THICKNESS MEASUREMENT OF SOLID HYDROGEN THIN FILM
FOR MUON CATALYZED FUSION
VIA ENERGY LOSS OF ALPHA PARTICLES

By

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Abstract

A novel target system for films of solid hydrogen isotopes has enabled unique experiments in muon catalyzed fusion. In order to understand the experimental data a knowledge of target thickness and uniformity is essential, but only indirect information was available. Conventional techniques for a thickness measurement do not apply, due to the limited available space and cryogenic requirements of the system. In this thesis, a method of thickness and uniformity measurement via the energy loss of alpha particles is presented. A critical review of the literature on the stopping powers of alpha particles was necessary, given no experimental data for solid hydrogen.

An absolute precision of ~5% at optimal condition was obtained in the thickness determination. The uncertainty in the relative measurements can be less than 1%. The average target thickness per unit gas input, weighted by the Gaussian beam profile of FWHM 20-25 mm is determined to 3.29±0.16 μg/(cm²-torr-litre). A significant non-uniformity in the thickness distribution was observed with an average deviation of about 7%. The linearity of deposited hydrogen thickness upon gas input was confirmed within the accuracy. The cross contamination from the other side of the diffuser nozzle is found to be less than 0.8 × 10⁻³ with 90% confidence level. The result is compared to a Monte Carlo study to understand deposition mechanism.

The importance of the stopping process in the alpha-sticking problem in muon catalyzed D-T fusion is discussed in detail. The physical phase effect of the stopping power of hydrogen may partly explain the discrepancy in the sticking values between theory and experiment at high densities. The concept of a new experiment to measure directly the sticking probability at high density is proposed. This offers certain advantages over
LAMPF/RAL measurements. A Monte Carlo simulation of the experiment is performed. A very preliminary result from a test run is presented.
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Chapter 1

INTRODUCTION

A novel target system of solid thin films has been developed at TRIUMF for the experiments studying the reaction of muonic hydrogen isotopes. Of the main interest with this new system is the study of processes in muon catalyzed fusion ($\mu$CF). Some unique measurements have been conducted and many more will come in the near future. This thesis will concentrate on characterizations of the solid thin targets in terms of thickness and uniformity. Following the introduction in this chapter, the principle will be discussed in Chapter 2. Chapter 3 treats the energy loss of the charged particles in detail. The experiments and data analysis is described in detail in Chapter 4 and 5, respectively. The results will be presented and discussed in Chapter 6. Chapter 7 is devoted to the applications of the knowledge of stopping processes to other problem in $\mu$CF.

1.1 Muon Catalyzed Fusion

Since its discovery in cosmic rays in 1937, the muon has played an important role in our understanding of nature. The muon has been extensively studied to determine its own properties and interactions as well as for a probe to reveal the nature of other particles and nuclei. Despite these efforts over 50 years, its existence itself is still considered to be one of the biggest mysteries in modern science. The muon has a mean life of 2.2 $\mu$s which is the second longest for all unstable particles (under weak decay) discovered so far. Similarly to the neutron, the longest lived unstable particle, it provides a rich variety of applications in diverse areas of science including condensed matter, medical
and archeological research, and perhaps, energy production.

Although its fundamental properties and interactions are of tremendous interest, much of the behaviour of a negative muon can simply be described, for our purpose regarding catalysis of nuclear fusion, by that of a heavy electron. When a muon penetrates matter, it gets rapidly thermalized and captured by an atom replacing an orbiting electron. Since the mass of muons is roughly 207 times that of electrons, the binding energy of the muonic atoms is about 207 times larger and the dimension 207 times smaller than that of ordinary atoms. A muonic hydrogen atom can approach another nucleus to a much closer distance than a normal atom or a bare nucleus can, because it is a compact neutral object. When a muonic hydrogen collides with other atoms, a muonic molecular ion may be formed. Again, its dimension is about 200 times smaller than diatomic molecular ions such as $\text{H}_2^+$. The striking difference in the muonic molecule is, however, that because of the closer internuclear distance, the tunneling of nuclear wave functions is enhanced exponentially and nuclear fusion may occur very rapidly, except of course for the $\mu\mu\mu$ case. After the fusion, most of the time the muon is released and participates in another series of processes leading to fusion.

Fig. 1.1 illustrates the simplified cycling processes in one of the most interesting systems, a deuterium - tritium mixture. A negative muon incident on a $\text{D}_2/\text{T}_2$ target gets captured by either d or t forming $\mu\text{d}$ or $\mu\text{t}$, respectively. The muon in $\mu\text{d}$ transfers to a triton due to its deeper Coulomb potential. A muonic molecule $\mu\text{d}$ $\mu$ $\text{t}$ is then formed mainly via the resonance mechanism which will be described later. Almost immediately after the $\mu\text{d}$ $\mu$ $\text{t}$ formation the fusion takes place producing a neutron and an alpha particle with an energy release of 17.6 MeV. A small fraction (~ 0.6%) of muons get attached to the $\alpha$ after the fusion. This probability is called the (effective) alpha sticking coefficient.

---

1In fact, initially ~ 1% of muons stick, but about 1/3 of them get stripped from the helium nuclei due to collisions with the atoms in the target.
Figure 1.1: Simplified diagram of muon catalyzed fusion cycle in a D$_2$/T$_2$ mixture.

$\omega^{\mu l}$, and is known to give the most stringent constraint for practical application of $\mu$CF to energy production. Nevertheless, the fusion yield of more than 100 per muon is reported in many independent experiments and active research is continuing around the world.

Muonic molecular formation, as well as $\alpha$-sticking, are the two most important processes in $\mu$CF. The process originally considered is a capture of muonic atoms on one of the nuclei in the hydrogen molecule, where the binding energy of the muonic molecule is released and transferred to an Auger electron. It is, in general, for any hydrogen isotope system, written as:

$$ \mu x + YZ \rightarrow [(x\mu y)z\epsilon]^+ + e, \quad (1.1) $$

where lower case $x$, $y$ and $z$ are the nuclei of hydrogen isotopes and the upper case represents an atom. This process is rather slow and a muon could only catalyze, at most, a few fusions in its lifetime. Another process was proposed by Vesman[1] to explain the
unexpectedly high fusion rate in the $d\mu d$ system. The process, in general, can be written as:

$$\mu x + YZ \rightarrow [(x\mu y)zee]^*.$$  \hspace{1cm} (1.2)

In the Vesman mechanism, because of the existence of a loosely bound state in the muonic molecule, the binding energy is absorbed as rotational and vibrational excitation quanta in the 6-body molecular complex. The muonic molecule $x\mu y$ and the hydrogen nucleus $z$ constitute the two nuclei of the compound molecule, with the symbol $^*$ referring to its excited state. For the $d\mu t$ system the reaction rate is, indeed, enhanced by about 100 times by the resonance process. Due to the delicate mechanism of the resonance, the molecular formation process shows a strong temperature dependence. Hence, the total fusion rate depends on temperature as well. It is interesting to note that in $\mu$CF phenomena, a nuclear reaction which releases millions of electron volts of energy, is affected by a temperature which typically has an energy scale of milli-electron volts. Here we see a beautiful interplay of nuclear and atomic/molecular physics. Thus, detailed understanding of molecular formation is of essential importance in $\mu$CF research.

1.2 Experiments with Hot Muonic Atoms from Cold Targets

History tells us many of the discoveries in science were serendipitous. Often people were looking for one thing, and found something else that was totally unexpected. The muonic hydrogen experiments at TRIUMF have a similar story. They were originally designed to search for the emission of $\mu p$ into vacuum in its excited state, for a precision test of Quantum Electrodynamics. To the dismay of the group, this was not realized. After several runs, however, it was found that energetic $\mu d$ atoms, instead of $\mu p$, were emitted in large amounts. Thus, the source of a neutral “beam” of hot $\mu d$ was discovered.

The importance of isolated muonic hydrogen in vacuum was immediately realized as
a tool for research in \( \mu \)CF and related processes. Following this discovery, a new versatile target system for solid thin layers was developed for the study of a variety of the reactions of muonic hydrogen isotopes.

Fig. 1.2 illustrates the schematic view of the target system. Two thin target foils made of 51 \( \mu \)m gold are supported by 1.6 mm copper frames. They are connected to the cold head of the cryostat which is cooled to 3 K by pumping on liquid helium. Target spacing can be varied from 15 mm to 40 mm. A 90 K copper thermal shield protects the system from radiation heating. All the copper present in the system is gold plated in order to reduce the emissivity.

The hydrogen is introduced through the gas deposition mechanism (diffuser) which can be inserted between the two target support frames. It has many small holes through which the gas is released towards either one of the target foils. The gas condenses and solidifies when it reaches the cold foil surface. The system is kept in an ultra-high vacuum of \( 10^{-8} - 10^{-10} \) torr except during the gas deposition when the pressure goes up to a few times \( 10^{-6} \) torr. All three isotopes of hydrogen, as well as other gases such as neon, can be used in the targets, both pure and in mixtures. Although the handling of tritium requires special attention due to its radioactivity, it is an essential ingredient in the high cycling muon catalyzed processes.

One of the main goals in the E613 experiment at TRIUMF using the target system mentioned above is the energy-dependent measurements of reaction processes via the time of flight method. Fig. 1.3 illustrates one such measurement, where energy-dependent molecular formation rate can be investigated in the following manner. The muon beam is stopped in solid protium (\( ^1\text{H}_2 \)) with a concentration of tritium of one part in a thousand (\( C_t = 10^{-3} \)). In this upstream target, muonic protium \( \mu p \) is formed and the negative muon is transferred to a triton forming \( \mu t \). Upon the transfer the difference in the binding energies gives the \( \mu t \) a kinetic energy of \( \sim 45 \) eV in the laboratory frame.
Chapter 1. INTRODUCTION

Figure 1.2: Schematic view of the solid hydrogen target system, taken from Ref. [2].
The $\mu t$ loses energy via elastic collisions on protium.

In general, for low energy collisions where the $l = 0$ partial wave is dominating, the scattering cross section can be written as

$$\sigma \sim \frac{4\pi}{k^2} \sin\delta_0,$$

where $\delta_0$ is the phase shift and $k$ the wave vector of the projectile. When $\delta_0$ has a value of $n\pi$ ($n = 1, 2, 3, ...$), the cross section goes to nearly zero. This is what is known as the Ramsauer-Townsend effect and was first discovered in low energy electron scattering on rare gas atoms[3]. For the Ramsauer-Townsend effect to occur, the presence of an attractive potential is necessary in order to produce a rapid change in the phase of the wave function. A repulsive potential alone cannot cause the effect, since the phase change in the potential is slow and a strong potential would be required to shift the phase by $\pi$. This would result in an increase in contributions from higher partial waves, therefore even though the $l = 0$ partial cross section goes to zero, the total scattering cross section still remains finite. Note that in the present $\mu t + p$ system, despite the presence of a strong repulsive potential at very small inter-nuclear distance, the scattering amplitude is dominated by the contribution from the attractive potential at larger inter-nuclear distances. Hence due to the Ramsauer-Townsend effect, the elastic scattering cross section of $\mu t$ on protium drops by several orders of magnitude at a $\mu t$ energy of the order of 1 eV. This results in the protium being nearly transparent for $\mu t$, and $\mu t$ is emitted into the neighboring vacuum with a velocity of the order of a few mm/μs. Thus we have a neutral “beam” of energetic muonic tritium.

When the $\mu t$ “beam” is incident on the second target foil (downstream foil) which holds a thin layer of D$_2$ the $\mu t$ interacts with D$_2$ to form the muonic molecule d$\mu t$. Formation of the d$\mu t$ molecule can be detected because of the almost immediate fusion reaction which occurs, producing an alpha particle and a neutron.
Figure 1.3: Measurement of energy-dependent molecular formation rate via time of flight experiment.
A recent theoretical calculation predicts strong resonances in $d\mu t$ formation at an energy of the order of 1 eV\[4\]. The energy range corresponds to a temperature of $\sim 10,000$ K and is inaccessible by conventional targets. The target system described here gives a unique way to measure the molecular formation cross section in the predicted resonant energy region. Furthermore, it provides event by event energy information, via the time of flight method, allowing an energy-dependent measurement of the cross sections. Given the distance of two target foils, the time interval between the entry of the muon and the fusion reaction gives the velocity, hence energy, of the $\mu t$, except for the unknown angle of emission. The angular dispersion of the emitted $\mu t$ can be reduced by using a collimating device. The time taken by the muon to be emitted after stopping, and the time taken for fusion after the molecule is formed, are both very fast compared to the time of flight, and can be neglected. A recent Monte Carlo study indicates the importance of the contribution from the $\mu t + d$ elastic scattering process\[5\]. This has to be carefully considered in extracting the molecular formation cross section.

The emission of a $\mu t$ in vacuum was observed for the first time in the December 1993 run. The principle of the measurement has been proven, which yielded a number of unambiguous fusion events with time of flight information\[6\]. The measurement with optimized conditions has just taken place in July-August 1994. The analysis of the result is now in progress.
Chapter 2

TARGET THICKNESS MEASUREMENT

2.1 Motivation and Goal

For any experiment in science, having a good understanding of the experimental system is essential. Our solid hydrogen isotope target system is no exception to this rule. Knowledge of the target thickness and uniformity is important, in particular, for the measurements of molecular formation and scattering cross sections, since it limits the precision of the measurements. The uncertainty in the thickness directly propagates to the final results. Also for X-ray measurements, the thickness of the layer affects the absorption of photons, which is an important correction for the absolute intensity.

As mentioned earlier, the target we use in the beam experiments includes all three isotopes of hydrogen and their mixtures, as well as other elements such as neon. The thickness ranges from a few $\mu$g/cm$^2$ to a few mg/cm$^2$, corresponding to a few hundreds of nanometers to a few mm for hydrogen.

The goal of precision for the $\mu$CF experiments discussed in section 1.2 is perhaps 10%, therefore a few percent accuracy in the thickness measurement should be aimed at. It should be noted, however, that if the variation of the thickness in the film is greater than the uncertainty of the thickness measurement, it is the former that dominates the uncertainty of the final result. Also, there is some uncertainty over the control of gas input in the deposition process. This becomes increasingly important for very thin targets. Hence the pursuit of better precision in the thickness measurement would be
less important, in the case where the above factors are dominating.

In addition to the E613 experiment at TRIUMF, the present measurement may give some insights to other experiments which use solid thin films as a target. A novel method on slow negative muon production via $\mu$CF proposed by Nagamine[7] uses a similar target and gas deposition mechanism[8, 9]. The uncertainty in the target film thickness is considered as one of the possible causes for disagreements between the recent measurements and the simulation[10]. Another example is an experiment on low energy kaon-nucleon interactions at DAΦNE, a $\phi$ factory. Olin et al. proposed the use of a solid hydrogen target inside the collider beam pipe[11]. The unique target system, combined with tagged, mono-energetic kaons from $\phi$ decay, is expected to provide unprecedented statistics in a low background environment for low energy kaon studies. Furthermore, the possibility of utilizing a solid target for the measurement of $\alpha$ sticking processes, probably the most important processes in terms of the practical application of $\mu$CF, is currently being investigated. The present measurement is hoped to provide useful information for these ongoing or potential experiments.

2.2 Previous Measurements

2.2.1 Pressure and Volume

Prior to the present measurement, some information on our target thickness was available. One is described here and the others in following sections.

The first measurement was done as follows. The hydrogen gas was deposited onto a cold foil while pumping the system with a cryopump. After the vacuum system was closed, the cryostat was warmed up to evaporate the frozen hydrogen. By measuring the total pressure, combined with the knowledge of the total volume of the system, one can estimate the amount of gas which actually sticks inside the system. This method tells
us that roughly 83% of the gas stays inside the system[12]. However, it does not provide the information on whether the gas sticks to the target foil at which the muon beam is directed. It could have been deposited anywhere in the vacuum system.

2.2.2 Muon Stops

Another piece of information came from the muon stopping signals. 99.9% of the muons which stop in the hydrogen decay and emit an energetic electron. It can be detected by an array of wire chambers, and the position of the decay can be estimated by tracing back the electron track. The spectrum of the decay electrons from muons stopping in hydrogen has a characteristic time constant of about 2.2 μs, and can be distinguished from the ones stopped in heavier material in the system, which have much smaller life times due to the capture on a nucleus via the semi-leptonic weak interaction. By looking at the yield of the decay electrons one can estimate how much hydrogen is on the target foil. This method, however, is subject to a large systematic uncertainty and it is very difficult to obtain the absolute thickness.

2.2.3 Cross Contamination

If the hydrogen molecule does not stick to the cold foil at first contact, it could bounce around the diffuser and stick to the other cold foil, causing cross contamination of the targets. This would make the experiments using two target foils of different compositions such as the one described in section 1.2 impossible.

The cross contamination was checked by observing the yield of μd emission. Since the mechanism of emission is based on the subtle condition of the Ramsauer Townsend resonance, its yield is very sensitive to any contamination on the surface. The μd emission target was first prepared in the upstream foil, and emission was observed. The thick deuterium target was then deposited in the down stream foil. If there was a significant
contamination from the downstream target on the upstream one, it would affect the emission yield of $\mu d$. The comparison with the case where a known amount of $D_2$ was intentionally put on top of the emission target, gives the upper limit of cross contamination to be less than $\sim 1\%$.

All the information discussed in this section is rather indirect. None of it gives the absolute thickness nor the uniformity. This leads us to perform a specific experiment for more direct thickness measurement, and its principle is described in the following section.

### 2.3 Principle of the Method

There are a number of conventional ways for measuring the thickness of thin films, such as optical interferometry and microscopy. However, the spatial limitations and cryogenic requirements of the target system do not allow such methods.

For condensed gases, a few methods have been reported. For example, Sørensen et al. used a quartz crystal oscillator to measure the thickness of solid hydrogen films [13] for the measurement of ranges of keV electrons[14]. This is a common technique for ordinary films made from evaporation. From the frequency change of oscillation, the amount of gas frozen on the quartz is determined. However, they found at least 40% non-linearity in the frequency change–thickness relation with a $D_2$ film as thin as 40 $\mu m$. The signal from the oscillator also deteriorated with increasing film thickness and it finally stopped oscillating. They attributed this to the smallness of the densities of hydrogen and deuterium. It should be recalled that our target thicknesses range up to 1 mm or more. Also it would not be possible to test the uniformity of films in this method, unless multiple crystals are used.

Chu et al. measured the thickness of argon, oxygen and CO$_2$ films with the Rutherford
Backscattering (RBS) method[15]. This popular technique for ordinary thin films is known to give fairly accurate results, provided there is an accurate calibration sample[16]. However, due to the small cross section for backward scattering, a dedicated accelerator is necessary for this measurement to provide sufficiently intense beam of ions.

We will use a method which meets the requirements and limitations imposed by the target system and still is relatively simple: By depositing a film directly on top of a radioactive source, and by measuring the residual energy of the transmitted particles, the energy loss of the particles is obtained. The energy loss can be related to the thickness of the film using the stopping power of charged particles in the material. Direct deposition provides measurements with better accuracy and wider dynamic ranges. The uniformity of the film can be easily determined by using an array of sources and sampling the different positions. The knowledge of the energy loss process of charged particles in matter is essential for this method. This will be discussed in detail in the following chapter.
Chapter 3

ENERGY LOSS OF CHARGED PARTICLES

The energy loss process of charged particles has been studied since the beginning of the century. A precise understanding has been demanded not only in nuclear physics, but also in medicine, biology, material science, device research and many other fields. These processes for charged particles include electronic excitation, ionization, nuclear collision, Cherenkov radiation and bremsstrahlung.

Heavy\(^{1}\) charged particles in matter lose the energy mainly via inelastic interactions with the bound electrons of the medium (electronic stopping power). Elastic Coulomb collisions in which recoil energy is imparted to atoms (nuclear stopping power) becomes important at very low velocity. For example, the nuclear stopping power contributes more than 1\% of total stopping power only below 150 keV in hydrogen for alpha particles\(^{17, 18}\). Radiative energy loss due to emission of bremsstrahlung (radiative stopping power) is significant for electrons and positrons, but negligible for heavier particles, since it is inversely proportional to the square of projectile mass. Other processes such as nuclear reactions and Cherenkov radiation have also only negligible contributions for heavy particles unless extremely relativistic. The latter is included in the standard formula of stopping power mentioned below.

The following sections review different aspects of the energy loss mechanism. Section 3.1 treats relativistic particles in terms of the Bethe-Bloch formula. The lower energy region is discussed in section 3.2. The effect of physical phase on energy loss is

\(^{1}\)By “heavy" it is meant that the mass of the particle is comparable to or greater than that of the nucleus.
considered in section 3.3.

3.1 Bethe-Bloch Theory

For moderately energetic particles, the electronic energy loss per unit path of the particle is derived in the first order Born approximation which is commonly known as the Bethe-Bloch formula for stopping power. This is described in many introductory nuclear physics textbooks, e.g., see [19]. The theory considers a particle interacting with an isolated atom of harmonic oscillators[20] and the assumptions include that the electron is moving slowly with respect to the incident particle and the projectile has a large mass compared to the electron. With two commonly used corrections the formula is written as:

\[
\frac{dE}{dx} = 4\pi N_A r_e^2 m_e^2 c^2 z^2 Z \frac{1}{\lambda} \left( \ln \frac{2 m_e c^2 \gamma^2 \beta^2}{I} - \beta^2 - \frac{\delta}{2} - \frac{\sum_i C_i}{Z} \right)
\]

with

\[
4\pi N_A r_e^2 m_e^2 c^2 = 0.3071 \text{ MeV cm}^2/\text{g}
\]

where

- \(N_A\) : Avogadro’s number
- \(r_e\) : classical electron radius
- \(m_e\) : electron mass
- \(z\) : charge of incident particle in units of \(e\)
- \(\beta = \frac{v}{c}\) of the incident particle
- \(\gamma = \frac{1}{\sqrt{1-\beta^2}}\)
- \(Z\) : atomic number of absorbing material
- \(A\) : atomic weight of absorbing material
- \(I\) : mean excitation potential
- \(\delta\) : density correction
3.1.1 Mean Excitation Potential

The mean excitation potential $I$ is the prime parameter in the formula, and the nature of the target materials is concentrated in this number. It is theoretically defined for gases\[21\] as:

$$\ln I = \frac{\int_0^\infty \frac{df}{dE} \ln E dE}{\int_0^\infty \frac{df}{dE} dE},$$

where $df/dE$ is the density of optical dipole oscillator strength ($f$) per unit energy of excitation ($E$) above the ground state. The oscillator strength is proportional to the photo-absorption cross section, but use of this is justified only for dilute gases for which there is only a weak correlation between the positions of the electrons in the medium\[22\]. For condensed matter, instead, $I$ is expressed in terms of the dielectric-response function $\epsilon(\omega)$,

$$\ln I = \frac{2}{\pi \omega_p^2} \int_0^\infty \omega \text{Im}[-1/\epsilon(\omega)] \ln(\hbar \omega) d\omega,$$

where $\omega_p$ is the electron plasma frequency, which describes the collective response of electrons to a disturbance. $\epsilon(\omega)$ is defined in $D = \epsilon(\omega)E$, and is generally a complex number. For non-magnetic materials, it can be related to the refractive index\[22\]. The classical damped harmonic oscillator model is known to give the same value of $\epsilon(\omega)$ as the corresponding quantum mechanical calculation\[23\].

The value of the mean excitation potential depends on the electronic structure of the material, and is very difficult to calculate accurately except for the simplest atomic gases. Empirical formulas for the $Z$-dependence of $I$ exist\[19, 24, 25\], e.g. $I/Z = C_i$ : shell correction for $i$ th shell

Some important points are discussed in following sections.
Chapter 3. ENERGY LOSS OF CHARGED PARTICLES

$9.76 + 58.8/Z^{1.19}$ for $Z \geq 13$, but it is known that $I$ does not depend on $Z$ smoothly, due to the effects of the atomic shell structures.

Most of the time values for $I$ have been determined for each element from actual stopping power measurements by fitting the data to equation 3.1. For the element for which data is not available, the $I$ value has to be deduced from semi-empirical interpolation. For some simple gaseous materials, when optical data are abundant, $I$ values can be determined by fitting the experimental polarizability to get semi-empirical oscillator-strength distributions.

Determination of $I$ values is one of the main tasks for authors of stopping power tables. The most recent compilation of $I$ values was made by Berger and Seltzer in preparation for the stopping power table of electrons and positrons\cite{22, 26}. Since the $I$ value is independent of projectile, it can be applied to heavy charged particles as well. They quote values as accurate as 1–2% for Al and Ag, for example. However, it is worth mentioning that Sabin et al. recently commented\cite{27} that it is impossible to determine the experimental mean excitation energies with precision better than $\sim 5$ eV for Al. They claim an experimental value of $I$ with less than $\sim 5\%$ uncertainty is useless as long as other correction terms such as shell, Barkas and Bloch corrections are not known accurately. As will be mentioned, these corrections are obtained by fitting the experimental stopping powers. Therefore, the experimental $I$ value depends on the choice of the corrections.

3.1.2 Density and Shell Correction

The correction due to the density effect $\delta$, also called the polarization correction, is only important at very high energy. The polarization in the medium atoms caused by

\footnote{The Particle Data Book cites the latter article, which is much more difficult to obtain yet contains less information. In the opinion of the present author, the former should be cited instead.}
the projectile perturbs the electron field, reducing the stopping power. A semiempirical formula was developed by Sternheimer and values obtained by employing claimed up-to-date values of $I$ are found in Ref. [25, 28]. The shell correction $C_i$ is to compensate the effect that electrons in inner shells of atoms do not participate in the energy loss processes of the projectile. This is more important at lower energy. Again, this is not precisely known and a semiempirical formula must be employed[29, 30].

### 3.1.3 Higher Order Correction

Higher order terms in the projectile charge $z$ are sometimes used for the correction due to departures from the first Born approximation. The Barkas correction is proportional to $z^3$, giving the different stopping powers between positively and negatively charged particles. This is named after Barkas who first observed in the 1950's that the range of negative pions is longer than that of positive pions. This is still an active field of research both experimentally and theoretically. For example, the antiproton proton stopping power in hydrogen below 120 keV was recently measured at Low-Energy Antiproton Ring at CERN by Adamo et al.[31]. The maximum in the antiproton stopping power was about 60% of that for proton, showing a significant Barkas effect.

The Bloch correction takes into account the perturbation of the wave functions of the atomic electrons due to the incident particles (quantum mechanical impact-parameter method). This is derived without the use of the first-order Born approximation and the correction is important only at large projectile velocities. With this correction added, the high energy limit of equation 3.1 approaches the classical stopping formula of Bohr.
3.2 Low Energy Region

3.2.1 Break Down of the Bethe-Bloch Theory

With corrections described above, the Bethe-Bloch formula is known to give results accurate to a few percent for protons and alpha particles of energies down to $\beta \sim 0.05$ ($\sim 10$ MeV for alphas, $\sim 2.5$ MeV for protons). However, many of the assumptions in the Bethe-Bloch formula start to become inadequate at lower energies. In the low energy limit, stopping power in the Bethe-Bloch formula equation 3.1 is inversely proportional to the kinetic energy of the projectile. However, after a certain maximum, the actual stopping power decreases with decreasing energy. Clearly, even with various corrections, the theory of Bethe-Bloch, which proves so successful in the relativistic regions, breaks down at low energies. This is the case typically below $\beta \sim 0.05$.

An additional complication for projectiles with $Z > 1$ is that, at low velocities, ions start to have partially bound electrons, and their effective charge states are no longer equal to $Z$. Reduced charge states result in lower stopping powers. This effect is more significant at lower velocities, and the ions are finally neutralized at very low velocity. According to Ziegler et al., despite a long controversy, a consensus seems to exist which claims that protons always exist as bare nuclei with an effective charge equal to one, at least in the condensed targets\[20\]. However, it is pointed out by Senba that in noble gases (in gaseous phase) nearly 100% of protons with initial energy of 100 MeV experience an electron capture process at least once by the time they slow down to 1 MeV\[32, 33\].

3.2.2 Varelas-Biersack Formula

Below $\beta \sim 0.05$, no satisfactory theoretical prediction of stopping powers is available. Therefore an empirical fit to the experimental data is the only way. Varelas and Biersack proposed a phenomenological formula for low energy stopping powers with five
parameters\[34\]:

\[
\frac{1}{S} = \frac{1}{S_{\text{low}}} + \frac{1}{S_{\text{high}}},
\]

(3.4)

where \( S \) is the electronic stopping power and \( S_{\text{low}} \) (low energy stopping) is

\[
S_{\text{low}} = A_1 E^{A_2}
\]

(3.5)

and \( S_{\text{high}} \) (high energy stopping) is

\[
S_{\text{high}} = \frac{A_3}{E} \ln(1 + \frac{A_4}{E} + EA_5)
\]

(3.6)

This formula is used by several authors to bridge between the Bethe-Bloch region and the very low energy region where some theoretical guide exists. At the very low energies, namely of the order of keV, stopping powers proportional to the projectile velocities are predicted by the free electron gas model. Many stopping power tables assume this relationship or a similar one\[^3\].

### 3.3 Effect of the Physical Phase

Because the present work involves solidified gases, it is important to consider the effect of the physical phase on stopping powers. Unfortunately, no experimental data exists, to the author’s knowledge, for solid hydrogen and a charged particle with energies of interest to us. Therefore, in this section, we will discuss existing studies of the issue in detail.

#### 3.3.1 Overview

An obvious effect of the physical phase on stopping powers is the polarization effect at relativistic energies due to the density change (the density effect in the Bethe-Bloch

\[^3\]A recent measurement reports, however, a departure from the velocity proportionality\[35\].
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formula). This is theoretically rather well understood as discussed in section 3.1. However for protons it reaches the 1% level only above 500 MeV, so clearly it is negligible for us. A channeling effect occurs in the crystalline structure of solids. In a single crystal material, it can reduce stopping power by as much as 30% in a certain direction, but our targets are likely to be multi-crystal, so no observable effect is expected. In fact, the measurements of stopping powers of a few MeV $^4$He ions in frozen gases by Chu et al., who used a pin hole deposition mechanism, report no such effects after measuring at several different angles[15].

The phase effects can be expected, at least in the Bethe-Bloch region, to reveal themselves in a higher value of $I$, the mean excitation potential. Experimental photoabsorption studies, as well as theoretical considerations show general upward shifts in the dipole oscillator strength distributions in solids compared with gases. Changes in outer electron arrangements may lead to changes in electronic excitation levels as aggregation occurs. For example, in Berger and Seltzer’s compilation of mean ionization potentials[22], gaseous and liquid hydrogen have different $I$ values, namely, 19.2 ± 0.4 eV for molecular gaseous hydrogen and 21.8 ± 1.6 eV for liquid hydrogen. The liquid value is obtained by reanalyzing data from the year 1952 and has a large uncertainty. Notice stopping power depends only logarithmically on the mean excitation potential, therefore the change in the stopping power $\Delta S/S$ should be smaller than the change in mean excitation potential $\Delta I/I$, that is $\Delta S/S \leq \Delta I/I$.

Early studies on phase effects were done in water and organic material using $\alpha$ particles. For a review, see [36]. They are conflicting in the magnitude, the sign and even in the existence of such effects. Later studies show a consistent tendency of significant differences for $\alpha$ particles in the energy region of 0.3 to a few MeV. A survey of stopping power data in hydrocarbons and related materials was done by Thwaites and Watt[37], and they concluded that stopping powers in gases are greater than in solids.
and liquids. A similar comparison by Ziegler et al.[38] describes the averaged ratio of \((\text{experimental/theory})_{\text{gas}}/(\text{experimental/theory})_{\text{solid}}\). The ratio is greater than unity for \(\alpha\) particles below \(\sim 4\) MeV in low Z material, and increases to as high as 1.4 at 0.5 MeV.

Recent reviews[39, 40] conclude the existence of 5–10% physical effects for protons and alpha particles at maximum stopping power energies in organic and similar materials. It is also suggested that for compound materials, the chemical binding significantly changes the stopping power, hence causing a break down of Bragg’s additivity rule \(^4\).

### 3.3.2 Heavy Ions

Stopping powers of heavier ions are measured in heavy ion accelerators such as GSI, Orsay and Chalk River. Although it is not directly relevant to our measurement, it may be instructive to look at the phase effects for heavy ions. Significant gas-solid effects are reported for 2-6 MeV/nucleon Cu, Kr and Ag projectiles[41], the gas stopping powers being lower than those of solid media. The effect is higher for the lighter degraders and heavier projectiles. The effect is explained to be due to an enhancement of the effective charge, namely the ionic charge state in a solid degrader. Because of the shorter time interval between successive collisions, the ionization rate in solid is higher, resulting in a higher effective charge. The effect was negligible in Ne and Ar[42]. It should be noted, however, that their measurements were done only in gases, and comparisons with solids rely on the theoretical calculations by one of the authors of the paper. On the other hand, Geissel et al. actually measured stopping powers of 3.6–7.9 MeV/u uranium both in gases and in solids of various z[43]. They found up to 20 % greater stopping in solid than in gas. Note that although these author call these “density effects,” it should not

\(^4\)It states that the stopping power of a compound is given by the average of the stopping powers of each element weighted by their composition.
be confused with the density effect corrections in the Bethe-Bloch formula, which gives smaller stopping powers for a higher density medium when the projectile is relativistic.

3.3.3 Condensed Gases

Several measurements exist for the stopping powers of frozen gases. For low energies, the situation is rather confusing. Børjesen et al. reported [44] a factor of two lower stopping power for 1–2 keV protons in solid nitrogen than gas. It was also shown [45] that, while the electronic stopping power of keV protons in solid H₂ and D₂, and of keV deuterons in solid H₂ is closely identical to that in a gas, for deuterons in solid D₂ the stopping power is only a half as large. It should be noted that the D₃⁺ ion was used for the latter, after they found no difference between H⁺, H₂⁺ and H₃⁺. For a review of stopping power for keV light ions in condensed molecular gases, see Børjesen [46].

In the MeV range, stopping powers for frozen gases were measured by Chu et al. in solid argon, oxygen and carbon dioxide [15]. They found a 5% lower stopping power in the solid from 0.5–1 MeV than in the gas, although no significant phase effect from 1 to 2 MeV was observed. Solid argon was also measured by Besenbacher et al., who found no phase effect within a 3% uncertainty [47].

As we have seen in this section, the current status of the phase effect appears rather inconclusive and thus it may give a dominating contribution to the uncertainty of our thickness measurements. However, a few points are worth mentioning. Most of the reported phase effects at least agree that the largest effect occurs at energy of stopping power (around 0.5–1 MeV for alpha particles in hydrogen) or lower, whereas the initial energy of an americium alpha source is about 5.5 MeV, which is almost in the Bethe-Bloch region where a better theoretical prediction exists. Most of our measurements center around 4–5 MeV, and we rarely go below 2.5 MeV. The specific choice of stopping
power values will be discussed in section 5.2.

In concluding this chapter, it can be said that the study of energy loss processes of charged particles is still an ongoing science. In fact, more than 200 papers were published on stopping power measurements from 1978 to 1987[48]. There are many open questions including chemical binding effects, physical phase effects and Bragg's additivity rule for compounds. For instance, a recent volume of *Nuclear Instruments and Method B* is devoted to aggregation and chemical effects in stopping[49], reflecting the demand for a more and more precise understanding for the applications of radiation in radiology, material studies and device fabrication, to name a few. Nevertheless, the current state of knowledge should serve our purpose for measurements of target thickness. Details of the experimental methods and the analysis are discussed in the following chapters.

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^5^It is interesting to note that the survey of the geographical distribution of papers shows Canada ranks near the top of the list. This is largely due to the contribution of the Chalk River group.
Chapter 4

EXPERIMENT

4.1 Apparatus

4.1.1 Target System

The cryogenic solid hydrogen target system used in the present experiment is the same as the one used for the beam experiments described in section 1.2, except for one of the target foils as discussed below. Shown in Fig. 4.1 are the schematic views of the experimental set up. Americium 241 is electrodeposited on the gold plated oxygen free copper plate to form an array of spot sources. The americium is covered with a thin gold layer for safety purposes. The spot diameter is less than 3 mm and 5 spots are separated by 10 mm center to center.

The spot sources were custom-manufactured by Isotope Product Laboratory\(^1\). This plate replaces the upstream target foil for the beam experiment. The target plate is cooled to approximately 3K, and solidifies the hydrogen gas onto it when it is introduced through the diffuser mechanism. Alpha particles penetrating through the hydrogen film are detected by a passivated, implanted planar silicon detector (Canberra, model FD/S-600-29-150-RM, serial number 12913), which is mounted on the top of the diffuser.

The silicon detector is collimated such that it only accepts the alphas from one spot source at a time. Furthermore, the collimator consists of an array of small holes (diameter 1 mm) in order to reduce the angular dispersion of the alpha beam. The collimated

\(^{1}\)3017 N. San Fernando Blvd, Burbank, CA
Figure 4.1: Schematic views of the experimental set up.
detector can move vertically to allow a measurement of the thickness at five different positions by detecting the alpha particles from each of the five spot sources. The profile of alpha counts in the silicon detector versus vertical position of detector for a bare target (with no hydrogen) is shown in Fig. 4.2.

This proves that we see only one source spot at a time, avoiding that complication in interpreting the data.

4.1.2 Silicon Detector

A silicon detector is used for the energy spectroscopy of alpha particles. The silicon surface barrier detector (SSB) is probably the most common detector used for charged particle spectroscopy. It is, in principle, a reverse biased diode. Instead of an np junction as in normal diodes, the junction is formed between a metal and semiconductor, typically gold and p-type silicon, creating a Schottky barrier[23]. Ion-implanted detectors have similar structure, but instead of a metal contact, acceptor ions are implanted by an accelerator to form p-type silicon at the surface. They offer some advantages over the SSB such as low leakage current and small dead layer, which contribute to a better energy resolution. They are also known to provide a more robust surface than an SSB which is very sensitive to surface contamination.

The detector used for the present experiment has an active area of 600 mm$^2$ (diameter 27.6 mm), and thickness of 150 $\mu$m with a dead layer of 50 nm. It is operated with reverse bias of 30 V, which fully depletes the detector. In fact, 10 to 15 volts would be sufficient to deplete the thickness corresponding to the range of 5.5 MeV alpha particles but appropriate bias gives better energy resolution due to the larger depletion depth which results in a lower capacitance. This would also provide a better time resolution because of the faster charge collection, although it is not of much importance in the present experiment.
Figure 4.2: Alpha counts versus vertical position of the silicon detector. Each peak corresponds to one of the five source spots. The detector is collimated such that it sees only one spot at a time.
Hydrogen in a semiconductor is known to influence its properties, and has become one of the hot topics in condensed matter physics\cite{50,51}. In particular, $\mu$SR, another remarkable application of the muon, has proven to be an almost exclusive probe of isolated hydrogen-like atoms in semiconductors\cite{52,53}. Thus, a solid state detector is normally considered to be incompatible with hydrogen gas. For example, in the measurements of stopping powers of twelve different gases, Bimbot et al. used a special target configuration for hydrogen gas\cite{42}. Our silicon detector, however, performs without significant degradation in a hydrogen rich, low temperature ($\sim 90$ K) environment. (It has proven to work satisfactorily even in a tritium environment in our December 1993 run\cite{6}.)

The silicon detector had a resolution of $\sim 30$ keV for 5.5 MeV alpha particles as determined with an americium source at room temperature. Cooling down to a lower temperature improves the resolution, and at 90 K it reaches $\sim 20$ keV. The leakage current dropped from 0.1 $\mu$A to nearly zero ($\ll 0.01$ $\mu$A). In the meantime the apparent energy for the same source drops significantly due to the increase in the band gap. This drift in gain becomes a source of the uncertainty for the final results as described in section 5.3.

4.1.3 Electronics and Data Acquisition System

The electronics diagram is shown in Fig. 4.3. The signal from the silicon detector is divided into an energy and a timing output with a charge sensitive preamplifier (Canberra 2003BT). The preamplifier works as a charge to voltage converter providing a positive polarity pulse to the energy output as well as providing a negative polarity fast differentiated pulse to the timing output. It should be noted that, due to a 110 M$\Omega$ resistor in series with the detector, the actual bias applied to the detector is reduced depending on the leakage current. The energy signal from the preamplifier is further amplified with a linear spectroscopy amplifier. An 8000 channel ADC (Analog to Digital Converter)
converts the analog voltage height into a digital signal, which can then be recorded with a CAMAC/VDACS data acquisition system described below.

The trigger signal for the data acquisition starts with the timing output from the preamplifier. After the pulse has gone through a timing filter amplifier and discriminators, an anti-coincidence is required to avoid pile up of signals before the system completes the signal processing. This is done by the hardware inhibit signal from the gate generator (HI Gate in the figure) and output register signal from the CAMAC. The trigger signal then gives gates for the ADC (Gate 1, 2) and the Starburst in CAMAC. It should be noted that the Starburst accepts negative TTL logic, therefore conversion from NIM logic to TTL is necessary. Also, ADC gates require TTL signals, which can be obtained from the outputs of the gate generators.
Figure 4.4: Alpha particle energy spectrum of a high resolution americium source for energy calibration. Three peaks are clearly separated.

The energy threshold is set by adjusting both the gain of the timing filter amplifier and discriminator threshold for the timing signal from the preamplifier. The data was collected with a VAXstation using the TRIUMF program VDACS (Vax Data Acquisition System)\cite{54}. The data acquisition procedure is specified by a file written in TWOTRAN language\cite{55}. A TWOTRAN file, after being compiled on the VAXstation, is downloaded to the Starburst, which actually collects the data and sends them to the VAXstation. On- and off-line analysis was done by the MOLLI program\cite{56} with the use of the FIOWA histogramming package\cite{57}. Fig. 4.4 illustrates the typical resolution of the detector/data acquisition system. Three peaks of an americium alpha source are clearly separated.

4.1.4 Calibration and Noise

The ADC channel numbers were converted to a proper energy scale by the calibration obtained with another high resolution americium alpha source (Fig. 4.4) and a precision
pulse generator. A pulse signal from the pulse generator, which simulates the real signal from the silicon detector, is given to the test input of the preamplifier as in Fig. 4.3. An example of pulser input versus ADC channel number is plotted in Fig. 4.5. To determine the offset of the electronics system, particularly of the ADC, the data points are fitted with the function \( y = ax + b \), where \( y \) is the ADC channel number, \( x \) is the pulse amplitude setting and \( a, b \) are fitted parameters. The fit gives a reasonable \( \chi^2 \) which verifies the linearity of the electronics. The precision pulse generator is assumed to be linear in its output. The parameter \( b \) gives the offset, that is the ADC channel number corresponding to zero energy. Then the proper energy \( E \) can be written as, \( E = \kappa(y - b) \), where \( \kappa \) can be obtained by substituting the calibration source energy 5.486 MeV (with no material covering it) and the corresponding ADC channel number.

Various sources of noise include nearby mechanical and electrical apparatus, such as
the turbo molecular pump, the mechanical pump, cryopump, quadrupole mass spectrometer and ion gauge. The sources of noise are both microphonic and electronic. It was also found that cables for the thermometer, when connected to the voltage meter, cause a huge random noise. There were some other occasional, large fluctuations whose sources could not be identified, possibly from the mini cyclotron near the experimental area.

4.2 Experimental Runs

Before the measurements with solid hydrogen, tests of the silicon detector, electronics and data acquisition were conducted with the test vacuum chamber. Optimization of bias voltage and amplifier shaping time as well as set up and debugging of the data acquisition hardware and software was achieved. The following two separate runs were conducted in the Muonic Hydrogen Group working area located in the Meson Hall at TRIUMF.

4.2.1 Series 1

The first series of runs was mainly devoted to the verification of the principle of the method. One hundred litres of liquid helium was consumed to give about 40 hours of measurement time, running continuously. It takes roughly 4-10 hours, depending on liquid helium flow rate, to cool down the cryostat from room temperature to 3 K. While cooling down, the leakage current drops to nearly zero (\(\ll 0.01 \mu\text{A}\)) and the voltage drop across the resistor in the preamplifier becomes negligible. Therefore the bias voltage has to be readjusted to prevent the break down of the detector. It was realized that the source position shifted by as much as 2.5 mm due to thermal contraction when the cryostat cooled. This is illustrated by the plot of source array profiles in Fig. 4.6.

Thickness measurements were done in the three different targets. The amount of
Figure 4.6: Shift in the position of source spots due to thermal contraction of the target system.
molecular gas introduced to the target is quoted in units of torr-litre, where one torr-litre corresponds to the number of molecules in 1 litre of gas at a pressure of 1 torr at room temperature (~295 K). The three measured targets included a gas injection of 20 torr-litres, 20 torr-litres deposited on top of the existing 20 torr-litre target (giving a total of 40 torr-litres) and separately deposited 150 torr-litres. Five spots were measured in each target except the 20 torr-litre target where only 3 spots were measured. The source spot position \( y \) is noted in terms of its vertical distance with respect to the center of the target foil, hence the beam. Therefore we have source spots at \( y = -20, -10, 0, 10 \) and 20 mm. The gas deposition was conducted typically at a rate of 1 torr-litre per second for thick target deposition. For thin targets, the rate was much slower.

### 4.2.2 Series 2

In the second run, the diffuser position was changed by 2.5 mm to compensate the thermal contraction effect (Fig. 4.6). Different deposition conditions such as deposition rate and with or without cryo-pumping were tried to check the effect on thickness and uniformity. The gas input used here was 150, 300, and 400 torr-litres. Also in this run, various measurements were tried as follows.

**Cross Contamination**

Cross contamination was tested in the following way, where all the measurements were done at \( y = 0 \) mm (center spot). First a large amount of gas (1000 torr-litres) was deposited on the other target foil (downstream target foil) and the alpha particle energy from the source on the upstream foil was measured. If there is any cross contamination, a corresponding energy shift of the alpha particles should be observed. One torr-litre of gas was then deposited on the upstream target foil to compare the energy shift. Finally
another 1000 torr liters of gas was deposited on top of the existing target on the down stream foil and the thickness of the up stream foil was checked again.

**Neon Targets**

Neon targets with gas input of 7.5 and 20 torr-litres were also measured. Neon films were used in beam experiments for the measurement of muon transfer rates. It is important to know the absolute thickness and uniformity to interpret the muonic X-ray data correctly.

**Another Method of Deposition**

Another set of targets was made in a very different way. Instead of blowing hydrogen gas directly onto the cold target foil, the diffuser was completely removed away from the foil, and the gas was introduced into the system with all pumping valves closed. 300 torr-litres of gas was used for this measurement. Relatively high vacuum pressure was kept ($\sim 10^{-4}$ torr) during introduction of the gas to allow interaction between the gas molecules. The gas is expected to be deposited onto all the cold parts in the system including the target foils. The measurement was originally aimed as a test for the DAΦNE experiment proposed by Olin et al.[11]. Its implication for the uniformity of our target is discussed in section 6.5.
Chapter 5

ANALYSIS OF DATA

5.1 Determination of Thickness

Shown in Fig 5.1 is an example of the energy spectra of alpha particles penetrating through the hydrogen film. Each peak represents alphas from the target with different amounts of gas let in, namely 0, 150, and 300 torr-litres. Clearly, one can observe alpha particles losing more energy when going through the target with the larger amount of injected hydrogen. Also, the peak becomes wider due to the energy straggling effect.

Notice that the bare target spectrum with no hydrogen has an asymmetric shape with a smaller second peak and a low energy tail. This is partly due to the energy loss in the gold overlayer on top of the americium, which was added for safety purposes. Backscattering of alpha particles from the gold substrate is also possible. Single scatterings with large energy loss from the collimator material as well as hydrogen can account for the low energy tails. Each bare spot has a slightly different energy and peak shape, probably due to variations in thickness of the sources and gold overlayers. It is this asymmetric and irregular shape of the peaks that makes it difficult to fit the data with a normal function such as a Gaussian. Instead, as recommended by Hanke and Laursen[58], we determine the mean channel values \(<N>\) from the distribution function \(f(N)\) in the ADC spectra:

\[
<N> = \frac{\int_{<N> - \epsilon}^{<N> + \epsilon} f(N) N dN}{\int_{<N>-\epsilon}^{<N> + \epsilon} f(N) dN},
\]

(5.1)

where \(\epsilon\) is a finite cut off value. The effect of the value of \(\epsilon\) upon the final results
Figure 5.1: Alpha particle energy spectra with different thicknesses of hydrogen film. The numbers indicate the amount of hydrogen gas injected in units of torr-litre.
is discussed in the section 5.3. \( <N> \) is then converted to the energy scale by the calibration which was described in section 4.1.4.

Given the range of alpha particles as a function of energy, \( \text{Range}(E) \), the thickness of the target can be obtained by:

\[
\text{Thickness} = \text{Range}(E_{\text{init}}) - \text{Range}(E_{\text{fin}}),
\]

(5.2)

where \( E_{\text{init}} \) is the initial energy of the alpha particles and \( E_{\text{fin}} \), the energy after penetrating through the target. This avoids the complication of numerically integrating stopping power cross sections. A correction due to the “wiggled” path of the particle may be necessary and will be considered later. The specific choice of energy-range values is discussed in the following section.

### 5.2 Energy-Range Tables

It is very important to use an accurate stopping power and/or range table in determination of the thickness by energy loss. A few energy-range tables published in the past decades, plus more recent ones are discussed and their values compared in this section.

#### 5.2.1 Northcliffe and Schilling (1970)

An energy-range table by Northcliffe and Schilling[59] was once considered as a standard. With emphasis on heavy ions, they compiled stopping power and range tables for ions of all atomic numbers at energies of 0.01–12 MeV/amu with no uncertainty specified. However, there now exist many experiments which disagree with the table by large amounts. For example, Bianco and Richer found a 35% deviation in the stopping power of alpha particles in \( \text{D}_2 \) gas from the table[60]. Geissel et al. claim that heavy ion stopping powers in light elements are underestimated by 15% in the table[43].
5.2.2 Ziegler (1977)

At present the most commonly used stopping power table for alpha particles is probably the one by Ziegler[17]. He divided the projectile energy into three regions. These are the high energy region, or Bethe-Bloch region, where the Bethe-Bloch formula is used to obtain the stopping powers, the low energy region, where velocity proportionality is assumed, and the intermediate energy region, where the experimental data are fitted to the parameterization discussed in section 3.2 (equations 3.4-3.6).

Ziegler also included in his table empirically estimated values of stopping power in condensed gases by interpolating from the elements for which solid data are available. Approximately 40% lower stopping power for solid hydrogen over the gas is given for alpha particles of 1 MeV. More recent measurements[15, 47], however, disprove such large phase effects in other condensed gases which are similarly predicted by the same table. Therefore its accuracy for solid hydrogen is dubious.

The proton range table by Janni[30] is widely used in the intermediate energy physics community. The emphasis is put on the statistical treatment of the experimental information in terms of least-square curve fitting as well as determining mean ionization potentials. All the range values are tabulated with their uncertainties. In (gaseous) hydrogen, for example, he quotes a 2% error for 5 MeV protons.

5.2.3 Ziegler, Biersack and Littmark (1985)

For material studies and device technology, the use of Ziegler, Biersack and Littmark’s tabulation for solids[20] together with their Monte Carlo code\footnote{TRIM (TRansport of Ions in Matter). It is available from James F. Ziegler, IBM-Research, 28-0, Yorktown, NY 10598 USA} is very common. It is known to give a good agreement with experiments for materials commonly used, such Si and GaAs. For solid targets for which there are few experimental data, an “interpolation
Chapter 5. ANALYSIS OF DATA

procedure” is used. This is based on the assumption that the deviation between the experimental and theoretical stopping powers $\kappa$

$$
\kappa = \frac{S_{\text{exp}}}{S_{\text{theory}}}
$$

depends smoothly on the target’s atomic number. However, extrapolation to atomic number 1 may be less reliable. The accuracy of 5% is quoted for alpha particles.

5.2.4 ICRU (1993)

The latest table on stopping powers and ranges is compiled by the International Commission of Radiation Units and Measurements (ICRU)[18]. The mission of ICRU is explained by Inokuti et al.[61] to recommend internationally acceptable values of physical quantities relevant to radiation measurements and radiological dosimetry. The report committee on stopping power consists of members such as Berger, Inokuti, Anderson, Bichsel, Seltzer, Thwaites, Watt and Sternheimer to name a few. These authors are frequently cited in this thesis and can be considered as the experts in the field of stopping power studies. The report contains the stopping powers and ranges of protons, alpha particles and negative pions. In the case of alpha particles in hydrogen, electronic stopping power for energies higher than $\sim$2 MeV are calculated by the Bethe-Bloch theory with various corrections such as shell, density and deviation from first-order Born approximation. Mean excitation energies were taken from Ref. [22], which was originally prepared for the ICRU report on electron and positron stopping powers. For alpha particle energies lower than 1 MeV, the Varelas-Biersack formula (equations 3.4-3.6) is used with new coefficients compiled by Watt[62], based on the available experimental data. The energy region between 1 and 2 MeV is interpolated by a cubic spline function. The ranges are calculated in the continuous-slowing-down approximation(csda). In this approximation,
energy-loss fluctuations are neglected, and charged particles are assumed to lose their energy continuously along their tracks at a rate given by the stopping power, as expressed in

\[ R_0(E_0 \rightarrow E_{st}) = \rho \int_{E_{st}}^{E_0} \left[ S_{\text{elec}}(E) + S_{\text{nuc}}(E) \right]^{-1} dE, \quad (5.3) \]

where \( R_0 \) is the csda range of a particle slowing down from an initial energy \( E_0 \) to an energy at which particles are considered to be stopped \( E_{st} \). In the ICRU table, \( E_{st} \) is taken to be 10 eV. The csda range is generally larger than the average penetration depth \( P_{av} \), which is the projection of the range on the initial direction of the particle track due to its “wigging” caused by multiple scattering. In the table the detour factor \( \zeta \), which is defined as the ratio \( P_{av}/R_0 \), is given to take this effect into account. Thus, equation 5.2 can be modified to:

\[ \text{Thickness} = \zeta(E_{int}) \cdot \text{Range}(E_{int}) - \zeta(E_{fin}) \cdot \text{Range}(E_{fin}), \quad (5.4) \]

where \( E_{int} \) and \( E_{fin} \) are the initial and final energies of the alpha particle respectively.

### 5.2.5 Comparison of Tables

Figures 5.2-5.3 compare the stopping powers of alpha particles in hydrogen from the various tables discussed above. Shown in Fig. 5.2 are stopping powers in gaseous hydrogen. As one can see, Northcliffe and Schilling[59] deviate significantly from Ziegler[17] and ICRU[18], whereas the latter two are in good agreement.

Fig. 5.3(a) compares stopping of solid hydrogen predicted by Ziegler(1977)[17] and Ziegler et al. (1985)[20] with that of gas by ICRU. For Ziegler et al. (1985), only electronic stopping power is plotted, but nuclear stopping is negligible for our purposes. While Ziegler(1977) shows a large phase effect, ICRU and Ziegler et al. (1985) are also significantly different at the stopping power maximum and at very low energy. However, differences between the latter two are rather small in Fig. 5.3(b), where the energy of
Figure 5.2: Comparison of alpha particle stopping power in *gaseous* hydrogen by ICRU[18], Ziegler[17] and Northcliffe and Schilling[59].
Figure 5.3: Comparison of predicted solid hydrogen stopping powers by Ziegler et al. (1985)[20] and Ziegler(1977)[17] with the gaseous hydrogen stopping power by ICRU[18].
Table 5.1: Comparison of the hydrogen thickness between TRIM-92 and ICRU corresponding to the same energy loss of alpha particles.

interest is plotted on a linear scale. At 2 MeV the difference is less than 3%, and we rarely go down below 2.5 MeV in our measurement. Note that from Fig. 5.3(b) it is clear that the difference in range is even smaller, since the stopping power curves are integrated to obtain the range.

The thicknesses determined by these two tables for the same energy loss are compared in the following way. For ICRU, equation 5.4 was used with the csda range and detour factor which are in the table. On the other hand, the Monte Carlo code TRIM-92\(^2\) which employs the electronic as well as nuclear stopping power from Ziegler \textit{et al.} (1985) was utilized for comparison. As shown in Table 5.1 they agree with each other within 1-2% for alpha energy loss of up to \(~4\) MeV (5.5 MeV\textrightarrow1.5 MeV). This is clearly within our limit of precision.

Thus it seems reasonable, at least in the energy region of our interest, to use either ICRU or Ziegler \textit{et al.} (1985). There is no compelling reason to use Ziegler’s

\(^2\)The latest version of TRIM released in December 1993. It is also available from J. F. Ziegler. See footnote in page 41.
old values (1977). In fact, Ziegler et al. themselves suggest that their older one is less accurate [20]. For reasons of convenience for our thickness determination we shall use the ICRU table, which provides the integrated ranges. The tabulated values are interpolated with fourth order polynomials, and equation 5.4 is used to obtain the target thickness.

5.3 Uncertainties

There are several sources of systematic uncertainties in the determination of the target thickness, other than the statistical uncertainty which is negligible most of the time. These include the knowledge of stopping powers, detector calibration, computation of mean energy and angular dispersion of alpha particles.

5.3.1 Stopping Powers and Ranges

In most cases, the dominant contribution to the systematic error comes from the knowledge of stopping powers. This is illustrated in Fig. 5.4, where variation of the thickness due to the stopping power value is shown in the case of ICRU and Northcliffe and Schilling. As large as a 25% difference can be observed. However, we can safely disregard Northcliffe and Schilling as discussed in section 5.2. The ICRU table [18] claims an uncertainty of ~1-2% at 4 MeV, and ~2-5% at 1 MeV for the stopping power of alpha particles. This is of course in gaseous hydrogen, and its application to solid requires caution. Ziegler et al. [20] quote an average accuracy of 5%. This is the average deviation of their values from 3290 available data points on alpha particle stopping power. In either case, there is no legitimate way to determine the uncertainty in the stopping power for solid hydrogen, since there exist no experimental data. However, since the above two tables agree fairly well in the energy region of interest, we shall quote 5% uncertainty in the stopping power, hence the range including the phase effect. This can be improved,
Figure 5.4: Variation of the thickness due to the use of different range tables. ICRU[18] and Northcliffe and Schilling[59] are compared.
in the future, if the stopping power of solid hydrogen becomes available.

5.3.2 Peak Energy Determination

To make a thickness measurement, one must measure two energies, before and after the deposition of the film. Computation of the centroid as well as the calibration of the silicon detector can be a source of systematic uncertainty in determining such energies.

Computation of Centroid

Some arbitrariness in the cut-off value $\epsilon$ in the integral in equation 5.1 in taking the centroid may affect the results. The influence of the choice of $\epsilon$ on the thickness was tested by changing $\epsilon$. This was found to be less than $\sim 5$ keV for bare, 20 torr-litre and 40 torr-litre targets, $\sim 10$ keV for 150 torr-litre, $\sim 30$ keV for 300 torr-litre and $\sim 60$ keV for 400 torr-litre targets. Due to the lower energy tail and increasing width of the peak, determination of a centroid is subject to a larger systematic uncertainty for increasing thickness of the target.

Detector Calibration

The calibration of the silicon detector was found to be very sensitive to its temperature. This effect is accounted for by the change in the average energy required to create an electron-hole in the silicon as the temperature shifts. Namely, the band gap in silicon increases as the temperature decreases. With a few exceptions, where we suffered an accidental large temperature change in the detector, the calibration of the detection system is taken to be accurate to 2 channels ($\sim 0.5\%$).

The above two uncertainties are, however, negligible in the measurement of thick targets compared to the uncertainty in the stopping power. They become important
only in thin (≤40 torr-litre) targets.

5.3.3 Random Noise

Occasionally the appearance of significant noise distorted the spectrum, resulting in a shift in the centroid. The effect can be as large as 10 keV in the peak energy determination. This was monitored by the signal from the test pulser, simultaneously recorded with the alpha spectrum, which is sensitive to such noise. When substantial noise is found, a larger uncertainty in the centroid is quoted accordingly.

Other uncertainties such as path difference due to angular dispersion of alpha particles and the uncertainty in gas handling are estimated to be negligible compared to the above sources.
Chapter 6

RESULTS AND DISCUSSION

6.1 Uniformity

6.1.1 Thickness Profile

Series 1

Fig. 6.1 shows the thickness profile from the first series of runs. Data from three different targets with gas input of 20, 40 and 150 torr-litres are plotted. The 40 torr-litre target was made by depositing 20 torr-litres on the existing 20 torr-litre target. Data at \( y = -20 \) and \( -10 \) mm do not exist for the 20 torr-litre targets. The error bars include the uncertainty in the stopping power as well as other errors. Hence, the relative uniformity can be determined more accurately.

Series 2

A typical thickness profile from the second series of runs is shown in Fig. 6.2. The diffuser position was displaced by 2.5 mm in Series 2 as mentioned in section 4.2. Data from targets with 150 torr-litre and 300 torr-litre gas input are plotted. Similarly to Fig. 6.1, the error bars include the stopping power uncertainty.

A few remarks can be made on the thickness profiles. First, there is a drop in thickness at both edges. Second, the shape is asymmetric with respect to the center of the beam. These non-uniformities will be discussed in section 6.7, but first we shall look at results from other measurements.
Figure 6.1: Target thickness profile from Series 1. The error bars include the uncertainty in the stopping power.
Figure 6.2: Typical thickness profile from Series 2.
6.1.2 Effect of Deposition Condition

Diffuser Position

The effect of the diffuser position on the thickness profile can be directly compared in Fig. 6.3. It compares 150 torr-litre targets deposited at the nominal diffuser position (Series 1) and the diffuser inserted a further 2.5 mm (Series 2). The uncertainty due to the stopping power is not plotted, since we are interested in the relative change in the thickness. Furthermore, systematic uncertainties in taking the centroid are expected to be equal, therefore the error bars overestimate the uncertainty if only relative change is of interest. We observe a significant difference in profile. However, the slope around the center of the target (y from -10 to 10 mm) does not seem to be affected by the position of diffuser. The problem of non-uniformity is further discussed in 6.7.2.

Cryo-pumping

The effect of cryo-pumping during the depositions is illustrated in Fig. 6.4. Gas input of 150 torr-litres is used for both depositions. There is no significant difference within the accuracy of measurement. This suggests that most molecules stick to the cold surface at first contact.

6.2 Linearity in Target Deposition

The targets used in our beam experiments range from a few $\mu$g/cm$^2$ to a few mg/cm$^2$. It is important to know the relation between amount of the injected gas and the deposited target thickness. It can be studied by plotting the two quantities. Fig. 6.5 shows such a relation between injected gas in torr-litres and thickness of the film at one of the source spots ($y = 0$ mm, where $y$ is the vertical displacement from beam center). The first three data points, at 20, 40 and 150 torr-litres, are taken from Series 1. After confirming
Figure 6.3: Thickness profile with different diffuser positions. Series 1 corresponds to the nominal diffuser position, and Series 2 is with diffuser inserted a further 2.5 mm. The error bars do not include the uncertainty in the stopping power.
Figure 6.4: Thickness profile with the cryo-pump port open and closed during target deposition.
Figure 6.5: Test of linearity of deposition. The line fitted to the data points is also plotted.

that the values of thickness for 150 torr-litres agree between Series 1 and Series 2 at the $y = 0$ mm spot, as seen in Fig. 6.3, the data from Series 2 (at 300 and 400 torr-litre) were plotted on the same graph. The 40 torr-litre and 300 torr-litre targets were made by two separate depositions of a 20 and 150 torr-litre target respectively, whereas a single deposition was performed for the 20 torr-litre, 150 torr-litre and 400 torr-litre targets.

The line in the figure is a least-squares fit to the data points, which proves linearity of depositions. This allows us to extrapolate our results to thicker targets which cannot be directly measured by the present method due to the limited range of 5.5 MeV alpha
particles. It should be recalled that our standard targets for muonic hydrogen isotope production are made with \( \sim 1000 \) torr-litres of injected gas.

The fit gives the conversion relation

\[
\text{Thickness (\( \mu g/cm^2 \))} = (3.38 \pm 0.17) \times \text{H}_2 \text{ Input (torr—litres)},
\]

for the central spot, \( y = 0 \) mm. Since the fitted line goes through all the data points, uncertainty is quoted only from that of the stopping power (5%).

### 6.3 Cross Contamination

The cross contamination was checked by depositing as much as 1000 torr-litres of gas from the other side of the diffuser to the downstream foil. It was measured only at the central spot, \( y = 0 \) mm. The first trial was rather inconclusive, giving the thickness of \( 1.4 \pm 3.2 \, \mu g/cm^2 \) on the upstream foil. With improved calibration and stable temperature, we obtained \( 1.62 \pm 0.84 \, \mu g/cm^2 \) of cross deposition, assuming no random noise effect (section 5.3.3). The systematic uncertainties in computing the centroid described in section 5.3 are negligible, since the energy loss is very small and there is very little change in the peak shape. The error in calibration is also neglected here, because measurements were made consecutively with a very short interval and the detector gain was stable. Thus, the quoted uncertainty comes only from statistics. With the conversion factor derived in section 6.2, \( 3.38 \, \mu g/(cm^2\cdot\text{torr—litre}) \), 1000 torr-litres correspond to \( \sim 3.4 \, mg/cm^2 \). Comparing this with the above value, we get \( 0.48 \pm 0.25 \times 10^{-3} \) cross contamination, or an upper limit of \( 0.8 \times 10^{-3} \) with 90% confidence level.

To check the sensitivity of our measurement to a small amount of gas such as is being discussed in this section, 1 torr-litre of hydrogen was deposited. This gives us \( 3.07 \pm 0.90 \mu g/cm^2 \) with uncertainty similarly derived as above. The result is consistent with the conversion factor obtained in section 6.2.
6.4 Neon Targets

Fig. 6.6 shows the thickness profile of neon targets with quantities of 7.5 and 20 torr-litres. Although there are only three spots measured in each target, they suggest a profile similar to the hydrogen targets. The conversion factor for neon obtained from a 20 torr-litre target is $32.2 \pm 2.0 \, \mu g/(cm^2\cdot\text{torr-litre})$ for the central spot.
6.5 Alternative Deposition

A proposed experiment at a $ \Phi $ factory involves a solid hydrogen target directly deposited on the collider beam pipe[11]. Feasibility of the concept was tested by the present method of thickness measurement. As already described in section 4.2, 300 torr-litres of hydrogen gas was introduced to a closed vacuum system with the diffuser completely removed from the cold target foil. This simulates the actual gas deposition on the beam pipe.

The result, shown in Fig. 6.7, illustrates the thickness profile of hydrogen deposited on the cold target foil. The uncertainty is relatively large due to the small amount of the deposited gas. In addition, the measurement suffered from some random noise and gain drift in the detector. Nevertheless, considering that the error bars include the stopping power uncertainty, the results indicates a uniform distribution. The weighted average thickness is found to be $39.4 \pm 2.1 \, \mu g/cm^2$. It should be noted that this is a very small thickness, given the gas input of 300 torr-litres. The conversion factor is $0.13 \pm 0.01 \, \mu g/(cm^2 \cdot torr \cdot litre)$, which should be compared to $3.38 \, \mu g/(cm^2 \cdot torr \cdot litre)$ for normal deposition at the central spot.

Given the non-uniformity observed in normal deposition, this deposition method may be applied to our beam experiment to provide a more uniform target. Obviously, the gas sticks to other cold surfaces in the system, therefore the cross contamination is unavoidable. However there are certain situations where it may be tolerated.

6.6 Effective Thickness for Beam Experiment

One of the main objectives of the present work is to determine the target thickness for the beam experiments. Having observed the non-uniformity in the target profiles, it is not sensible to take the straight average of five points. Because the muon beam has a rather narrow distribution, the peripheral spots contribute less. Thus the average weighted by
Figure 6.7: Alternative deposition: Hydrogen gas is introduced to the entire cryostat with vacuum pump closed. The gas may stick uniformly to all the cold surfaces and walls.
the beam distribution profile should be considered. With \( w_i \) being the weighting factor at the \( i \)th source spot, the effective average thickness \( \bar{t}_{\text{eff}} \) is given by

\[
\bar{t}_{\text{eff}} = \frac{\sum w_i t_i}{\sum w_i}.
\]  

(6.2)

where \( t_i \) is the target thickness at the \( i \)th source spot. It is also convenient to define effective standard deviation, \( \sigma_{\text{eff}} \), of the thickness distribution as a measure of the target uniformity:

\[
\sigma_{\text{eff}} = \sqrt{\frac{\sum w_i (t_i - \bar{t}_{\text{eff}})^2}{\sum w_i}}.
\]  

(6.3)

This should not be confused with the uncertainty in the average thickness. One could measure the average thickness to a very high precision, but still have a large non-uniformity, hence a large \( \sigma_{\text{eff}} \). However, this is important because the uncertainty in the final result of the scattering cross sections in the beam experiment will be related to the \( \sigma_{\text{eff}} \). Also, it is a useful measure when we compare the uniformity between the targets with different thickness distributions.

The full width at half maximum (FWHM) of the beam profile is estimated to be 20–25 mm from imaging by a multi wire proportional chamber array[2]. It is generally a difficult inverse problem to reconstruct the actual distribution from the projected image, with many factors involved such as resolution, solid angle and efficiency. Here we shall assume that the beam distribution is Gaussian, and calculate the weighted average and the standard deviation of the thickness distributions for different FWHM of the beam profile. The thicknesses from 150 torr-litre targets from both Series 1 and Series 2 (diffuser 2.5 mm further inserted) are used. The FWHM of the beam distribution is varied from 10 mm to 50 mm by 5 mm steps. The result is summarized in Table 6.1.

More digits than are significant are shown in the table to improve interpolation. It can be seen that the weighted average is rather insensitive to the FWHM of the beam profile. In both Series 1 and Series 2, the variation is less than 5% between beam FWHM.
| Beam FWHM (mm) | Series 1 | | Series 2 | |
|----------------|----------|----------------|----------|
|                | $\bar{t}^{eff}$ | $\sigma^{eff}$ ($\mu g/cm^2$) | $\bar{t}^{eff}$ | $\sigma^{eff}$ ($\mu g/cm^2$) |
| 10             | 497.7    | 16.8            | 501.2    | 15.2            |
| 15             | 496.2    | 26.7            | 498.8    | 24.0            |
| 20             | 493.7    | 34.2            | 495.6    | 30.6            |
| 25             | 490.8    | 40.1            | 492.0    | 35.7            |
| 30             | 487.9    | 44.6            | 488.4    | 39.4            |
| 35             | 485.4    | 48.0            | 485.4    | 42.1            |
| 40             | 483.3    | 50.4            | 482.9    | 44.0            |
| 45             | 481.6    | 52.3            | 480.7    | 45.4            |
| 50             | 480.1    | 53.8            | 479.0    | 46.5            |

Table 6.1: Effective average of target thickness $\bar{t}^{eff}$ and standard deviation of the distribution $\sigma^{eff}$, weighted by beam profile for Series 1 and Series 2 (diffuser 2.5 mm up) for various FWHM of beam distribution. A Gaussian beam profile is assumed.
value of 10 mm and 50 mm. However, as naturally expected, the standard deviation of the effective thickness profile changes more rapidly with the beam width. There is about a factor of three difference between the FWHM of 10 mm and 50 mm. Series 2 systematically shows a smaller standard deviation than Series 1, indicating that Series 2 has a more uniform target.

For a realistic estimate, we take the average of the values for FWHM 20 mm and 25 mm. This gives an effective thickness of 492±25 μg/cm² for Series 1, and 494±25 μg/cm² for Series 2. The uncertainty of these value is dominated by the absolute errors in the thickness measurements, since the variation of the average value due to the beam width is only about 1%. The standard deviations of the thickness distributions are 37 μg/cm² and 33 μg/cm² for Series 1 and Series 2, respectively. These correspond to roughly 7% non-uniformity in the targets. The effective conversion factors, that is the effective target thickness per unit gas input, are 3.28±0.16 μg/(cm²-torr-litre) and 3.29±0.16 μg/(cm²-torr-litre) for Series 1 and Series 2 respectively, and they are nearly independent of the beam width. It should be noted, however, these values are the effective thickness for the upstream target. For the downstream target, it is the μt rather than μ spatial distribution that is important. Therefore, the thickness profile should be weighted by the distribution of incident μt. The μt atoms have a large angular dispersion due to the mechanism of emission, but they can be made into a “beam” if a collimating device is used. To estimate the μt beam distribution we must rely on the Monte Carlo calculation which includes details of the kinetics as well as target geometry. Nevertheless, the standard deviation is around 10–11% of the mean value even for FWHM 50 mm, which is close to the upper limit of the μt beam width since the target diameter is about 60 mm.

It may be instructive to consider an “efficiency” of the deposition, that is the ratio of the average thickness with respect to the case where the deposited area is the same as the
physical area of the diffuser. At 295 K, one torr-litre of hydrogen molecules ($^1\text{H}_2$) weighs 108.7 μg. Given the diameter of the diffuser, 6.0 mm, the thickness for 100% efficiency is 3.78 μg/(cm²-torr-litre). If we compare this with the effective average thickness (3.29 μg/(cm²-torr-litre)), then we obtain 87% deposition efficiency. If we compare with the straight (non-weighted) average of 3.11 μg/(cm²-torr-litre)), the efficiency becomes 82%.

It is interesting to note, however, that the thickest target spot ($y = -10$ mm) has an efficiency as high as 95%.

### 6.7 Mechanism of Gas Deposition

In this section, the mechanism of the gas deposition is discussed based on what we have learned from the above measurements.

#### 6.7.1 Monte Carlo Simulation of Gas Deposition

A Monte Carlo simulation was conducted to gain a better understanding of the gas deposition process. The following assumptions were made in the calculation:

- The gas molecules diffuse isotropically from the diffuser.
- The sticking probability of the molecule onto the cold foil is 100%.
- There is no interaction between the gas molecules.
- The perforation structure of the diffuser is neglected.

The result is shown in Fig. 6.8(a) and 6.8(b). The former shows the surface plot of the 2-dimensional distribution of deposited gas. The latter is a 5 mm wide slice in the horizontal direction. This cut roughly corresponds to the detector acceptance of alphas from the sources and should be compared with the measured thickness profile. We notice
Chapter 6. RESULTS AND DISCUSSION

(a) Thickness Profile of Deposited Gas

(b) Thickness Profile in Vertical Direction

Figure 6.8: Monte Carlo simulation of gas deposition.
that with the above idealized condition, \( \sim 25\% \) drop at both edges is observed. This at least partly explains the measured non-uniformity, in particular, the drop at the edges of the target thickness profile. However, this does not explain the asymmetry in the profile.

### 6.7.2 Non-uniformity

The non-uniformity in the target can be partly explained by the finite size of the diffuser. The acceptance of the peripheral spots on the target foil with respect to the diffuser is smaller than that of the central points, unless the diffuser is very large compared to the target foil. This is confirmed by the Monte Carlo simulation above.

On the other hand, the cause of the asymmetric target thickness profile is not too obvious. A few possible explanations exist. For example, it is possible that there is an asymmetry in the perforation structure of the diffuser. In particular, the diameter of the small holes might slightly vary from one to another. If this happens in a systematic way, it could cause the observed asymmetry in the thickness profile. Another plausible explanation is that since the hydrogen gas is introduced from the bottom part of the diffuser, more gas comes out from the bottom than the top, hence the asymmetry in the profile. In fact, the preliminary results from the recent measurements with a different diffuser supports this model. However, because the diffusers are not identical, this interpretation requires caution. Another possibility is the migration of hydrogen due to gravity.

### 6.8 Summary of the Method

#### 6.8.1 Performance

As we have seen, the method for film thickness measurement via energy loss of alpha particles proves to give a reasonably good accuracy. For a hydrogen film of the thickness of a few hundreds \( \mu g/cm^2 \) or larger, the accuracy is limited by the knowledge of the
stopping power. If accurate stopping power data are provided in the future, the intrinsic
uncertainty of the method can be less than 1%. The present method applies to the
thickness range of a few $\mu g/cm^2$ to a few $mg/cm^2$, although both very thin and very
thick targets are subject to larger uncertainties.

One of the advantages of the present method is its relative simplicity compared with
other methods such as Rutherford Backscattering, which requires a dedicated accelerator.
Also the uniformity can be easily determined by moving the detector, which is very
difficult for other methods.

It should now be possible to apply the present method to other experiments where thin
solid targets are required. Typical examples are an experiment on slow $\mu^-$ production[7,
8, 9] and a low energy kaon scattering experiment[11].

6.8.2 Possible Improvements

The upper limit of the thickness that can be measured with present method is determined
by the range of the alpha particles. This can be improved by using an alpha source whose
energy is higher than americium. For example, $^{212}$Po provides alpha particles of energy
as high as 8.8 MeV. This would also reduce the uncertainty in the stopping power, since
the relevant energy region moves closer to the Bethe-Bloch region where the theoretical
prediction is more reliable.

The measurement of the stopping power of solid hydrogen is not only useful for our
purpose of improving the accuracy in the thickness measurement, but also very important
for the fundamental understanding of the energy loss process as well as for radiological
applications since hydrogen is a major constituent of the human body. It also plays a
significant role in the alpha-sticking problem, as becomes apparent in the next chapter.
It would be possible to measure the stopping power from the energy loss, if one can make
the solid hydrogen film of a known thickness. Hardy suggested a possible method to
prepare this by using a chamber of known base area and with a wall with a temperature
gradient[63]. This, if achieved, would undoubtedly help resolve the frustrating situation
of the study of the physical phase effect, and deserves further investigation.

6.8.3 Thickness Measurement in Beam

After the establishment of the present method, it was realized that a similar thickness
measurement could be done during the actual beam experiments. Of course, the muon
beam is more “expensive” than an americium source, so it is not very practical when we
already have a system which is satisfactory. However, it may be worthy of a consideration,
and in fact, it turned out to be the case.

The idea is to use muon catalyzed fusion reactions in a D-T mixture target layer as
a source of mono-energetic alpha particles\(^1\). By depositing a sample layer on top of the
source layer, and by measuring the energy shift of the alpha particles, one can determine
the thickness of the former in the same way as we have done. This original concept
was modified by a suggestion of Knowles[65] to use the layer of protium with \(10^{-3}\)
tritium concentration to cause more fusions in the source layer. Further consideration
lead to an optimal target arrangement as in Fig. 6.9. The muon is stopped in the first
layer, and the formation of the \(\mu p\) and the transfer to the \(\mu t\) take place rapidly. Due to
the Ramsauer-Townsend effect, the \(\mu t\) is emitted to the adjacent layer, where the fusion
reaction provides a 3.5 MeV alpha particle. The thickness of the sample layer is measured
by the energy loss of the alpha particle. The efficiency can be improved by using the
\(D_2/T_2\) mixture in the source layer to allow the cycling of \(\mu CF\) reactions.

The few differences which exist in the conditions between the actual beam exper­
iments and the source thickness measurements are considered to be negligible, but if

\(^1\)A similar method using protons from D-D fusion is proposed independently by Strasser[64].
Figure 6.9: Thickness measurement in beam, by using the \( \mu \text{CF} \) reaction as a source of the mono-energetic alpha particles.
there appears to be any doubt in the future, the thickness measurement in beam de-
scribed above can provide the information. A possible effect on the deposition efficiency
might come from the radiation heat load of the tritium in the target or the difference in
the thickness of the Au foils on which hydrogen is deposited.

Nevertheless, the use of the muon beam is probably not justifiable only for the thick-
ness measurement, given the situation that we can do as well or better in off-line exper-
iments. However, this same target configuration turns out to give interesting opportuni-
ties for measurements of very important parameters in \( \mu \)CF, as we shall see in the next
chapter.
Chapter 7

STOPPING POWERS AND ALPHA-STICKING IN $\mu$CF

The importance of the alpha particle stopping power in hydrogen in $\mu$CF is not confined to the target thickness measurement. In fact, the stopping power is deeply related to the alpha-sticking process, one of the most important in $\mu$CF. We shall begin the discussion with the review of the sticking problem. Its relevance to the stopping power in terms of the reactivation, as well as a possible new experiment, are described in the sections that follow.

7.1 Brief Review of the Sticking Problem

Contrary to the initial expectation of Alvarez[66], it is not the short lifetime of the muon that gives the upper limit on the efficiency of $\mu$CF. Even if the muon were stable, one would not obtain enough energy to achieve economical break-even according to our present understanding, as long as there remains the alpha-sticking process. Therefore, this process has attracted the greatest attention of the $\mu$CF researchers around the world in recent years. A good understanding has been gained for D-D fusion, but for the D-T system there are discrepancies among different groups as well as between theory and experiment. We will concentrate on the D-T case, which is the most interesting in terms of practical applications.

As briefly described in Chapter 1, the alpha-sticking is a process where the muon gets
attached to the helium nucleus ($\alpha$) after the fusion reaction:

$$d\mu t \rightarrow \mu\alpha + n.$$  \hfill (7.1)

After the muons stick to the helium nuclei, some of them get reactivated, or “stripped” from the nucleus, in collisions with the target atoms. The effective alpha-sticking probability $\omega^{eff}_{s}$ is written as

$$\omega^{eff}_{s} = \omega^{0}_{s}(1 - R),$$ \hfill (7.2)

where $\omega^{0}_{s}$ is the initial sticking and $R$, the reactivation probability.

### 7.1.1 Theory

The calculation of the initial sticking and the reactivation should be distinguished, since they are two separate processes. As for the initial sticking, there exists a remarkable agreement amongst many authors in recent years. It is based on the framework of the sudden approximation[67], that is the fusion process is considered to be very fast compared to the time scale of the muonic motion. The sticking of a $\mu^{-}$ to a given state $k$ of muonic helium is then given by the overlap of the initial and final wave functions,

$$P_{k} = |<\psi^{f}_{k}|\psi^{i}>|^2,$$ \hfill (7.3)

where $\psi^{i}$ is the normalized initial wave function of the $d\mu t$ molecule in the limit of zero internuclear distance, and $\psi^{f}_{k}$ is the final wave function of the recoiling $\mu\alpha$. Accurate calculation of the muonic molecule wave function is a complex task, but recent works using at least six different mathematical methods converge to a consistent value of $\omega^{0}_{s}$. The latest calculations incorporate the nuclear effect, and give the value $\omega^{0}_{s} = (0.92 \pm 0.01)$%[68].

The reactivation occurs mainly via two processes:
**Ionization** The ionization of the $\mu^-$ from the helium nucleus in collision with a hydrogen isotope $x$, followed by its atomic capture on another hydrogen isotope $y$. ($x$ and $y$ could be different isotopes in a mixture.)

$$\mu^- + x \rightarrow \mu^- + \alpha + x,$$

$$\mu^- + y \rightarrow \mu^- y.$$  

\[(7.4)\]

**Transfer** The exchange reaction between the alpha and a hydrogen isotope $x$.

$$\mu^- + x + \alpha \rightarrow \mu^- x$$ 

\[(7.5)\]

At higher densities ionization through the multi-step excitation becomes increasingly important. These processes are calculated by scaling the similar cross sections in ordinary atomic collisions, $p + He^+$ to the muonic system. The most recent calculation predicts $R$ to be 0.27 at density $\phi = 0.05$ and 0.34 at $\phi = 1.2$ with about 10% uncertainty, where $\phi$ is the liquid hydrogen density[69].

Combining the values of $\omega_s^0$ and $R$, we obtain the theoretical value for the effective sticking $\omega_s^{eff}(th)$ to be

$$\omega_s^{eff}(th) = 0.67 \pm 0.03\% \ (\phi = 0.05)$$  

\[(7.6)\]

$$\omega_s^{eff}(th) = 0.61 \pm 0.03\% \ (\phi = 1.2).$$  

\[(7.7)\]

A slight density dependence comes from that of the reactivation probability.

### 7.1.2 Experiments

Fig. 7.1 summarizes the current status of the sticking problem. A few remarks deserve mention. First, there is a discrepancy between two major experiments. The 1986 LAMPF data show a strong density dependence, while PSI data are only slightly dependent on
Figure 7.1: Summary of experimental results of the effective sticking $\omega^{eff}_s$ as a function of the density $\phi$, plotted with theoretical predictions, where $\phi$ is the liquid hydrogen density ($4.25 \times 10^{22}$ atoms/cm$^3$), taken from Ref. [68].
the target density\textsuperscript{1}. Second, all the experimental values, except the LAMPF data at low density, are substantially smaller than the theoretical values.

Let us take a look at the methods used in these experiments. Most of the early experiments use the neutron method, where the time evolution of the detected neutrons is fitted to the model describing the kinetics of the reactions. The main difficulty with this method is that the sticking probability is one of the many parameters in the formula and a complete understanding of the detailed kinetics of the fusion cycle is required at least in the steady state. A typical example of a process yet to be understood is a scavenging of the muon due to transfer to $^3\text{He}$ and $^4\text{He}$, which accumulate slowly in the target from the beta decay of tritium and as a product of the fusion reaction, respectively. In addition, this method requires an absolute calibration of the neutron detector. This is difficult to do to a high accuracy, and is suggested to be one of the sources for the discrepancy between the experiments\textsuperscript{68}. Thus, the neutron method is model-dependent and indirect in determining the sticking probability.

The X-ray method provides more direct information on the sticking process. It detects the characteristic X-ray of the muonic helium atom formed after a fusion event. A fraction of the sticking occurs to excited states of muonic helium and it may de-excite via emission of an X-ray. The experimental difficulty arises from a large bremsstrahlung background from tritium beta decay. Therefore the experiments were done at a low tritium concentration at PSI. On the other hand, the pulsed beam structure of KEK enabled the measurement to be done at a high tritium concentration.

Although the X-ray method offers a more direct observation of the sticking than the neutron method, the extraction of the sticking value from the observed X-ray intensities relies on detailed knowledge of the population of the excited states of $\mu\alpha$. The initial population of each state is determined by the partial sticking fraction, but the state

\textsuperscript{1}The analysis of 1986 LAMPF data may be unreliable at low densities\textsuperscript{70}. 
can be further populated by the collisional excitations from the lower states, or can be depleted by ionization. In addition, processes such as Auger transitions, Stark mixing and multi-step collisions have to be taken into account correctly. This, on the other hand, means that the method can provide very useful information on the kinetics of the $\mu\alpha$ as well as the sticking. An experiment with an improved accuracy is planned at the RIKEN muon channel at Rutherford Appleton Lab in the UK, and the results are eagerly awaited.

The most direct method of measuring the sticking probability is the detection of the charged particles $\mu\alpha$ and $\alpha$. The sticking probability can be determined from its very definition:

$$\omega_s = \frac{N_{\mu\alpha}}{N_{\mu\alpha} + N_{\alpha}}$$

(7.8)

where $N_{\mu\alpha}$ and $N_{\alpha}$ are the number of each species. In this method, the serious systematic uncertainties involved in the indirect methods in extracting $\omega_s$ from the complex series of processes are avoided. No knowledge of the obscure processes such as muon scavenging to impurities or excitation and de-excitation of $\mu\alpha$ is required. The experiments with this method have been done in two ways. The detection of the $\mu\alpha/\alpha$ by a surface barrier detector placed outside a low density ($\phi \sim 0.001$) gas target, was first achieved at LAMPF[71, 72] and later at RAL[73]. Because of the small reactivation in the low density gas target, this method is more sensitive to the initial sticking, $\omega_s^0$. Despite these pioneering efforts, the experiments suffered from many problems, some of them intrinsic to the method, and only preliminary results have been published so far:

$$\omega_s^0(exp) = 0.80 \pm 0.15(stat.) \pm (0.12)(syst.)\%$$

(7.9)

$$\omega_s^0(exp) = 0.69 \pm 0.40(stat.) \pm (0.14)(syst.)\%$$

(7.10)

This will be discussed in further detail in section 7.3. Another way was originally developed at St. Petersburg for the D-D fusion experiment, for which they used the
hydrogen ionization chamber as a target as well as an active detector. Two recent theses report the result of the PSI measurement of D-T sticking[74, 75] using a similar chamber. This provides the most direct and model-independent value of the effective sticking at medium density ($\phi \sim 0.17$):

$$\omega_{eff} = 0.565 \pm 0.046\text{(stat.)} \pm 0.025\text{(syst.)} \%.$$  \hfill (7.11)

The combined uncertainty gives 9.3% relative accuracy. With a reactivation model assumed, the relative uncertainty reduces to 6.6%.

The current status of the sticking problem in the D-T system may be summarized as follows[68]. At medium density ($\phi \sim 0.17$), the experimental effective sticking value is two standard deviations lower than the theory. At high density ($1.0 \leq \phi \leq 1.4$), the experimental values are lower by three standard deviations. If we assume the theoretical initial sticking $\omega_s^0 = 0.92 \pm 0.01\%$, which appears rather reliable as discussed above, the reactivation probability must be increased by 30%. We will take a careful look into the problem of the reactivation in the following chapter.

### 7.2 Stopping Power and the Reactivation

The stopping process of $\mu\alpha$ in the target medium is very important in the reactivation problem, because it competes with the stripping process. $R$, the reactivation probability at the end of slowing down is calculated[76] from

$$R = 1 - e^{\lambda I},$$

$$I = \int_{E_f}^{E_i} \frac{\sigma_{str} dE}{S},$$  \hfill (7.12)

where $E_i$ is the initial energy, $E_f$ is the threshold energy for the stripping reaction ($\sim$few keV), $\sigma_{str}$ is the stripping cross section, and $S$, the stopping power of $\mu\alpha$. Since the $R$ depends exponentially on $I$, it is very sensitive to the cross sections for stripping and
the stopping power. Because the $\mu\alpha$ is a tightly bound compact object of charge one, its stopping power is assumed to be equivalent to that of protons of the same velocity in most calculations. Thus the proton stopping power tables such as those of Anderson and Ziegler\cite{77} and Janni\cite{30} are often used. It should be pointed out that there exists the following problem, even if the above assumption is correct. As we have seen in detail in Chapter 3, no experimental data on the stopping power is available in the condensed phase of hydrogen for heavy charged particles for the energy of interest. The phase effect is known to become more important at energies of the maximum stopping power ($\sim 1$ MeV) and lower, but in the present case, one needs the stopping powers for the energy from 3.5 MeV all the way down to a few keV\(^2\).

The latest calculation on the reactivation probability was done by Stodden \textit{et al.}\cite{69}, and this is claimed to be the most accurate calculation with about 10\% uncertainty in $R$. They used the stopping powers from Anderson and Ziegler\cite{77} and assume their error to be 10\% over the entire density range. It should be pointed out that all the PSI neutron data at high density (Fig. 7.1) are taken in the liquid or solid phase\cite{78, 79}. Given the rather controversial situation on the phase effect, the above uncertainty in the stopping power may well be underestimated at densities above $\phi \sim 0.95$. In fact, their final error bar in the calculation of $R$ at high density ($\phi = 1.2$) is 20\% smaller than that at low density ($\phi = 0.05$), which is in the opposite direction to the reliability of the stopping powers. Thus, the failure to take the phase effect into account in the reactivation calculation may, at least in part, explain the larger discrepancy in the effective sticking values between theory and experiment at high density\(^3\).

\(^2\)For the case of our thickness measurement, we had a rather fortunate situation, that is we were only interested in the stopping powers of the energy region between 5.5 MeV and about 3 MeV, which is not too far from the Bethe-Bloch region where the theoretical prediction is more reliable.

\(^3\)"Unrealistic" parameterization for density dependent stopping powers\cite{80} to explain LAMPF's sticking data, which shows a very strong dependence on density, is not relevant (nor acceptable) here. We are concerned about the discrete change in the stopping power due to the physical phase transition.
One obvious remark from equation 7.12 is, if the stopping power of the $\mu\alpha$ were reduced for any reason, the efficiency of the fusion reactions would increase due to the increased reactivation probability. This may be the key for achieving the practical use of $\mu$CF. More complete understanding of the stopping process of charged particles could open up new opportunities.

It is interesting to note that the injection of frozen hydrogen pellets is considered the leading candidate for re-fueling tokamak thermonuclear fusion reactors[81, 82]. It is one of the attempts to solve the problem of how to deposit atoms of fuel deep within the magnetically confined, hot plasma. Here again, the energy loss process in solid hydrogen will become important.

With these in mind, the measurement of the stopping power of hydrogen in the condensed phase appears more than justifiable from the $\mu$CF point of view, as well from an interest in the basic atomic physics. Feasibility of film growth suggested by Hardy[63] should be seriously considered.

7.3 Direct Measurement of the Sticking Probability at High Density

7.3.1 Introduction

A struggle with the frustrating situation of the stopping power problems (Chapters 3 and 5), together with the realization of the potential of the $\mu$CF reaction as a source of alpha particles (Section 6.8.3), motivated some new considerations. After a recently published review by Petitijean[68] attracted the author’s attention to the sticking problem, an idea came for a very ambitious series of experiments; the first systematic study of the reactivation process, and a model-independent measurement of the intial sticking.
Despite the challenging goal, at least the first phase of the experiment\textsuperscript{4} appears feasible, which comes as a natural extension of the previous work at LAMPF/RAL[72, 73]. The goal of the initial phase is the first direct measurement of the sticking probability at high density ($\phi \sim 1.4$). As discussed in section 7.1, detecting the fusion products directly is the most unambiguous method for determining the alpha-sticking probability. The previous direct measurements have been tried at medium density ($\phi = 0.17$) and very low density ($\phi \sim 10^{-3}$). Given the controversy over the density dependence, the direct measurement at a high density will definitely be very important. Furthermore, this method can provide a less model-dependent measurement of the initial sticking $\omega^0_\phi$, with a relatively small correction ($\lesssim 10\%$) required. Of course, the success in this first phase is a prerequisite for the more ambitious experiments that might follow.

\subsection{Description of Experiment}

Fig. 7.2 shows a schematic view of the proposed experiment. Notice that the target configuration is exactly the same as the one for our thickness measurement in beam (Fig. 6.9). But, while we were interested only in the alphas before, this time we want also to look at mu-alphas that come out of the source layer.

The principle of the method can be briefly described as follows. The muon is stopped in the emitter layer, where it goes through an atomic capture by a proton and transfer to a triton to form $\mu t$, which then travels a macroscopic distance due to the Ramsauer-Townsend mechanism as described before. The $\mu t$ is stopped in the source layer, and forms $d\mu t$ which fuses almost immediately. The fusion produces a neutron and an $\alpha$ or a $\mu\alpha$, the fraction of the latter being smaller by more than 100 times. Although the energies of the $\alpha^{++}$ and $\mu\alpha^+$ are very close to each other (3.54 MeV and 3.46 MeV,

\textsuperscript{4}An idea similar to the first phase of the present experiments is independently proposed by P. Kammel[83].
Figure 7.2: Schematic top view of a direct measurement of the alpha-sticking in a solid target.
respectively), the difference in the stopping powers permits us to separate them by the energy loss in the degrader layer. Recalled that the stopping power is proportional to the square of the projectile charge in the Bethe-Bloch formula. The $\mu\alpha$ being singly charged, there is about a factor of four difference in the stopping power between $\mu\alpha$ and $\alpha$. It should be emphasized, however, that for the purpose of separating the two species, knowledge of their absolute stopping powers in solid hydrogen is not important, but only the fact that they differ significantly.

In order to gain in the counting rate, a few things can be tried. The $D_2/T_2$ mixture may be used in the source layer which allows the cycling reactions. Due to the diffusion of the $\mu t$ out of the source layer, a large cycling is unlikely, but some improvement in the rate is expected. In certain cases, it may be advantageous to use the protium layer with $\sim 10^{-3}$ tritium concentration as the degrader. Considering that the emission of the $\mu t$ is isotropic, this acts as another emitter layer, as well as the degrader, emitting the $\mu t$ backward into the source layer. The increase in the fusion rate can be significant for thick degraders.

The fast fusion neutrons are detected in the liquid scintillation counters. Demanding collinear coincidence between $\alpha/\mu\alpha$ and the neutron (Si and N1) is an extremely powerful technique in reducing the background as we will see later. The existence of the neutron counter, N2 in Fig. 7.2 is useful for the estimation of the accidental background, as well as for monitoring real (physics related) backgrounds such as protons from D-D fusion following the D-T fusion.

The uniformity of the target layers, in particular of the degrader layer, might be important in order to achieve a high energy resolution. Unfortunately, as we have seen in this thesis, there is a significant non-uniformity in our target. If this turns out to be the limitation for the energy resolution, the alternative deposition described in section 6.5 may be applied to provide a more uniform target. However, the relatively high deposition
pressure ($\sim 10^{-4}$ torr) may cause some problem when depositing multi-layered targets. Off-line measurements should be conducted to test the possibility.

It is convenient to generalize the effective sticking and reactivation in equation 7.2 and define

$$\omega^{\text{eff}}_s(T) = \omega^0_s(1 - R(T)), \tag{7.13}$$

where $\omega^{\text{eff}}_s(T)$ and $R(T)$ are a function of the thickness ($\rho x$) of the material that the $\mu\alpha$ goes through. Hence,

$$\omega^{\text{eff}}_s(T)|_0 = \omega^0_s, \tag{7.14}$$

$$\omega^{\text{eff}}_s(T)|_{r_{\mu\alpha}} = \omega^{\text{eff}}_s, \tag{7.15}$$

$$R(T)|_0 = 0, \tag{7.16}$$

$$R(T)|_{r_{\mu\alpha}} = R, \tag{7.17}$$

where $r_{\mu\alpha}$ is the range of the $\mu\alpha$ in the given material. The symbols on the right hand sides of equations refer to their conventional uses.

The direct observable in this method is the effective sticking at the degrader thickness $T_d$, namely $\omega^{\text{eff}}_s(T_d)$. If $T_d$ can be made such that the reactivation probability $R(T_d)$ is small, then this method will come close to measuring the initial sticking $\omega^0_s$.

By changing the thickness of the degrader layer, a systematic study of the reactivation process can be performed for the first time. However, our sensitivity to the stripping process is limited to medium energy or higher ($\geq 0.5\text{MeV}$).

### 7.3.3 Comparison with LAMPF/RAL Experiments

Apart from the difference in the density at which measurements are done, the present method offers certain advantages over the LAMPF/RAL experiments. The experiment
at LAMPF suffered from a very low muon stop rate. Only ~0.05% of the incident muons were stopped in the target at 490 torr, and the rest stopped in the walls producing a large background. Although improved, still only 1% was stopped at RAL. We have confirmed in our experiments that we can stop more than 30% of the muons in our solid targets, providing a better signal to background ratio.

In the gaseous targets, since the fusion can occur anywhere within the target cell, the amount of target material that the $\alpha$ or the $\mu\alpha$ must go through before reaching the detector can vary substantially, resulting in large fluctuations in the energy loss. On the other hand, in our system the fusion occurs only in the spatially well “confined” source layer, therefore the energy loss which occurs in the degrader is well defined. This will result in much better energy resolution for the two species.

The spatial “confinement” of the fusion source offers a further advantage. Since the source layer is isolated from the degrader, we can control the energy loss process of the $\alpha$ and $\mu\alpha$ by adjusting only the degrader, without affecting the dynamics in the fusion layer. This is in contrast to the LAMPF experiments, where the measurements were done at two different densities; low density to allow both $\alpha$ and $\mu\alpha$ to reach the detector, and high density to allow only $\mu\alpha$ to reach the detector. We now know the existence of highly non-linear processes in density, the typical example being the epithermal molecular formation which we are trying to measure in E613. Thus, the interpretation of the data becomes very difficult when the different densities are compared, due to the change in the molecular/atomic dynamics in the target. Since we confine the dynamics in the source layer, we can control the processes after the fusion rather freely without affecting the fusion process itself.

Another major problem with the LAMPF/RAL measurement is the diffusion of tritium out of the target cell window. This can cause a few things. One is the reduction in the target tritium concentration, hence the decease in fusion rate. This could create
a problem in the normalization of the data. Another is the diffusion of tritium into the solid state detector, resulting in the deterioration of the resolution. To prevent the latter, the double enclosure with two windows and D$_2$ buffer gas is used in their target. This causes extra stripping in these materials before the $\mu\alpha$ reaches the detector, which adds to the systematic uncertainties in obtaining the initial sticking probability. Also, because of the presence of the diffused tritium in the buffer D$_2$ gas, some fusion reactions take place there. This creates a serious background, because they all resemble a $\mu\alpha$ event due to the small energy loss$^5$. In our target system, however, the silicon detector is adjacent to the solid target, with nothing but ultra-high vacuum between them. It has proven to work very well in the tritium environment in past beam runs. Therefore the protection of the silicon detector with a buffer gas or a coating with aluminum dioxide, as investigated for the RAL experiment$^{[85]}$ is not necessary. Of course, the amount of tritium present in our target is much smaller, about 10 Ci, compared to 750 Ci for the RAL experiment.

7.3.4 Monte Carlo Simulations

Method

In order to illustrate the feasibility of the proposed experiment, Monte Carlo calculations were performed. The prime objective is to gain a qualitative understanding with a simplified model, therefore quantitative values in the result should not be taken too seriously.

The calculations were performed with the following assumptions. The $\alpha$ and $\mu\alpha$ originate uniformly from the source layer, 15 torr-litre ($\sim$105 $\mu$g/cm$^2$ for D$_2$)$^6$ in thickness. According to a recent theoretical study$^{[5]}$ and our recent experimental runs (July-August

$^5$A Monte Carlo study shows that a timing resolution of 1.2 ns, for both the silicon and neutron detectors combined, is required to separate this background$^{[84]}$. The above resolution appears rather unrealistic for a conventional alpha and neutron detection system.

$^6$The conversion is based on the preliminary result for a new diffuser system.
Figure 7.3: Monte Carlo simulation for the direct measurement of alpha-sticking with various degrader thicknesses. The alpha counts are compressed by factor of 20 for a comparison.
1994), this thickness is sufficient to stop most of the energetic $\mu t$ atoms which are emitted into the source layer from the emitter layer. Both the $\mu\alpha^+$ and $\alpha^{++}$ were assumed to originate uniformly from the source layer with an initial energy of 3.5 MeV and lose energy in the rest of the source layer as well as in the degrader layer. The alpha particle stopping powers were taken from the ICRU tables for gaseous hydrogen[18]. For mu-aldas, simply one fourth of the alpha particle stopping power was taken for the same energy. Straggling and multiple scattering in layers were ignored, but a separate calculation by TRIM-92 7 shows that it contributes only $\sim$1.5% with the energy loss of 1 MeV. Also, the reactivation of the muon is not taken into account. It is estimated to be less than 5% for a moderately thin degrader. All the solid hydrogen layers are assumed to be uniform. The source radius is taken to be 10 mm, though the actual size may be larger than this. The detector radius is 10 mm unless otherwise specified. The target and the detector are separated by 45 mm, center to center, as in the actual apparatus.

**Results and Analysis**

The histograms in Fig. 7.3 show the energy spectra of two charged species, $\alpha$ and $\mu\alpha$, in a silicon detector with various thicknesses of the degrader layer. The $\alpha$ peak is compressed by a factor of 20 for an easier comparison. With a moderate amount of the degrader (between $\sim$ 40 torr-litre (140 $\mu$g/cm$^2$) and $\sim$ 80 torr-litre (280 $\mu$g/cm$^2$)), two peaks can clearly be separated. The most direct measurement of sticking can be achieved in this region by simply counting the number of the two species detected. With a degrader of 20 torr-litre (140 $\mu$g/cm$^2$) or less, the separation of the two is rather difficult. This region can be investigated by using muonic X-ray and/or nuclear capture gamma ray, as will be discussed in section 7.4. At larger thicknesses of degrader, alphas no longer make their way through and most of them stop or fall below the detector threshold. In this region,

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7See footnote in page 46
the analysis become slightly more complicated than the medium thickness case, but we can still hope to obtain relatively accurate results. Details of analysis methods for these cases are discussed below, but there is an interesting application for this region, as well (see section 7.4).

Fig. 7.4 illustrates how to deal with one of the worst scenarios with this method. That is, the case where the broadening of two peaks is so large, for any reason, that we cannot hope to resolve the α and μα peak in a single spectrum. One obvious thing is to follow the example of the RAL experiment[73].

Since they could only observe μα, they determined the initial sticking from

\[ \omega_s^0 = \frac{N_c}{BN_n} \cdot f \]  

(7.18)

where \( f \) is some correction factor for the stripping. The number \( N_c \) is μα–neutron coincidences and \( N_n \), the singles neutrons. The factor \( B \) is the ratio of the solid angle for collinear coincidence detection to that of neutron detector itself. The equation 7.18 follows from the definition of \( B \). They tried to calculate \( B \) from Monte Carlo calculations but found it to be rather sensitive to the beam parameters such as width and divergence.

But we can do better, even in this case, in the following way. First, we measure all α–n and μα–n coincidence events with no degrader \( (N_{\alpha+\mu\alpha}) \). Also we record the singles neutron events \( n1 \). Then, we put a degrader of thickness \( T_d \) sufficient to stop all the alphas (320 torr-litre, or 1120 μg/cm², for example), and count the μα–n events \( (N_{\mu\alpha}) \) and singles neutrons \( n2 \). Finally, we simply take the ratio of the two numbers normalized by singles neutrons to get the sticking value at degrader thickness \( T_d \):

\[ \omega_s^{eff}(T_d) = \frac{N_{\mu\alpha}/n1}{N_{\alpha+\mu\alpha}/n2} \]  

(7.19)

8By demanding the electron decay signal in plastic scintillators surrounding the target after the neutron signal, we can obtain a very clean fusion neutron signal.
Figure 7.4: Determination of the sticking probability from two separate measurements. Counts in each spectrum will be normalized to the single neutrons.
After all, this turns out to be the same thing as actually measuring the coincidence efficiency $B$, rather than calculating by Monte Carlo using uncertain beam parameters. It should be pointed out that, if $B=1$, which is the case when the silicon detector is sufficiently closer to the target than the neutron detector, $\alpha$–n coincidence measurements provide a very accurate *absolute* calibration of the neutron detector, which is generally a very difficult task.

There are many parameters which could influence the energy resolution of the present method. It is important to understand what those are to achieve the optimal condition. Histograms in Fig. 7.5 show one such investigation. The radius of the silicon detector was varied, and its effect on the spectrum plotted here. As the detector size is increased, the two peaks become wider, and in particular, the lower energy tails grow. This is quite natural because a larger detector can accept particles coming out in a larger angular dispersion, hence suffering a wider range of energy losses. As a realistic consideration, we presently have two kinds of silicon detectors with radius of 13.8 mm, and 25.2 mm respectively. While the solid angle of the latter is almost four times larger, the energy resolution shown in the histogram is rather unsatisfactory. The use of a collimating device may help both to gain in the event rate as well as to a good resolution. It can be relatively easily mounted on top of the diffuser. Another set of calculations shows that the resolution is rather insensitive to the thickness of the source layer. A thicker source layer could provide the higher cycling rate, due to the smaller probability of the $\mu t$ escaping out of the layer. Further investigation for the optimization of the conditions is certainly necessary.

**7.3.5 Test Measurement**

We were fortunate in the 1994 July/August beam time to have a few hours to spend on a test measurement of the proposed experiment. The schematic top view of the detector
Figure 7.5: Influence of the silicon detector size on the peak width in the energy spectrum is plotted. $R_d$ refers to the detector radius. A degrader of 70 torr-litre (245 $\mu$g/cm$^2$) is used in the calculations.
arrangement used in this measurement is shown in Fig. 7.6. The target layer configuration is very similar to Fig. 6.9 except we used only D_2 in the source layer.

Before discussing the result, it should be emphasized that the geometry is far from optimal, because of the very wide dispersion of particles coming into the silicon detector. Some of the particles suffer more energy loss than others. Also the data were taken only for a few hours, as more or less a ‘fill-in’ measurement right before a maintenance day. So we should not be discouraged too much, even if the result is not too convincing.

Fig. 7.7 can illustrate the power of the coincidence technique by comparing the silicon detector singles on the left side with the collinear Si–n coincidence spectra on the right side. The huge background at low energy has almost disappeared with the coincidence demanded. Histograms on the top row are taken without any degrader, and the bottom with 70 torr-litre (245 μg/cm²) Degrader. The horizontal axis represents the energy in the silicon detector and one channel corresponds to approximately 1 keV. The bottom right spectrum is taken with 70 torr-litre degrader with a collinear Si–n coincidence. The peak energy indicated by an arrow is consistent with the energy loss of μα in 70 torr-litre hydrogen emitted at an angle of about 80 degrees from perpendicular, which is an average angle from the target to the silicon detector (see Fig. 7.6). If the alpha particle was traveling at the same angle, however, it would not penetrate into even half of the 70 torr-litre hydrogen layer. Hence, events in the peak are μα candidates.

The background level can be studied from Fig. 7.8. The coincidence events in a collinear detector pair as well as non-collinear pairs are plotted. As seen in Fig. 7.6, the (Si2&N1) pair are the only collinearly aligned detectors, so counts in any other coincidence pairs are background to us. Fig. 7.8 shows very low background in non-collinear pairs, and the peak in (Si2&N1) appears statistically significant.

However, it seems rather premature to claim the observation of μα from only this one measurement. For example, non-uniformity of the target at the edges may allow
Figure 7.6: Schematic top view of the test run for the sticking experiment. Target configuration is similar to Fig. 6.9. Coincidence was taken between neutron detector N1 and silicon detector Si2.
Figure 7.7: Data from a test run for the sticking experiment. The data were taken only for a few hours and the target geometry was far from optimal. The horizontal axis is the Si energy in channels, where one channel corresponds to approximately 1 keV. On the left sides are the Si detector singles, and on the right, the Si2–N1 collinear coincidence events. In the bottom right, candidate events for $\mu\alpha$ are seen. The peak position is consistent with the estimated energy loss.
alpha particles to escape from the degrader layer, and imitate \( \mu \alpha \) events. But, at least, the feasibility of the direct sticking measurement in the solid hydrogen target has been successfully demonstrated.

### 7.3.6 Discussion

#### Rate Estimates and Precision

The incident muon rate \( R_\mu \) is typically \( 5 \times 10^3 / \text{s} \) in 12 cm\(^2\) at 27 MeV/c, with \( \delta p/p = 0.04 \). The muon stopping fraction \( f_\mu \), is known to be approximately 0.3. According to a Monte Carlo study of Markushin[5], the fusion yield per stopped muon, \( Y_f \), is about 0.06 with source layer thickness of \( \sim 15 \text{ torr-litre} \), which is consistent with our experimental data.

For the silicon detector of 25 mm radius at 45 mm from the target foil, the solid angle \( \epsilon_{si} \) is \( \sim 7.5\% \). The NE213 liquid scintillation neutron counter has \( \sim 20\% \) intrinsic efficiency \( \epsilon_n \), and we take the geometrical efficiency for the Si–n coincidence \( \epsilon_c \), that is the ratio of the solid angle of for collinear coincidence detection to that of the silicon detector itself, to be 0.3. Assuming effective sticking \( \omega_{\epsilon}^{eff}(T_d) \) of \( \sim 0.8\% \), allowing \( \sim 10\% \) reactivation in the degrader \( (R(T_d) = 0.1) \), the overall coincidence event rate for \( \mu \alpha–n \) coincidence is

\[
R_c = R_\mu \cdot f_\mu \cdot Y_f \cdot \omega_{\epsilon}^{eff}(T) \cdot \epsilon_{si} \cdot \epsilon_n \cdot \epsilon_c. \tag{7.20}
\]

The event rate can be estimated to be \( \sim 3 \times 10^{-3} / \text{s} \) or 1500 events in a 5 day period. Even if we assume a signal to background ratio of 1:1, which seems rather unlikely from the test run, this corresponds to \( \sim 3.5\% \) statistical uncertainty. This is without considering the cycling reactions, which may improve the rate by an order of magnitude or more.

The sources of systematic uncertainties include the correction due to the reactivation in the target and the energy dependence of the detection efficiency of the silicon detector. As for the former, the correction itself appears to be less than 10\% at a moderate thickness of the degrader, so even if we allow 30\% uncertainty in the reactivation correction, the
Figure 7.8: Comparison of collinear and non-collinear Si-n coincidence. The top left (Si2&N1) is the only collinearly aligned pair. Coincidence events in other pairs are background. For the arrangement of the detectors, see Fig. 7.6.
contribution to the final result is 3%. Besides, we can check the reactivation calculation by changing the degrader thickness. For the silicon detector efficiency, we plan to make an off-line measurement.

If the clear separation of the two peaks in one spectrum is achieved, as in the Monte Carlo calculation, we can, in principle, expect the most accurate measurement of the initial sticking, with the precision almost comparable to the PSI ionization chamber measurement.

### 7.3.7 Readiness

It should be emphasized that the proposed experiment, at least in its initial phase, does not require a major rebuild of the existing apparatus for E613. Turning the diffuser mechanism and a target foil by 45 degrees should not be a problem, since the former has a rotational symmetry, and the latter can be easily bent if we remove the second target foil that we do not use.

Testing of the deposition and the measurement of thickness in an off-line experiment is necessary before the beam run, but otherwise we do not foresee the need for much preparation in terms of hardware.

Of course, more refined Monte Carlo simulations are necessary to determine the optimal conditions.

### 7.4 Towards the Future

Although the method described above provides a fairly accurate value for the initial sticking probability, it is not totally free from theoretical assumptions about the reactivation process. A potential method for a model-independent measurement of the initial sticking by directly measuring the reactivation probability, together with other applications of
Chapter 7. STOPPING POWERS AND ALPHA-STICKING IN $\mu$CF

our apparatus is discussed in this section. (What will be proposed here are, in reality, severely limited by the event rates at least with TRIUMF intensity, but let us be a little imaginative, in ending the thesis!)

One of the routine methods of detecting the presence of negative muons is the observation of muonic X-rays, or similarly nuclear capture gamma-rays. Unfortunately, the original idea of using the muonic silicon X-rays from the silicon detector to identify $\mu\alpha$ for the initial sticking measurement, turned out to be not very useful, because the probability of the stripping process in silicon is dependent on the initial energy of the $\mu\alpha$, and its correction would be somewhat dependent on the knowledge of $\mu\alpha$ kinetics. We instead proposed to use gold and look for the 356 keV muonic capture gamma rays.

Let us assume that from the initial phase of the sticking measurement we know $\omega_s^{\text{eff}}(T_d)$, the effective sticking with degrader thickness $T_d$. Recall that

$$\omega_s^{\text{eff}}(T_d) = \omega_s^0 (1 - R(T_d)). \quad (7.21)$$

Our objective here is to determine $\omega_s^0$ by measuring $R(T_d)$, the reactivation probability at degrader thickness $T_d$.

The apparatus we propose is the same as the initial phase (Fig. 7.2), except we have a germanium X-ray detector instead of the silicon, and we place a gold plate in front of it to strip off the muons from mu-alphas and then observe the characteristic gamma rays (and X-rays) from gold. It was shown by Cohen[86] that the muon in $\mu\alpha$ with initial energy of 3.5 MeV gets completely stripped by the time the $\mu\alpha$ travels 10 $\mu$m in gold, or by the time it loses energy to 2.5 MeV.

First, with a target with no degrader, we measure the number of 356 keV gold capture gammas (for example) from the muon that was stripped off from $\mu\alpha$. Coincidence with a fusion neutron collinear with $\mu\alpha$ emission is demanded to clean up the signal. Let us
call this number $Y_0$, which can be written

$$Y_0 = N_f \cdot \omega_s^0 \cdot \epsilon_0,$$

(7.22)

where $N_f$ is a normalized number of all fusion events and $\epsilon_0$ is the detection efficiency including germanium, neutron and coincidence solid angle.

Second, we add a degrader of thickness $T_d$, which must be the same thickness as the previous measurement where $\omega_s^{eff}(T_d)$ was obtained. We again measure the gold capture gamma rays in coincidence with the neutron ($Y_1$), which is now written

$$Y_1 = N_f \cdot \omega_s^0 \cdot (1 - R(T_d)) \cdot \epsilon_1,$$

(7.23)

where $\epsilon_1$ is the detection efficiency for this time. If $T_d$ is not too large, i.e., the energy of the $\mu\alpha$ is not too low, it appears safe to assume the probability of the muon getting stripped from $\mu\alpha$ in gold is 100% according to Cohen[86]. Since other efficiencies cancel with each other, we have $\epsilon_0 = \epsilon_1$, so we can simply take the ratio of the observed gamma-neutron coincidence counts (normalized to singles neutrons) $Y_1$, $Y_0$ to obtain

$$1 - R(T_d) = \frac{Y_1}{Y_0}.$$

(7.24)

It should be noted that the above assumption is much less dependent on the theoretical model of the $\mu\alpha$ kinetics, upon which the calculation of the reactivation probability $R$ depends heavily. It can also be tested, for example, using a beam of muonic helium which will be described later. Thus, combining the above value of $R(T_d)$ with the $\omega_s^{eff}(T_d)$, we can determine the nearly model-independent value of the initial sticking $\omega_s^0$.

The last thing proposed in this thesis is the use of $\mu$CF as a source for a beam of muonic helium. In our standard three-layered target arrangement, if we put a degrader that is thick enough to stop all the alphas, we obtain a “beam” of muonic helium with
rather well defined energy. An example of the energy distribution was already shown in Fig. 7.4. This would be complimentary to a keV muonic helium source, proposed by Nagamine[87], since our beam typically has energy of order of 1 MeV, although the intensity is much lower. New “Atomic Physics” experiments with muonic helium beam may become possible. Some interesting examples include the test of Cohen’s prediction on the stripping process in gold foil and the study of transfer process to neon which can be deposited directly on top of the beam source layer. It may also be possible to obtain a muonic $^3$He beam from the sticking in the D-D fusion reactions, which could be interesting for muon capture studies.

All the discussion in this section ignores the problem of the event rates, and may be rather unrealistic, at least with the muon intensity of TRIUMF. However, some of them may become possible in future muon facilities. At least the initial phase of the sticking experiment, which was discussed in detail in section 7.3, appears feasible, and a research proposal for TRIUMF Experiment Evaluation Committee is currently being prepared.
Chapter 8

CONCLUSION

The energy loss of alpha particles was utilized to determine the thickness and uniformity of solid hydrogen and other frozen gas targets for muon catalyzed fusion experiments. The energy of alpha particles from an array of americium sources was measured by a silicon detector. By moving the detector and measuring at different positions, the uniformity of the films was determined. For the conversion of the measured energy loss into the thickness, the latest stopping power and range table by ICRU[18] was employed, after critical review of the many tables.

An accuracy of about 5% was achieved at a few hundred $\mu$g/cm$^2$, which is limited by the stopping power uncertainty. The linear relation between the hydrogen thickness and the amount of injected gas was confirmed within the accuracy. This enables the extrapolation of the results beyond the range of measurement accessible by the present method. The cross contamination is found to be less than $0.8 \times 10^{-3}$ with 90% confidence level. A significant non-uniformity in the target profile was observed. This could be partly explained by a solid angle effect, which is consistent with a Monte Carlo study. However, an asymmetry in the thickness distribution is not clearly understood. While cryo-pumping during the deposition does not affect the target profile within the accuracy, the position change of the diffuser by 2.5 mm gives a significant difference in the thickness distributions. The effective thickness per unit gas input is determined to be $3.29 \pm 0.16$ $\mu$g/(cm$^2$-torr-litre) and the average non-uniformity (the weighted standard deviation of
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The thickness distribution) is about 7% for a Gaussian beam distribution of FWHM 20–25 mm. The neon targets had similar thickness profile to the hydrogen. An alternative method of deposition is also described, which may improve the uniformity of the target. The present method of thickness measurement can now be applied to other experiments where solid thin targets are required.

The importance of the stopping process in the alpha-sticking problem in muon catalyzed D-T fusion was discussed in detail. The physical phase effect of the stopping power of hydrogen may partly explain the discrepancy in the sticking values between theory and experiment at high densities. The concept of a new experiment to measure directly the sticking probability at high density was proposed. This offers certain advantages over LAMPF/RAL measurements. A Monte Carlo simulation of the experiment was performed. A very preliminary result from a test run is presented. Preparation of the research proposal for TRIUMF Experiments Evaluation Committee is in progress.
Bibliography


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