D-DS*DS) ELSE CAREA = AA0 ENDIF ENDIF RETURN END FUNCTION INTT(X,Y,N,DX)

Determines the interval at which X belongs. Used with Spline
 calculations.

IMPLICIT REAL*8(A-H,O-Z) REAL*8 Y(N) INTT = 1 J = N-1 30 K = (INTT+J)/2 IF(X.LT.Y(K))J=K IF(X.GE.Y(K))INTT=K IF(J.NE.INTT+1)GOTO 30 DX = X - Y(INTT) RETURN END

DOUBLE PRECISION FUNCTION QAINT(X, ICODE) IMPLICIT REAL*8(A-H.O-Z) INCLUDE 'H.COM1' INCLUDE 'H.COM4' IF(ICODE.LT.3)THEN I = INTT(X, DMT, NSP, DX)ELSEIF(ICODE.GE.11) THEN I = INTT(X,Z,NZ,DX)ELSE I = INTT(X,CZ,NSP,DX)ENDIF IF(ICODE.EQ.1)THEN QAINT = ZMT(I)+ZMTQ(I)*DX+ZMTR(I)*DX*DX+ZMTS(I)*DX**3 ELSEIF(ICODE.EQ.2)THEN QAINT = SMT(I)+SMTQ(I)*DX+SMTR(I)*DX*DX+SMTS(I)*DX**3 ELSEIF(ICODE.EQ.3)THEN QAINT = PA(I)+PQ(I)*DX+PR(I)*DX**2+PS(I)*DX**3 С QAINT = CS(I)+CQ(I)*DX+CR(I)*DX**2+CT(I)*DX**3 ELSEIF(ICODE.EQ.4)THEN QAINT = US(I)+UQ(I)*DX+UR(I)*DX**2+UT(I)*DX**3 ELSEIF(ICODE.EQ.5)THEN QAINT = VS(I)+VQ(I)*DX+VR(I)*DX*DX+VT(I)*DX**3ELSEIF(ICODE.EQ.6)THEN QAINT = ES(I)+EQ(I)*DX+ER(I)*DX**2+ET(I)*DX**3 ELSEIF(ICODE.EQ.7)THEN QAINT = QAN(I)+QANQ(I)*DX+QANR(I)*DX**2+QANS(I)*DX**3 ELSEIF(ICODE.EQ.8)THEN QAINT = DQAN(I)+DQANQ(I)*DX+DQANR(I)*DX**2+DQANS(I)*DX**3 ELSEIF(ICODE.EQ.9)THEN QAINT = PAN(I)+PANQ(I)*DX+PANR(I)*DX**2+PANS(I)*DX**3 ELSEIF(ICODE.EQ.10)THEN QAINT = DPAN(I)+DPANQ(I)*DX+DPANR(I)*DX**2+DPANS(I)*DX**3 ELSEIF(ICODE.EQ.11)THEN QAINT = QZA(I)+QZAQ(I)*DX+QZAR(I)*DX**2+QZAS(I)*DX**3ELSEIF(ICODE.EQ.12)THEN QAINT = RAD(I)+RADQ(I)*DX+RADR(I)*DX**2+RADS(I)*DX**3 ELSE WRITE(7,*)' ERROR IN CODE IN QAINT, ICODE = ',ICODE ENDIF RETURN END

DOUBLE PRECISION FUNCTION PMOR(X)

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' AC = RHO*UMF*TV/(RHOP-RHO)/GRAV/DI IF(PSI.NE.1.)AC=AC*PSI*(5.*PSI**3-7.57*PSI**2+4.09*PSI-.516) PHIA = 7.18*(AC-DI/DCOL) + 1.07IF(AC.GT.0.07)WRITE(8,*)' P-MOR NOT VALID, AC>.07 = ',AC XX1 = 1./(X/DCOL+1.)XQT = .1BB1 = 2.*(XX1-2.*XQT)-2.*(1.-XQT)+4.*(1.-XQT)**2/PHIA CC1 = (XX1-2.)*(XX1-2.*XQT) - 4.*(1.-XQT)**2*XX1/PHIA Y1 = -BB1/2, + DSQRT(BB1**2-4.*CC1)/2. C Y2 = -BB1/2. - DSQRT(BB1**2-4.*CC1)/2. $DPF0 = -DPF^*X/H$ PMOR = (1.-Y1)*DPF0RETURN END

SUBROUTINE PLOTA2

IMPLICIT REAL*8(A-H,O-Z) **DIMENSION IPAK(800)** REAL AX(30),P(30),PAV(30) INCLUDE 'H.COM1' CALL DSPDEV('PLOT') CALL NOBRDR CALL AREA2D(6.0,6.0) CALL PHYSOR(1.,2.) CALL THKCRV(0.03) CALL XREVTK CALL YREVTK CALL COMPLX CALL YAXANG(0.) CALL XNAME('Height, m\$',100) CALL YNAME('Pressure in Annulus, kPa\$',100) CALL YTICKS(3) CALL XTICKS(2) C CALL HEADIN('Pressure Profile in Annulus\$',-100,-3,1) CALL GRAF(0, 1, 4, 0, 3, 1.3) CALL SCLPIC(2.) CALL FRAME PMORH = PMOR(H)/1000.AX(1) = 0.P(1) = PMORH - PMOR(0.D0)/1000.DO 30 | = 2,21 $ZZZ = H^{(1-1)/20}$. AX(I) = ZZZP(I) = PMORH - PMOR(ZZZ)/1000.**30 CONTINUE** CALL CURVE(AX,P,21,0) CALL ENDGR(0) CALL ENDPL(0) END

SUBROUTINE PLOTS1

IMPLICIT REAL*8(A-H,O-Z) DIMENSION IPAK(500) REAL AX(20), V(20), VAJ(20), AX2(2) INCLUDE 'H.COM1' CALL DSPDEV('PLOT') CALL NOBRDR CALL AREA2D(6.0,6.0) CALL PHYSOR(1.,2.) CALL THKCRV(0.03) CALL XREVTK CALL YREVTK CALL COMPLX CALL YAXANG(0.) CALL XNAME('Height, m\$',100) CALL YNAME('Air Velocity in Spout, m/s\$',100) CALL YTICKS(3) CALL XTICKS(2) CALL GRAF(0, 1, 36, 0, 10, 50.) CALL FRAME CALL SCLPIC(2.) CALL HSPOUT DX = (H-HB)/20. START = HB DO 20 | = 1,20XXX = START + DX*I AX(I) = XXXV(I) = QAINT(XXX,4)20 CONTINUE CALL CURVE(AX,V,20,0) CALL ENDGR(0) CALL ENDPL(0) CALL DSPDEV('PLOT') CALL NOBRDR CALL AREA2D(6.0,6.0) CALL PHYSOR(1.,2.) CALL THKCRV(0.03) CALL XREVTK CALL YREVTK CALL COMPLX CALL YAXANG(0.)

CALL XNAME('Height, m\$',100) CALL YNAME('Concentration in Spout, kg/m3\$',100) CALL YTICKS(3) CALL XTICKS(2) CALL GRAF(0, 1, 36,0, 05, 15) CALL FRAME CALL SCLPIC(2.) DO 23 I = 1.20 XXX = START + DX*I AX(I) = XXXV(1) = QAINT(XXX,3)23 CONTINUE CALL CURVE(AX,V,20,0) CALL ENDGR(0) CALL ENDPL(0) END

SUBROUTINE STREAM

* This pgm determines the stream functions in the annulus using

- * the boundary condition
- * str = 0 @ wall
- * d(str)/dz = 0 @ annulus surface
- * P = function given by Morgan @ spout-annulus interface

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' INCLUDE 'H.COM4' DIMENSION STR(101, 101), VEL(5, 101, 101), DPMOR(101) DIMENSION RW(101), PMORG(101), PERG(101, 101) REAL X1, X2, Y1, Y2, RSIZ, ZSIZ, XSTEP, YSTEP REAL AAR(6), BB(20,6), AAZ(20), PLX(21,101) COMMON /GRIM/ PLX, AAZ, AAR, BB, NA, NS, IR0, IZ0, NR COMMON WORK(50000) COMMON /FSTRSB/ F1,F2,RX,DELR,DELRZ2,ILIN DATA NRM, EPS, MAXIT/51, 1.D-9, 7000/ |L|N = 1NRM = 20 ALPHA = 0.7ALPHAM=1.-ALPHA ALP2 = ALPHAALP2M = 1. - ALP2F1 = CK1F2 = CK2RS = DS/2.RB = DB/2. R0 = DCOL/2. IF(DCOL.GT.0.24) R0 = R0/0.81 NR = NRM + 1NRP = NR + 1NRMM = NR - 2DELR = (R0-RS)/NRM DELR2 = DELR**2 DELZ = DELR/TAN30 R(1) = RS|Z0 = 1|IR0 = 1IF(RS.GE.RB)THEN Z(1) = (RS-RB)/TAN30 + HB

NZS = 3ELSE IR0 = IR0 + 1RW0 = DELR +RS 2 IF(RW0.GT.RB) THEN Z(1) = (RW0-RB)*TAN30 + HBIF(Z(1).GT.DELR) THEN 4 Z(1) = Z(1) - DELZ|Z0 = |Z0 + 1|IF(Z(1).GT.DELZ) GOTO 4 ENDIF NZS = |Z0 + 1|WRITE(8,*)'HERE DS<DB, IR0 = ',IR0 GOTO 6 ENDIF RW0 = RW0 + DELR IR0 = IR0 + 1GOTO 2 ENDIF 6 CONTINUE IR0M = IR0 - 1 IROMM = IROM - 1IZOM = IZO - 1IZOP = IZO + 1DELRZ = DELR*DELZ DELRZ2 = (DELR/DELZ)**2 NZP = 18 Z0 = Z(1) + DELZ*NZPNZP = NZP + 1IF(Z0.LT.H) GOTO 8 NZ = NZP - 1NZM = NZ - 1NZMM = NZ - 2WRITE(8,*) ' N, NR, NZ, ALPHA, DELR, Z1= ', NRM, NR, NZ, ALPHA, DELR, Z(1) WRITE(8,*) 'DS,DB,F1,F2,H,DCOL= ',DS,DB,F1,F2,H,DCOL PMORH = PMOR(H)PMORG(1) = PMORH - PMOR(Z(1))RW(1) = RB+(Z(1)-HB)*TAN30DO 10 I=2,NZ Z(I) = Z(I-1) + DELZRW(I) = RB+(Z(I)-HB)*TAN30IF(Z(I),LT,HB)RW(I) = RB

IF(RW(I).GT.R0) RW(I) = R0RDIVBM = 1. - 1./RDIVBPMORG(I) = PMORH-PMOR(Z(I))ENDIF **10 CONTINUE** || = |Z0|JJ = IR0DPMOR(1) = (4.*PMORG(2)-PMORG(3)-3.*PMORG(1))/2./DELRDPMOR(NZ) = (PMORG(NZ-2)-4.*PMORG(NZM)+3.*PMORG(NZ))/2./DELR DO 50 I = 1,NZP DO 15 I = 2.NZMDO 40 J = 1. NRP DPMOR(I) = (PMORG(I+1)-PMORG(I-1))/2./DELR STR(I,J) = 0.**15 CONTINUE** PERG(I,J) = PMORG(1)17 FORMAT('I.PMORG,DPMOR = './5G12.5) DO 18 | = 1.NZ С WRITE(8,17)I, PMORG(I), DPMOR(I) || = || + 1|С C 18 CONTINUE JJ = JJ + 1С STOP STR(I,J) = 0.DO 20 J = 2.NR ENDIF IF(J.LT.IR0)R(J) = R(J-1) + DELRDO 30 K = 1, 5 IF(J.GE.IR0)R(J) = RW(IZ0+J-IR0)VEL(K, I, J) = 1.C WRITE(8,*)'R(J),RW,NR = ', R(J),RW(J),(R(J)-RS)/DELR 30 CONTINUE 40 CONTINUE 20 CONTINUE DELZS = H - Z(NZ)STR(I,NR)=0. ZDIVS = DELZS/DELZ **50 CONTINUE** WRITE(8,*)'DELZS, DELZ, DELR = ', DELZS, DELZ, DELR ZDIVSP = 1. + ZDIVS 55 CONTINUE ZDIVSM = 1. - 1./ZDIVS DO 1000 ITER=1, MAXIT DNM = 2.*(1.+DELRZ2)DIFMAX=0. DNMS = 2.*(1.+DELRZ2/ZDIVS) RX = RSE1 = (1.+DELR/2.)/DNME2 = (1.-DELR/2.)/DNME3 = DELRZ2/DNME4 = E1*DNM/DNMS IF(IR0.EQ.1)THEN E5 = E2*DNM/DNMS DR1 = -STR(2,NRP)/2./DELR E6 = 2.*DELRZ2/ZDIVSP/DNMS DR1M = -STR(1,NRP)/DELR IF(IR0.GT.1) THEN DZ1 = STR(3,1)/2./DELZDELZB = Z(1)ZDIVB = DELZB/DELZ ZDIVBP = 1. + ZDIVBIF(IR0.EQ.2)THEN C7 = 2.*C1 - C5DELRB = RB - RSELSE + DELRB = RB - IROM * DELR - RS ENDIF RDIVB = DELRB/DELR STR(2,NRP) = -C0*2.*DELRRDIVBP = 1. + RDIVBVEL(1,2,1) = DR1

IF(R(J).LT.RW(I))STR(I,J) = (RW(I)-R(J))*Z(I)*1.D-3IF(II.EQ.I.AND.JJ.EQ.J)THEN * If the spout diameter is larger than the cone neck DZR = (VEL(1,3,1)-DR1M)/2./DELZ CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C0 = DPMOR(2)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))STRNEW = ALPHAM*STR(2,1)+ALPHA*(C5*STR(2,2) + C7*STR(2,NRP)+C2*DELRZ2* (STR(3,1)+STR(1,1))+C4*DELR2)/2./C6

DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(2,1)))

VEL(2,2,1) = DZ1

STR(2,1) = STRNEW

DZ1 = STR(2,1)/DELZ

STR(2,NRP) = - C0*DELR

* If the spout diameter is less than the cone neck

C0 = DPMOR(1)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1M**2))

ELSEIF(IR0.EQ.2) THEN DR1 = -STR(1,NRP)/2./DELRB + DZ1 = STR(2,1)/2./DELZBDZR = (STR(2,NRP)/2./DELR-DR1+DR1/ZDIVB)/2./DELZ CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C0 = DPMOR(1)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))STRNEW = ALPHAM*STR(1,1)+ALPHA*((C1-C3*DELRB/2.)* + STR(1,NRP)+2.*C2/ZDIVBP*DELRZ2*RDIVB**2*STR(2,1) + + C4*DELRB**2)/2./(C1+C2/ZDIVB*DELRZ2*RDIVB**2) ELSE DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(2,1))) * If the second r node exist, calculate str at I = 1 STR(2,NRP) = -C0*2.*DELRBVEL(1,1,1) = DR1VEL(2,1,1) = DZ1DR1 = (STR(1,2)-STR(1,NRP))/2./DELRSTR(1,1) = STRNEWDZ1 = (STR(2,1)-STR(1,1)+STR(1,1)/ZDIVB)/2./DELZDZR = (VEL(1,2,1)-DR1+DR1/ZDIVB)/2./DELZ IF(IZ0.GT.2) THEN DO 60 I = 2,1Z0M CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C0 = DPMOR(1)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))|P = | + 1IM = I - 1C7 = 2.*C1 - C5DR1 = -STR(I,NRP)/2./DELRB STRNEW = ALPHAM*STR(1,1)+ALPHA*(C5*STR(1,2) + C7*STR(1,NRP)+2.*C2*DELRZ2/ZDIVPB* DZ1 = (STR(IP,1)-STR(IM,1))/2./DELZ+ DZR = (STR(IP,NRP)-STR(IM,NRP))/4./DELRB/DELZ STR(2,1)+C4*DELR2)/2./(C1+C2*DELRZ2/ZDIVB) + CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(1,1))) STR(1,NRP) = STR(1,2) - C0*2.*DELR C0 = DPMOR(I)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))STRNEW = ALPHAM*STR(I,1)+ALPHA*((C1-C3*DELRB/2.)* VEL(1,1,1) = DR1STR(I,NRP)+C2*DELRZ2*RDIVB**2*(STR(IP,1)+STR(IM,1)) VEL(2,1,1) = DZ1+ + + C4*DELRB**2)/2./(C1+C2*DELRZ2*RDIVB**2) STR(1,1) = STRNEWDIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(1,1))) IF(IR0M.LE.2) GOTO 80 STR(I,NRP) = - C0*2.*DELR DO 70 J = 2, IR0MMVEL(1,1,1) = DR1JP = J + 1JM = J - 1VEL(2,1,1) = DZ1DR1 = (STR(1,JP)-STR(1,JM))/2./DELRSTR(I,1) = STRNEWCONTINUE DZ1 = (STR(2,J)-STR(1,J)+STR(1,J)/ZDIVB)/2./DELZ60 ENDIF DZR = (VEL(1,2,J)-DR1+DR1/ZDIVB)/2./DELZ RX = R(J)| = |Z0|

DR1 = -STR(I,NRP)/2./DELR DZ1 = (STR(IP,1)-STR(IM,1))/2./DELZDZR = (VEL(1,IP,1)-VEL(1,IM,1))/2./DELZ CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C0 = DPMOR(I)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(I,1)+ALPHA*(C5*STR(I,2) + C7*STR(I,NRP)+C2*DELRZ2* (STR(IP,1)+STR(IM,1))+C4*DELR2)/2./C6 DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(I,1))) STR(I,NRP) = -C0*2,*DELRVEL(1, 1, 1) = DR1VEL(2,1,1) = DZ1STR(1,1) = STRNEW

IP = 1 + 1

|M = | - 1

Appendix III: Computer Program Listings

CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6)

C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(1,J)+ALPHA*(C5*STR(1,JP) + C7*STR(1,JM)+2.*C2*DELRZ2/ZDIVBP* + STR(2,J)+C4*DELR2)/2./(C1+C2*DELRZ2/ZDIVB) + DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(1,J))) VEL(1,1,J) = DR1JP = J + 1VEL(2,1,J) = DZ1JM = J - 1STR(1,J) = STRNEW70 CONTINUE 80 CONTINUE DR1 = (RDIVBM*STR(1,IR0M)-STR(1,IR0MM))/2./DELR RX = R(J)DZ1 = (STR(2,IR0M)-STR(1,IR0M)+STR(1,IR0M)/ZDIVB)/2./DELZ DZR = (VEL(1,2,IR0M)-DR1+DR1/ZDIVB)/2./DELZ RX = R(IR0M)CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) STRNEW = ALPHAM*STR(1,IR0M)+ALPHA*((2.*C1/ZDIVBP+C3*DELRB/2.) + + *STR(1,IR0MM)+2.*C2*DELRZ2/ZDIVBP* STR(2,IR0M)+C4*DELR2)/2./(C1/RDIVB+C2*DELRZ2/ZDIVB + + -RDIVBM*C3*DELRB/4.) DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(1,IR0M))) VEL(1,1,IR0M) = DR190 VEL(2,1,IR0M) = DZ1100 STR(1,IR0M) = STRNEW * If the second r node exist, calculate str at 1 > I > IZ0 IF(IZ0.EQ.2) GOTO 120 DO 110 I = 2.1Z0MIP = I + 1|M = | - 1|DR1 = (STR(1,2)-STR(1,NRP))/2./DELR+ DZ1 = (STR(IP,1)-STR(IM,1))/2./DELZDZR=(STR(IP,2)-STR(IP,NRP)-STR(IM,2)+STR(IM,NRP))/4./DELRZ RX = RSCALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C0 = DPMOR(I)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))C7 = 2.*C1 - C5110 CONTINUE STRNEW = ALPHAM*STR(I,1)+ALPHA*(C5*STR(I,2) + 120 CONTINUE DR1 = (STR(IZ0,2)-STR(IZ0,NRP))/2./DELR C7*STR(I,NRP)+C2*DELRZ2* + (STR(IP,1)+STR(IM,1))+C4*DELR2)/2./(C1+C2*DELRZ2) + DZ1 = (STR(IZ0P, 1)-STR(IZ0M, 1))/2./DELZDIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(I,1))) DZR = (VEL(1,IZ0P,1)-VEL(1,IZ0M,1))/2./DELZ

STR(I,NRP) = STR(I,2) - C0*2.*DELRVEL(1,1,1) = DR1VEL(2, 1, 1) = DZ1STR(I,1) = STRNEWIF(IR0M.LE.2) GOTO 100 DO 90 J = 2.1R0MMDR1 = (STR(I,JP)-STR(I,JM))/2./DELRDZ1 = (STR(IP,J)-STR(IM,J))/2./DELZDZR=(STR(IP,JP)-STR(IP,JM)-STR(IM,JP)+STR(IM,JM))/4./DELRZ CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(I,J)+ALPHA*(C5*STR(I,JP) + C7*STR(I,JM)+C2*DELRZ2* (STR(IP,J)+STR(IM,J))+C4*DELR2)/2./(C1+C2*DELRZ2) DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(I,J))) VEL(1, I, J) = DR1VEL(2, I, J) = DZ1STR(I,J) = STRNEWCONTINUE CONTINUE DR1 = (RDIVBM*STR(I,IR0M)-STR(I,IR0MM))/2./DELR DZ1 = (STR(IP,IR0M)-STR(IM,IR0M))/2./DELZ DZR = (VEL(1, IP, IROM)-VEL(1, IM, IROM))/2./DELRZ RX = R(IR0M)CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(I,IROM)+ALPHA*((2.*C1/RDIVBP+ C8*DELRB/2.)*STR(I,IR0MM) + C2*DELRZ2* (STR(IP,IR0M)+STR(IM,IR0M))+C4*DELR2)/2./ (C1/RDIVB+C2*DELRZ2-RDIVBM*C3*DELRB/4.) DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(I,IR0M))) VEL(1,I,IR0M) = DR1VEL(2,I,IROM) = DZ1STR(I,IROM) = STRNEW

DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(IZ0,IR0M)))

RX = RS

CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C0 = DPMOR(IZ0)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(IZ0,1)+ALPHA*(C5*STR(IZ0,2) + C7*STR(IZ0,NRP)+C2*DELRZ2* (STR(IZ0P,1)+STR(IZ0M,1))+C4*DELR2)/2./(C1+C2*DELRZ2) DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(IZ0,1))) STR(IZ0,NRP) = STR(IZ0,2) - C0*2.*DELRVEL(1, IZ0, 1) = DR1VEL(2, IZ0, 1) = DZ1STR(IZ0,1) = STRNEWIF(IR0M.LE.2) GOTO 135 DO 130 J = 2, IR0MM JP = J + 1JM = J - 1DR1 = (STR(IZ0,JP)-STR(IZ0,JM))/2./DELRDZ1 = (STR(IZ0P,J)-STR(IZ0M,J))/2./DELZDZR = (VEL(1,IZ0P,1)-VEL(1,IZ0M,1))/2./DELZ RX = R(J)CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(IZ0,J)+ALPHA*(C5*STR(IZ0,JP) + C7*STR(IZ0,JM)+C2*DELRZ2* + (STR(IZ0P,J)+STR(IZ0M,J))+C4*DELR2)/2./(C1+C2*DELRZ2) + DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(IZ0,J))) VEL(1, IZ0, J) = DR1VEL(2,IZ0,J) = DZ1 STR(IZ0,J) = STRNEW130 CONTINUE 135 CONTINUE DR1 = -STR(IZ0,IR0MM)/2./DELR DZ1 = (STR(IZ0P,IR0M)-STR(IZ0M,IR0M))/2./DELZ DZR = (VEL(1,IZ0P,IR0M)-VEL(1,IZ0M,IR0M))/2./DELZ RX = R(IR0M)CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(IZ0,IR0M)+ALPHA*(C5*STR(IZ0,IR0) + + C7*STR(IZ0,IR0MM)+C2*DELRZ2* (STR(IZ0P,IR0M)+STR(IZ0M,IR0))+C4*DELR2) + /2./(C1+C2*DELRZ2) +

VEL(2, IZ0, IR0M) = DZ1STR(IZ0,IR0M) = STRNEW ENDIF DO 140 I = NZS.NZM IP = 1 + 1IM = I - 1DZ1 = (STR(IP,1)-STR(IM,1))/2./DELZDR1 = (STR(I,2)-STR(I,NRP))/2./DELRDZR = (STR(IP,2)-STR(IM,2)-STR(IP,NRP)+STR(IM,NRP))/4./DELRZ RX = RSCALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C0 = DPMOR(I)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))C7 = 2.*C1-C5STRNEW = ALPHAM*STR(I,1)+ALPHA*(C5*STR(I,2) + C7*STR(I,NRP) + + C2*DELRZ2*(STR(IP,1)+STR(IM,1))+C4*DELR2)/2./C6 DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(I,1))) VEL(1, 1, 1) = DR1VEL(2,1,1) = DZ1STR(1,1) = STRNEWSTR(I,NRP) = STR(I,2) - C0*2.*DELRDO 137 J = 2,NRM JP = J + 1JM = J - 1IF(RW(I).LT.R0.AND.R(JP).EQ.RW(I)) THEN DZ1 = STR(IP,J)/2./DELZDR1 = -STR(I, JM)/2./DELRDZR = (STR(I,J)-STR(IP,JM))/4./DELRZ RX = R(J)CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C7 = 2*C1 - C5STRNEW = ALPHAM*STR(I,J)+ALPHA*(C5*STR(I,JP) + C7*STR(I,JM) + C2*DELRZ2*(STR(IP,J)+STR(IM,J))+C4*DELR2)/2./C6 DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(I,J))) VEL(1,I,J) = DR1VEL(2, I, J) = DZ1STR(I,J) = STRNEW **GOTO 140** ENDIF DZ1 = (STR(IP,J)-STR(IM,J))/2./DELZDR1 = (STR(I,JP)-STR(I,JM))/2./DELR

VEL(1, IZ0, IR0M) = DR1

+STR(IM,JM))/4./DELRZ + RX = R(J)CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(I,J)+ALPHA*(C5*STR(I,JP) + C7*STR(I,JM)+C2*DELRZ2* + (STR(IP,J)+STR(IM,J))+C4*DELR2)/2./C6 DIFMAX = DMAX1(DIFMAX, DABS(STRNEW-STR(I, J)))VEL(1,I,J) = DR1VEL(2, I, J) = DZ1STR(I,J) = STRNEW137 CONTINUE **140 CONTINUE** DZ1 = (STR(NZ,1)-STR(NZM,1))/2./DELZDR1 = (STR(NZ,2)-STR(NZ,NRP))/2./DELRDR1P = (STR(NZ,2)-STR(NZP,NRP))/2./DELRDZR = (DR1P-VEL(1,NZM,1))/2./DELZ RX = RS CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C0 = DPMOR(NZ)*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2))C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(NZ,1)+ALPHA*(C5*STR(NZ,2) + C7*STR(NZ,NRP)+2.*C2*DELRZ2/ZDIVSP* + STR(NZM,1)+C4*DELR2)/2./(C1+C2*DELRZ2/ZDIVSP) DIFMAX = DMAX1(DIFMAX, DABS(STRNEW-STR(NZ, 1))) STR(NZ,NRP) = STR(NZ,2) - C0*2.*DELR C0 = -PMORG(NZ)/DELZS*RS/(F1+F2/RS*DSQRT(DZ1**2+DR1**2)) STR(NZP,NRP) = STR(NZ,2) - C0*2.*DELRVEL(1,NZ,1) = DR1VEL(2, NZ, 1) = DZ1STR(NZ,1) = STRNEWSTR(NZP,1) = STR(NZ,1)DO 150 J = 2,NRM JP = J+1JM = J-1DZ1 = (STR(NZ,J)-STR(NZM,J))/2./DELZ DR1 = (STR(NZ, JP)-STR(NZ, JM))/2./DELRDZR = (DR1-VEL(1,NZM,J))/2./DELZ IF(RW(I).LT.R0.AND.R(JP).EQ.RW(I)) + DZR=(STR(NZ,J)-STR(NZ,JM))/4./DELRZ RX = R(J)

DZR = (STR(IP,JP)-STR(IM,JP)-STR(IP,JM))

CALL FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6) C7 = 2.*C1 - C5STRNEW = ALPHAM*STR(NZ,J)+ALPHA*(C5*STR(NZ,JP) + C7*STR(NZ,JM)+2.*C2*DELRZ2/ZDIVSP* STR(NZM,J)+C4*DELR2)/2./(C1+C2*DELRZ2/ZDIVSP) DIFMAX = DMAX1(DIFMAX,DABS(STRNEW-STR(NZ,J))) VEL(1,NZ,J) = DR1VEL(2,NZ,J) = DZ1STR(NZ,J) = STRNEWSTR(NZP,J) = STR(NZ,J)IF(RW(NZ).LT.R0.AND.R(JP).EQ.RW(NZ))GOTO 160 150 CONTINUE **160 CONTINUE** C IF(ITER/100*100.EQ.ITER)WRITE(8,170)ITER,STR(NZ,1),STR(NZ,5) C + STR(NZ,10),STR(NZ,15),STR(NZ,NR),DIFMAX 170 FORMAT(15,6G12.3) IF (DIFMAX.LT.EPS)GOTO 2000 1000 CONTINUE WRITE(6,*)' Convergence Failure - DIFMAX = ',DIFMAX 2000 IF(ILIN.EQ.0)THEN ILIN = 1**GOTO 55** ENDIF || = |Z0|JJ = IR0DO 410 I = 1,NZ IPP = 1 + 2IP = 1 + 1IM = 1 - 1IMM = 1 - 2PERG(I,1) = PMORG(I)VEL(3,1,1) = VEL(2,1,1)/RSVEL(4,1,1) = -VEL(1,1,1)/RSIF(IR0.EQ.1.AND.I.EQ.1) THEN VEL(3,I,1) =FDIF(1,STR(1,1),STR(2,1),STR(3,1),DELZ)/RS VEL(4,I,1) = STR(I,NRP)/DELR/RS **GOTO 410** ENDIF IF(IR0.EQ.2.AND.I.LT.IZ0)GOTO 410 DO 400 J = 2,NRM JMM = J - 2JM = J - 1
IF(JMM.EQ.0) JMM = NRP

IF(IR0.EQ.1.AND.J.EQ.2.AND.I.EQ.J) THEN VEL(3,I,J) = FDIF(1,STR(I,J),STR(IP,J),STR(IPP,J),DELZ)+ /R(J) VEL(4,I,J) =-FDIF(3,STR(I,J),STR(I,1),STR(I,NRP),DELR) + /R(J)|| = || + 2JJ = JJ + 2**GOTO 410** ELSEIF(II.EQ.I.AND.JJ.EQ.J) THEN VEL(3,I,J) =FDIF(1,STR(I,J),STR(IP,J),STR(IPP,J),DELZ) + + /R(J) VEL(4,I,J)=-FDIF(3,STR(I,J),STR(I,JM),STR(I,JMM),DELR) + /R(J)|| = || + 1JJ = JJ + 1**GOTO 410** ELSE + VEL(3,I,J) = VEL(2,I,J)/R(J)VEL(4, I, J) = -VEL(1, I, J)/R(J)IF(I.LT.II.AND.J+1.EQ.JJ) GOTO 410 ENDIF 400 CONTINUE VEL(3,I,NR) = 0.VEL(4,I,NR) = -FDIF(3,STR(I,NR),STR(I,NRM),STR(I,NRMM),DELR) /R(NR) **410 CONTINUE** || = |Z0|JJ = IR0MJUN = 0WRITE(8,*)'NZ,NR = ',NZ,NR DO 430 | = 1,NZ + IPP = 1 + 2IP = I + 1|M = | - 1|IMM = 1 - 2IF(I.EQ.1.AND.IR0.EQ.1) GOTO 430 IF(I.LT.IZ0.AND.IR0.EQ.2) GOTO 430 DO 420 J = 2,NRM JMM = J - 2JM = J - 1IF(JMM.EQ.0) JMM = NRP +

UAR = VEL(3,I,J)UZ = VEL(4,I,J)IF(UAR+UZ.LE.0.)WRITE(8,*)'ZERO AT*',I,J,NZ JP = J + 1IF(I.EQ.NZ) THEN DURR = FDIF(2, VEL(3, I, JP), VEL(3, I, JM), XX, DELR)DUZR = FDIF(2, VEL(4, I, JP), VEL(4, I, JM), XX, DELR)DURZ = (ZDIVSM*VEL(3,I,J)-VEL(3,IM,J))/2./DELZ DUZZ = (VEL(4,I,J)-VEL(4,IM,J))/2./DELZVEL(5,I,J)=F2*(UAR**2*DURR+UAR*UZ*(DUZR+DURZ) +UZ**2*DUZZ)/ DSQRT(UAR**2+UZ**2) ELSEIF(IR0.EQ.1.AND.J.EQ.2.AND.I.EQ.J) THEN DURR = (VEL(3,I,J)-VEL(3,I,JM))/DELRDURZ = FDIF(1, VEL(3, I, J), VEL(3, IP, J), VEL(3, IPP, J), DELZ)DUZR = (VEL(4,I,J)-VEL(4,I,JM))/DELRDUZZ = FDIF(1, VEL(4, I, J), VEL(4, IP, J), VEL(4, IPP, J), DELZ)VEL(5,I,J)=F2*(UAR**2*DURR+UAR*UZ*(DUZR+DURZ) +UZ**2*DUZZ)/ DSQRT(UAR**2+UZ**2) 11 = 11 + 2 JJ = JJ + 2**GOTO 430** ELSEIF(II.EQ.I.AND.JJ.EQ.J) THEN DURR = FDIF(3, VEL(3, I, J), VEL(3, I, JM), VEL(3, I, JMM), DELR)DURZ = FDIF(1,VEL(3,I,J),VEL(3,IP,J),VEL(3,IPP,J),DELZ)DUZR = FDIF(3, VEL(4, I, J), VEL(4, I, JM), VEL(4, I, JMM), DELR)DUZZ = FDIF(1, VEL(4, I, J), VEL(4, IP, J), VEL(4, IPP, J), DELZ)IF(J.EQ.2.AND.IR0.EQ.2) THEN DURR = (VEL(3,1,J)-VEL(3,1,JM))/DELRDUZR = (VEL(4,I,J)-VEL(4,I,JM))/DELRENDIF VEL(5,I,J)=F2*(UAR**2*DURR+UAR*UZ*(DUZR+DURZ) +UZ**2*DUZZ)/ DSQRT(UAR**2+UZ**2) || = || + 1JJ = JJ + 1**GOTO 430** ELSE DURR = FDIF(2, VEL(3, I, JP), VEL(3, I, JM), XX, DELR) DURZ = FDIF(2, VEL(3, IP, J), VEL(3, IM, J), XX, DELZ)DUZR = FDIF(2, VEL(4, I, JP), VEL(4, I, JM), XX, DELR)DUZZ = FDIF(2, VEL(4, IP, J), VEL(4, IM, J), XX, DELZ)VEL(5,I,J)=F2*(UAR**2*DURR+UAR*UZ*(DUZR+DURZ) +UZ**2*DUZZ)/ DSQRT(UAR**2+UZ**2)

IF(I.LT.II.AND.J+1.EQ.JJ) GOTO 430

ENDIF

|M = | - 1|

420 CONTINUE UAR = VEL(3,I,NR)UZ = VEL(4, I, NR)IF(UAR+UZ.LE.0.)WRITE(8,*)'ZERO AT ',I,NR IF(MJUN.EQ.0)THEN DURR = FDIF(3, VEL(3, I, NR), VEL(3, I, NRM), VEL(3, I, NRMM), DELR)DURZ = FDIF(1, VEL(3, I, NR), VEL(3, IP, NR), VEL(3, IPP, NR), DELZ)DUZR = FDIF(3,VEL(4,I,NR),VEL(4,I,NRM),VEL(4,I,NRMM),DELR) DUZZ = FDIF(1, VEL(4, I, NR), VEL(4, IP, NR), VEL(4, IPP, NR), DELZ)VEL(5,I,NR)=F2*(UAR**2*DURR+UAR*UZ*(DUZR+DURZ)) + +UZ**2*DUZZ)/ DSQRT(UAR**2+UZ**2) MJUN = 1ELSEIF(I.EQ.NZ) THEN DURR = FDIF(3, VEL(3, I, NR), VEL(3, I, NRM), VEL(3, I, NRMM), DELR) DURZ = FDIF(3, VEL(3, I, NR), VEL(3, IM, NR), VEL(3, IMM, NR), DELZ) DUZR = FDIF(3, VEL(4, I, NR), VEL(4, I, NRM), VEL(4, I, NRMM), DELR)DUZZ = FDIF(3,VEL(4,I,NR),VEL(4,IM,NR),VEL(4,IMM,NR),DELZ) VEL(5,I,NR)=F2*(UAR**2*DURR+UAR*UZ*(DUZR+DURZ) +UZ**2*DUZZ)/ DSQRT(UAR**2+UZ**2) + ELSE DURR = FDIF(3,VEL(3,I,NR),VEL(3,I,NRM),VEL(3,I,NRMM),DELR) DURZ = FDIF(1,VEL(3,IP,NR),VEL(3,IM,NR),XX,DELZ) DUZR = FDIF(3, VEL(4, I, NR), VEL(4, I, NRM), VEL(4, I, NRMM), DELR)DUZZ = FDIF(1,VEL(4,IP,NR),VEL(4,IM,NR),XX,DELZ) VEL(5,I,NR)=F2*(UAR**2*DURR+UAR*UZ*(DUZR+DURZ) ÷ +UZ**2*DUZZ)/ DSQRT(UAR**2+UZ**2) ENDIF 430 CONTINUE EPSP = EPS*PERG(1,1) WRITE(8,*)'EPS, EPSP = ', EPS, EPSP ANG = 1./6. RDIVC = 2.*DCOS(ANG)**2 - 1. ZDIVC = 2.*DCOS(ANG)*DSIN(ANG)*TAN30 DO 7000 ITER = 1.MAXIT || = |Z0|JJ = IR0DIFMAX=0. DO 450 | = 1,NZ IP = 1 + 1

IF(I.LT.IZ0.AND.IR0.EQ.2) GOTO 450 DO 440 J = 2.NRJM = J - 1JP = J + 1IF(I.EQ.NZ) THEN E4 = (1.+DELR/2./R(J))/DNMS $E5 \approx (1.-DELR/2./R(J))/DNMS$ PERNEW=PERG(I,J)*ALP2M+ALP2*(E4*PERG(I,JP) + E5*PERG(I,JM)+E6*PERG(IM,J)+VEL(5,I,J)*DELR2/DNMS) + DIFMAX = DMAX1(DIFMAX,DABS(PERNEW-PERG(I,J))) PERG(I,J) = PERNEW IF(II.EQ.I.AND.JJ.EQ.J) PERG(I,JP) = 0.ELSE E1 = (1.+DELR/2./R(J))/DNME2 = (1.-DELR/2./R(J))/DNMPERNEW=PERG(I,J)*ALP2M+ALP2*(E1*PERG(I,JP) + E2*PERG(I,JM)+E3*(PERG(IM,J)+PERG(IP,J)) + +VEL(5,I,J)*DELR2/DNM) + DIFMAX = DMAX1(DIFMAX,DABS(PERNEW-PERG(I,J))) PERG(I,J) = PERNEWENDIF IF(IR0.EQ.1.AND.J.EQ.2.AND.I.EQ.J) THEN P2 = PERG(I,JM)P1 = PERG(IM, JM)PERG(IM,J) = P1 + (P2-P1)*ZDIVC|| = || + 2JJ = JJ + 2**GOTO 450** ELSEIF(II.EQ.I.AND.JJ.EQ.J) THEN IF(J.GT.2) THEN JMM = J - 2P1 = PERG(IM,JM) + (PERG(IM,JMM)-PERG(IM,JM))*RDIVC P2 = PERG(I,JM) + (PERG(I,JMM)-PERG(I,JM))*RDIVC PERG(IM,J) = P1 + (P2-P1)*ZDIVCELSE P2 = PERG(I,JM)P1 = PERG(IM, JM)PERG(IM,J) = P1 + (P2-P1)*ZDIVCENDIF

IF(J.EQ.NR) PERG(I,NRP) = PERG(I,NRM)

|| = || + 1

IF(I.EQ.1.AND.IR0.EQ.1) GOTO 450

```
ELSEIF(I.LT.II.AND.JP.EQ.JJ)THEN
        PERG(I,JP) = PERG(I,J)
        GOTO 450
      ENDIF
 440 CONTINUE
     PERG(I,NRP) = PERG(I,NRM)
 450 CONTINUE
C IF(ITER/10*10.EQ.ITER)WRITE(8,170)ITER.PERG(NZ,1),PERG(NZ,5)
C + PERG(NZ,10), PERG(NZ,15), PERG(NZ,NR), DIFMAX
   IF (DIFMAX.LT.EPSP)GOTO 8000
7000 CONTINUE
   WRITE(6,*)' CONVERGENCE FAILURE II - DIFMAX = '.DIFMAX
8000 CONTINUE
   NSP = NZP
   NSPM = NSP - 1
   DDX = (H-Z(1))/(NSP-1.)
   JJ = IR0
   DO 510 | = 1. NZ
     IP = 1 + 1
     IF(I.EQ.1.AND.IR0.EQ.1) THEN
      QZA(I) = 0.
      JJ = 2
       GOTO 510
     ELSEIF(I.LT.IZ0.AND.IR0.EQ.2) THEN
       QZA(I) = PI*VEL(4,I,J)*(RS+RB)/2.*(RB-RS)
       GOTO 510
     ENDIF
     QZA(I) = 0.
     DO 500 J = 1, JJ - 1
     JP = J + 1
      QZA(I)=QZA(I)+PI*(VEL(4,I,J)*R(J)+VEL(4,I,JP)*R(JP))/2.*DELR
 500 CONTINUE
     JJ = JJ + 1
     IF(JJ.GT.NR) JJ = NR
 510 CONTINUE
   CALL MYSPLN(Z,QZA,QZAQ,QZAR,QZAS,NZ,NDS)
   CZ(1) = Z(1)
   PAN(1) = PMORG(1)
   QAN(1) = 0.
```

JJ = JJ + 1

GOTO 450

WRITE(8,*)'DDX = ',DDXDO 490 I = 2,NSPM CZ(I) = CZ(I-1) + DDXPAN(I) = PMORH - PMOR(CZ(I))QAN(I) = QAINT(CZ(I), 11)**490 CONTINUE** CZ(NSP) = HPAN(NSP) = 0.QAN(NSP) = QZA(NZ)DPAN(1) = (4.*PAN(2)-PAN(3)-3.*PAN(1))/2./DELR DPAN(NSP) = (PAN(NSP-2)-4.*PAN(NSPM)+3.*PAN(NSP))/2./DELR DQAN(1) = (4.*QAN(2)-QAN(3)-3.*QAN(1))/2./DELRDQAN(NSP) = (QAN(NSP-2)-4.*QAN(NSPM)+3.*QAN(NSP))/2./DELR DO 520 I = 2,NSPM DPAN(I) = (PAN(I+1)-PAN(I-1))/2./DELRDQAN(I) = (QAN(I+1)-QAN(I-1))/2./DELR**520 CONTINUE** CALL MYSPLN(CZ, QAN, QANQ, QANR, QANS, NSP, NDS) CALL MYSPLN(CZ.DQAN.DQANQ.DQANR.DQANS.NSP.NDS) CALL MYSPLN(CZ, PAN, PANQ, PANR, PANS, NSP, NDS) CALL MYSPLN(CZ, DPAN, DPANQ, DPANR, DPANS, NSP, NDS) DO 6040 I = 1.NZ DO 6030 J=1,NR PLX(J,I) = PERG(I,J)6030 CONTINUE 6040 CONTINUE RETURN 1111 WRITE(8,5000)((STR(I,J),J=1,NR,2),I=1,NZ) C WRITE(8,5000)((PERG(I,J),J=1,NR,2),I=1,NZ) C WRITE(8,5000)((VEL(1,I,J),J=1,NR,2),I=1,NZ) C WRITE(8,5000)((VEL(2,I,J),J=1,NR,2),I=1,NZ) C WRITE(8,5000)((VEL(3,I,J),J=1,NR,2),I=1,NZ) С WRITE(8,5000)((VEL(4,I,J),J=1,NR,2),I=1,NZ) С WRITE(8,5000)((VEL(5,1,J),J=1,NR,2),I=1,NZ) RSIZ = 3.0ZSIZ = (Z(NZ)-Z(1))*2.5/(R0-RS) + 0.5X1= 0.02 XSTEP = 0.02X2 = R(NR)Y1 = DNINT(100.*Z(1))/100.

YSTEP = 0.04

Y2=Z(NZ)IF(R0.GT.0.121) THEN RSIZ = 3.5 $ZSIZ = (Z(NZ)-Z(1))^{3.0}(R0-RS) + 0.5$ XSTEP = 0.04ENDIF 5000 FORMAT(11G12.3) CALL DSPDEV('PLOT') CALL AREA2D(RSIZ,ZSIZ) CALL COMPLX CALL XNAME('Radius, m',1) CALL YNAME('Height, m',1) CALL GRAF(X1,XSTEP,X2,Y1,YSTEP,Y2) CALL FRAME CALL BCOMON(50000) DO 5020 I = 1.NZ DO 5010 J=1,NR PLX(J,I) = STR(I,J)*1.E55010 CONTINUE 5020 CONTINUE CALL CONMAK(PLX,NR,NZ,'SCALE') CALL CONLIN (0,'SOLID','LABEL',2,5) CALL CONLIN (1, 'DASH', 'NOLABELS', 1, 3) CALL CONANG(90.) CALL RASPLN(0.25) CALL CONTUR(2,'LABELS','DRAW') CALL ENDPL(0) CALL DONEPL CALL DSPDEV('PLOT') CALL AREA2D(RSIZ,ZSIZ) CALL COMPLX CALL XNAME('Radius, m',1) CALL YNAME('Height, m',1) CALL GRAF(X1,XSTEP,X2,Y1,YSTEP,Y2) CALL FRAME CALL BCOMON(50000) DO 5040 I = 1,NZ DO 5030 J=1,NR PLX(J,I) = PERG(I,J)5030 CONTINUE **5040 CONTINUE** CALL CONMAK(PLX,NR,NZ,'SCALE')

CALL CONLIN (0,'SOLID','LABEL',2,5) CALL CONLIN (1,'DASH','NOLABELS',1,3) CALL CONANG(90.) CALL RASPLN(0.25) CALL CONTUR(2,'LABELS','DRAW') CALL ENDPL(0) CALL DONEPL END SUBROUTINE FSTR(DZ1,DR1,DZR,C1,C2,C3,C4,C5,C6)

```
IMPLICIT REAL*8(A-H,O-Z)
COMMON /FSTRSB/ F1,F2,RX,DELR,DELRZ2,ILIN
IF(ILIN.EQ.1)THEN
 A2STR = DZ1**2 + DR1**2
 IF(A2STR.LE.0.)WRITE(8,*)' YEP, A2STR = ',A2STR
 A1STR = DSQRT(A2STR)
 C1 = F1*RX/F2*A1STR + A2STR + DR1**2
 C2 = C1 - DR1^{**}2 + DZ1^{**}2
 C3 = -(F1/F2+2./RX*A1STR)*A1STR
 C4 = 2.*DZR*DR1*DZ1
 C5 = C1 + C3 + DELR/2.
 C6 = C1+C2*DELRZ2
 RETURN
ENDIF
C1 = 10.
C2 = 10.
C3 = 1.
C4 = 1.
C5 = C1 + C3 + DELR/2.
C6 = C1+C2*DELRZ2
END
```

DOUBLE PRECISION FUNCTION FDIF(K,X,Y,Z,DEL)

* K: 1 = FORWARD 2 = CENTRAL 3 = BACKWARD

```
IMPLICIT REAL*8(A-H,O-Z)

IF(K.EQ.1) THEN

FDIF = (-3.*X+4.*Y-Z)/2./DEL

ELSEIF(K.EQ.2) THEN

FDIF = (X - Y)/2./DEL

ELSEIF(K.EQ.3) THEN

FDIF = (3.*X - 4.*Y + Z)/2./DEL

ELSE

WRITE(8,*)' Error in FDIF code, code = ',K

ENDIF

END
```

DOUBLE PRECISION FUNCTION QAERG(X)

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' QAERG = PI*QAINT(X,11)*QAINT(X,12) END

SUBROUTINE PLOTMT

IMPLICIT REAL*8(A-H,O-Z) EXTERNAL SS **INCLUDE 'H.COM1'** INCLUDE 'H.COM4' **DIMENSION STOT(10000)** REAL XP1(10000), PR1(10000), XP2(100) N = NSPVAH = VS(N)*(1.-ES(N))/((DCOL/DS)**2-1.)/(1.-VOID) DMT(N) = DCOLZMT(N) = CZ(1)DMT(1) = DSZMT(1) = HDO 10 I = 2,N-1 J = | -1ZMT(N-J) = CZ(I)XXX = DCOL**2-DS**2*VS(I)/VAH*(1.-ES(I))/(1.-VOID) IF(XXX.LE.0)WRITE(8,*)'XXX,CZ(I),VS(I),VAH,ES(I)', + XXX,CZ(I),VS(I),VAH,ES(I) DMT(N-J) = DSIF(XXX.GT.0.) DMT(N-J) = DSQRT(XXX)IF(DMT(N-J).LE.DS) DMT(N-J) = DS+0.001*+ (H-ZMT(N-J))/H **10 CONTINUE** WRITE(8,*)'ZMT NOT OK ' CALL MYSPLN(DMT,ZMT,ZMTQ,ZMTR,ZMTS,N,NDS) WRITE(8,*)'ZMT OK ' SMT(1) = 0.DO 20 I = 2, N SMT(I) = 0.IF(ZMT(I-1).GT.HCOAT) CALL NC4AD(SS,ZMT(I),ZMT(I-1), + 0.5D-1,SMT(I),NFC) SMT(I) = SMT(I-1) + SMT(I)WRITE(7,*)I, DMT(I), ZMT(I), SMT(I) С 20 CONTINUE C WRITE(7,*)1, DMT(1), ZMT(1), SMT(1) C CALL MYSPLN(DMT,SMT,SMTQ,SMTR,SMTS,N,NDS) С

Randomly choose particle landing position and start the
 simulation
 UWT = PI/6.*DPU**3*RHOU

SURE = WS*UWT/WU VEXIT = WU/(1.-XS)/RHOP VTOT = VS(N)*(1.-ES(N))*PI4*DS**2 VCIRC = VTOT - VEXIT PEXIT = VEXIT/VTOT PTOT = VTOT/VCIRC WRITE(7,8)SURE, VCIRC, VEXIT, PEXIT, PTOT 8 FORMAT(' SURE, VCIRC, VEXIT, PEXIT, PTOT = '.5G12.5) MMM = 1000LFEED = 1 $C1 = DS^{**2}$ CALL DSPDEV('PLOT') CALL ATRANS CALL NOBRDR CALL COMPLX CALL AREA2D(5.5.5.) CALL XNAME('Sulfur Content\$',100) CALL YNAME ('Cumulative Probability\$',100) CALL XREVTK CALL YREVTK C CALL HEADIN('Sulfur Content Distribution\$',-100,3,1) CALL YAXANG(0.) CALL GRAF(0.,0.1,.8,0.0,.20,1.) CALL THKCRV(3) CALL SCLPIC(2.) 47 CONTINUE IF(LFEED.EQ.1) THEN DFEED = DCOL-0.06 $CB1 = DFEED^{**2}$ $CC = DCOL^{**2} - CB1$ C2 = CB1 - C1 $C3 = DCOL^{**2} - C1$ XFEED = WU/RHOU/VTOT*C3/CC IF (XFEED.GT.1.) THEN WRITE (8,*) 'XFEED too large, inc. feed area, XF=',XFEED STOP ENDIF CA = C2 + (1.-XFEED)*CCCB = CA/(1.-XFEED)CB2 = -C2/(1.-XFEED)XFM = C2/(C2+(1.-XFEED)*CC)WRITE(8,113) C1,C2,C3,CA,CB,CB1,CB2,DFEED,XFEED,XFM

113 FORMAT('C1.C2.C3.CA.CB, CB1.CB2.DFEED,XFEED,XFM = '/ + 5G12.5/5G12.5) ELSEIF(LFEED.EQ.0) THEN $C = DCOL^{**2} - C1$ ELSE WRITE(8,*) 'Wrong Feed Code, LFEED = '.LFEED STOP ENDIF WRITE(7,*)'XFM, XFEED, DFEED = ', XFM, XFEED, DFEED XRAND = RAND(831.)DO 390 I1 = 1,MMM XRAND = FRAND(1.0)XRAND = XRAND*PTOT IF(LFEED.EQ.1) THEN DMTI = DSQRT(CB1+XRAND*CC)ELSE DMTI = DSQRT(C1+XRAND*C) ENDIF II = INTT(DMTI,DMT,NSP,DX) STOT(I1)=SMT(II)+(SMT(II+1)-SMT(II))*DX/(DMT(II+1)-DMT(II)) С STOT(I1) = QAINT(DMTI,2)WRITE(8,71)XRAND.DMTI.STOT(11) С IF(STOT(11),LT.0.) STOT(11) = 0.С 391 XRAND = FRAND(1.0)XRAND = XRAND * PTOT IF(XRAND.GT.1.0)GOTO 390 IF(LFEED.EQ.0) THEN DMTI = DSQRT(C1+XRAND*C) ELSE IF(XRAND.LE.XFM) THEN DMTI = DSQRT(C1+XRAND*CA)ELSE DMTI = DSQRT(CB1+CB2+XRAND*CB) ENDIF ENDIF II = INTT(DMTI,DMT,NSP,DX) SMTI = SMT(II) + (SMT(II+1)-SMT(II))*DX/(DMT(II+1)-DMT(II))С SMTI = QAINT(DMTI,2)WRITE(8,71)XRAND, DMTI, STOT(11) С C 71 FORMAT('XRAND,DMTI,STOT = ',3G12.5)

C IF(SMTI.LT.0.) SMTI = 0. STOT(I1) = STOT(I1) + SMTI

GOTO 391 390 CONTINUE SUM1 = 0. SUMUD = 0.C WRITE(7,*)'I, STOT, XP1 :: UWT = ',UWT' DO 557 I1 = 1, MMM SUM1 = SUM1 + STOT(I1)XP1(I1) = STOT(I1)/(STOT(I1)+UWT)WRITE(7,*)11, STOT(11),XP1(11) С 557 CONTINUE SAVG = SUM1/(SUM1+UWT*MMM) SUM2 = 0.DO 395 I = 1.MMM SUM2 = SUM2 + (SAVG-XP1(I))**2 XP1(I) = STOT(I)/(STOT(I)+UWT)*XS/SAVG TANHC = (0.19823 - XP1(I))/XP1(I)/(0.715183 - XP1(I)) $PARTUD \approx (DTANH(TANHC)+1.)/2.$ IF(XP1(I).GT.0.715183) PARTUD = 0.SUMUD = SUMUD + PARTUD 395 CONTINUE UDC = SUMUD/MMM $XUDC = UDC^{(1.-XS)}$ $STD = (SUM2/(MMM-1))^{**.5}$ CALL CUMP(MMM,XP1,PR1,UWT) DO 397 I = 1,50 PRINT(3, NPRINT, I) = XP1(I*20)397 CONTINUE WRITE(7,*)'S AVERAGE = ',SAVG,' ST.DEV. = ',STD WRITE(7,*)'UWT = ', UWT,' UREA DIS = ', UDC,' XUDC = ', XUDC CALL CURVE(XP1,PR1,MMM,0) CALL DASH C IF (LFEED.EQ.1) GOTO 48 IF (LFEED.EQ.1) GOTO 51 LFEED = 1**GOTO 47 48 CONTINUE** DO 49 JJ = 1.40 PR1(JJ) = JJ/40.READ(4,*)SC1 XP1(JJ) = SC1/100.XP2(JJ) = SC2/100.С **49 CONTINUE**

CALL CURVE(XP1,PR1,40,-1) C CALL MARKER(5) C CALL CURVE(XP2,PR1,40,-1) **51 CONTINUE** CALL FRAME CALL ENDGR(0) CALL ENDPL(0) C REWIND (4) 555 CONTINUE CALL DSPDEV('PLOT') CALL NOBRDR CALL COMPLX CALL AREA2D(5.5.5.) CALL XNAME('Height, m\$',100) CALL YNAME('Coating Amount per Cycle, kg\$',100) CALL XREVTK CALL YREVTK C CALL HEADIN('Density of Coating amount/c\$',-100,3,1) CALL YAXANG(0.) IF(HCOAT.LE.0.)CALL GRAF(0.,0.05,0.3,0.0,.25E-4,1.E-4) IF(HCOAT.GT.0.)CALL GRAF(0.,0.05,0.3,0.0,.2E-6,.1E-5) CALL THKCRV(3) DO 567 I = 1,N XP1(I) = ZMT(I)PR1(I) = SMT(I)PRINT(2,NPRINT,I) = SMT(I)567 CONTINUE CALL CURVE(XP1, PR1, N, 0) CALL FRAME CALL ENDGR(0) CALL ENDPL(0) RETURN END

SUBROUTINE PROBTR(A,B,IC)

* Initialization routine for probability plot for *disspla

ENTRY TRANS(A,B,IC) CALL YPRTRN(A,B,IC) RETURN END

SUBROUTINE CUMP(M,X1,Y1,WTU)

* Sorts and calculates cumulative #'s for plotting

REAL*8 WTU REAL X1(10000), Y1(10000) DO 10 I=1,M-1 DO 10 J=1,M-I IF(X1(J).GT.X1(J+1))THEN $\dot{S}A\dot{V}E = X1(J+1)$ X1(J+1) = X1(J)X1(J) = SAVE ENDIF **10 CONTINUE** DO 20 I=1,M Y1(I) = FLOAT(I)/FLOAT(M)C WRITE(8,*)'PR, SC = ',Y1(I),X1(I) C X1(I) = X1(I)/(X1(I)+WTU)20 CONTINUE RETURN END

SUBROUTINE PRINTO

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' INCLUDE 'H.COM4' DO 10 I = 1.3 IF(I.EQ.3) THEN NPR0 = 50ELSE NPR0 = NSPENDIF DO 20 J = 1, NPR0IF(I.EQ.3)WRITE(11,100)J,(PRINT(I,K,J),K=1,NPRINT) IF(I.EQ.2)WRITE(11,100)ZMT(J),(PRINT(I,K,J),K=1,NPRINT) IF(I.EQ.1)WRITE(11,100)CZ(J),(PRINT(I,K,J),K=1,NPRINT) 20 CONTINUE **10 CONTINUE** 100 FORMAT(2X,50G12.4) STOP END

DOUBLE PRECISION FUNCTION SS(X)

* Finds the amount of sulfur a particle picks up in the spout

$$\begin{split} & \text{IMPLICIT REAL*8(A-H,O-Z)} \\ & \text{INCLUDE 'H.COM1'} \\ & \text{I} = \text{INTT}(X,CZ,11,DX) \\ & \text{V} = \text{QAINT}(X,5) \\ & \text{E} = \text{QAINT}(X,6) \\ & \text{U} = \text{QAINT}(X,6) \\ & \text{U} = \text{QAINT}(X,3) \\ & \text{IF}(V.LE.0.D0.OR.X.LT.HCOAT)THEN \\ & \text{SS} = 0. \\ & \text{RETURN} \\ & \text{ENDIF} \\ & \text{SS} = \text{PI4*C*ETA}(\text{DABS}(U-V))^*\text{DABS}(U-V)^*\text{DP**2/V} \\ & \text{RETURN} \\ & \text{END} \\ & \text{END} \end{split}$$

SUBROUTINE INITIA

- * This subroutine assigns the following constants to be used
- * in the spouted bed hydrodynamics and coating calculations:
- * PI = pi PI4 = pi/4
- * THETA = half cone angle
- * WMA = molecular weight of air
- * RHOSM = density of monoclinic sulfur
- * RHOSR = " " rhombic sulfur
- * RHOU = " " urea
- * DPU = averaged diameter of urea
- * GRAV = gravitational constant
- * UNIVC = universal constant
- * DCOL = column diameter
- * HB = the distance from the shuttle to the bottom of the cone
- * DB = the diameter of cone base
- * TAN30 = tangent of 30 degrees
- * VOID = loose packed voidage of the bed.
- * NDS = dimension size for the arrays in SPLINE
- * DAT = nozzle opening for air
- * AAT = area of flow through the nozzle cap

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1' NDS = 101 PI = DACOS(-1.D0)PI4 = PI/4.THETA = PI/6. TAN30 = DTAN(THETA)WMA = 29.0 RHOSF = 1780. RHOSM = 1960. RHOSR = 2007. RHOU = 1335. DPU = 0.00216 **GRAV = 9.8** UNIVC = .08206 DNI = 0.0029 DAT = 0.002921 AAT = PI4*DAT**2 HB = .01DB = .04

VOID = 0.42 PSI = 1.

* Voidage calculations.

- C H = 0.25 + 0.017
- C XS = 0.31
- C WB = 6.02273
- C = V = CAL(9,H,H)
- $C = VP = WB^{*}((1.-XS)/RHOU+XS/RHOSR)$
- C VOID = (V-VP)/V RETURN

END

SUBROUTINE EOS(XT, DENSTY, VISCOT)

- * This subroutine calculates density and viscosity of air.
- * Parameters are:
- * T = Temperature in Kelvin as INPUT
- * D = Will contain density data in Kg/Cubic Metres AS OUTPUT
- * V = " viscosity " Pa/s or kg.m/s AS "

IMPLICIT REAL*8(A-H,O-Z) INCLUDE 'H.COM1'

- * Density calculated from the ideal gas law. DENSTY = 1.*WMA/UNIVC/XT
- * Viscosity calculated from the correlation by Neufeld (1972) given in
- * The Properties of Gases and Liquids (1977).

TSTAR = XT/78.6

OMEGA = 1.16145/TSTAR**0.14874 + 0.52487*EXP(-0.7732*TSTAR)

+ + 2.16178*EXP(-2.43787*TSTAR)

VISCOT = 10.43676*XT**.5/OMEGA*1.E-7 RETURN

END

DOUBLE PRECISION FUNCTION CAL(I,X,T)

- * This subroutine provides calibration correlations for
- * I = 1: Spouting air rotameter 1 (the larger of the two)
 - 2: " " 2 (the smaller " ")
 - 3: Atomizing air rotameter
 - * 4: Sulfur rotameter
 - * 5: Urea Feeder
 - * 6: S-type pitot tube
 - * 7: Smaller pitot tube
 - * 8: Effective shutter diameter at the spouting air inlet
 - 9: Bed volume
- * Parameters are
- I = Type of equipment as given above
- X: for I = 1 to 4, X = float position (4 lower ball)
 - ^5, X = feeder setting
 - ^6 to^7, X = water manometer height difference in meters
 - ^8, X = diameter of shutter inlet, metres
 - ^9, X = bed height, metres
- T = density of air where the measurement was taken
- * ^ dummy value of T may be given for they are not used

* Units of CAL:

- * for I = 1 to 3: cubic meters/s
- 4 to 5: kg/s
- * 6 to 7: m/s
- * 8: m
- * 9: cubic metres

IMPLICIT REAL*8(A-H,O-Z) IF(I.EQ.2)THEN IF(X.LE.0.D0)THEN CAL = 0.D0 RETURN ENDIF Q = 0.0194 + 0.0058196*X - 1.12D-5*X*X + + 2.9982D-8*X**3 CAL = Q/T/60. RETURN ELSEIF(I.EQ.1)THEN

IF(X.LE.0.D0)THEN

RETURN ELSEIF(I.EQ.3)THEN IF(X.LE.0.D0)THEN CAL = 0.D0RETURN ENDIF С Q = (32.3 - 636.12/(X-41.2))/1000./60.С $CAL = Q^{1.2/T}$ CAL = (0.811+0.2085*X)/60./T/1000. RETURN ELSEIF(I.EQ.4)THEN IF(X.LE.0.D0)THEN CAL = 0.D0RETURN ENDIF CAL = (0.0117605*X - 0.047784)/60. RETURN ELSEIF(I.EQ.5)THEN CAL = (.749 - 1.485*X + .452*X*X)/60. RETURN ELSEIF(I.EQ.6)THEN CAL = 0.827*DSQRT(2*9.8*998.*X/T) RETURN ELSEIF(I.EQ.7)THEN CAL = 1.06*DSQRT(2*9.8*998.*X/T) RETURN ELSEIF(I.EQ.8)THEN R = 23./32.*.0254 IF(X.GT.R*2.)THEN PRINT*, ' *** DI BIGGER THAN POSSIBLE *** ' STOP ENDIF PI = DACOS(-1.D0)A1 = 1.+DCOS(PI/5.)A2 = 1.-DCOS(PI/5.)**2A = A2+A1**2

Q = 0.10089 + 0.0147172*X + 1.55D-5*X*X

+ 8.3131D-8*X**3

CAL = 0.D0

RETURN

CAL = Q/T/60.

ENDIF

+

C = (X-R)**2-R*RXL1 = (-B + DSQRT(B*B-4.*A*C))/2./AXL2 = XL1*DCOS(PI/5.)XL3 = DSQRT(XL1**2-XL2**2)XL4 = DSQRT(R*R-XL3**2)С XL5 = R - XL4A3 = 5.*XL2*XL3 + 5.*(DASIN(XL3/R)*R*R -+ XL3*XL4) CAL = DSQRT(4./PI*(A3-.8107318D-4))CAL = DSQRT(4./PI*(A3-1.*0.8107318D-4)) RETURN ELSEIF(I.EQ.9)THEN С PI4 = DACOS(-1.D0)/4.С ANG = 30. С DB = 0.04С HB = 0.01С AB = PI4 * DB* DBС VB = HB*AB С PHI = ANG/180.*PI4*4. С HN = DB/2./DTAN(PHI)С VN = HN*AB/3. С D = 0.24С A = PI4*D*DС HC = D/2./DTAN(PHI)С VC = A*HC/3.С VP = VC - VNС HT = HC-HN+HBС VT = VP+VB HT = 0.18320509VT = 0.31323016532D-2 A = 0.4523893780696D-1 $VA = A^{(X-HT)}$ CAL = VA + VTRETURN ENDIF PRINT *, '*** UNKNOWN CODE IN CALIBRATION SUB. ***' STOP END

B = -2.*A1*(X-R)

SUBROUTINE PTIN(L,N,A,B,C)

* This subroutine finds the minimum and maximum values of arrays A and B as well as determine the axis scales of plots. Parameters are L = no. of divisions in axis' N = no. of data in A and B A,B = array of x and y data respectively C = array containing the output values 1,6 = minimum x- and y-axis values respectively 2.7 = length of " " " " scales 3.8 = maximum " " " " values 4.9 = minimum value in A and B 5,10 = maximum " " " " REAL A(N), B(N), C(10) AM = A(N)BM = B(N)AS = A(1)BS = B(1)DO 10 I=1,N IF(AM.LT.A(I))AM=A(I) IF(BM.LT.B(I))BM=B(I) IF(BS.GT.B(I))BS=B(I) IF(AS.GT.A(I))AS=A(I) **10 CONTINUE** ADEL = (AM - AS)/LFACT=1. IF(ADEL.LT.10.)THEN 1=1 1000 FACT = 10.**I IF(ADEL*FACT.LT.10)THEN | = | + 1GOTO 1000 ENDIF ELSEIF(ADEL.GE.100)THEN 1 = -1 2000 FACT = 10.**1 IF(ADEL*FACT.GT.100)THEN |=|-1 **GOTO 2000** ENDIF

ENDIF ADEL = INT((ADEL*FACT+5.)/5.)*5/FACT A1 = INT(AS/ADEL)*ADEL IF(A1.LT.0.)A1 = A1-ADEL $A3 = ADEL^{+}L+A1$ IF(A3.LT.AM)A3=A3+ADEL BDEL = (BM - BS)/L FACT=1. IF(BDEL.LT.10.)THEN 1=1 3000 FACT = 10**I IF(BDEL*FACT.LT.10)THEN |=|+1 **GOTO 3000** ENDIF ELSEIF(BDEL.GE.100)THEN 1=1 4000 FACT = 1./10.**I IF(BDEL*FACT.GT.100)THEN | = | + 1**GOTO 4000** ENDIF ENDIF BDEL = INT((BDEL*FACT+5,)/5,)*5/FACT B1 = INT(BS/BDEL)*BDEL IF(B1.LT.0.)B1 = B1-BDEL B3 = BDEL*L+B1 IF(B3.LT.BM)B3=B3+BDEL C(1) = A1C(2) = ADELC(3) = A3C(6) = B1C(7) = BDELC(8) = B3C(4) = ASC(5) = AMC(9) = BSC(10)= BM RETURN END

SUBROUTINE MYPLO(L,M,N,A,B,XN,YN,T,C)

- * This subroutine plots data given in A and B. Multi plots in
- * one graph are possible per call. Parameters are
- * L = total number of data to be plotted
- * M = array containing the specifications of number & type of data
- * to be plotted, ie. 0=no symbols,just a line, 1=every point
- * connected by a line, -1=every point not connected by a line
- * N = array containing the number of data in the respective plots
- * A,B = x and y data
- * XN,YN,T = titles of x- and y-axis and plot
- * C = array containing the scales of the axis' usually determined
- * from PTIN subroutine

REAL A(L), B(L), C(10), X(200), Y(200) DIMENSION M(10), N(10) CHARACTER*50 XN, YN, T CALL DSPDEV('PLOT') CALL NOBRDR CALL AREA2D(6.0,6.0) CALL PHYSOR(1.,2.) CALL THKCRV(0.03) CALL XREVTK CALL YREVTK CALL BLSYM CALL YAXANG(0.) CALL XNAME(XN, 100) CALL YNAME(YN, 100) CALL YTICKS(2) CALL XTICKS(2) CALL COMPLX CALL HEADIN(T, 100, -3, 1) CALL GRAF(C(1),C(2),C(3),C(6),C(7),C(8)) C CALL GRAF(C(6),C(7),C(8),C(1),C(2),C(3)) CALL FRAME **MARK = 14** DO 10 J=1,N(1) X(J) = A(J)Y(J) = B(J)**10 CONTINUE** CALL MARKER(2) CALL CURVE(X,Y,N(1),M(1))

 $\begin{array}{l} \mbox{IF}(N(10).LE.1) \mbox{GOTO} \ 200 \\ \mbox{NP} = 0 \\ \mbox{DO} \ 20 \ I=2, \mbox{N}(10) \\ \mbox{NP} = \mbox{N}(I-1) + \mbox{NP} \\ \mbox{DO} \ 30 \ J=1, \mbox{N}(I) \\ \mbox{X}(J) = \mbox{A}(J+\mbox{NP}) \\ \mbox{Y}(J) = \mbox{B}(J+\mbox{NP}) \\ \mbox{30 CONTINUE} \\ \mbox{CALL MARKER}(\mbox{MARK+I}) \\ \mbox{CALL CURVE}(\mbox{X}, \mbox{Y}, \mbox{N}(I), \mbox{M}(I)) \\ \mbox{20 CONTINUE} \\ \mbox{200 CALL ENDGR}(0) \\ \mbox{CALL ENDPL}(0) \\ \mbox{RETURN} \\ \mbox{END} \end{array}$

SUBROUTINE MYSPLN(X,Y,Q,R,S,N,NDS) KI=KI0+I Interpolation using cubic splines with fitted end points, 20 CONTINUE Input: X Array of independent x-values A4=TAB(1) Y Array of dependent y-values B4=TAB(M) N Number of data points * Calculate H(I) Output: Q,R,S Coefficients of cubic spline equations DO 50 I=1,NM 50 H(I)=X(I+1)-X(I) IMPLICIT REAL*8(A-H,O-Z) REAL*8 Y(N),X(N),H(200),A(200),B(200),C(200),D(200),TAB(200) REAL*8 Q(NDS),R(NDS),S(NDS) A(1) = 0.D0B(1)=-H(1)* Write an error message if N < 2*M. C(1) = H(1)M=4 IF(N.LT.M)THEN DO 60 I=2,NM PRINT*, 'ORDER OF DIFFERENCES GREATER THAN NO OF DATA' IP=1+1 STOP IM=I-1 ELSEIF(N.LT.2*M)THEN A(I)=H(IM)PRINT*, 'NO OF END PTS TO BE FITTED EXCEED NO OF DATA PTS.' STOP C(I) = H(I)ENDIF A(N)=H(NM)* Find coefficients A4 and B4 using divided difference method. B(N)=-H(NM)* The method considered here only makes use of m end points on C(N) = 0.D0* either end. NM=N-1 MP=M+1 MM=M-1 KI0=N-M 11=1 DO 10 I=1.NM DO 70 I=1,NM IF(I.LE.MM.OR.I.GT.KI0)THEN IP=1+1 TAB(II)=(Y(I+1)-Y(I))/(X(I+1)-X(I))11=11+1 ENDIF RETURN **10 CONTINUE** END 10=11-M DO 20 J=3.M DO 20 I=1.MP-J KM=10+1

TAB(KM)=(TAB(KM+1)-TAB(KM))/(X(KI+J-1)-X(KI))TAB(I)=(TAB(I+1)-TAB(I))/(X(I+J-1)-X(I))* Coefficients of tridiagonal equations D(1)=3.D0*H(1)*H(1)*A4 B(I)=2.D0*(H(IM)+H(I))60 D(I)=3.D0*((Y(IP)-Y(I))/H(I)-(Y(I)-Y(IM))/H(IM)) D(N)=-3.D0*H(NM)*H(NM)*B4 * Call Thomas algorithm to solve tridiagonal set CALL TDMA(A,B,C,D,R,N) * Determine Q(I) and S(I) Q(I)=(Y(IP)-Y(I))/H(I)-H(I)*(2.*R(I)+R(IP))/3.70 S(I)=(R(IP)-R(I))/(3.*H(I))

SUBROUTINE TDMA(A,B,C,D,X,N)

Thomas algorithm

IMPLICIT REAL*8(A-H,O-Z) DIMENSION A(N),B(N),C(N),D(N),X(N),P(201),Q(201) NM=N-1 P(1)=-C(1)/B(1)Q(1)=D(1)/B(1)DO 10 I=2,N IM=I-1 DEN=A(I)*P(IM)+B(I)P(I)=-C(I)/DEN 10 Q(I)=(D(I)-A(I)*Q(IM))/DEN X(N)=Q(N)DO 20 II=1,NM 1=N-11 20 X(I)=P(I)*X(I+1)+Q(I) RETURN END

SUBROUTINE NC4AD(F,A,B,EPS,SUM,N)

* This subroutine utilies the Newton-Cotes four panel method * to integrate the function F between the limits A and B with an * accuracy of EPS. The integrated area is return as SUM and the * total number of function evaluation is returned as N. IMPLICIT REAL*8(A-H,O-Z) DIMENSION H(20), TOL(20), SR(20), XR(20), F1(20), F2(20), F3(20) DIMENSION F5(20), F6(20), F7(20), F8(20), F9(20), F4(20) * Initialization: set convergence tolerance slightly lower than * the calculated value. IMAX=20 N=5 SUM=0.D0 X1=A H(1)=(B-A)/4.D0 TOL(1)=63.D0*EPS * Calculate the step sizes and the tolerances at each level * up to 20 levels. DO 10 I=2,IMAX IM=I-1 H(I)=H(IM)/2.D0TOL(I)=TOL(IM)/2.D0 **10 CONTINUE** HIM=H(IMAX)/2.D0 XR(1)=B F1(1)=F(A) F3(1)=F(A+H(1))F5(1)=F(A+2.D0*H(1)) F7(1)=F(B-H(1)) F9(1)=F(B) * Calculate the total area in level 1. S=H(1)/22.5D0*(7.D0*(F1(1)+F9(1))+32.D0*(F3(1)+F7(1)) + +12.D0*F5(1)) i=1 20 N=N+4

 $\begin{array}{ll} F6(l)=F(X1+2.5D0*H(l)) \\ F8(l)=F(X1+3.5D0*H(l)) \\ SL=H(l)/45.D0*(7.D0*(F1(l)+F5(l))+32.D0*(F2(l)+F4(l)) \\ 1 & +12.D0*F3(l)) \\ SR(l)=H(l)/45.D0*(7.D0*(F5(l)+F9(l))+32.D0*(F6(l)+F8(l)) \\ 1 & +12.D0*F7(l)) \end{array}$

* If tolerance not met, divide the LHS area in two and redo

* the calculations at 20. IF(DABS(SL+SR(I)-S).GT.TOL(I))THEN

IM=I I=I+1 IF(I.GT.IMAX)THEN WRITE(8,70)X1 RETURN ENDIF S=SL F1(I)=F1(IM) F3(I)=F2(IM) F5(I)=F3(IM) F7(I)=F4(IM) F9(I)=F5(IM) XR(I)=X1+4.D0*H(I) * If the convergence criteria is satisfied, then add the area * and restart the calculations with the next section.

ELSE

SUM=SUM+SL+SR(I) X1=X1+4.D0*H(I) DO 40 J=1,I

IF(DABS(X1-XR(J)).LT.HIM) GOTO 50

40 CONTINUE

50 I=J IF(I.EQ.1)RETURN IM=I-1 S=SR(IM) F1(I)=F5(IM) F3(I)=F6(IM) F5(I)=F7(IM) F7(I)=F8(IM) F9(I)=F9(IM)

ENDIF

GOTO 20 70 FORMAT(1X,'warning - Integration Fails Beyond X = ',G12.5) END

Appendix IV: Calibration Results

In this section calibration results are provided for air, sulfur and urea flowmeters, and pitot tubes used in this work (and described in Chapter 3). The rotameters for the spouting air were calibrated against standard ASME orifice meter using 1/2" and 1 1/4" orifice sizes. The rotameter for the atomizing air was calibrated against Stainless Steel Wet Test Meter (manufactured by Fisher Scientific; capacity: 680 L/h; Model Type 63115). Sulfur and urea flowmeters were calibrated from direct measurements of the flow rates using a stopwatch and weight balance. Pitot tubes were calibrated against a standard ASME pitot tube in a 0.30 x 0.41 by 3.7 m long wind tunnel attached to an Axial Flow Fan (size 15; type B-VANE; manufactured by Canadian Blower & Forge Co. Ltd.).

The calibration results are shown in Figures IV-1 to IV-8. The solid lines in the figures represent the calibration equations as they were used in all calculations in this work. The calibration equations were derived using either linear or non-linear regressions using UBC NLP (Vaessen, 1984) available under the UBC MTSG (Runnals, 1989) main frame operating system, and are listed in Table IV-1. Table IV-1: Calibration equations for flowmeters and refractometer.

Figure	e <u>Flowmeter</u>	Calibration Equation
1	Spouting air rotameter 1	$Q_s\left(\frac{\text{kg}}{\text{min}}\right) = 0.0194 + 0.00482R_R - 1.12 \times 10^{-5}R_R^2 + 3.00 \times 10^{-8}R_R^3$
2	Spouting air rotameter 2	$Q_s\left(\frac{\text{kg}}{\text{min}}\right) = 0.0101 + 0.0147R_R - 1.55 \times 10^{-5}R_R^2 + 8.31 \times 10^{-8}R_R^3$
3	Atomizing air rotameter	$Q_a(L/min) = 0.811 + 0.209 R_R$
4	Sulfur rotameter	$W_s(\text{kg/min}) = 0.0118R_R - 0.0478$
5	Urea feeder	$Q_s(\text{kg}/\text{min}) = 0.749 - 1.49R_R + 0.452R_R^2$
6	1/16" pitot tube	C_o (Equation (5.6)) = 1.06
7	S-type pitot tube	C_o (Equation (5.6)) = 0.827
8	Refractometer	$C_u(g/mL) = 7.20\Delta(R_I) - 63.5\Delta(R_I)^2 + 3240\Delta(R_I)^3$
		where $\Delta(R_I) = R_I - 1.3345$; 1.3345 = R_I for distilled water

 R_R = Rotameter reading or urea feeder setting; R_I = Refractive index reading.



Figure IV-1: Calibration curve for the lower capacity spouting air rotameter.



Figure IV-2: Calibration curve for the higher capacity spouting air rotameter.



Figure IV-3: Calibration curve for the atomizing air rotameter.



Figure IV-4: Calibration curve for the sulfur rotameter.



Figure IV-5: Calibration curve for the urea feeder.



Figure IV-6: Predictions using the calibrated value of C_o for the static-pitot tube.



Figure IV-7: Predictions using the calibrated value of C_o for the S-type pitot tube.



Figure IV-8: Calibration curve for the Abbey refractometer.