SONOCHEMICAL REACTOR DESIGN BASED ON
ELECTROSTATIC FILM TRANSDUCERS

by

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We accept this thesis as conforming
to the required standard

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April 1994

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Abstract

Sonochemistry is a relatively new field of chemistry which uses intense sound energy to influence chemical reactions. Laboratory investigations have indicated that a number of commercially important chemical processes can be improved by sonochemistry techniques. However, there does not appear to be any sonochemical reactor designs which are feasible for use at the industrial scale. All the reactor designs proposed thus far utilize piezoelectric or magnetostrictive transducers for generating the sound energy. These transducers are widely used for other sound applications, but their properties make construction of an industrial-scale sonochemical reactor complex and expensive. A new sonochemical reactor design based on electrostatic film transducers is introduced here which may present a breakthrough in the economic feasibility of industrial sonochemical processing. A working prototype of this reactor was not achieved because the maximum sound pressure which could be obtained from simple electrostatic film transducer prototypes was only a fraction of that required for cavitation. An acoustic model of the reactor was developed and experimentally confirmed which showed that the reactor sound pressure was limited primarily by excessive damping in the transducer.

Commercialization of this technology will first require the construction of a high-pressure electrostatic film transducer with minimal damping. This could then be followed by lab-scale reactor manufacture, pilot plant evaluations, and finally industrial-scale implementation.
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Sonochemistry is an emerging field of chemical processing technology which utilizes intense sound energy to enhance chemical reactions. Research in sonochemistry began near the turn of the century with the development of the first sources of high-pressure sound. Laboratory investigations have shown that a number of commercially important reactions can be improved by the use of sonochemistry. In the past decade, sonochemistry research has been rapidly expanding, led by new applications in the synthesis of fine chemicals and pharmaceuticals. [1]

The chemical effects of high-pressure sound are due primarily to a phenomenon known as cavitation. Cavitation is the formation of small vapour bubbles in a liquid subject to large negative pressures. In sonochemistry, cavitation is generated by the large negative pressures associated with the rarefraction portion of intense sound waves. Extreme temperatures and pressures of the order of 5000 °C and 500 atm. are generated inside collapsing cavitation bubbles. Also, powerful shock waves caused by collapsing bubbles break up solid particles and cause vigorous mixing of the liquid. These and other effects combine to enhance certain chemical reactions. [1]

Intense sound energy used in sonochemistry is produced in a device known as a sonochemical reactor. Fig. 1.1. shows a simple sonochemical reactor which is used for laboratory experiments. It consists of a rectangular tank containing the liquid chemicals with electroacoustic transducers mounted on the walls. The transducers
Introduction

convert electrical energy into sound energy which is then radiated into the liquid. The sound causes cavitation, which in turn stimulates the reaction of chemicals in the liquid. Typically, the sound used is in the frequency range of 10 - 100 kHz and at sound pressures in excess of one atmosphere.

Figure 1.1. Basic Sonochemical Reactor

A number of sonochemical reactor designs have been proposed to implement sonochemistry on an industrial scale. Most of these designs use either piezoelectric or magnetostrictive transducers. These transducers are well-proven and commonly used for other acoustical applications; but they are impractical for use in an industrial-scale sonochemical reactor because of three main problems: 1) Their bulky, and in some cases fragile, nature makes construction and maintenance of the reactor awkward and costly; 2) Their high acoustic impedance makes efficient coupling to liquids difficult; and, 3) Their high mechanical Q requires that they always be operated precisely at resonance, which further complicates their design. As a result of these, and other problems, none of these previous designs has been successful commercialized.

This thesis presents a new sonochemical reactor design based on electrostatic
film transducers. Electrostatic film transducers have properties which are entirely different from those of magnetostrictive and piezoelectric transducers. These unique properties make this new sonochemical reactor intrinsically free of the deficiencies faced by previous reactor designs and may for the first time allow the construction of a commercially viable industrial-scale sonochemical reactor.

In Chapter 1 some basic concepts of acoustics, cavitation, sonochemistry, and sonochemical reactor design will be reviewed. Chapter 2 explains the general operating principles behind electrostatic transducers and the design of electrostatic film transducers. The mechanisms which limit the maximum sound pressure possible from an electrostatic transducer are discussed in detail because of their major importance to the feasibility of this technology. Chapter 3 describes a number of different, but similar, designs for a sonochemical reactor based on electrostatic film transducers. Chapter 4 reports the results of a prototyping effort to construct an electrostatic film transducer powerful enough for use in the sonochemical reactor designs described in Chapter 3. Chapter 5 discusses an acoustic model of the reactor which is used to determine the effect of various reactor parameters on overall performance and provide insight into methods of optimization. Chapter 6 outlines potential strategies for commercialization the new reactor technology; and Chapter 7 gives some conclusions from the work.
1.1 Acoustical Fundamentals

1.1.1 Wave Equation

At low sound pressures, the acoustic behaviour of most fluids is linear and can be derived from the following simple analysis.[2] Referring to Fig. 1.2, a one-dimensional fluid element is shown where \( p \) is the pressure; \( \xi \) is the liquid particle displacement at an initial longitudinal position \( x \); \( S \) is the area of the fluid element perpendicular to the \( x \) direction; and \( \rho \) is the fluid density. The corresponding pressure and liquid particle displacements at position \( x + dx \) are \( p + \partial p / \partial x \, dx \) and \( \xi + \partial \xi / \partial x \, dx \) respectively.

\[ \begin{align*}
\begin{array}{c}
\text{Figure 1.2. One-Dimensional Fluid Element} \\
\begin{array}{c}
\text{x} \\
p \\
\xi \\
\text{x + dx} \\
p + \partial p / \partial x \, dx \\
\xi + \partial \xi / \partial x \, dx
\end{array}
\end{array}
\end{align*} \]

Applying Newton's second law to the forces acting on this fluid element yields

\[ - S \frac{\partial p}{\partial x} \, dx = \rho S dx \frac{\partial^2 \xi}{\partial t^2} \]
Chapter 1 Literature Review

For negligible changes in fluid density $\rho$ this becomes

$$-\frac{\partial p}{\partial x} = \rho \frac{\partial^2 \xi}{\partial t^2}$$

The bulk modulus $B$ of a fluid is defined by

$$\frac{\partial p}{\partial x} = B \frac{\partial^2 \xi}{\partial x^2}$$

Combining these equations yields the one-dimensional wave equation

$$\frac{\partial^2 p}{\partial x^2} = \frac{1}{c^2} \frac{\partial^2 p}{\partial t^2}$$

The general three-dimensional wave equation is usually given in terms of the acoustic velocity potential $\varphi$ as

$$\nabla^2 \varphi + k^2 \varphi = 0$$

where $v = \nabla \varphi$ and $p = -\rho \frac{\partial \varphi}{\partial t}$. For the simple case of plane wave propagation into an infinite fluid medium (also called free-field propagation), solutions to this equation are of the form

$$p = Ae^{i(\omega t - kx)}$$

where $\omega = 2\pi f$ and $k$ is the classical attenuation due to viscous and thermal conduction losses. [3] A case commonly seen in sonochemistry is cylindrical wave propagation where the solutions involve combinations of Bessel functions. These solutions will be used later in Chapter 5 for the modelling of the reactor.

1.1.2 Acoustic Impedance and Intensity

The impedance $z$ of an acoustic system is defined as

$$z = \frac{p}{v}$$

where $p$ is pressure and $v$ is velocity. For example, the acoustical impedance of a fluid
in free-field sound propagation is \( z = \rho c \), where \( \rho \) is the liquid density and \( c \) is the wave speed (i.e. speed of sound) in the liquid.

If sound waves encounter a change of medium, e.g. a wall, then reflection can occur. Fig. 1.3 shows a one-dimensional model of an interface between two mediums with acoustic impedances of \( z_1 \) and \( z_2 \) respectively.

The fraction of the incident sound pressure reflected \( R \), and transmitted \( T \), at this boundary for normal incidence is found by

\[
R = \frac{z_1 - z_2}{z_1 + z_2}; \quad T = 1 - R
\]

Thus, a wall of acoustic impedance similar to the fluids on either side of it will be nearly transparent to sound. For example, when the fluid is water, low density polymers such as LDPE and RTV Silicone are suitable for use as acoustically transparent barriers. [4] Higher impedance materials such as metal can also be nearly acoustically transparent if they are thin enough. [5]

The intensity of a sound wave is defined as the sound power per unit area radiated in the direction of propagation. Using the definition for acoustic impedance \( z \),
the intensity $I$ (W/m$^2$) is given by

$$I = \frac{p^2}{2z} = \frac{v^2z}{2}$$

where $p$ and $v$ are the peak pressure and velocity of the sound wave respectively. A related quantity is the average sound energy density $E = I/z$ (W/m$^3$).

1.1.3 Standing Waves and Fluid Resonance

If sound waves are reflected by an impedance interface back towards their source, there will be more than one wave occupying the same space. The waves travelling forward and the waves travelling backward are superimposed on each other. The result is a wave which no longer appears to be travelling, and hence is termed a standing wave. Mathematically, the superposition leads to the following equations for sound pressure and velocity of a plane wave:

$$p_x = (p_r + p_i) \cos(kx)e^{j\omega t}$$
$$v_x = (v_r + v_i) \sin(kx)e^{j\omega t}$$

where $x$ is the direction of propagation: $p_r, p_i, v_r, v_i$ are the incident and reflected pressure and velocity components respectively.

![Figure 1.4. Standing Wave](image)

Fig. 1.4 shows a standing wave between two reflecting walls excited by a
transducer attached to one of them. Analogous to the case of a vibrating string, the longitudinal standing wave exhibits a series of nodes and antinodes which repeat at intervals of $\lambda/2$ where $\lambda = c/f$ is the wavelength of sound at a frequency $f$. One can see here that the velocity and pressure standing waves are exactly 90 degrees out of phase; the velocity nodes correspond to the pressure antinodes and vice versa.

The standing wave shown in Fig. 1.4 can be interpreted as one of the resonant modes of this acoustic system. Since fluids have both elasticity and mass, they exhibit resonant behaviour. A one dimensional fluid medium of length $L$ will resonate at frequencies corresponding to wavelengths that satisfies the relation $n\lambda/2 = L$ where $n$ is an integer. The mode shapes of these resonant frequencies are precisely those described in the equations above. If the walls containing the fluid are not perfectly rigid (as is usually the case) they become part of the acoustic system and influence its resonant characteristics.

In a resonant acoustic system the only energy supplied by the sound source (i.e. transducer) is that necessary to overcome damping losses in the system. This differs from free-field propagation in that there is no net transport of sound energy. The *quality factor* "Q" of a resonance is the ratio of sound energy stored in the system to the sound energy dissipated in one period of oscillation. Thus, the Q gives a measure of the degree of damping in the acoustic system. When the damping is low, the Q is high, and high sound pressures can be generated with a relatively small amount of sound energy.
1.1.4 Liquid Damping

At low sound pressures, the main sources of damping in a liquid are viscous and thermal conduction losses which can be predicted quite well using classical sound attenuation theory. [6] At high sound pressures, liquids experience dramatic increases in damping with the onset of cavitation and other non-linear phenomenon such as:

1) Sawtooth Waveforms: Changes in liquid properties (i.e. density, bulk modulus) with pressure cause the wave speed to become faster at high pressure points than at lower pressure points. The peaks of the wave try to "catch up" with the troughs, and the originally sinusoidal wave assumes a sawtooth shape as shown in Fig. 1.5. The sawtooth waveform causes some of the sound energy to be carried by higher spectral frequencies, and since classical attenuation losses increase as the square of frequency, this leads to a major increase in damping for the sawtooth sound wave as compared to the sinusoidal sound wave. In addition, large pressure gradients on the face of the sawtooth cause large temperature gradients, which in turn increase losses due to thermal conduction. [7]

Figure 1.5. Sawtooth Waveform at High Sound Pressure (after Rozenberg)
2) Acoustic Streaming: High damping caused by the sawtooth waveforms and cavitation lead to large sound energy gradients in the liquid. These gradients cause the liquid to flow (or stream) in the direction of the sound energy propagation (i.e. away from the sound source). Streaming also occurs on a microscopic scale in the region surrounding collapsing cavitation bubbles. Both types of streaming lead to viscous losses which further increase damping. [7]

1.2 Cavitation

1.2.1 Cavitation Formation

Cavitation bubbles originate from small nucleation points in a liquid. These nucleation points are typically small bubbles, on the order of 1 μm in diameter, which are attached to dust particles contained in the liquid.[6] These bubbles are then expanded into cavitation bubbles during the negative pressure portion of a sound wave. This explains the initiation of cavitation. However, once cavitation has begun, there are about $10^5$ times more cavitation bubbles than nucleation points. Thus, the progression to fully developed cavitation must involve a chain reaction. [7]

It is theorized that the implosion of the first cavitation bubbles creates large numbers of hot microbubbles which then serve as the nucleation points for the cavitation bubbles formed in the next acoustic cycle. Thus, the number of cavitation bubbles continually increases until a steady-state maximum has been reached. Because there are more nucleation points at steady-state, this suggests that the sound pressure required to sustain cavitation should be lower than that required to initiate it. This has been experimentally confirmed. [7]
1.2.2 Cavitation Threshold

Insight into the threshold sound pressure required to produce cavitation can be obtained by examining the unstable force equilibrium acting on a cavitation bubble (see Fig. 1.6) given by the following relation:

\[ P_g = P_0 + \frac{2\sigma}{R_0} \]

where \( P_g \) is the pressure of the gas contained inside the bubble, \( P_0 \) is the ambient pressure in the surrounding liquid, \( R_0 \) is the instantaneous radius of the bubble, and \( \sigma \) is the liquid surface tension. If the ambient pressure in the liquid (\( P_0 \)) is lowered (i.e. in the rarefraction portion of a sound wave), then the bubble will expand. [8]

![Figure 1.6. Unstable Force Equilibrium on a Cavitation Bubble](image)

To obtain spontaneous rapid growth of a bubble of radius \( R_b \), the negative sound pressure \( P_a \) must exceed a value known as the Blake Threshold Pressure [8] given by

\[ P_a = P_0 + \frac{8\sigma}{9} \left[ \frac{3\sigma}{2[P_0 + (2\sigma/R_b)]R_b^3} \right]^{\frac{1}{2}} \]

Once a microbubble has been subjected to a negative pressure exceeding the Blake Threshold Pressure as described above, it may grow by one of two processes:
rectified gaseous diffusion or vapourization. Which process will dominate depends on the microbubble's initial radius $R_0$, and the ratio of acoustic pressure ($P_T$) to ambient hydrostatic pressure ($P_0$). For air-saturated water, and a sound frequency of 20 kHz, $P_T/P_0$ is plotted versus $R_0$ in Fig. 1.7.[8] This graph defines three regions of interest. In region $X$, the sound pressure is insufficient to support bubble growth by either mechanism, and nothing happens. Above the threshold line $AB$ is region $Y$, where rectified gaseous diffusion occurs. Gas dissolved in the liquid diffuses into the bubble during the expansion cycle, and diffuses out of the bubble during the contraction cycle.

However, since the diffusion is proportional to the surface area of the bubble, more gas enters the bubble during expansion when the bubble is large than is removed on compression when the bubble is smaller, and thus the microbubble radius ($R_0$) increases with each successive acoustic cycle. Two possibilities now exist: if the microbubble was initially at a state such as point $S$, then it will grow until it enters the

Figure 1.7. Cavitation Threshold for Air Saturated Water (after Ronald)
region Z; or, if it is at state such as point T, it will grow indefinitely until it either
collapses due to surface instability, or floats out of solution. The oscillation and
continual growth of gaseous microbubbles over many acoustic cycles is often referred
to as **stable cavitation**.

Microbubbles in region Z will be subject to rapid expansion due to vapourization
of the surrounding liquid and a **transient cavitation** bubble will be formed. This
expansion will occur entirely during the one sound rarefraction phase. As the sound
wave enters the compression phase, the cavitation bubble will become unstable and
undergo violent collapse. Both transient and stable cavitation are responsible for
chemical effects observed in sonochemistry. [9]

The graph shown in Fig. 1.7 defines the transient cavitation threshold for a
specific frequency and set of liquid properties. Variation in the cavitation threshold
with changes in these parameters are explained below.

1.2.3.1 Effect of Acoustic Frequency on Cavitation Threshold

Fig. 1.8 shows the variation in cavitation threshold with frequency of sound
waves for both aerated and deaerated water. In this graph, the cavitation threshold is
given as a function of acoustic intensity, as opposed to acoustic pressure which was
used in Fig. 1.7. The cavitation threshold curve for deaerated water is noticeably higher
than that of normal aerated water because it contains fewer nucleation sites and thus
the chain reaction required to initiate cavitation is slowed.

Fig. 1.8 shows data for the specific case of water under certain conditions, but
the general trend is similar for most liquids: The cavitation threshold is constant up to
about 50 kHz at which point it rises very rapidly with frequency and continues to do so on up into the MHz range.

This rapid increase above 50 kHz is the reason most commercial uses of cavitation, such as ultrasonic cleaning use frequencies as low as possible without entering the audible range where they would cause excessive noise. Cavitation does not occur at high frequencies (i.e. > 5 MHz) because the rarefraction cycle in the sound wave is too short to allow the formation of a cavitation bubble.[7]

![Graph of cavitation threshold vs. frequency for water](image)

**Figure 1.8. Variation of Cavitation Threshold with Frequency for Water (after Mason)**

Laboratory investigations suggest that chemical effects of cavitation depend on the frequency of sound used. Each reaction has its own optimal frequency for cavitation energy to be most efficiently converted into chemical effects. [10] However, there are no sonochemical reactors, even at the lab-scale, which can operate in a
frequency tunable mode, and so the determination of this optimal frequency has been very difficult with existing equipment.

1.2.3.2 Effect of Liquid Properties on Cavitation Threshold

A number of liquid properties affect the cavitation threshold. The more important of these and their effects are summarized in Table 1.1.

<table>
<thead>
<tr>
<th>Liquid Property</th>
<th>Effect of Increase in Property on Cavitation Threshold</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface Tension</td>
<td>Raises</td>
<td>For example, the addition of a surfactant (i.e. soap) to the liquid will lower its surface tension and therefore lower the cavitation threshold.</td>
</tr>
<tr>
<td>Viscosity</td>
<td>Raises</td>
<td>The cohesive forces in a viscous liquid are much larger than in a non-viscous liquid. Since it is the cohesive forces which the acoustic pressure must overcome, one expects that an increase in viscosity of a liquid will raise the cavitation threshold.</td>
</tr>
<tr>
<td>Vapour Pressure</td>
<td>Lowers</td>
<td>High vapour pressure liquids tend to have a lower cavitation threshold. However, the cavity implosions in low vapour pressure liquids are less violent and therefore produce weaker sonochemical effects.</td>
</tr>
<tr>
<td>Ambient Temperature</td>
<td>Lowers</td>
<td>Higher temperatures raise the vapour pressure of a liquid and therefore also lower its cavitation threshold. The fact that sonochemical effects are reduced with increased temperature is contrary to conventional reaction mechanics as may require cooling of the process liquid.</td>
</tr>
<tr>
<td>Ambient Pressure</td>
<td>Raises</td>
<td>Higher ambient pressures (Po) raise the threshold for cavitation because a larger acoustic pressure (Pt) is now required to get the same ratio of Pt/Po as shown in Fig. 1.7 (p. 12)</td>
</tr>
</tbody>
</table>

Table 1.1. Effect of Liquid Properties on Cavitation Threshold [13]

1.2.3 Acoustical Effects of Cavitation

Cavitation in a liquid causes its behaviour to deviate significantly from that predicted by the linear acoustic theory introduced in Sec. 1.1. One of these changes is a reduction of liquid impedance.[7] Fig. 1.9 shows a plot of liquid impedance (expressed as a fraction of characteristic impedance ρc) vs. the sound pressure (expressed as the voltage applied to transducer). As the sound pressure increases
beyond the cavitation threshold, liquid impedance drops to nearly a tenth of its nominal value. This drop is explained by the fact that a cavitating liquid becomes "softer" due to the inclusion of highly compressible gas pockets. A decrease in liquid impedance means that both the sound pressure and intensity produced by the transducer will be less than that expected based on linear theory for the same acoustic velocity. This must be corrected for in the design of a sonochemical reactor.

\[ \frac{\rho_x c_x}{\rho_0 c_0} \]

Figure 1.9. Liquid Impedance vs. Transducer Voltage (after Rozenberg)

1.2.4 Erosion Effects of Cavitation

Cavitation implosions which occur near solid surfaces cause microscopic erosion. This is the principle behind ultrasonic cleaning. In ultrasonic cleaning equipment the sound pressure is quite low, and the duration of exposure short, so cavitation causes only minimal amounts of erosion. With the slightly higher sound pressures and much longer durations of exposure found in sonochemistry, erosion becomes a serious problem.
Thus, in designing a sonochemical reactor, it is desirable to limit intense cavitation to a volume of the liquid away from the equipment surfaces. This can be achieved by focussing the sound energy as will be discussed later. Where contact cannot be avoided, metals such as ASTM 316 grade stainless steels or titanium are typically used to resist cavitational erosion.[11]

1.2.5 Chemical Effects of Cavitation

Some aspects of sonochemistry which make it of interest to the chemical process industry are summarized in Table 1.2.

<table>
<thead>
<tr>
<th>Benefits of Sonochemistry</th>
</tr>
</thead>
<tbody>
<tr>
<td>• Increased Reaction Rates</td>
</tr>
<tr>
<td>• Higher Quality Products</td>
</tr>
<tr>
<td>• Lower Temperatures and Pressures Required</td>
</tr>
<tr>
<td>• New Reaction Pathways</td>
</tr>
</tbody>
</table>

Table 1.2. Benefits of Sonochemistry

The chemical effects of cavitation have been partially attributed to the extreme temperatures and pressures which exist inside the cavitation bubble during collapse.[12] If the collapse is assumed to be adiabatic, the maximum pressure ($P_{\text{max}}$) and temperature ($T_{\text{max}}$) present in the cavitation bubble occur at the end of its collapse and are found by the following expressions:

$$P_{\text{max}} = P_g \left[ (P_0 (\gamma - 1)/P_g) \right]^{\gamma/\gamma-1}$$

$$T_{\text{max}} = T_0 P_0 (\gamma - 1)/P_g$$

Here, $P_0$ is the ambient pressure of the liquid; $P_g$ is the pressure in the cavitation bubble just before collapse; $\gamma$ is the ratio of specific heats for the vapour in
the bubble; and $T_0$ is the ambient temperature. $P_g$ may be approximated by the vapour pressure of the liquid.[13]

From these equations one can see that as the pressure in the bubble prior to implosion ($P_g$) increases, the amount of energy released by the implosion also increases. Thus, the chemical effects of cavitation can be improved by raising $P_g$ through pressurization of the process liquid. However, higher sound pressures will now be required to produce cavitation (see Fig. 1.8).

The chemical effects of cavitation appear to depend heavily on the number of cavitation bubbles ($N$) present in the liquid. [7] Fig. 1.10 shows the variation in number of cavitation bubbles ($N$) with the voltage squared ($V^2$) applied to the transducer of a sonochemical reactor. The sound pressure and intensity are approximately linearly related to the voltage squared for the transducer used in obtaining this data. Also plotted in this graph is the amount of sonoluminescent light emitted by the liquid which is a measure of the sonochemical activity occurring in the cavitation bubbles.
The two curves match quite well if the appropriate scale is chosen for the ordinate axis. Operation of a sonochemical reactor at the peak of these curves, where there is a maximum number of cavitation bubbles, will give optimal conversion of sound energy to chemical effects.

The mechanism for chemical effects of cavitation can be roughly categorized as follows: If all reagents are in the liquid phase then the reaction is said to be 
homogeneous. Otherwise, if one or more of the reagents is a solid, or there is a solid catalyst present, then the reaction is said to be heterogeneous.

Homogeneous reactions are affected as follows: 1) vapour contained inside the imploding cavity experiences high pressures and temperatures which cause the formation of reactive species such as OH⁻; 2) chemical species in the liquid surface surrounding the bubble will be exposed to, and subsequently react with, the reactive
species generated in the bubble; and, 3) shock waves generated by the supersonic collapse of the bubble wall causes intense shear forces in the nearby liquid which can break apart polymer chains.

*Heterogeneous* reactions are affected as follows: 1) bubbles imploding on or near the surface of the solid cause mechanical erosion which continually cleans the surface of reaction products, thereby enhancing reactivity; and, 2) small solid particles may be ruptured by shock waves, increasing reagent surface area, and reactivity.

1.3 Conventional Electroacoustic Transducers

The main component of a sonochemical reactor is a device which converts electrical energy into acoustic energy, the *electroacoustic transducer*, or *transducer* for brevity. A wide range of transducers operating on different principles are available for use in sonochemistry and a recent review of these is given in [14]. Of the transducers available, the two types which are used extensively in ultrasonic cleaning equipment, and lab-scale sonochemical reactors, are *piezoelectric* and *magnetostrictive* transducers.

1.3.1 Piezoelectric Transducers

*Piezoelectric* transducers are made from materials which expand and contract in an applied electric field, and thereby create sound from an electrical signal. A number of naturally occurring crystals such as quartz and Rochelle salt exhibit a piezoelectric effect and these were some of the first materials used to make transducers. Natural piezoelectric materials have become obsolete by the advent of synthetic piezoelectric materials such as barium titanate and PZT which give much
higher performance.

Piezoelectrics such as barium titanate and PZT are manufactured by first molding the desired shape of the transducer from a specially blended ceramic which does not yet exhibit piezoelectric properties. The ceramic blank is then polarized at high temperature with a very strong electric field to produce a crystal structure which is piezoelectric. Because piezoelectric ceramics are "artificially" piezoelectric, they are susceptible to losing their piezoelectric effect. This happens gradually over time, but the process is accelerated when the material is exposed to high temperatures. Once the piezoelectric effect is lost, it cannot be recovered without repolarization.

Even if a piezoelectric transducer is operated in a relatively cool environment, heat generated by internal damping can create high temperatures in its core. This fact limits the amount of power which can be handled (i.e. transduced) by piezoelectric materials.

Piezoelectric transducers exhibit mechanical properties which one would expect from ceramic materials; they are rigid and fragile. This makes the manufacture and installation of large transducers, which is necessary for industrial-scale sonochemical reactors, very difficult and expensive. The fragility of the transducer also makes it susceptible to fracture on impact during operation or maintenance. A further mechanical problem with piezoelectric ceramics is that they are prone to mechanical fatigue when subjected to large tensile stresses. This problem is partially solved by the use of a sandwich transducer construction where the piezoelectric
material is held in compression between two metal blocks bolted together as shown in Fig. 1.11.

![Piezoelectric Sandwich Transducer](image)

**Figure 1.11. Piezoelectric Sandwich Transducer**

The efficiency of converting electrical energy into sound energy is high in piezoelectric materials when they are operated in a resonant mode. For resonance to occur, the transducer must be driven at a frequency such that the transducer is integer multiples of a half-wavelength thick. At low frequencies, the large thickness required is obtained by using a sandwich construction of the type mentioned above wherein the metal blocks form part of the half-wavelength required. The disadvantage of using sandwich transducers is that they typically have a lower operating efficiency.[15]

The rigidity of piezoelectric ceramics gives them a high acoustic impedance. For transmission of the sound energy into lower impedance mediums such as liquids, an impedance matching layer of some type is often used to obtain a better coupling between the transducer and the liquid. An impedance matching layer (see Fig. 1.12) is designed to be an odd number of quarter wavelengths in thickness and have an impedance \( z_m \) which is the geometric mean of the transducer \( z_t \) and the liquid \( z_l \); i.e. \( z_m = (z_t z_l)^{0.5} \).
Acoustic impedance matching layers reduce system efficiency by introducing additional transmission losses. They also increase manufacturing costs, and therefore are preferably avoided.

1.3.2 Magnetostrictive Transducers

Magnetostrictive transducers operate similar to piezoelectric transducers with the distinction that they are made from materials which expand and contract in response to a magnetic field as opposed to an electric field. Magnetostrictive transducers are made from ferromagnetic materials such as Nickel, Ferrite, Cobalt-Nickel, etc. When operated at mechanical resonance these materials also have high efficiency, although typically not quite as high as piezoelectric materials like PZT. As with piezoelectric materials, power capacity is limited by mechanical fatigue and high core temperatures due to heat generated by internal damping. Magnetostrictive transducers are also subject to eddy current and magnetic hysteretic losses which increase rapidly with frequency. This imposes a practical upper limit of around 50 kHz for the operation of magnetostrictive transducers, and ideally should be below 20 kHz.
Table 1.3 lists some common piezoelectric and magnetostrictive materials and their reported efficiencies.[16]

<table>
<thead>
<tr>
<th>Transducer Material</th>
<th>Efficiency (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Piezoelectrics</td>
<td></td>
</tr>
<tr>
<td>Barium Titanate</td>
<td>8.4</td>
</tr>
<tr>
<td>Lead Metaniobate</td>
<td>32</td>
</tr>
<tr>
<td>PZT-5A</td>
<td>70</td>
</tr>
<tr>
<td>PZT-4</td>
<td>65</td>
</tr>
<tr>
<td>Magnetostrictives</td>
<td></td>
</tr>
<tr>
<td>Nickel Alloys</td>
<td>30-65</td>
</tr>
<tr>
<td>Nickel Ferrites</td>
<td>90</td>
</tr>
</tbody>
</table>

Table 1.3. Electroacoustic Efficiencies of Common Transducers [16]

1.4 Sonochemical Reactor Design

The main objective in the design of a sonochemical reactor is to produce a device with the lowest total cost per unit of production. The total cost of a reactor can be broken down into two components: capital costs and operating costs.

Operating costs are all the costs incurred after the reactor has been commissioned and is in operation. Energy consumption and maintenance are the two main operating costs for a sonochemical reactor. Capital costs reflect the costs of purchasing and installing the reactor equipment itself, and the costs of any additional plant (i.e. structures, piping, equipment) required to incorporate the device into the process. A cost example given in [14] suggests that capital costs are likely to be of the same order of magnitude as operating costs over a ten year operating lifetime.
The design criteria relevant to minimization of reactor costs are as follows:

1.4.1. Energy Consumption

Sonochemical reactors convert electrical energy into chemical energy through a series of stages such as those shown in Fig. 1.13.

![Diagram of energy conversion stages in a sonochemical reactor]

*Figure 1.13. Energy Conversion Stages in a Sonochemical Reactor*

In the first stage, electrical energy from a standard power grid (e.g. 60 Hz) enters a *generator*. The generator is basically comprised of an amplifier and transformer which produce a high voltage (e.g. 2 kV) signal at the desired frequency (e.g. 10kHz). This electric signal is fed to the transducer (e.g. piezoelectric) which converts it into sound waves. The sound waves are then radiated into the process liquid, either by direct contact with the liquid, or by transmission through the reactor wall. The geometry of the reactor wall, which forms the boundary to the process liquid, determines the nature of the sound field which is generated. A cavitation zone occurs in the high pressure region of the sound field where the desired chemical effects occur.
At each stage in the conversion process, energy is lost from the system in the form of heat. This author estimates the following efficiencies for each stage in a typical reactor: generator: 90%; transducer: 80%; reactor wall: 50%; acoustic field: 60%; and cavitation zone: 60%; then the overall conversion efficiency is roughly 13%.

The majority (87%) of the energy supplied to the reactor is being lost as heat which serves no useful purpose, and may cause operational problems with reactor components such as the transducer. Some steps which can be taken at each stage to minimize the losses of energy and improve the overall efficiency of the reactor are as follows:

1.4.1.1 Generator

The losses in the generator will be minimized when the power factor of the generator’s load is as close to unity as possible. This corresponds to minimizing the amount of reactive power circulating the electrical system. Reactive power is zero when the electrical system is operated at resonance. Generator designs are available which incorporate reactive tuning elements (i.e. inductors) to ensure the circuit operates at resonance.

1.4.1.2 Transducer

As mentioned earlier, transducer losses are minimized when it operates at its mechanical resonant frequency.

1.4.1.3 Reactor Wall

For simplicity of construction and maintenance, transducers are often situated on the exterior of the reactor wall containing the process liquid. In this case the wall
of the reactor must serve two functions: First, it must contain the process liquid in a confined volume; and, second, it must allow passage of sound energy from the transducer into the process liquid. These are conflicting requirements because it is desirable to have the wall very strong (and therefore rigid) to satisfy the first purpose, but precisely the opposite is necessary to satisfy the second. Thus, reactors which use transducers on the exterior of the reactor wall always face an inherent trade-off between efficiency and durability.

1.4.1.4 Sound Field

The sound field is determined by the geometry of the acoustic boundaries in the reactor, which are usually just the reactor walls. A focussing sound field efficiently converts the sound energy radiated through the reactor wall into a zone of intense cavitation. Another advantage of a focussing geometry is that cavitational erosion is minimized because the highest sound pressure occurs in a volume away from the equipment surfaces. Of the focussing geometries available, this author believes the cylindrical one is best because it gives a uniform flow pattern for the process liquid, unlike a sphere for instance. However, a cylindrical reactor is difficult to construct using conventional piezoelectric or magnetostrictive transducers.

1.4.1.5 Cavitation Zone

As mentioned earlier in Sec.1.2.5 the chemical effects of cavitation depend on the number of cavitation bubbles occurring in the cavitation zone. From Fig. 1.10 it is apparent that there exists some power level at which the transducers should be driven for generating the maximum number of cavitation bubbles. In certain applications a
catalyst or other source of activation energy (e.g. UV light) could be placed in the focal region to further enhance the use of cavitation energy.

1.4.2 Maintenance Costs

The cost of maintaining a sonochemical reactor includes not only the direct cost of servicing defective reactor components, but also the cost of plant down-time and production interruption. Often it is these latter components which constitute the majority of the maintenance cost. Maintenance costs are best minimized by making the reactor components as durable as possible so that fewer breakdowns occur. Where maintenance is inevitable, the reactor should be designed such that repairs can be executed simply, and with a minimum amount of down-time for the equipment.

The destructive capability of cavitation on equipment surfaces has been cited as a potential maintenance problem by companies looking at the use of industrial-scale sonochemical reactors.[10] As mentioned earlier, a reactor sound field which focusses the acoustic energy away from the equipment surfaces may prevent this problem from occurring.

1.4.3 Capital Costs

The capital cost of the reactor depends primarily on the ease with which it can be manufactured. Complex reactor designs for obtaining marginal improvements to efficiency are not likely to be feasible. Additional capital cost incurred by the plant (i.e. space, structure, piping, etc) required to accomodate the reactor will be reduced by a reactor design which is compact and requires a limited amount of piping. For example, a reactor system which utilizes one hundred small reactors operating in
parallel requires more plant than a system using several large reactors, and therefore will have a much higher total capital cost.

1.5.4 Prior Art Sonochemical Reactor Designs

A number of designs for sonochemical reactors have been proposed in the literature. Most of these designs use a flow-through duct arrangement for carrying the process liquid through the active zone of the reactor. [13] A flow-through duct allows for uniform processing of the liquid reagents, and is more generic in its industrial applicability. The geometry of the duct is typically cylindrical to produce a focusing effect for reasons which have already been mentioned in Sec. 1.4.1.4. Other duct geometries which have been proposed are pentagonal or hexagonal to allow for simpler mounting of sandwich transducer modules.

In general, piezoelectric transducers are used because these are the most efficient, and also can be readily manufactured into a variety of shapes and sizes. Given a cylindrical duct geometry and the use of piezoelectric transducers, there are essentially two methods of combining them to produce a sonochemical reactor:

1) The transducer is manufactured in the shape of cylindrical ring which completely envelops the process liquid and is located within the interior of the reactor wall as shown in Fig. 1.14. [26]; or,

2) The transducer is mounted as a sandwich module on the exterior of the reactor wall as shown in Fig. 1.15. [27]
Chapter 1 Literature Review

Figure 1.14. Sonochemical Reactor with Cylindrical Transducer Element

The approach shown in Fig. 1.14 can efficiently generate a cavitation zone but suffers the following problems: a) The diameter and length of the transducers is limited by manufacturing difficulties due to the brittle nature of piezoelectric ceramics. This limits the volumetric capacity and/or maximum sound pressures which can be generated; and, b) The transducer material is subject to large tensile stresses and thus is susceptible to mechanical fatigue.

The approach shown in Fig. 1.15 does not experience these two problems because: a) The size of the reactor is not limited by the size of the transducers; if a larger reactor is required, it simply means more transducer modules must be used; and, b) The transducers are held in compression by the sandwich module and thus are not as susceptible to fatigue. However, the penalty for using this approach is that it is less efficient because the transducer must radiate the sound energy through the reactor wall where large losses can occur due to the impedance mismatch. The resulting sound field is not well focussed into the center, and most of the cavitation occurs near the reactor wall. This results in further losses in efficiency and may also
make the reactor prone to cavitation erosion.

Figure 1.15. Sonochemical Reactor with Modular Transducer Elements

Variations on the design shown in Fig. 1.15 have been proposed in order to improve the coupling efficiency between the transducers and the process liquid. One approach attempts to correct the efficiency problem by energizing resonant modes in the reactor shell. [16] Another approach uses a buffer liquid between the transducer and the wall of the reactor to conduct the sound energy. Here, sound energy losses on the order of 60% have been reported to occur at the interface of the buffer liquid and the reactor wall [17].

One can conclude, with the exception of the most recent designs proposed, that the lack of commercial success of these prior art designs is indicative of their poor economic feasibility.
Chapter 2

Electrostatic Film Transducers

One of the first reported uses of an electrostatic transducer for the production of high-pressure sound was in 1915 by French physicist Paul Langevin in a prototype sonar device for detecting submarines. Soon after its construction, Langevin found that a piezoelectric transducer made of quartz was able to radiate far more power. With the discovery of piezoelectrics, and later magnetostrictives, the use of electrostatic transducers for high-pressure sound generation was abandoned.[18]

High-pressure electrostatic transducers re-emerged in the mid-1980's with the invention of the electrostatic film transducer. These electrostatic film transducers have unique properties which make them ideal for use in a sonochemical reactor.

The basic principles of electrostatic transducers and the factors limiting their pressure will be discussed in Sections 2.1, 2.2, and 2.3. Specific electrostatic film transducer designs will be discussed in Sec. 2.4.

2.1 Operating Principles of Electrostatic Transducers

\[\text{Figure 2.1. Components of an Electrostatic Transducer}\]
In their most basic form, electrostatic transducers consist of two electrode plates compliantly spaced and electrically insulated from each other. When an alternating voltage is applied across the plates, electrostatic pressures are created which in turn generate sound in an acoustic system. The basic components of an electrostatic transducer are shown in Fig. 2.1.

The electrostatic pressure developed between the two oppositely charged parallel electrodes is given by the following relation:

\[ P_e = \frac{\epsilon V^2}{2d_e^2} = \frac{\epsilon E^2}{2} \]

where \( V \) is the voltage, \( d_e \) is the effective electrode separation, and \( E = V/d \) is the electric field between the electrodes, and \( \epsilon \) is the permittivity constant = \( 8.85 \times 10^{-12} \) N/V². The effective electrode separation \( d_e \) is given by

\[ d_e = \sum_n \left[ \frac{d_1}{\kappa_1} + \frac{d_2}{\kappa_2} + \frac{d_3}{\kappa_3} + \ldots + \frac{d_n}{\kappa_n} \right] \]

where \( d_n \) is the thickness of each layer of dielectric material with a dielectric constant \( \kappa_n \). Materials with a higher dielectric constant contribute less to the effective thickness of the dielectric.

It has been assumed that the acoustic displacement of the electrodes \( \xi \), where \( \xi = \frac{V/2\pi f}{2\pi f} \), is small compared to the electrode separation. This assumption may not be valid for the high sound amplitudes (e.g. \( \xi = 2 \mu m \)) used in sonochemistry. However, this approximation is sufficient for the purposes of this investigation.

If the signal voltage applied to the transducer electrodes is an alternating sinusoid of frequency \( f \), and voltage \( V_s \), then the electrostatic pressure amplitude \( P_{ea} \)
developed is $P_e/2$ at a frequency $2f$ (see Fig. 2.2). This result comes from

$$(\varepsilon V/2d \times \sin\omega t)^2 = P_e (1 + \cos 2\omega t)/2 = P_{ea} (1 + \cos 2\omega t).$$

Since the mean electrostatic pressure is half the peak, then so also is the amplitude.

![Figure 2.2. Signal Voltage and Corresponding Electrostatic Pressure](image)

If an acoustic wave of the same frequency $f$ as the signal voltage is desired, this can be achieved by superimposing the alternating signal voltage $V_s$ on a DC Bias voltage $V_{dc}$ about an order of magnitude larger (i.e. $V_{dc} >> V_s$) (see Fig. 2.3).

![Figure 2.3. Signal Voltage and Corresponding Electrostatic Pressure with DC Bias](image)

In this case, the electrostatic pressure amplitude $P_{ea}$ is approximated by [19]

$$P_{ea} = \frac{\varepsilon V_s V_{dc}}{d^2}$$

### 2.2 Dynamic Behaviour of an Electrostatic Transducer

The oscillating electrostatic pressure in the transducer supplies sound energy to the acoustic system it is a part of. Like all acoustic systems, the nature and magnitude of the sound pressure developed depends on the dynamic properties of
the entire system. For electrostatic transducers, the dynamic properties can be represented by a single degree of freedom model such as the one shown in Fig. 2.4. The mass, stiffness, and damping elements of the radiating electrode are designated $m_t$, $k_t$, and $c_t$ respectively.

![Figure 2.4. Model for an Electrostatic Film Transducer](image)

The total dynamic impedance $z_t$ of the transducer is then

$$z_t = c_t + j \left( \frac{k_t}{\omega} - m_t\omega \right)$$

where $\omega = 2\pi f$, is the circular frequency.

For the simple case of free-field propagation the transfer function between electrostatic pressure amplitude $P_{ea}$ and sound pressure $p$ is

$$\frac{p}{P_{ea}} = \frac{z_t}{z_l + z_t} = \frac{\rho c}{c_t + j \left( \frac{k_t}{\omega} - m_t\omega \right) + \rho c}$$

where the free-field impedance of the fluid $z_l = \rho c$.

### 2.3 Maximum Electrostatic Pressure

As shown earlier, the maximum electrostatic pressure which can be generated by an electrostatic transducer is proportional to the square of the maximum electric field. Limitations on the maximum electric field which can be applied are discussed in the following sections:
2.3.1 Dielectric Strength of Gases: Paschen's Law

For the dielectric of the transducer to be compressible, a substantial fraction of its volume must be gas. The insulating strength of a gas is determined by Paschen's law. [26] Paschen's law states that the breakdown voltage $V_b$ of a gas, is a function only of the product of gas pressure $P$ and the distance between the electrodes $d$, i.e.

$$V_b = f(Pd)$$

For air, the behaviour of this function has been experimentally determined and is given by the Paschen curve in Fig. 2.5.

![Paschen Curve for Air](image)

Figure 2.5. Paschen Curve for Air

The Paschen curves for other gases are similar to that of air. Most have the characteristic of a breakdown voltage steadily declining as $Pd$, the Paschen Product is reduced until it reaches a minimum value, termed the Paschen Minimum. Below the Paschen minimum, the breakdown voltage begins to rise again, climbing with a much
steeper slope than above the Paschen minimum. Using Paschens law, one can
determine the breakdown voltage of any combination of gas pressure and electrode
spacing.

If one assumes that the gas contained in the dielectric is at atmospheric
pressure, the maximum electric field obtainable as a function of electrode spacing
can be calculated. This is done for air in Fig. 2.6. Here one can see dramatically the
rapid increase in electric field strength with decreasing electrode spacing.

![Electric Field Strength vs. Electrode Spacing for Air at 1 atm.](after Saums et. al)

Figs. 2.5 and 2.6 show the measured dielectric strength of air. There exists a
certain class of gases, called electronegative gases, which are more resistant to
ionization than air. Table 2.1 shows a comparison of the relative dielectric strength of
some electronegative gases to air at a given value of Pd.
Table 2.1. Comparison of Electronegative Gases to Air at same Pd (after Saums)

The use of electronegative gases can yield substantial improvement in dielectric strength over air. However, there are difficulties associated with the manufacture and operation of a transducer using them, such as sealing the transducer, incompatibility with transducer components, etc.

2.3.2 Electron Field Emission

Looking at Fig. 2.6, one might wonder what happens at very small values of d. Does the electric field strength become infinite? As intuition would suggest, this is not the case. Paschen’s law is based on theory that electrode gaps break down due to ionization of the gas contained between them. If the Paschen product $Pd$ is very small, then the mean free path between gas molecules also becomes very small, and breakdown by ionization is difficult. However, before ionization ceases to be a problem, another breakdown mechanism will become dominant, electron field emission.

Theory predicts that at electric fields of the order $5 \times 10^8 \text{ V/m}$, electrons will be drawn from the negative electrode (cathode) and accelerated toward the positive electrode (anode). Upon striking the anode, positive ions will be discharged which are
in turn accelerated back towards the cathode. The process expands geometrically until breakdown occurs. In practice, field emission can cause breakdown in *nominal* electric fields $E_n = V/d$ as low as $2 \times 10^7 \, \text{V/m}$. [21] Field emission occurs at nominal fields much lower than those predicted by theory because of *field intensification* near sharp irregularities on the surface of the electrodes (see Fig. 2.7).

![Figure 2.7. Field Intensification due to Electrode Surface Irregularities](image)

Here the *local* electric field $E_i = \mu E_n$ is increased by a field intensification factor $\mu$. Even electrode surfaces which have been highly polished using mechanical buffing or electropolishing can have field intensification factors as high as 100. [21] Two methods of improving the field emission strength of electrodes are:

1) **Solid Dielectric Coatings:** Application of a high field strength solid dielectric such as mica or polyester (i.e. Mylar) to the electrode surfaces may increase the ultimate field strength of the transducer by up to 70% [20]. This added strength is probably due to the straightening out of the concentrating field lines before they reach the surface of the electrode. Another effect of surface dielectric coating is a reduction in pre-breakdown currents known as *dark currents*. This may improve the long term durability of the transducer. Note that because the transducer is using AC voltage, the electrodes alternate being the cathode, and thus both surfaces must be coated.
2) \textit{In Situ} Electrode Conditioning: With a large resistance placed in series with the transducer, a voltage (DC or AC) is applied, at first low and then increased incrementally. The transducer is left for some period of time at each increment before progressing to the next level. The field emitting sites on the electrodes are eroded away slowly due to the limited current flow. After a number of increments, the maximum breakdown voltage will be obtained. \cite{22}

It appears from the literature that the maximum nominal field attainable between two well-prepared electrodes is likely to be no more than $5 \times 10^8 \text{ V/m.}$ \cite{23} This corresponds to a maximum electrostatic pressure amplitude of about 5.5 atm.

2.3.3 Dielectric Strength of Solids

The solid dielectric materials used to maintain the void spaces between the transducer electrodes must be selected so that they have a dielectric strength at least as high as the breakdown strength of the voids due to gas ionization or field emission. There are two aspects of dielectric strength for solids: \textit{bulk} dielectric strength, and \textit{surface} dielectric strength. The bulk dielectric strength is the strength of the solid to an electric field applied across its thickness. Values for bulk dielectric strength ($E$) in some common insulating materials are given in Table 2.2 along with their dielectric constant ($\kappa$) and the product of the two ($E\kappa$). ($E\kappa$) is the figure of merit because materials with a higher dielectric constant will support a higher nominal electric field ($V/d$) for the overall transducer. This follows from the fact that the $D$ ($= E\kappa$) field in the solid dielectric and the air gap of the transducer must be equal.

The bulk dielectric strength of solid materials displays a similar dependency on
Chapter 2 Electrostatic Film Transducers

thickness as Paschen's law for gases: the thinner the dielectric is, the higher is its strength. Thus, the bulk dielectric strength reported for a material should indicate at what thickness the measurement was taken. However this is not always done for insulators which are typically used in thick sections anyhow. The dielectric strengths noted with an asterisk (*) in Table 2.2 were not provided with a sample thickness in their reference.

<table>
<thead>
<tr>
<th>Material</th>
<th>Dielectric Strength (E) MV/m for 25μm thick sample</th>
<th>Dielectric Constant k</th>
<th>Product (E x k) MV/m</th>
</tr>
</thead>
<tbody>
<tr>
<td>Muscovite Mica [28]</td>
<td>200</td>
<td>6.5</td>
<td>1300</td>
</tr>
<tr>
<td>Mylar [23]</td>
<td>280</td>
<td>3</td>
<td>840</td>
</tr>
<tr>
<td>Kapton [23]</td>
<td>303</td>
<td>3</td>
<td>909</td>
</tr>
<tr>
<td>Paper [22]</td>
<td>50*</td>
<td>2.2</td>
<td>110</td>
</tr>
<tr>
<td>Silicone Rubber [28]</td>
<td>20*</td>
<td>3</td>
<td>60</td>
</tr>
<tr>
<td>Urethane Rubber [28]</td>
<td>20*</td>
<td>7.8</td>
<td>156</td>
</tr>
<tr>
<td>Natural Rubber [22]</td>
<td>24*</td>
<td>2.6</td>
<td>62.4</td>
</tr>
</tbody>
</table>

Table 2.2. Bulk Dielectric Strength of Selected Materials

The surface dielectric strength is the strength of a solid to an electric field applied across its surface. This strength is typically lower than the bulk dielectric strength for the same material, although specific data on this parameter is scarce.

2.3.4 Corona Damage

Another potential source of breakdown for solids is corona damage. Corona is the pre-breakdown ionization of gas near a dielectric surface. The subsequent
bombardment of the dielectric surface with ions causes heat and/or erosion which ultimately leads to breakdown. This mechanism can cause certain types of dielectrics to fail at electric fields one order of magnitude below their bulk dielectric strength. For example, the corona strength of a 25 \( \mu \)m thick film of Mylar is quoted by its manufacturer as \( 1.6 \times 10^7 \) V/m which is an order of magnitude less than its bulk dielectric strength of \( 2.8 \times 10^8 \) V/m.[23] Certain materials, such as mica and silicone, are highly resistant to corona, and their dielectric strength is not as limited by it.[22]

2.4 Electrostatic Film Transducer Designs

In the late 1980's, two electrostatic transducer designs were proposed which partially address the aforementioned problems of dielectric breakdown, while still maintaining a reasonable dielectric compliance, and thus may be capable of producing the high electrostatic pressures required for sonochemistry.

All of these transducer designs use a thin film of porous elastomeric as a dielectric to support the spacing of the two electrodes. High compressibility is achieved because of the large gas fraction and the resilient elastomer structure. High electric field strength is achieved because the gas spaces are made thin enough that a small \( Pd \) can be obtained. Because these transducers have the properties and appearance of a polymer film, they are called electrostatic film transducers, or EFT for short.

One of the EFT designs disclosed by Whitehead et. al. in [27], called EMEET, is shown in Fig. 2.8. This design uses a series of elastomeric strips or nodules to directly define the spacing between the electrodes. The spacing \( d \) between the
electrodes can be made to whatever distance \( d \) desired to give the required stiffness and ease of manufacture. The void space can be evacuated to reduce the gas pressure \( P \) and thus the Paschen product \( Pd \) to obtain the necessary electric field strength. Evacuation allows the use of larger electrode spacings \( d \) and also substantially reduces problems due to corona damage. Field emission resistance can be improved by the coating of the electrodes with a dielectric layer.

![Figure 2.8. EMEET Electrostatic Film Transducer (after U.S. Pat. 4,883,765)](image)

An EFT design disclosed by Kirjavainen in [28], called ETMF, is shown in Figure 2.9.

![Figure 2.9. ETMF Electrostatic Film Transducer (after U.S. Pat 4,654,546)](image)
This design consists of a closed cell foam of flat, biaxially oriented, bubbles in a polymer matrix. The bubbles are at atmospheric pressure but can be made very thin so they have a high dielectric strength. The thinness of the polymer matrix gives it a high dielectric strength as well. The structure of the polymer matrix is very flexible and the compliance of the transducer depends only on the compressibility of the filling gas. [19]
Chapter 3

EFT Sonochemical Reactor

3.1 Description

Using the electrostatic film transducer (EFT) technology described in Chapter 2, a new sonochemical reactor design has been developed by this author and his thesis supervisor Dr. A. B. Dunwoody, called the EFT reactor.

Like previous reactor designs, the EFT reactor uses a cylindrical flow-through geometry. A basic embodiment of the reactor, shown in Fig. 3.1a, consists of a structural cylinder for carrying the process liquid with the transducer adhered directly to the interior surface of the cylinder wall. The transducer, with the addition of some type of protective coating, is in direct contact with the process liquid. The sound waves generated by the transducer are focussed along the axis of the cylinder where they cause intense cavitation.

If the protective layer of the transducer is insufficient (i.e. due to excessive process liquid temperatures), then the use of an additional interior wall is required.
This embodiment is shown in Fig. 3.1b. The space between the transducer and the inner wall is filled with a coupling liquid which transmits the sound energy generated by the transducer to the process liquid. The coupling liquid can be circulated through a cooling system if necessary to ensure it remains at a temperature compatible with the transducer. A coupling liquid with a high cavitation threshold liquid is preferably used as this will reduce unwanted cavitation which lowers efficiency. The interior wall is constructed so as to be acoustically transparent.

The embodiments shown in Figs. 3.1a and 3.1b require that the transducer be located on the interior surface of the reactor wall. Although these embodiments are simple to construct, they may not be acoustically optimal. If it is desired to place the transducer at a different radial distance then the embodiments shown in Figs. 3.2a and 3.2b can be used.

![Diagram](image)

**Figure 3.2a.**

**Figure 3.2b.**

Similar to the inner cylinder just described for use in confining the process liquid, this additional support cylinder is also made to be acoustically transparent. The space between the support cylinder and the inner surface of the reactor wall is filled with a coupling liquid. This liquid may also be used to as a coolant to maintain a
desired temperature for the transducer. In Fig. 3.2b the design has been augmented with an inner wall for confining the process liquid to a selected volume. A coupling liquid once again fills the space between the inner wall and the transducer support wall for transmitting the acoustic energy therebetween. In all embodiments which use an additional interior cylinder, the pressure of coupling liquid will have to match that of the process liquid. This can be achieved by allowing a portion of the interior wall to deform in response any pressure fluctuations. For example this could be some type of flexible diaphragm.

Fig. 3.3 shows an example of how the embodiment in Fig. 3.2a might look like in longitudinal cross-section. Here, the reactor has been connected to the process piping system by flanges.

![Figure 3.3. Example Longitudinal Cross-Section](image)

In the reactor embodiments described above, the transducer is shown continuous about the circumference of the cylinder, and completely enveloping the process liquid. Manufacturing the device may be simpler if the transducer is divided into a number of discrete segments. This approach is shown in Fig. 3.4 for the
embodiment previously described in Fig. 3.1a. A segmented transducer reactor will perform similar to its continuous transducer counterpart provided that the circumferential spacing between the segments remains small compared to a wavelength.

![Figure 3.4. EFT Reactor With Segmented Transducer](image)

In specific processing applications it may be useful to combine the use of cavitation with other sources of activation energy such as UV light or catalysts. This can be efficiently achieved by placing the catalyst or UV light source along the longitudinal axis of the reactor as exemplified by Fig. 3.5.

![Figure 3.5. EFT Reactor With UV Light Source](image)

When higher sound pressures are required (i.e., for processing high cavitation threshold liquids), then it may be desirable to use a spherically focussing reactor geometry such as the one shown in Fig. 3.6. In this reactor, process liquid is carried
into and out of the spherical chamber by a split two-way pipe. Coupling liquid fills the space between the exterior of the pipe carrying the process liquid and the surface of the transducer so that acoustic energy may be efficiently transmitted therebetween. A grommetted joint seals the process liquid pipe and the reactor wall together to prevent leakage of the coupling liquid.

![Figure 3.6. Spherical EFT Reactor](image)

### 3.2 Advantages of EFT Reactor Over Previous Designs

First of all, unlike the fragile cylindrical piezoelectric transducers used in previous reactors (Fig. 1.14 for example), electrostatic film transducers are flexible and durable. This flexibility allows them to be easily conformed into a cylindrical shape which requires no elaborate mounting arrangements and thus reduces manufacturing costs. Shipping, handling, and maintenance costs are all also reduced by the to the simple, durable nature of these devices.

Secondly, the direct coupling of the transducer to the liquid in an EFT reactor eliminates the reactor wall losses associated with reactors which use sandwich transducers located on the exterior of the reactor wall. (Fig. 1.15 for example). The
problem of high sound pressures causing cavitation on the surface of the reactor wall adjacent to the transducer is also eliminated. This reduces both acoustic decoupling of the transducer and cavitationsal erosion of the reactor wall.

Finally, the low mechanical Q of electrostatic film transducers allows the reactor to operate efficiently at frequencies slightly off resonance. This eliminates the need for expensive phase-locked-loop tuning generators required to maintain piezoelectric transducers driven at resonant frequency when slight changes in the acoustic system occur such as liquid temperature, viscosity, etc. For laboratory-scale reactors, the low mechanical Q of these transducers has another advantage: the reactor can be operated effectively at a range of frequencies. This allows sonochemistry researchers the ability to easily determine the optimal frequency of sound to use for each specific chemical reaction, something that was prohibitively costly in the past.
A prototype EFT Sonochemical Reactor capable of generating cavitation in water is a key step in proving the feasibility of this technology. To achieve this goal, an electrostatic film transducer capable of generating approximately 1 atm of sound pressure in water is required.

4.1 Electrostatic Film Transducer Prototypes

The author’s design of a suitable prototype electrostatic film transducer was governed by the following two physical constraints: 1) the stiffness of the transducer must be as low as possible; and, 2) the dielectric must have an electric field strength of at least $2 \times 10^8$ V/m. In addition, there were two economic constraints: 3) the available generator equipment could supply a maximum of 2 kV (peak) to the transducer electrodes; and, 4) the prototype must be realizable with about $10,000$ of resources.

4.1.1 ETMF Prototypes

Prototype transducers using the ETMF design were obtained from a Finnish company, Messet Oy. These transducers were made using a polypropylene matrix and air as the bubble gas. The total dielectric thickness of the film is about 28 μm and the individual bubbles are about 1 μm thick. The bubbles make up about 50% of the uncompressed film volume. Electrodes were a thin metal layer deposited on the surface of the film.[19]
The maximum voltage that could be applied to the ETMF transducer was found to be around 800 V (peak). At this voltage, very little acoustic pressure (~ 0.3 kPa) was produced in the test system (which is described later in section 4.4) and so the use of this transducer to generate cavitation would not be possible.

4.1.2 EMEET Prototypes

Using the principle of the EMEET transducer design, fine textured elastomer surfaces, thin open cell foams, or elastomeric papers containing a substantial fraction of their volume as gas could be used as a compressible dielectric material. The inclusion of the gas fraction is essential since solid materials have almost no compressibility in the range of pressures being considered here.

Because of the voltage limitation mentioned earlier, the effective thickness of the dielectric could not be greater than 10 μm. Also, the material must have high elasticity and high dielectric strength. A search for suitable dielectric materials in the trade literature revealed a number of possible candidates, but almost all of them were far too thick to be useful.

The one exception was fine tissue paper, normally used in multiple layers for wrapping delicate merchandise. Samples of this paper obtained from a local giftshop were approximately 25 μm thick as measured using a micrometer. Although this material was also far too thick to produce the required electric field at 2 kV, it was decided that so long as the material was able to hold 2kV, the purchase of a new generator capable of a higher voltage might be justified to allow the production of higher electric fields.
Chapter 4  Electrostatic Film Transducer Prototyping

The two basic transducer designs developed using the tissue are shown in Fig. 4.1. The first design is comprised of one sheet of the tissue paper only. The tissue is sandwiched between the aluminum coatings of two 25 μm thick sheets of aluminized mylar whose conductive coating acts as an electrode. Unfortunately, microscope inspection of the tissue paper showed that there were "holes" of up to 1 mm in diameter where there were no fibers present due to the non-uniform distribution of fibers during manufacture of the paper. These "holes" might allow one electrode to contact the other and thus cause a short circuit.

To eliminate this as a potential source of failure, a second design was used in which one of the sheets of aluminized mylar was reversed so that the 25 μm thick layer of mylar was now part of the dielectric. The mylar acts to insulate the electrodes completely and thereby prevent short-circuiting across the "holes" in the tissue. However, this tissue/mylar dielectric has two problems associated with it: 1) the increase in dielectric thickness to 33 μm (κ_{mylar} = 3) reduces the maximum electric field which may be applied with the 2 kV available from the generator to about 6 x 10^7 V/m; and, 2) the mylar itself may act as a cause of breakdown for the dielectric due
bulk dielectric failure or corona damage.

It was found in the first sets of experiments that the tissue dielectrics shown in Fig. 4.1 were not compliant enough. In an effort to reduce the stiffness of the dielectric, prototypes were constructed using an elastomeric adhesive bonded to the tissue layer (see Fig. 4.2). The elastomeric adhesive came as a sheet approximately 50 \( \mu \text{m} \) thick (\( \kappa = 3 \) assumed) supported on both sides by release paper. It was hoped that the tissue fibers embedded into the flexible adhesive would reduce the stiffness of the transducer.

![Figure 4.2. Elastomer Adhesive Dielectric.](image)

Prototypes were also constructed where the 1 mil (25 \( \mu \text{m} \)) thick Mylar insulator was replaced with a 75 \( \mu \text{m} \) thick Kapton (\( \kappa_{\text{Kapton}} \approx 3 \)) insulator. It was hypothesized that a thicker insulating film would be less susceptible to creasing during the transducer construction process. Creases create points at which the electrostatic field will be locally enhanced and thus could provide a mechanism for premature failure of the polymer insulator which would in turn lead to overall dielectric failure in the transducer. Kapton was used for the thicker insulator because it was readily available whereas the same thickness in Mylar was not.
4.1.3 Multiple Dielectric Layers

Another approach to reducing the stiffness of the transducer was to use multiple layers of dielectric and electrodes stacked together as shown in Fig. 4.3. The stiffness of the multi-layer transducer is reduced in direct proportion to the number of layers used. For example, if five transducer layers are stacked together, the composite transducer will have an effective stiffness which is one-fifth of the individual transducer layers. This follows directly from the rule of mechanics for stiffness elements in series.

The effect of using multiple dielectric layers was tested using the basic tissue/mylar dielectric. Transducers made of 3, 5, 7, and 11 layers of dielectric were constructed. To minimize the dielectric thickness, each dielectric layer was independently energized.

![Figure 4.3. Multiple layer Electrostatic Film Transducer](image)

4.1.4 Evacuation of Dielectric Gas

The tissue paper used in the dielectric designs just described creates air pockets which may be up to the thickness of the paper (25 μm) in depth. According to Fig. 2.5, air at 1 atm with this thickness should have a dielectric strength of 1.2 x
10^7 V/m, which is far too low. However, if the air pressure is decreased to 0.08 atm, the dielectric strength rises to 3.2 x 10^8 V/m which is adequate. A standard roughing pump can draw vacuums on the order of 0.01 atm very easily, and so this technique was used to increase the dielectric strength of the transducer prototypes.

The evacuation of the dielectric gas is also helpful in reducing the possibility of breakdown due to corona damage of the paper fibers or the mylar insulator. The potential for field emission breakdown is unknown since the *in situ* microsurface of the aluminized mylar electrodes was not investigated.

### 4.3 Transducer Construction

All the transducers were constructed to a size of 20 cm x 15 cm. The layers of aluminized mylar and tissue were stacked and 20 cm long strips of household grade aluminum foil 20 cm wide were placed in contact with the respective electrodes for later connection to the generator output terminals. The transducer was then laminated using a conventional pinch roller laminating machine. The pinch rollers of the laminating machine removed any wrinkles and excess air in the layers of the transducer. The plastic laminating jacket also provided a simple and robust means of sealing the transducer from the liquid test environment. The aluminum foil electrodes extended well beyond the transducer layers and thus could be connected to without destroying the air-tight seal on the transducer.

In the transducers which were to be evacuated, the tissue paper layers were extended about 3 cm beyond the conductor layers on one side to allow passage of air out to the vacuum port which was later cut in the laminating jacket. Vacuum cups
machined from aluminum were glued to the laminating jacket over these holes and sealed using RTV silicone. Two vacuum ports were placed on each transducer, one was connected to the vacuum pump, and the other was connected to a pressure gauge. The two ports, placed as far apart as possible, were used to ensure that the entire transducer was in a vacuum, and not just the region around the suction port. The layout of the transducers prior to testing is shown in Fig. 4.4.

![Diagram of Prototype Transducer Layout](image)

**Figure 4.4. Prototype Transducer Layout**

### 4.4 Equipment and Apparatus

The transducers were set-up for testing in the apparatus shown in Fig. 4.5. A rectangular plexiglas tank filled with water served as the acoustic medium. This system was used in lieu of a cylindrical tank because of difficulties encountered in bending the transducers into a cylindrical shape. The transducer was attached to a 3 mm thick brass plate which was subsequently leaned against the side wall of the tank. A vacuum roughing pump was placed on a nearby table and connected to the transducer via the vacuum ports described above.
An Atlantic Research Corp. model BC-30 hydrophone (see Table 4.1 for characteristics) was used to sense the sound pressure in the water in front of the center of the transducer halfway towards the opposing wall. The hydrophone was held in position by placing its mounting sleeve inside a 30 cm length of 1 cm diameter aluminum pipe held with a ring-stand. The interior of the aluminum pipe was flooded to minimize its effect on the acoustic system.

<table>
<thead>
<tr>
<th>Operating Bandwidth</th>
<th>1 Hz to 40 kHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sensitivity</td>
<td>0.2 volts/psi</td>
</tr>
<tr>
<td>Capacitance</td>
<td>0.008 microfarads</td>
</tr>
<tr>
<td>Linearity</td>
<td>± 2% to 100 psi</td>
</tr>
</tbody>
</table>

**Table 4.1. Hydrophone Characteristics**

Noise-free cable supplied with the hydrophone was used to connect it to an oscilloscope where the output voltage amplitude was read. Because the frequency of the acoustic waves was double the electrical driving frequency (see Sec. 2.1), electromagnetic pick-up did not substantially influence the hydrophone readings. The hydrophone system was not calibrated.
The transducer was driven by the circuit shown in Fig. 4.6. A sinusoidal signal was generated by a manually tunable oscillator. This signal was amplified by a high-capacity audio amplifier, and then transformed to the operating voltage with a 20:1 turns ratio transformer. Bandwidths for the amplifier and transformer were 20 Hz - 40 kHz and 500 Hz - 10 kHz respectively. The amplifier and the transformer were both capable of handling at least 5 kW of apparent power. The voltage applied to the transducer was measured with a voltmeter connected to a 100:1 voltage divider circuit. This circuit had an error of +/- 10% over the frequency range of 5 - 10 kHz.

4.5 Transducer Test Procedure

Each transducer prototype was tested to determine the maximum sound pressure generated in the test tank before breakdown of the transducer. It was apparent from a frequency sweep with one of the transducer prototypes that the test system was exhibiting substantial resonant behaviour, as expected. If all of the tests were done at the same frequency, the output would vary substantially based on the dynamic properties of the transducers and how they influenced the resonant frequencies of the acoustic system. It was decided to use the best (i.e. one which
yielded the most sound pressure for a given voltage) resonant frequency for each transducer within the operating band of 10 - 30 kHz. The sound frequency which gave the best output for most tests was around 20kHz.

The following simple procedure was followed for each transducer test: Step 1: Using a low constant signal voltage (i.e. 150 V) sweep for the frequency which gives the highest sound pressure at the hydrophone. Step 2: At this frequency, incrementally increase the voltage until transducer failure occurs, noting the sound pressure reading from the oscilloscope at each interval. This procedure yielded the breakdown voltage for the transducer and the maximum sound pressure produced, which was invariably that occurring just prior to dielectric breakdown.

4.6 Experimental Results

4.6.1 Single-Layer Transducers

The data measured from the various transducer prototypes constructed are shown in Table 4.2. The first column gives the type of construction used for the dielectric. The second column lists the gas pressure in the air pockets between the fibers in the tissue paper (e.g. 0.01 atm is evacuated). The maximum nominal electric field (V/d) obtained in the transducer prior to electrical breakdown is given in the third column. This electric field is calculated based on the measured thickness of the dielectric and the maximum voltage measured. In the fourth column, the electrostatic pressure theoretically generated by this electric field is calculated based on the formulas described in Sec. 2.1. The fifth column contains the sound pressure obtained just prior to dielectric breakdown, which as expected, was the maximum
produced by a given transducer for the range of voltages over which it was tested. The electrostatic pressure amplitude ($P_{ea}$) (see Sec. 2.1) theoretically generated by the electroic field at dielectric breakdown is calculated and shown in column 5. The ratio of sound pressure to electrostatic forcing pressure amplitude ($P_a/P_{ea}$) is the most important figure of merit here and is calculated in the final column. This transfer function ratio ($P_a/P_{ea}$), which shows how much sound pressure is being produced for a given amount of electrostatic pressure, will be large if: a) the transducer is well coupled to the dominant resonant modes of the acoustic cavity; and, b) the overall damping in the acoustic system is not large.

To maximize the coupling of the transducer to the dominant modes of the acoustic cavity, the reactive impedance of the transducer should be as small as possible, thus giving it a "broadband" performance. Since the reactive impedance of electrostatic film transducers is dominated by stiffness (mass effects are negligible), the transducer's coupling to the resonant modes is expected to increase with decreasing transducer stiffness.

The damping of the acoustic cavity comes from three main sources: 1) damping in the transducer itself; 2) radiation losses from the cavity into the surrounding air; and, 3) damping of the sound in the liquid due to viscous losses. For the present purposes of finding a suitable electrostatic film transducer prototype, the concern is with minimizing the damping from the transducer, although this is largely beyond control at this time.
From Table 4.2, one observes that prototype #6 recorded the highest sound pressure of 3.9 kPa. This was followed closely by prototype #1 with a sound pressure of 3.4 kPa. The dielectric in these transducers was at atmospheric pressure. The remaining transducers were evacuated to a dielectric gas pressure of approximately 0.01 atm. As expected, evacuation increased the dielectric strength. However, the pressure transfer function was much lower than those without evacuation. For example, prototype #3 had a dielectric strength of $5.6 \times 10^7$ V/m, which produces an alternating electrostatic pressure of 6.9 kPa, but only 1.1 kPa of sound pressure was actually produced.

The (Pa/Pea) numbers show that there is a definite reduction in the pressure transfer function for those transducers which have been evacuated. This reduction is due to the compression preload on the transducer which substantially reduces its compliance (or increases it's stiffness). The reduced compliance arises from one or
both of the following:

1) The aluminized mylar spanning between the paper fibers deflects in a plate bending mode for no evacuation. When the atmospheric pressure is applied, the limits of plate bending are soon reached and the deflection is now due to a membrane stress mode which is much less compliant than for a plate bending mode. [24]

2) The cylindrical cellulose fibers which make up the tissue become compressed into a more elliptical shape with the pressure of the evacuation. The elliptical fibers have a much lower compliance than the cylindrical ones and thus the overall compliance of the transducer is reduced.

4.6.2 Multiple-Layer Transducers

If the performance of evacuated tissue transducers is being limited by the stiffness of the tissue dielectric then transducers constructed of multiple layers should produce higher sound pressures (see Sec. 4.1.3). To confirm this hypothesis, transducers using 3, 5, 7, and 11 layers of tissue/mylar dielectric were tested. The tissue/mylar dielectric was used instead of the apparently higher dielectric strength tissue because it was easier to construct using the single-sided aluminized mylar available, and it was also less susceptible to electrical shorting by the mechanism previously explained.

The performance of transducers consisting of multiple layers of a Tissue / Mylar dielectric is shown in Table 4.3 in a similar manner as the data presented in Table. 4.2.
Chapter 4  Electrostatic Film Transducer Prototyping

<table>
<thead>
<tr>
<th>Prototype #</th>
<th>Number of Dielectric Layers</th>
<th>Electric Field @Breakdown MV/m</th>
<th>Electrostatic Pressure @ Breakdown (kPa)</th>
<th>Sound Pressure @ Breakdown (kPa)</th>
<th>Pa/Pea</th>
</tr>
</thead>
<tbody>
<tr>
<td>3A</td>
<td>3</td>
<td>30.3</td>
<td>2.0</td>
<td>7.6</td>
<td>3.8</td>
</tr>
<tr>
<td>3B</td>
<td>3</td>
<td>18.2</td>
<td>0.7</td>
<td>2.4</td>
<td>3.3</td>
</tr>
<tr>
<td>5A</td>
<td>5</td>
<td>18.2</td>
<td>0.7</td>
<td>3.8</td>
<td>5.2</td>
</tr>
<tr>
<td>5B</td>
<td>5</td>
<td>26.1</td>
<td>1.5</td>
<td>7.8</td>
<td>5.2</td>
</tr>
<tr>
<td>5C</td>
<td>5</td>
<td>13.6</td>
<td>0.4</td>
<td>1.6</td>
<td>3.9</td>
</tr>
<tr>
<td>7A</td>
<td>7</td>
<td>18.2</td>
<td>0.7</td>
<td>5.2</td>
<td>7.1</td>
</tr>
<tr>
<td>7B</td>
<td>7</td>
<td>21.2</td>
<td>1.0</td>
<td>5.2</td>
<td>5.3</td>
</tr>
<tr>
<td>11A</td>
<td>11</td>
<td>10.6</td>
<td>0.2</td>
<td>1.8</td>
<td>7.3</td>
</tr>
<tr>
<td>11B</td>
<td>11</td>
<td>24.2</td>
<td>1.3</td>
<td>5.5</td>
<td>4.3</td>
</tr>
</tbody>
</table>

Table 4.3. Multiple Layer Transducer Performance Data

The last two columns of data in Table 4.3 are plotted in Fig. 4.8. The maximum value for each transducer type (i.e. number of dielectric layers) has been used.

Figure 4.8. Performance vs. Number of Transducer Layers

The graph on the left in Fig. 4.8 shows that increasing the transducer compliance by increasing the number of transducer layers substantially increases the
pressure transfer function (Pa/Pea). This suggests, as anticipated, that the coupling of the transducer to the dominant modes of the test tank acoustic cavity is better for lower stiffness impedances of the transducer.

However, although the pressure transfer function is increasing with the number of dielectric layers, the graph on the right in Fig. 4.8 shows that the breakdown field is dropping after about five dielectric layers, and thus the maximum sound power is actually decreasing. This drop in breakdown field is likely due to the fact that wrinkling of the tissue paper dielectric during transducer construction becomes a progressively more troublesome problem as the number of dielectric layers increases. Wrinkles in the tissue generate gas pockets which are much larger than the thickness of the tissue, and thus are more prone to breakdown according to Paschen's law (see Sec. 2.3.1).
In this chapter, an acoustic model of a basic sonochemical reactor is developed and experimentally confirmed. The purpose of this model is two-fold: first, it provides a physical understanding of how the reactor functions; and, second, it allows one to optimize the reactor parameters for maximum transfer of power from the transducer to cavitation.

5.1 Model Assumptions

A sonochemical reactor operates at such high sound pressures that its behaviour is highly non-linear. However, for this model standard linear theory is assumed in order to make the modelling problem more tractable. It is hoped that the results obtained, although not accurate, will be indicative of the general behaviour of the system. Namely, the optimal parameters predicted by the linear model should be similar to the optimal parameters for an actual non-linear reactor. The viscosity of the liquid has been neglected in this model since its effect is small compared to those associated with cavitation.

The sonochemical reactor design shown in Fig. 5.1 is the basis for the model. This reactor design is the same as the one shown earlier in Fig. 3.1a. It is very simple and thus will make both the analytical and experimental work much easier. The electrostatic film transducer is attached to the interior surface of a structural cylinder filled with liquid. The length of the cylinder is 2L and its inside radius is R.
Using a single degree of freedom model for the transducer, the reactor can be represented by the lump mechanical elements as shown in Fig. 5.2. Here, \( m \) is the mass of the structural cylinder wall, and \( k_c \) is its radial stiffness given by

\[
k_c = \frac{Et}{r^2}
\]

The stiffness and damping of the transducer are given as \( k_t \) and \( c_t \) respectively. It is assumed that the transducer mass is negligible at the frequencies of interest (10-20 kHz) and that the damping of the structural cylinder is negligible compared to that of the transducer.
transducer, one of which is integral with the structural cylinder wall. The acoustic pressure of the liquid acts on the diaphragm electrode the transducer. The boundary condition at both ends of the cylinder are pressure release surfaces (i.e. \( p = 0 \)). Because of symmetry, this model will also apply to the case of one end closed and one end open.

5.2 Development of the Analytical Model

For a cylindrical liquid column with pressure release boundary conditions at either end, it has been shown [25] that the velocity potential \( \varphi \) of the acoustic field will be of the form

\[
\varphi = \sum_b C_b \cos(M_b z) J_0(Y_b r)e^{i\omega t}
\]

where \( \varphi \) satisfies the standard wave equation

\[
\nabla^2 \varphi + k^2 \varphi = 0
\]

The wavenumber of the liquid \( k = \omega/c \) where \( \omega = 2\pi f \) and \( c \) is the wave speed of the liquid. To satisfy the pressure release boundary conditions the axial wavenumber \( M_b \) is defined as follows

\[
M_b L = \frac{\pi}{2}, \frac{3\pi}{2}, \frac{5\pi}{2}, \ldots, (2b - 1) \left(\frac{\pi}{2}\right)
\]

\[
M_b = \frac{(2b - 1)\pi}{2L}
\]

To satisfy the wave equation, the radial wavenumber \( Y_b \) is then

\[
Y_b = \sqrt{(\omega/c)^2 - M_b^2} \quad \left(1 \leq b \leq \frac{1}{2} + \frac{\omega L}{\pi c}\right)
\]

which is valid only over the interval shown because \( Y_b \) may not be complex. The pressure in the liquid is
\[ p = -\rho \frac{\partial \phi}{\partial t} \]
\[ = -j\rho \omega \sum_b C_b \cos(M_b z) J_0(Y_b r) e^{j\omega t} \]

A force balance applied to the system in Fig. 5.2 establishes a relation between the electrostatic pressure \( P_e \) and the sound pressure \( p \) as a function of the system elements which can then be manipulated to solve for the modal amplitude coefficients \( C_b \).

\[ C_b = \frac{P_e A(1)}{(2b - 1)\pi} \left[ \left( \frac{k_t}{j\omega} + c \right) Y_b J_1(Y_b r) - \left( 1 + \frac{k_t + j\omega c}{k_e - \omega^2 m} \right) j\omega \rho J_0(Y_b r) \right] \]

### 5.3 Experimental Confirmation of the Model

#### 5.3.1 Equipment and Apparatus

![Diagram of Reactor Prototype](image-url)

*Figure 5.3. Reactor Prototype*
A prototype reactor constructed to experimentally confirm the analytical model is shown in Fig. 5.3.

The prototype consists of a 280 mm long section of 124 mm ID, 5.5 mm thick aluminum pipe mounted upright and filled completely with water. A 4 mm thick silicone rubber sheet is glued to the end of the cylinder with RTV silicone adhesive to contain the liquid. Several layers of air bubble-pad used for packaging delicate items is placed between the bottom of the cylinder and the rigid table. The author believes the flexible silicone rubber on the bubble-pad is a good approximation to a pressure release end-condition.

An S28 ETMF sample supplied by Messet Oy, Finland was used as the transducer element. The transducer was first heat-laminated in a plastic jacket to seal it from the water which will be used to fill the cylinder. The lamination jacket glues directly to the surface of the transducer electrodes and thus there are no air inclusions which might introduce additional compliance. The laminated transducer was placed against the cylinder's inside wall by first wrapping it into a cylinder of the correct diameter, and then press-fitting it into the reactor cylinder. The small space (i.e. ~ 0.1 mm) remaining between the transducer and the cylinder wall became filled with water due to seepage. Although there is not a rigid bond between the transducer and the cylinder wall as described in the model, the shallow film of liquid between them behaves like one because its thickness is very small compared to the wavelengths of sound in question (70 - 140 mm).

The model BC-30 hydrophone (see Table 4.1 for details) used in the
electrostatic film transducer prototyping described in Chapter 4 was also used for this experiment. As before, the mounting sleeve of the hydrophone was inserted snugly into the inside of a length aluminum pipe which was then held in position using a ring-stand. The mounting pipe was again filled with water to minimize its effect on the acoustic system.

Electromagnetic interference between the transducer and hydrophone is minimized by using the electrode facing the water as a ground. A strip of aluminum foil connected to a separate ground circuit was also inserted into the water to reduce capacitive coupling between the transducer and the hydrophone. The transducer is driven by the same generator previously described in Fig. 4.6 and the output of the hydrophone is observed on the oscilloscope.

5.3.2 Experimental Results

Measurements were taken with the hydrophone placed at various depths along the longitudinal axis of the cylinder. By continuity, this axis always corresponds to a pressure antinode. With the transducer signal voltage held constant at a magnitude of 200V, the frequency of acoustic excitation (double the electrical frequency) was varied from 10kHz to 20kHz. Data on the sound pressure amplitude measured by the hydrophone was taken at intervals of 800 Hz. The sound pressure amplitude data was then plotted as a function of frequency.

Similar plots were made for comparison from data generated by the analytical model. The following properties were used for the aluminum cylinder: 2014-T6 aluminum $\rho = 2800 \text{ kg/m}^3$; and $E = 72 \text{ GPa}$ [25]. The stiffness of the transducer was
calculated using the following expression [19]
\[ k_t = \frac{\rho c^2}{ad} \]
where \( \rho \) and \( c \) are the density and wave speed of the gas in the transducer dielectric and \( a \) is the volume fraction of the transducer made up of gas. \( d \) is the thickness of the transducer dielectric. For the S28 sample of ETMF used in this experiment, \( \rho = 1.2 \text{ kg/m}^3 \), \( c = 340 \text{ m/s} \), \( a = 0.5 \), and \( d = 28 \mu\text{m} \) which yield a value of \( k_t = 9.9 \times 10^9 \text{ kg/m}^2\text{s}^2 \).[19]

The damping of the transducer \( c_t \), an unknown, was mathematically varied until the sharpness of the resonant peaks was similar to those obtained experimentally. This occurred at roughly \( c_t = 2.5 \times 10^5 \text{ kg/m}^2\text{s} \). The magnitude of the curves were then aligned by arbitrarily selecting the electrostatic pressure amplitude \( P_e \) used in the analytical model calculations. A \( P_e \) value of 11 Pa gave the best match. This is an order of magnitude less than the electrostatic pressure of 112 Pa which theoretically should have been present due to the 200 V signal applied.

The wave speed in the water was initially assumed to be the nominal value for water, namely \( c = 1500 \text{ m/s} \). However, due to the excess aeration of the liquid in the cavity from bubbles adhering the surfaces of the transducer and hydrophone support shaft a reduction was thought by the author to be in order. This would shift the predicted response slightly in terms of frequency and align it better with the experimental results. A value of \( c = 1350 \text{ m/s} \) gave the best match. It should be noted that some of the difference in the frequency spectrum of the experimental results may have been due to the non-ideal pressure release condition at the base of the prototype.
reactor.

Fig. 5.4 shows the curves obtained analytically and experimentally for the case $z = 0.09 \text{ m}, r = 0$ (cylinder axis). Only one other set of data (at $z = 0.04 \text{ m}, r = 0$) was obtained before the transducer suffered electrical breakdown.

![Graph showing predicted and experimental frequency responses](image)

**Figure 5.4. Experimental and Predicted Frequency Response for Reactor**

### 5.3.3 Discussion of Experimental Results

Apart from the fact that the predicted response was an order of magnitude larger than the response observed experimentally, the correlation between the two curves is quite good. The order of magnitude loss of sound pressure may have been due to a higher transducer stiffness than predicted, un-modeled behaviours of the system, or simply instrumentation errors. Data taken for $z = 0.04 \text{ m}, r = 0$ also agrees with the model, but the peaks are more damped and not quite as distinct.
A physical understanding of the resonant behaviour shown in Fig. 5.4 is found by first calculating the frequency $f_{m,b}$ of the coupled modes of the liquid column using the formula

$$f_{m,b} = \frac{c}{2\pi} \sqrt{M^2_b + \psi^2_m}$$

where $c$ is the wave speed (1350 m/s), $M_b$ are the axial wave numbers, and $\psi_m$ are the roots of the $J_0$ Bessel function. The frequencies for the first few modes are given in Table 7 and the shapes of the pressure modes are drawn in Fig. 5.5.

<table>
<thead>
<tr>
<th>Frequency (kHz)</th>
<th>b = 1</th>
<th>b = 2</th>
<th>b = 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m = 1$</td>
<td>8.7</td>
<td>19.0</td>
<td>29.6</td>
</tr>
<tr>
<td>$m = 2$</td>
<td>19.0</td>
<td>30.4</td>
<td>31.9</td>
</tr>
<tr>
<td>$m = 3$</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 7. Resonant Frequencies of Liquid Column

Referring back to Fig. 5.4, one sees that the first major resonance occurs at roughly 11 kHz. This is very close to the $b = 2, m = 1$ mode. The next major resonance is much broader and covers a range of 19 kHz - 21 kHz. This corresponds to the excitation of both the $b = 1, m = 2$; and $b = 2, m = 2$ modes. The small hump in the neighbourhood of 15 kHz is probably excitation of the $b = 3, m = 1$ mode. It is
important to remember that these are coupled modes. If the cylinder is very long relative to its diameter, then the factor $M_b$ becomes small relative to $\psi_m$ and the resonant frequencies correspond to purely radial modes (i.e. $m = 1, 2, 3, \ldots$). Conversely, if the cylinder is very short relative to its diameter, only axial modes (i.e. $b = 1, 2, 3, \ldots$) will be excited.

5.4 Optimization of Reactor Parameters

Since the reactor model appears to represent the behaviour of the real system quite well, it will now be used to explore what parameter changes can be made to increase the pressure (electrostatic to sound) transfer function of the reactor.

Decreasing the transducer damping $c_t$ will reduce power losses and thus magnify the quality of the resonance. With all other parameters held constant, an order of magnitude drop in transducer damping will increase the peak magnitude of the pressure transfer function by roughly the same ratio. A secondary benefit of less transducer damping is that less heat will be generated in the transducer where it may cause thermal fatigue of the dielectric material.

Fig. 5.6 shows a plot of the pressure transfer function for a transducer damping $c_t = 2.5 \times 10^4$ kg/m$^2$s which is an order of magnitude less than the value $c_t = 2.5 \times 10^5$ kg/m$^2$s used in Fig. 13. Reducing the stiffness $k_t$ of the transducer does not have much effect on the transfer function up to the practical limit of about $k_t = 1 \times 10^7$ kg/m$^2$s$^2$, which is about three orders of magnitude below the value used in the Fig. 5.4 calculation.

Changing the properties (elastic modulus, density) of the cylindrical shell
material does not affect the system significantly within the limits of suitable materials. For instance, if the shell material is changed from aluminum (\(E = 72\) GPa, \(\rho = 2800\) kg/m\(^3\)) to steel (\(E = 200\) GPa, \(\rho = 7860\) kg/m\(^3\)) in the model, there is very little effect on the transfer function. Changes in the thickness of the shell also did not have a substantial impact on the transfer function for values of practical interest (i.e. greater than 2 mm). This is not surprising since both the stiffness and mass of the cylinder scale linearly with its thickness.

![Predicted Pressure Transfer Function](image)

**Figure 5.6. Pressure Transfer Function for \(c_t = 2.5 \times 10^4\) kg/m\(^3\)/s.**

In summary, reducing transducer damping appears to be the best means of increasing the performance of this reactor.

For the EFT reactor design shown in Fig. 3.2a, where the transducer is not fixed to the wall of the reactor, selecting different radial positions for the transducer may provide another means of increasing reactor performance.
Chapter 6

Technology Commercialization

Issues pertaining to the commercialization of technology developed in this thesis are discussed in this chapter.

6.1 Intellectual Property Management

The intellectual property described in this thesis has been disclosed in U.S. Patent Application "Acoustic Liquid Processing Device" filed October 4, 1993. The main objective of the invention is the optimal use of electrostatic film transducers in a sonochemical reactor. It also discloses the preferred embodiments for the invention, and how these differ from the prior art.

All other intellectual property, such as improved transducer dielectric designs, are held as trade secrets. In order to remain trade secrets, due diligence must be exercised in ensuring that this information is not openly disclosed, or allowed to be openly disclosed to anyone. This precludes all obvious forms of display such as public seminars, conferences, etc. It also precludes the discussion with colleagues who are not bound by some form of non-disclosure agreement. The advantage of using trade secret protection is that one does not have to incur the expense of filing a patent application right away. Also, the technology can be held as a trade secret until its value has been proven by experimentation, and so the filing of patent applications on worthless ideas can be avoided.
6.2 Technology Development Plan

The development of an industrial-scale sonochemical reactor based on the technology described in this thesis is expected to go through the following phases:

6.2.1 High-Pressure Electrostatic Transducer Development

A sound pressure of nearly 0.1 atm was obtained from transducer prototypes constructed using minimal resources. With a properly engineered transducer dielectric, it is expected that a sound pressure in excess of the 1 atm required for cavitation can be produced. However, the prototyping of these designs requires specialized equipment and expertise which is not currently available to our research group.

It is thus necessary to find a strategic partner willing to assist in the prototyping and future development of the electrostatic film transducer. In return for this assistance, the strategic partner would be given a right of first refusal on an exclusive license for commercial manufacture of the transducer. Since there likely are numerous other applications for this new transducer technology, this license could have substantial value beyond sonochemistry, and thus provide the necessary motivation for a major commitment by this strategic partner.

6.2.2 Manufacture of Laboratory-Scale Reactors

Once a high-performance electrostatic film transducer has been developed by the strategic partner, the next objective will be to construct a number of bench-top reactors for use in sonochemistry research laboratories. The lab-scale reactors will be sold to research groups around the world. The performance of these reactors can
then be independently assessed by the chemists and chemical engineers working on the development of sonochemistry processes.

These researchers will be lured to the use of this reactor over its competitors for two key reasons: 1) The potential of this reactor technology to be scaled up to industrial use is readily apparent. This allows good results obtained in the lab to be easily analysed for their industrial feasibility, and thus makes sonochemistry research much more attractive to private labs. 2) This reactor design offers the capability to operate in a frequency tunable mode over nearly the entire acoustic spectrum of interest for sonochemistry (i.e. 10kHz - 100 kHz). This allows sonochemistry researchers to do something long desired, but never possible with other transducer technologies: accurately determine the relationship between chemical activation and frequency of sound energy. [10]

The manufacture and sale of these reactors will generate two vital things for the technology: (1) Awareness: People working in all areas of sonochemistry development will become aware of this reactor technology through mention of the lab-scale versions used in research work presented at conferences and in journal papers. (2) Credibility: The independent evaluation of the reactor performance by third parties will validate its capabilities.

In effect, the lab-scale reactors will generate a market demand for their industrial-scale counterpart.

6.2.3 Construction of Pilot Plant Reactors

Eventually a critical mass of interest and reactor performance will be reached
and there will be requests by industrial users to construct pilot plant reactors for testing the feasibility of specific sonochemical processes. The design of the reactor will need to be optimized for the given process characteristics (i.e. temperature, pressure, volume, etc.).

Numerous pilot plant scale projects may be undertaken at once as there is likely to be substantial overlap in the design expertise required to develop them. If problems relating to scale-up of both the reactors and processes can be successfully addressed, several industrial-scale sonochemistry processes will be created.

**6.2.4 Construction of Industrial-Scale Reactors**

The final step in the development cycle for this technology will be the design and construction of industrial-scale sonochemical reactors. This step is where the profits of the technology will be realized.

**6.3 Technology Transfer Vehicles**

A strategic development plan for the sonochemical reactor has been presented. The next step in management of this technology is the choice of a technology transfer vehicle or sequence of vehicles for implementing this development.

It has already been mentioned that the development of the electrostatic film transducer would be best handled by a strategic partner. The reason being that substantial specialized equipment and know-how would be required which likely could not be feasibly procured solely for the development of this transducer. A strategic partner already possessing these skills and equipment could carry out this
development for a much lower cost. Also, they could pursue sales of the transducer for other applications; an activity that might otherwise distract the group responsible for reactor development.

For the overall reactor development, the following three vehicles are possible:

6.3.1 New Venture

A new venture is created to continue investigation of the mechanical design of the reactor while overseeing the development of the electrostatic transducer by the strategic partner. Upon completion of a working transducer prototype, the venture would acquire manufacturing capability to produce the lab-scale reactors for sale to sonochemistry researchers. Manufacturing and design capabilities would be expanded to accommodate pilot plant and ultimately industrial-scale reactor construction.

6.3.2 Licensing

A license to the intellectual property of the reactor technology (i.e. patents, trade secrets) is sold to an interested company. The licence might also include a research contract to the inventors of the technology to continue work on the reactor design. All additional intellectual property generated by the inventors would then become the property of the licensee. In this way the licensee would not lose control of the technology due to a new development which made the original intellectual property obsolete.

6.3.3 Strategic Partnership

A new venture is created as described above. However, after the pilot plant
stage is completed and orders have been received for industrial scale reactors, another strategic partner is sought to take charge of manufacturing and installing the industrial scale reactors. The new venture would continue basic research on the reactor technology and carry out the design of critical components.

6.3.4 Relative Advantages of Each Vehicle

The basic characteristics of the three development vehicles from the perspective of the technology manager are compared in Table 6.1.

<table>
<thead>
<tr>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>New Venture</td>
<td>8-10 years</td>
<td>high</td>
<td>high</td>
<td>integral</td>
</tr>
<tr>
<td>Licensing</td>
<td>6-8 years</td>
<td>low</td>
<td>low</td>
<td>little</td>
</tr>
<tr>
<td>Strategic Partner</td>
<td>7-9 years</td>
<td>less than New Venture</td>
<td>less than New Venture</td>
<td>integral</td>
</tr>
</tbody>
</table>

Table 6.1. Comparison of Development Vehicles

The new venture vehicle carries higher risk than licensing because usually a substantial investment is required of the owners of the technology toward the financing of the new venture. This investment can take forms other than money. For example, if the technology owner is also the lead entrepreneur, this investment may be in the form of a reduced salary and other personal guarantees. The use of a strategic partner in conjunction with a new venture is likely to reduce the financial risk to the new venture as its capital requirements become smaller. However, a portion of the venture's return now must be given to the strategic partner, and this will reduce the potential financial reward.
Another important comparison is the level of involvement in the development of the technology by the technology manager. Where active involvement is desirable (i.e. the technology manager has specific expertise in the area) then the new venture is probably the best way to organize this. However, if for some reason (e.g. other commitments) this is not possible, and no suitable replacement entrepreneur(s) can be found to lead the venture, then there may be no choice but to use licensing.

The preference for development vehicle will undoubtably change over time no matter which vehicle is chosen at the outset. For instance, if a new venture is created and at some point during its life a company makes an attractive offer to license the reactor technology for a specific industrial application area (i.e. pulp and paper) then this might be done. Likewise, one or more strategic partnerships may be formed based on market geography or application area.
Chemists and process engineers have identified the need for a sonochemical reactor design which is feasible for use at the industrial scale. Numerous attempts have been made to develop an industrial-scale reactor using conventional piezoelectric and magnetostrictive transducers, but have been unsuccessful so far. To circumvent the problems faced by previous design attempts, a sonochemical reactor design based on entirely new transducer technology called electrostatic film has been developed.

The operating principles and pressure limitations of electrostatic film transducers are not well documented elsewhere in the literature. It was highlighted that very high electric fields are required to generate the pressures needed for cavitation. At these high electric fields numerous mechanisms of dielectric failure can be present such as gas ionization, electron field emission, corona, solid breakdown, surface breakdown, etc. Some dielectric designs proposed by others for dealing with these problems were presented.

The design of sonochemical reactors based electrostatic film transducers was discussed in detail. A cylindrical reactor geometry was selected because it focusses the sound energy into an intense zone where cavitation can be efficiently generated and also allows for uniform flow-thru processing. Two main embodiments were presented: 1) the transducer is attached directly to the interior surface of the reactor
cylinder; and, 2) the transducer is placed on an acoustically transparent inner cylinder so that it may drive the system from a more optimal location on the standing waves in the liquid.

The cost advantages of this reactor over conventional designs are as follows: 1) capital cost should be lower because the reactors are simple to manufacture and utilize low-cost electrostatic film transducers; 2) energy consumption is reduced due to much higher coupling efficiency between the transducer and the process liquid; and 3) less maintenance is required because intense cavitation, which causes erosion damage to equipment, is confined to a volume of process liquid away from equipment surfaces.

An attempt was made to build a prototype of this reactor capable of producing cavitation. The first step in this prototyping program required the construction of a suitable electrostatic film transducer. Tests were done on transducers made using inexpensive, readily available materials such as tissue paper and mylar. Significant progress was made in this experimental program but in the end the maximum acoustic pressure obtained was still one order of magnitude less than that necessary for cavitation.

A physical model of the reactor's acoustic behaviour was developed to calculate the transfer function frequency response between the transducer electrostatic pressure and the acoustic pressure in the reactor liquid. Frequency response data from experiments conducted on a prototype reactor agreed fairly well with those predicted by the model. The data showed that only two or three modes are
dominant over the frequency range 10 kHz - 20 kHz. The model was used to explore
the effect of system parameter changes on the pressure transfer function. Reduced
transducer damping was found to be the primary means of increasing this transfer
function, and thereby the reactor performance.

Finally, various aspects of commercialization of this technology have been
examined. The first step required will be the development of an electrostatic film
transducer capable of producing sufficient acoustic pressure to cause intense
cavitation. The second step has essentially three options: 1) Licence the technology;
2) Start a company to carry out development, manufacture, and distribution; and, 3)
Start a company to carry out development, but enlist a strategic partner to handle
manufacturing and distribution. The appropriate option to choose will depend on
factors such as interest in licensing, access to venture capital, and the availability of
suitable strategic partners.

Recommended future work is the development of an acoustic model for the
reactor which includes some of the key non-linear behaviours such as cavitation. This
model is necessary to fully understand the performance of the reactor and thus
determine its optimal parameters. Such a model would yeild a more detailed
specification of the transducer requirements in terms of stiffness and mass
impedances, damping, electric field strength, etc. Finally, the overall reactor
efficiency could be estimated and used to forecast the economic feasibility of a
commercial scale product.
References


[21] I. N. Slivkov et. al.; "Electrical Breakdown and Discharge in a Vacuum." Foreign Technology Division, WP-AFB, Ohio; 1972

[23] Dupont Mylar Technical Information; Dupont Canada Inc.; 1993


