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RADIATION INDUCED ACOUSTIC CAVITATION IN A SCINTILLATION LIQUID

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13. ABSTRACT

This study attempted to establish a time correlation between the incidence of radiation in a liquid upon which a sound field was impressed, and the subsequent cavitation event.

A scintillating liquid enclosed in a cavitation cell was considered ideal for such a study. An ionizing particle produced a scintillation event, and if of sufficient energy triggered a cavitation event. Both events are observable, and a time correlation could be established between them. Such a time correlation has not been established. Several problems of major import have been encountered. However, none of them are necessarily insurmountable. Suggestions are made for improvements which would allow this study to be carried to a fruitful conclusion.

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RADIATION INDUCED ACOUSTIC CAVITATION IN A SCINTILLATION LIQUID

A Trident Scholar Project Report

by

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Abstract

A study of radiation induced acoustic cavitation has been conducted with five resonators, two of which have been found particularly suitable for such a study. Various irradiating particles have been considered, specifically neutrons, alpha particles, and fission fragments. Finally, various liquids have been considered in this study, methanol, decalin, and toluene. Of these, only toluene has been found well suited to the task.

The purpose of the study was to attempt to establish a time correlation between the incidence of radiation in a liquid upon which a sound field was impressed, and the subsequent cavitation event. Current theories are not in agreement with respect to the time correlation to be expected. Some observers feel there is no appreciable delay between incidence of radiation and the cavitation event. Others maintain that there is an induction time of twenty minutes to an hour between events.

A scintillating liquid enclosed in a cavitation cell was considered ideal for such a study. An ionizing particle produced a scintillation event, and if of sufficient energy triggered a cavitation event. Both

events are observable, and a time correlation could be established between them.

Such a time correlation has not been established. Several problems of major import have been encountered. However, none of them are necessarily insurmountable. Suggestions are made for improvements which would allow this study to be carried to a fruitful conclusion. Such a conclusion would be invaluable in the solution of the exact nature of the radiation-induced cavitation event.

Preface

Cavitation is and has been a problem to the Navy for many years. For several reasons it is important that research in this field be conducted. Cavitation produces three effects of importance to the Navy. Cavitation damage to propellors, sonar domes, and the general underwater hull area of a ship necessitates regular repair and replacement of propellors and external mountings to the hull. In addition, cavitation damage to a propellor will increase the radiated noise level of a ship. Most of the radiated noise produced by a ship is caused by either cavitation or machinery. Indeed in the higher frequencies and at speeds above about five knots, cavitation noise is the dominant source of acoustic output[1]. The range of detectability depends on the level of this output. Finally, sonar output is cavitation limited. At negative pressures above the cavitation threshold, cavitation will occur, effectively interrupting sound transmission from and to a sonar device. In addition, acoustic cavitation on the dome will result in damage to its surface.

For all these reasons cavitation study needs to be carried on in the hope of finding the ultimate

solution to the problem of cavitation. Unfortunately hydrodynamic cavitation is not easily studied in the laboratory. Therefore, researchers have conducted their studies in the field of acoustic cavitation in the hopes that results obtained in such studies are applicable to the general problem of cavitation. This study is a basic one in the field of acoustic cavitation.

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I. INTRODUCTION

A. Background

Cavitation events fall into two basic categories or classifications, depending on the origin of the conditions which produce them. These are acoustic and hydrodynamic cavitation. Hydrodynamic cavitation is unimportant with respect to this study. It is caused by the flow of a liquid over a surface, producing regions of low or negative pressure on the reverse side of the surface. In these regions, a hydrodynamic cavitation event occurs when a bubble expands and collapses due to the pressure effects.

Acoustic cavitation may be defined as the growth and subsequent collapse of a bubble in a sound field in a liquid. There are two types of acoustic cavitation, gaseous and vaporous. Gaseous cavities are those in which the relative amounts of gas and vapor remain fixed through an acoustic cycle. Vaporous cavities are those in which the relative amounts of gas and vapor are not constant through an acoustic cycle due to evaporation and condensation of the host liquid at the cavity interface.[2] These cavities may be transient or stable, the transient cavities being the only ones of concern in this study. A gaseous cavity will produce the gaseous cavitation with its associated streamers of

bubbles and its characteristic hissing or "frying" sound. On the other hand vaporous cavities produce vaporous cavitation, in which a bubble suddenly collapses producing a sharp, audible click. This type of cavitation is also termed "hard" cavitation. This type of collapse was first described by Rayleigh who postulated a cavity which commenced its collapse as soon as it was formed in a liquid.[3] It is more likely that a cavity grows through several acoustic cycles until it reaches a critical radius and collapses violently.[4]

This critical radius is described by the equation:

$$R_C = \frac{4\sigma}{3|P_L - P_V|}$$

where σ is the surface tension of the bubble, P_L is the absolute pressure in the liquid, and P_V is the vapor pressure. Also,

$$P_L = P_A + P_E$$

where P_A is the acoustic pressure and P_E is the equilibrium or ambient pressure. If P_A is in the negative portion of the cycle, P_L may be negative, a condition necessary for acoustic cavitation to occur. When the critical radius is reached, the cavity becomes transient and collapses violently

producing the characteristic click and a flash of light known as sonoluminescence.

Ordinarily the nucleating agent for acoustic cavitation is a cavity of vapor and gas trapped in cracks or crevices of very small solid particles suspended in the liquid. This idea was originally set forth by Harvey.[5] Such a cavity if exposed to an acoustic field begins to oscillate and eventually becomes a transient cavity and collapses. The question arises as to what would occur if such particles, commonly known as motes, were removed from the liquid. If sufficient numbers of motes are removed, and the size of those remaining is sufficiently small, the threshold of acoustic cavitation will be increased. In other words, the liquid will require a larger acoustic pressure impressed upon it in order to produce cavitation. If all the motes could be removed from a liquid, acoustic pressure on the order of the tensile strength of the liquid would be required to induce cavitation. In this the liquid would literally be ripped apart by the acoustic field. For instance, CCl_4 has a tensile strength of 260 bars, but cavitation has been produced in CCl_4 at 1.75 bars, due to the presence of inhomogeneities in the liquid, i.e. motes.

In a liquid which has been filtered to remove these motes, a new cavitation threshold may be defined, that of radiation induced cavitation, first observed by Lieberman in acetone and pentane. He postulated that the motes were replaced by the nuclei of some of the heavier elements, carbon and oxygen, which acquired their energy through collision with incident neutrons.[6] The immediate interest in this study was a result of the work of Greenspan and Tschiegg, and that of Sette and Wanderlingh, all of whom worked in the field of radiation induced cavitation. The results produced by these two groups of researchers are at best not in complete agreement. For instance, Sette found that the radiation-induced threshold was not greatly affected by the motes present[7], whereas Greenspan suggested filtering all motes which would produce cavitation below a threshold of twice the radiation threshold of the liquid[8]. Furthermore, Sette and Wanderlingh found a so-called "cumulative effect," which predicted that long-lived microcavities were produced by the radiation which could produce cavitation at any later time. He also found that an induction time of several minutes or longer was required to lower the cavitation threshold after the incidence of radiation[9]. Greenspan and

Tschiegg on the other hand found no evidence of such an induction time.[10] Finch's study found the same effect that Sette found, with a shorter induction time of approximately half an hour at most.[11] Barger's work also supported Sette's hypothesis; he reported a time lag between the placement of a paraffin shield and the reduction of the cavitation rate.[12]

B. Purpose

The purpose of this study was two-fold. First, it was necessary to produce radiation-induced acoustic cavitation. Secondly, a time correlation was to be established between the incidence of radiation on a liquid in a sound field, and the subsequent cavitation event. To do this, a system had to be designed which would detect the radiation as well as the cavitation. A scintillating liquid was proposed and used as the medium for the experiment. Not only will radiation induce cavitation, but it will also produce scintillation in this liquid. The correlation was to be established between the scintillation event and the cavitation event. This correlation was expected to answer the question of whether or not radiation nuclei have a measurable half-life or not.

C. Scope

This project was not intended to be an elaborate investigation of this time correlation. Rather, it was intended that a workable apparatus be designed and constructed within the limits of the resources available, and that an estimate as to the correctness of the empirical results obtained by Greenspan's group and by Sette's group be established.

II. APPARATUS

A. Resonators

The first problem of major importance was the design and construction of an efficient resonator. Two types of resonators were possible, a standing wave system and a traveling wave system. A traveling wave system was described by Sette, but the intricacies of the focusing mechanism suggested that the standing wave resonators were more realistic. Because of the simplicity, of such a system, it was adopted. It has another major advantage in that such a system does not require large amounts of power to produce high negative pressures. Power needs are dictated by the losses of the system.

The cylindrical system used by Greenspan was considered ideal. It was small, portable, and easily

mounted to a phototube, which was to be used to observe the scintillation. Such a cylindrical system develops radially symmetric modes, which produce pressure maxima and minima along the axis of the cylinder. This radial symmetry is defined by the Bessel function, $J_0(r)$. This system also develops modes with an azimuthal dependence. These modes are unsatisfactory due to the fact that they develop maximum pressure on the walls. Satisfactory and unsatisfactory modes may be calculated, but they are easier to discover through trial and error.

The first cell constructed was built about a PZT-4 barium titinate transducer, O.D. 3 inches, I.D. 2.62 inches, and length 3 inches. The outer and inner surfaces of the transducer were silvered by the manufacturer; the ends were bare. A brass ring 0.5 inches in thickness was cemented to the upper end with a commercial epoxy, Marinetex, and a ring 0.38 inches in thickness was cemented to the lower end in a similar manner. These rings served several purposes. They were used to establish electrical contact, the upper ring being electrically continuous with the inner ground surface, and the lower ring being continuous with the outer drive surface. A groove was cut in the upper ring to provide for the mounting of an O-ring. Threads were cut on the outer

surface of the upper ring in order to allow a lid to be screwed down to form a seal with the O-ring. The lid was another brass ring, 0.25 inches in thickness, and threaded on the inner surface. Electrical contact between the lower ring and the outer cylinder surface was established with a conducting silver epoxy. Similarly, contact was made with the inner cylinder surface with a strip of silver epoxy between the upper ring and the silvered surface. To prevent a short circuit, it was necessary to remove a strip of silver from the outer surface near the upper ring. Holes were drilled in the upper and lower rings to serve as filling tubes. Brass nozzles were soldered over these tubes for ease of attachment of piping to be used in the filtering system. An aluminum plate, 0.08 inches thick was selected for use as the lower boundary due to its very small capture cross section for thermal neutrons, 0.24 barns. The plate was attached to the base of the transducer with RTV, a silicone base adhesive. The aluminum proved to be unsatisfactory due to the fact that it was readily pitted in an electrocatalytic reaction with water. The second choice for a base plate was stainless steel plate, also 0.08 inches thick. The slow neutron capture cross section for iron

(2.62 barns) is an order of magnitude larger than that of aluminum, but a compromise was necessary. Finally, a circular piece of glass, 0.08 inches thick was cemented to the lid, the 0-ring seal being made with this glass. The purpose of the removable lid was to allow the sound field to be sampled with a piezoelectric probe.

Several serious faults were incorporated into this design. The use of the silver epoxy was a mistake due to the fact that it was a rough surface, and cavitation occurred upon it, causing it to suffer damage and contaminate the liquid. The resonator was operated in a (1, 0, 1) mode, the breathing mode, at 30.1 KHz, and in the (3, 0, 1) mode at 51.9KHz. Initially the cell seemed efficient, producing cavitation in degassed, distilled water at an input voltage of 15 volts in the (3, 0, 1) mode. The threshold for degassed, distilled water is approximately 12 bars according to the data of Barger.[13] However, the cell's acoustic properties apparently deteriorated because an input voltage of 240 volts was eventually required to produce the same type of cavitation. A measurement of the mechanical Q of the transducer at 60.6KHz showed a half power band width of 560Hz. The Q of a system is defined as

$$Q = \frac{F_C}{BW}$$

where F_C is the center frequency of the resonance; BW is the half power bandwidth. Thus the Q of the mechanical system was 108. The mechanical Q of a PZT-4 transducer should be in the vicinity of 600.[14] Thus it was decided that the fittings on the transducer were lossy; that is, high power losses were present because of the brass rings, the 0-ring, and the steel and glass plates. The brass rings in particular clamped the motion of the transducer, and the 0-ring dissipated energy that otherwise would have driven the liquid enclosed. This design was subsequently abandoned.

Another transducer of the same type used previously was incorporated in the new design, which was intended to correct the deficiencies of the initial design. Specifically, the clamping of the rings and the 0-ring were to be eliminated. Also, it was desired that a close approximation to a pressure release boundary be made at the ends of the cylinder. The velocity potential, upon which the pressure is dependent, for such a system is given by

$$\phi = BJ_0(K_r r)\sin(n\pi z/L)$$

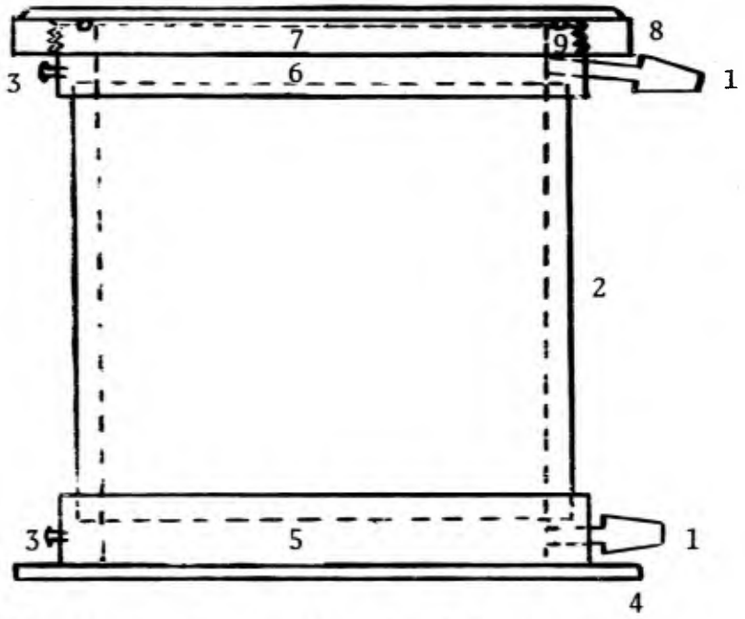


Figure 1, Resonator 1, Schematic

1. Fill Tubes
2. Piezoelectric cylinder
3. Electrical contacts
4. Base plate
5. Brass ring
6. Brass ring
7. Brass lid
8. Glass plate
9. 0-ring

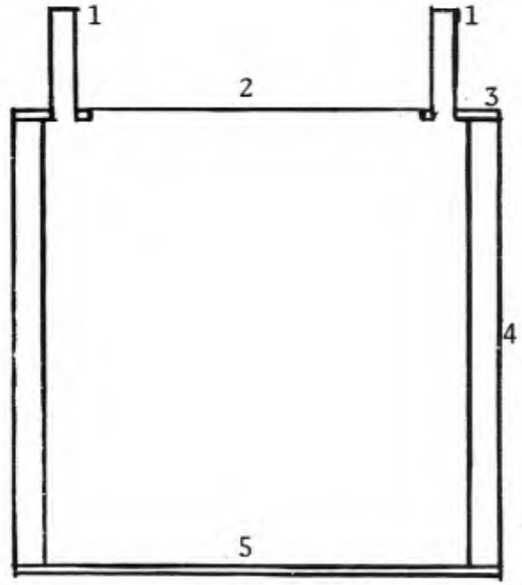


Figure 2, Resonator 2, Schematic Cross Section

Note the simplicity of this design compared to that of resonator 1.

1. Fill tubes
2. Stainless steel foil
3. Brass ring
4. Piezoelectric cylinder
5. Glass plate

where K_r is the radial wave number, n is an integer, and L is the length of the cylinder. It can be seen that the pressure will be zero when Z = 0 or L; that is, the pressure will be zero at the ends of the cylinder. This arrangement has numerous advantages. Primarily, it prevents cavitation on the boundary surfaces. Secondly, it reduces the disturbance of the sound field by the fill tubes.

Two circular strips of silver were removed from the outer surface to prevent a short circuit. These strips also served as the mounting points of the transducer in its cradle. A brass ring was again used at the upper end as a mounting platform for the fill tubes. The ring however, was only 0.05 inches in thickness rather than 0.5 inches as for the previous ring. Two holes were drilled in the ring and short lengths of copper tubing were soldered over them to serve as fill tubes. Since electrical contact was to be made with the exterior surface of the cylinder rather than with the brass rings, it was necessary to establish an electrical path across the bare ends of the transducer. To accomplish this, aluminum was sputtered on to these surfaces using a vacuum system. A large bell jar was evacuated to a pressure of approximately eight microns. Small strips of pure aluminum were heated on a tungsten filament

until they vaporized. Reaching the exposed surface of the cell the vapor condensed producing a thin layer of aluminum suitable for carrying current to the inner surface. The brass ring was then epoxied to the upper end of the cylinder, the epoxy protecting the aluminum from the enclosed liquid. Stainless steel foil, 12 to 75 μ thick was mounted to the brass ring with RTV. This provided the desired pressure release boundry and allowed for the transmission of neutrons. Glass was again used to provide the lower boundry of the cell; however, it was mounted directly to the cell with RTV and it was only .01 inches thick. Again the purpose of this was to simulate as closely as possible a pressure release boundary.

As expected, this cell proved to be a very efficient one, initially cavitating water at an input voltage of 9 volts. The mechanical Q of the transducer was measured at various frequencies as shown in the table on the following page.

Center Frequency (KHz)	Upper Half Power Point (KHz)	Lower Half Power Point (KHz)	Bandwidth (Hz)	Q
45.145	45.237	45.084	153	295
67.874	68.038	67.791	247	274
88.982	89.107	88.868	239	375
109.603	109.784	109.468	316	347
25.604	25.631	25.585	440	581

Table 1: Mechanical Q Factor for Second Resonator

This compares very favorably with the mechanical Q measured on the first cell. A subsequent measurement of the Q of the first cell at 25.1 KHz showed it to be 92. The system Q of the new cell was measured in the desired (3, 0, 1) mode at 50.380 KHz and found to be approximately 25,000; the half-power bandwidth was 2 Hz. The acoustic probe also showed that the pressure at the central maximum was twice that on the walls.

The aluminum sputtered on the surface to make electrical contact eventually deteriorated even though it was shielded by epoxy. Thin wires were soldered to the outer and inner surfaces to reestablish electrical contact. These wires, combined with a small crack which developed, reduced the system Q to 3100, which, though considerably lower than the original Q, was high enough to produce the desired cavitation.

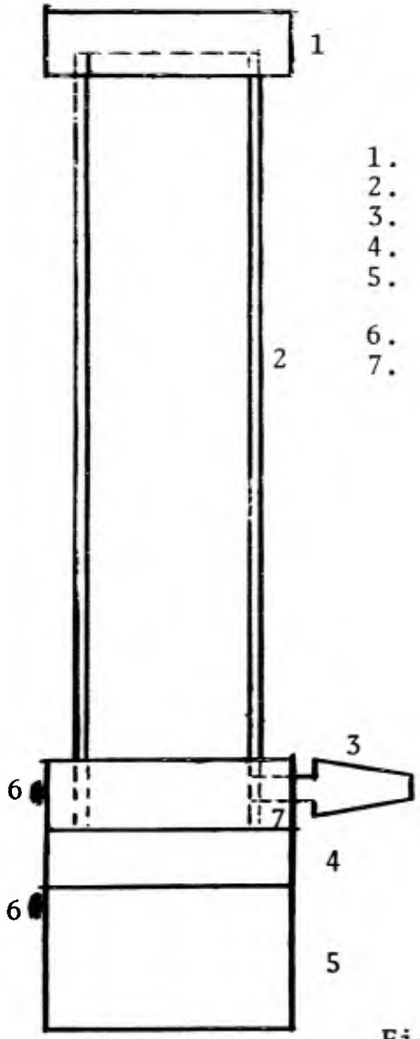
The third design used was an externally driven cell. This was more or less an auxiliary cell, used for probe calibration, and to attempt to produce neutron-induced cavitation in water. Greenspan mentioned the externally driven cell as ideal for use in the cavitation in water.[15] Water requires a very smooth wall or else cavitation will occur on the walls. Therefore the cell itself was a glass cylinder 6 inches length, with a diameter of 1.18 inches. The drive was provided by a disc transducer, silvered on both sides, 0.5 inches thick, and 1.5 inches in diameter. A brass disc 1 inch thick and 1.5 inches in diameter was epoxied to the base of the transducer with silver conducting epoxy. It served as a resonant frequency shifter and as an electrical contact to the transducer. A brass ring was fitted around the lower end of the glass cylinder, and the assembly was epoxied to the upper surface of the transducer with silver epoxy, electrical contact again being made through the brass ring. A drain tube was soldered over a hole drilled in the brass ring. This proved to serve a rather novel purpose which will be discussed in the appendix. Finally a brass ring was epoxied to the upper end of the glass tube to prevent it from cracking.

This cell exhibited a strange property in that its

mechanical Q was larger than the system Q. The mechanical Q at 5[illegible].780 KHz was 775 whereas the system Q at 52.909 KHz (the resonant frequency of the system) was only 422. This was possibly due to some sort of damping of the glass cylinder by the liquid. However, the system was suitable for the probe calibration.

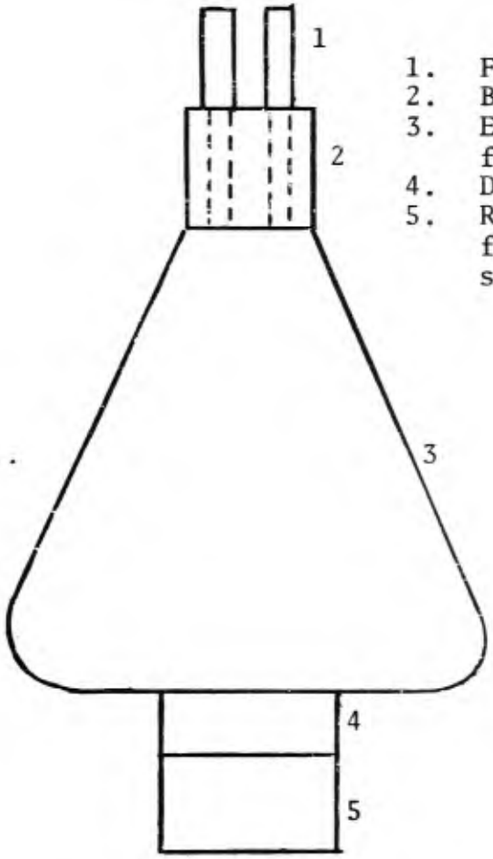
The fourth cell utilized was another PZT-4 transducer, O.D. 2 inches, I.D. 1.62 inches and length 2 inches. It was constructed in a manner identical to that of the second cell except that the ring used was a titanium alloy rather than brass.

This cell did not perform at all like the cell after which it was modeled. It was intended as a standby during the frequent down periods that were encountered due to the failure of the RTV and epoxy bonds. The system Q was measured at 45.308 KHz, and was found to be 487. Hard cavitation was never produced in this cell, regardless of the input voltage. Two things were undoubtedly responsible for the power loss which was responsible for this low Q. First, the stainless foil vibrated considerably, indicating power was being used to drive it. Secondly, the fill tubes were of the same size as those in the 3 inch cell, but they therefore occupied almost three times as much upper boundaryarea as in the large cell.



1. Brass ring
2. Glass cylinder
3. Drain tube
4. Disc transducer
5. Resonant frequency shifter
6. Electrical contacts
7. Brass sleeve

Figure 3, Resonator 3, Schematic



1. Fill tubes
2. Brass plug
3. Erlenmeyer flask
4. Disc transducer
5. Resonant frequency shifter

Figure 4, Resonator 5, Schematic

These tubes both interfere with the sound field, and they are lossy;[16] that is, they produce power losses. The tubes on the small resonator, occupying a much larger surface area, resulted in further power losses, rendering the cell unsuitable for use.

The fifth resonator used was an externally driven Erlenmeyer flask, also suggested by Greenspan's work.[17] The flask was a standard 250 ml Pyrex Erlenmeyer flask. To its base was epoxied a disc transducer of the type used in the externally driven cylinder. Electrical contact was made directly to the silvered surfaces of the transducer. A brass disc was cemented to the base of the transducer, acting as a resonant frequency shifter. A brass plug was epoxied inside the neck of the flask. Fill tubes were two short lengths of copper tubing epoxied over two holes drilled in the plug.

This cell could not be effectively coupled to the phototube; however, it produced many interesting and significant results. The exact pressure distribution inside the Erlenmeyer is difficult to predict due to its odd geometry. However, pressure maxima and minima occur along the conical axis, and cavitation was readily obtained. The biggest problem with the Erlenmeyer is the vibration of the glass which masks acoustic output at high levels of drive.

B. Filtering and Degassing

Filtering of the liquid was required to remove the motes present though distilled water was not filtered. Filtering of the liquid finally used was accomplished with a microbellows pump and fiberglass filters. Greenspan had mentioned in his work that he filtered his water extensively to remove motes. However, he also stated that in organic liquids such as toluene an extensive filtering process was not necessary due to the fact that organic liquids 'wet' the motes contained in them quite effectively, or else the motes themselves are soluble in the liquids. In particular, he mentioned that the particular liquid used, toluene, should require only about 15 minutes filtering with the fiberglass filter to remove any motes which would cause cavitation at a threshold of 2T or lower.[18]

Greenspan maintained that gas content of the liquid was not important in radiation induced cavitation. However, when degassing was not performed, the liquid invariably started to cavitate at a very low threshold, but the cavitation was gassey rather than hard. This gassey cavitation detuned the circuit and prevented the occurrence of hard cavitation. The original degassing and filtering system used was a closed one, designed

to allow for continuous flow of the liquid and for degassing at regular intervals. The microbellows pump, a liquid reservoir, and the cavitation cell were connected in series. The reservoir was sealed and could be pumped down with a water aspirator. This degassing served another purpose in that it increased the scintillation efficiency of the system by about 20%. A valve was installed which allowed argon to fill the space over the liquid in the reservoir after pumping ceased. This was also intended to increase the scintillation efficiency by about 30%.[19] Two main effects rendered this system unusable. In the degassing process, air bubbles coming out of solution were trapped in the cell and in the tubing connecting the three elements. Unfortunately air bubbles resonating in the cell will disturb the sound field, preventing cavitation. The air in the lines also produced a secondary effect, air locks in the pump, which caused it to cease pumping the liquid. Essentially what was done after this was to divorce the degassing and filtering systems from the cell and from each other. The liquid sample was first filtered, then degassed, and finally poured into the cell. Care had to be taken to prevent contamination of the liquid in the process of pouring. A magnetic stirrer proved to be

an invaluable aid in both the filtering and degassing stages.

C. Electronics and Experimental Arrangement

A 0 - 2 MHz oscillator provided the input signal for the cell. The signal was amplified by a 75 watt audio amplifier. A frequency counter connected in parallel measured the frequency of operation. The output of the amplifier was connected in series with the cell. A voltmeter measured input voltage. A capacitive tuning circuit was connected in parallel with the cell. Output was to be provided by a 3 inch phototube with S - 11 response. The amplified signal of the phototube was displayed on an oscilloscope; it was also the input for a pulse height analyzer, which was to be used to obtain a spectrum of the tube response under radiation.

The tuning circuit used was one described by Greenspan.[20] It consisted of two variable capacitors

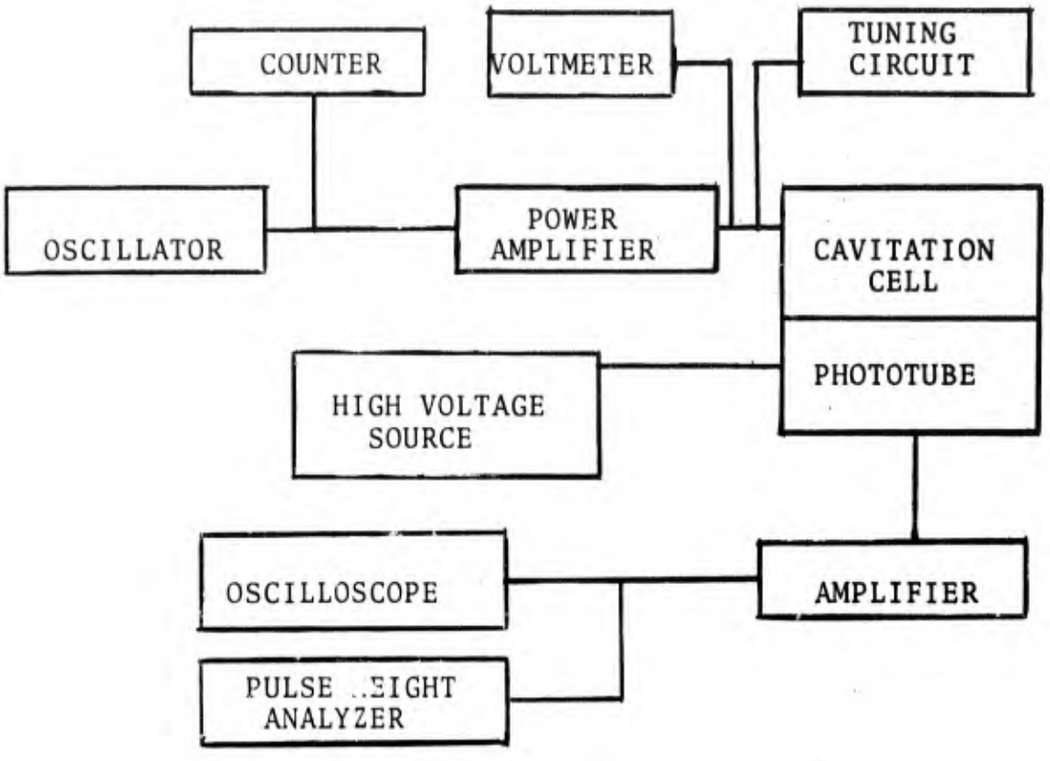


Figure 5, Schematic Block Diagram of the Experimental Appartus

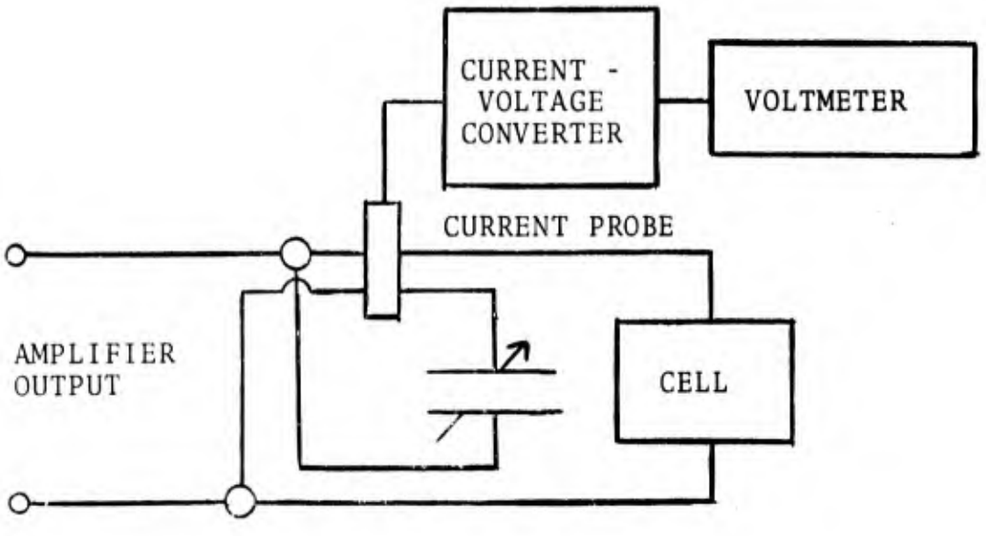


Figure 6, Schematic, Tuning Circuit

connected in parallel with the cell, a clip-on current probe, a current-to-voltage converter, and a voltmeter. The input from the amplifier was split, one branch going directly to the cell. The other branch was connected to tne ground terminal of the capacitor. The positive terminal of the capacitor was connected to ground. The probe was clipped onto the wire leading to the cell and to the wire from the positive terminal of the capacitor. Thus the probe measured the algebraic sum of the currents. At a frequency not a resonant frequency of the cell, the variable capacitors were tuned until the voltmeter registered the smallest possible output. After this operation the circuit was tuned for any frequency. At resonance, the current measured by the probe will increase sharply due to the large increase of motional current. System Q's were readily measured with this device. Prior to the use of this device, a variable inductor had been used for tuning; the variable inductor had an inherent fault in that it had to be retuned each

time the frequency was changed. This problem was eliminated with the tuning circuit.

An acoustic probe was used to determine the sound field existing in the resonator. This probe was a small, highly sensitive piezoelectric transducer attached to the tip of a rod which could be lowered into the liquid. The probe was coated with epoxy in order to protect it from the corrosive action of liquids and from cavitation damage. The major problem with this probe was the disturbance of the sound field it produced. Each time the probe entered the liquid, the sound field was immediately disturbed; so results obtained with the probe were approximate only.

Probe calibration was accomplished using a method described by Crum and Eller.[21] This method involved a measurement of the equilibrium position of a bubble of known size trapped in a sound field to determine pressure amplitude. The output voltage of the probe which is in the same sound field was recorded for the calibration. Bubble radius was determined by measuring the rise velocity of the bubble in the liquid. Using a digital computer the data was processed and calibration curves resulted.

Originally, it was intended that the oscilloscope

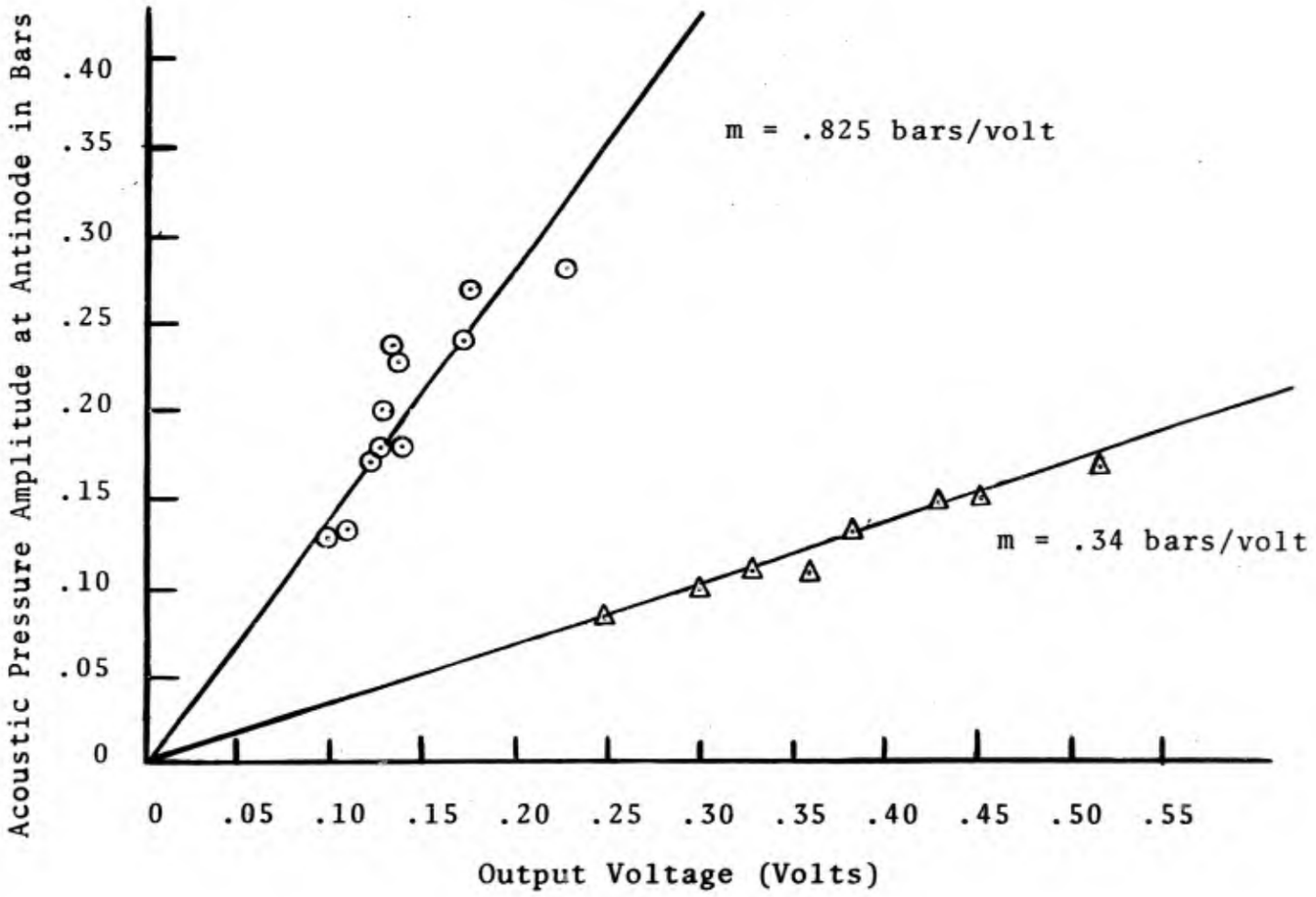


Figure 7, Probe Calibration Curve

Data taken at different times produced significantly different curves. Apparently the mechanical properties of the probe changed with time and use.

itself be used as the time correlator. Cavitation produces a phenomonen known as sonoluminescence; when a cavity collapses, the temperature of the cavity increases to about 5000°C and light is emitted. Hahn and Peacock[22] detected this sonoluminescence in CCl_3F with a photomultiplier tube. They reported light pulses with a decay time shorter than 10 nsec which occurred synchronous in time with the audible neutron induced cavitation click. West and Howlett,[23] using a pulsed neutron source and a phototube also observed the sonoluminescent flash. They found a rise time for the flash of less than 12 nsec. No mention is made of light intensity. However, they reported 1,376 counts in the time spectrum of the flash. The rise time for the scintillation flash is on the order of 5 nsec. Thus the phototube presentation expected was a single pulse, the scintillation event, followed by an avalanche of smaller pulses from the sonoluminescence. If this avalanche existed it was lost in the background of the phototube, because nothing of the above description was observed. Since the time interval between the incidence of radiation and the cavitation events could range from immediate response to a few minutes or an hour, it was decided that the video output of the scintillation, and the audio

output of the cavitation could be correlated by the observer. The expected output in this case was the scintillation flash appearing on the scope followed by the audible report of a cavitation event.

The phototube, the cavitation cell, and a paraffin filled box were enclosed in a light-tight box. A hole was drilled in the paraffin box to house the Pu-Be source. A solid plug of paraffin, thermalized the neutrons prior to their incidence on the steel foil. Two plugs were made to provide collimated beams of fast neutrons of different fluxes. The beams were directed toward the critical area of the cell, that is the axis of the cell where the largest negative pressures were sustained.

III. LIQUIDS

It was necessary to find a liquid suitable for cavitation, but which, at the same time, was chemically inert with respect to the materials used in the resonators, and finally which could act as a solvent for the standard scintillants available, PPO, P-Terphenyl, and the wavelength shifter, POPOP.

Decalin, a paint base, was originally considered due to its immediate availability. It had no adverse chemical effect on the materials used in the resonators.

Originally the experiment was to be conducted in the following manner. A thermal neutron was to be captured by a B^{10} nucleus which then decayed in the following reaction,

$$ {}_0n^1 + {}_5B^{10} \rightarrow {}_3Li^7 + \alpha, $$

the α-particle producing the scintillation and cavitation events. An attempt was made to find a boron compound soluble in decalin; no such compound was found, and decalin was discarded as the liquid medium for the experiment.

The next liquid considered was toluene, $C_6H_5CH_3$. Due to its low tensile strength, toluene was considered one of the "weak" liquids, such as the alcohols. Such liquids do not require large negative pressures to produce radiation induced cavitation; four or five bars is usually sufficient. It was also well suited for this experiment in that it "wetted" the motes contained in it to such an extent that extensive filtering was not required. Toluene is also a standard solvent used in the practice of liquid scintillation counting. It was also easily doped with various liquids, such as Trimethyl Borate, $B(OCH_3)_3$, to be used to produce the desired radiation. It was also

found that it readily acted as a solvent for a solution of methanol and uranyl nitrate or thorium nitrate, the uranyl nitrate producing fission events, and the thorium nitrate producing alpha particles. Solutions of uranyl nitrate and methanol containing up to 100 gm/liter uranyl nitrate could be dissolved in the toluene. Toluene proved to be the liquid which was used in most of the experimentation. The major problem with the toluene was the fact that it caused the RTV to swell during prolonged exposure, and it slowly attacked both the Marinetex and the silver epoxy bonds, causing them to fail eventually. The toluene also provided another reason for abandonment of the 0-ring design; it caused the 0-rings to swell.

An attempt was made to make use of the decalin by dissolving uranyl nitrate or thorium nitrate in acetone or methanol and then dissolving this solution in decalin. Methanol was found to be insoluble in decalin. Acetone is soluble in decalin, but when a solution of uranyl nitrate and acetone was introduced into the decalin, the uranyl nitrate was precipitated out of solution. Decalin was used only with fast neutron irradiation after it was found that uranyl nitrate would not remain in solution.

Methanol was also tested since uranyl nitrate

dissolved in it readily. However the scintillants did not dissolve in it; thus it could not be used.

IV. RADIATION

Radiation was of three types, alpha particles, neutrons, and fission fragments. Greenspan produced preliminary evidence that the cavitation threshold depended (inversely) upon the energies of the nucleating particles.[24] Thus, the thresholds for alpha irradiation were expected to be the highest since the alpha particle emitters used produced alpha particles of energies smaller than the fast neutron energies of the Pu-Be source. The thresholds for fission fragments were expected to be the smallest because the fission fragments carried by far the largest energies of the particles used.

The neutron source used was a 1 curie Pu-Be source with a flux of 1.6×10^6 neutrons/cm^2-sec. The energies of these neutrons ranged from 0 to 10.7 Mex. The neutrons were produced by the following (α,n) reaction:

$$_2He^4 + {}_4Be^9 \rightarrow {}_0n^1 + {}_6C^{12}.$$

The alpha-particles were provided by the $_{94}Pu^{239}$. In using polyenergetic neutrons, the assumption was made that the threshold observed was produced by the inter-

action of the most energetic neutrons, since they had the most energy to dissipate, and at a given impact parameter will produce recoil nuclei of the highest energy. A monoenergetic source would have been more desirable.

The fissionable material used, uranyl nitrate, produced spontaneous, fast, and slow fissions. The calculated rate of spontaneous fission for this material is 25 fissions/gm hour. The cell contained 4 grams of uranyl nitrate in solution, for a spontaneous fission rate of 100 fissions/gm hour. The spectrum curves showed, as expected, an increase in the number of events upon irradiation with neutrons. Fragment energies fall in the range 40-110 MEV, with peaks at approximately 65 MEV and 100 MEV.[25] Uranyl nitrate also produced alpha particles of energies 4.60 MEV and 4.19 MEV, as well as other alpha particles of other energies emitted by daughters in the Uranium decay chain. The spectrum peak was very broad due to the fact that many different energies were being counted, and because amplifier gain was high.

Thorium nitrate, the alpha particle emitter intended for use produced alpha particles of energies 4.01 MEV and 3.95 MEV. Such a salt is also likely to produce

alphas of various energies, because the salt is contaminated with daughters which emit alphas.

V. RESULTS

Work was initally performed with the first resonator design. The assumption was made that if the cell would cavitate clean, degassed water under normal conditions, i.e. no radiation present, it should have been able to produce sufficient stress in the scintillating liquids to cavitate them in the presence of nuclear radiation. Barger's value for the cavitation threshold of clean, degassed water at an ambient pressure of 1 atmosphere is 12 bars. Greenspan found an absolute threshold of 5 bars for radiation induced cavitation in ethanol.[26] Assuming toluene to be a 'weak' liquid comparable to ethanol, 12 bars should have been more than enough stress to cause the desired cavitation in toluene. Both toluene and decalin were tried in the first cylinder, without producing the desired cavitation. Gassey cavitation occurred on the walls of the cylinder in both the (3, 0, 1) mode and the (1, 0, 1) mode. Subsequent measurements showed that the negative pressure was indeed greater on the walls of the cell than in the center. It was also noted that the cell was heating up in operation, indicating that power transfer to the

liquid was not efficient; the cell was lossy and acted as a drain on the input power. Also, the host liquids for the scintillants, in particular, toluene, acted on the adhesives used, limiting the useful life of the cell before repair became necessary. The vibration of the cell also exhibited a detrimental effect in that continued vibration eventually caused epoxy bonds to deteriorate.

Preliminary experiments were conducted with known alpha sources, Pu^{210}, Pa^{234}, and Bi^{210}, producing alpha particles of energies 5.3 MEV, 2.32 MEV, and 1.19 MEV respectively. A determination of the response of the phototube to alpha particles was desired, as well as a calibration of the multichannel analyzer for particle energies in the range 0 - 10 MEV. As can be seen in the graph on the following page, the Bi^{210} peak was buried in the background, and the Pu^{210} and Pa^{234} peaks were quite pronounced. A similar plot of the spectrum of the uranyl nitrate solution showed a peak between the Pu^{210} and Pa^{234} peaks, as expected. In the vicinity of 1 MEV, the background became too high to distinguish any peaks. This was due to amplifier noise and counting of other radiation, such as gamma radiation.

Many attempts were made to produce cavitation with the alpha emitters; each attempt meeting with little success. The conclusion was made that with the amplifier

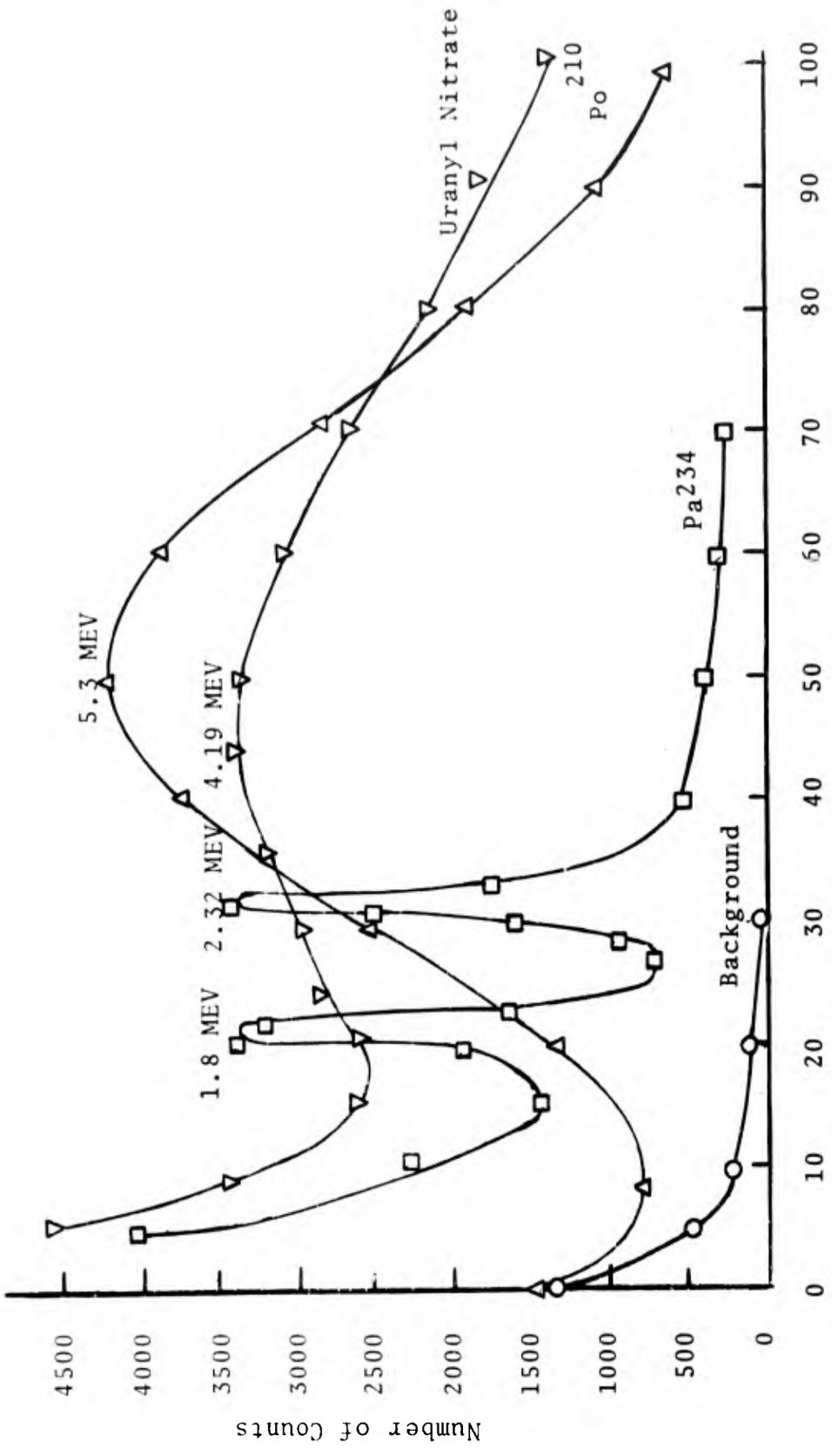


Figure 8, Spectrum curves of Background, Pa^{234}, Po^{210}, and Uranyl nitrate solution, showing peaks at 5.3 MEV, 4.14 MEV, 2.32 MEV, and 1.8 MEV.

used, the sound field was not sufficiently negative at its peak to reach the cavitation threshold for alpha induced cavitation. Alpha particles did not lend themselves well to the purpose of the experiment in any case due to the fact that their flux was so large that a single α-event could not be distinguished. Furthermore, because they were continuously emitted, there was no definite time at which it could be said the radiation commenced.

Uranyl nitrate on the other hand produced fission events at a rate of about two per minute. The individual scintillation flashes of these events were observed on the oscilloscope. Assuming linearity in the amplifiers amplifying the signal from the phototube, a rough estimate of the energies of the fission products was made. Calibration was impossible because there were no monoenergetic sources in the 40-100 MEV range available. In any case, the alpha peak in the uranyl nitrate spectrum showed a deflection of 0.4 cm, corresponding to a particle energy of 4.19 MEV. The scintillation flashes assumed to be produced by fission events showed deflections of 5 to 8 cm. Again assuming linearity, these deflections corresponded to energies in the range 52.5 - 84 MEV, energies which were indeed on the order of fission frag-

ment energies. Therefore, the assumption made appeared to have been valid.

Uranyl nitrate offered other advantages also, the most important of which was the reduced stress required to produce cavitation. Thus, even if the stress in the liquid were not high enough to produce alpha or neutron cavitation, it would be high enough to produce fission cavitation; negative pressures on the order of 2 bars, are sufficient for fission induced cavitation in a 'weak' liquid.[27] Again because monoenergetic particles were not available, exact thresholds could not be measured.

Since the fission events could be observed individually on the scope, they were much better suited for the time correlation experiment. The exact time of occurrence of any one event could be noted. Also, since cavitation events are single particle events,[28] it was only necessary to observe the scintillation event on the scope and to listen for the audio output of the cavitation event.

In resonator two, driven in the (3, 0, 1) mode with an input voltage of 40 volts, the first evidence of the radiation-induced cavitation was noted. Test samples of toluene driven in the same mode showed no evidence of the occurrence of cavitation, the reason being that the host

liquid wetted its motes or was very pure. The doped solution produced cavitation at the rate of 30 to 40 events per minute. These were very mild events, more like 'hard' cavitation than gassey cavitation in that they were single, distinguishable events. However, they were not as violent as those observed previously. It has been mentioned that alpha induced cavitation met with little success. These events may just have been alpha nucleated events. However, the results were not reproduced in subsequent runs. In any case they could not have been fission events because the frequency of occurrence was much too high. The neutron source was then placed in the light-tight box. No appreciable difference in cavitation rate was noted, and the nature of the cavitation remained the same.

In other runs on several occasions, hard cavitation was produced at input voltages in the range 20 - 30 volts. The threshold was inexact at best, as had been expected with the uranyl nitrate solution. The frequency of operation was 42.9 KHz. The events occurred at a rate of about 1.5 per minute and continued until the equipment was shut down. These were quite likely fission induced cavitations considering their rate of occurrence. However, no correlation was observed on the scope between

the scintillation events and the cavitation events. Neutrons incident on the liquid again had no effect on cavitation rate.

In other runs, cavitation occurred immediately when the threshold was reached at the rate of 6 - 8 events per minute. Again no correlation was observed between these events and the scintillation events. Input voltage at threshold was 30 volts, frequency 42.9 KHz. Within ten minutes cavitation had ceased completely and raising the input voltage only resulted in producing "gassey" cavitation. Neutron irradiation was then begun with no acoustic input. The cell was irradiated for twenty minutes, in an attempt to observe whether or not long-lived cavitation nuclei of the type described by Sette had been formed. When the power was again increased until the threshold observed previously had been reached, no cavitation resulted. It has not yet been ascertained just why this apparent incongruity in results occurred. Quite possibly it was due to slight heating of the cylinder in the latter runs after the hairline crack developed. Such heating caused frequency shifts which lowered the Q of the system.

Further tests on the threshold showed that the input voltage at threshold was reduced from 42 volts in an

undoped toluene sample to 22 - 26 volts in the doped solution. This further demonstrated that radiation induced cavitation was indeed occurring.

Test runs in this cylinder using undoped decalin and the neutron source produced no result other than the characteristic "gassey" cavitation which occurs at high drive.

In the final run made operating at a frequency of 43.4 KHz, a result was obtained which had been long awaited. Increasing the voltage in two volt steps of ten second duration, threshold was reached at 12 volts. The cavitation events were random at a rate of about 3 per minute. Observation of the scope showed the characteristic scintillations produced by fission fragments. During the period of observation three 'hard' cavitation events were heard immediately after high energy flashes appeared on the screen of the scope. In one apparent double fission event, four scintillation flashes occurred almost simultaneously, followed by the pings of two cavitation events. After these events, the cavitation ceased and even with neutron irradiation would not commence again. The frequency had drifted about 200 cycles and the Q of the system had fallen.

Cavitation of decalin with fast neutrons was attemped in the 2 inch cylinder at 35.1 KHz with no success. Gassey water produced only gassey cavitation; no vaporous or transient cavities were observed. Neutron irradiation did nothing to change this result. Degassed water with neutron irradiation proved to behave in the same manner. The uranyl nitrate solution also produced gassey cavitation in this cell. Hard cavitation was not produced in any of the liquids used in the cell. Apparently the cell had no good modes, as it produced only gassey cavitation on the walls and nothing along the axis.

The glass cylinder produced highly contradictory results. Filled with degassed water at an operating frequency of 52.3 KHz and an input voltage of 48 volts at threshold, a count rate of 8 counts per minute was established. Then the cell was subjected to neutron irradiation, and the count rate, rather than increasing as expected, decreased to 4 counts per minute. The only conclusion drawn from this result was that the neutrons were not responsible for the cavitation at all. The liquid obviously contained motes which were drowning any radiation effects. Motes would account for the decreasing count rate, in view of the fact that cavitation of

motes uses them up.

The Erlenmeyer flask, though it could not be coupled to the phototube, produced results which if nothing else confirmed previous observations. It also showed some interesting side effects. Degassed, doped toluene produced good sharp cavitation at a frequency of 38.6 KHz and an input voltage of 34 volts at threshold. Cavitation rate was initially 10 events/minute, decreasing with increasing time and eventually ceasing completely. To determine whether or not this was radiation induced cavitation a test sample of undoped toluene was introduced into the flask to the same liquid level as that for the doped sample. Frequency was identical and the voltage was set at 34 volts. Cavitation did not occur. The voltage was increased in increments of 2 volts and the sample began to cavitate at 50 volts. The cavitation was not as violent as that noted in the doped solution. In any case, the threshold was considerably reduced by doping with uranyl nitrate, making it plain that the cavitation was radiation induced.

The Erlenmeyer exhibited several interesting phenomena which will be commented upon briefly, due to the fact that they may lend themselves to future study. When the acoustic field was initially turned up to

previously determined threshold values and frequencies, hundreds of foam-like, white masses of bubbles formed in degassed toluene, eventually rising to the surface. At other times the entire volume of the liquid broke into large foam like streamers. Both of these processes lasted approximately 1 minute, and then stopped, leaving the liquid clear. Then hard cavitation began to occur randomly and continued for a period of time. The surface effects noted in the flask were quite interesting also. As the voltage was increased, fountains of liquid formed at the sides of the flask, held there by the sound field. Small fountains also appeared on the surface. Finally, in a water-methyl alcohol mixture, hard cavitation first occurred, followed by the formation of bubble streamers. These streamers broke apart, and their constituent bubbles formed larger bubbles of 0.12 inches diameter or larger, which were then stabilized at the pressure antinode along the axis of the flask. These bubbles remained stationary for approximately five seconds, then rose to the surface.

VI. DISCUSSION

A. Suggestions for Improvement of the Apparatus

The work of the project was severely hampered by equipment failures and malfunctions requiring long periods of dead time for repair and modification. The resonators, in particular proved to be extremely difficult to maintain since with each use they deteriorated to a greater extent. The epoxies and adhesives presented the main equipment problem in that the bonds made usually failed after a short period of time due to vibration of the cell and due to the action of the toluene. No particularly useful suggestion can be made to alleviate this problem except to say that better adhesives may exist. It was noted that the bonds lasted longer if care was taken to eliminate all air pockets from the adhesive between the two surfaces to be bonded.

The electric field on the cell itself produced interference with the phototube output. A shield of aluminum foil wrapped about the phototube circuitry and the use of coaxial cable alleviated this problem somewhat, but it is impossible to eliminate it completely because the phototube has to be mounted to the transducer.

The filtering and degassing system is mediocre. There is too much of a chance of contamination of the liquid. A closed system, of the type first tried is needed. This would eliminate the possibility of contamination of the liquid in pouring. Valves could be shut at both of the fill tubes and the cell could be removed from the system for experimentation without liquid spillage or contamination.

Another system which should be added to the electrical network is an automatic frequency control, which will automatically track the resonant frequency of the system, driving the system at its resonant frequency. This is absolutely necessary since the resonant frequency drifts enough during operation that the oscillator must be continually reset. In addition, another control gear for fine tuning should be added to the oscillator (not necessary if automatic frequency control is used). With high Q systems such as these, a one cycle change in frequency is often necessary. The oscillators available do not allow for such fine tuning.

B. Cavitation

The fact that radiation induced cavitation was produced with fission fragments, whose energies range from 40 - 110 MEV, and not with neutron irradiation

or alpha particles suggests the following:

1. The assumption that the degassed, distilled water was stressed to 12 bars may be wrong; it may have actually been cavitated at a lower threshold, a threshold below that of neutron induced cavitation but above that of fission induced cavitation.

2. The neutron source, located in a position 6 inches away from the cell, may have been far enough away so that the fast neutron flux at the cell may have been considerably reduced due to spherical spreading. The neutrons which did reach the cell may have been reduced in energy enough to raise the neutron-induced threshold. It should be noted that the work of Hahn and Peacock was conducted with the source mounted directly to the resonator. In any case, in all runs in which the neutron source was used, it seemed to have no effect on cavitation rate or threshold.

The results of the runs in which cavitation was produced with the uranyl nitrate suggest that the experimental results of Greenspan and Sette may both be applicable. As mentioned previously, the cavitation rate at threshold in runs with both the cylinder and the flask were initially high, in the range 6 - 10 cavitation events per minute. With time, this cavitation rate was reduced and eventually cavitation ceased. After it ceased, changing the frequency or increasing the voltage had no effect. This suggests that during the period in which the cell was not being operated, non-filterable, long-lived cavitation nuclei were being created in the liquid. When threshold was reached, these nuclei were producing the cavitation and being exhausted, causing the cavitation to cease. This result supports the conclusions of the work of Sette.

In the run in which cavitation was observed immediately after the scintillation flash, a somewhat different procedure was followed. Instead of gradually increasing the voltage until a threshold was reached, it was increased in 2 volt increments, and the output was monitored for 10 seconds. The input voltage at threshold in this case was only 16 volts, as opposed to thresholds in the range 20 - 30 volts in the other runs.

This suggests that cavitation on the proposed long-lived nuclei required more negative pressure than cavitation on the newly formed nuclei. It also suggests that the nuclei, after they are formed, lose some of their "strength" with time, requiring a greater threshold. The newly formed nuclei on the other hand, retain all their energy and thus produce cavitation at a lower threshold.

The results of the final run also suggest that a time correlation of the type suggested by the work of Greenspan exists. No evidence of an induction time for radiation induced cavitation was noted.

VII. CONCLUSIONS

An apparatus suitable for this work has been developed which shows significant promise of success with some refinements. Data taken was considered sufficient to make suggestions as to what occurs upon the incidence of radiation on a liquid upon which a sound field has been impressed. However, it was not sufficient to make any definite conclusions. It appears as if cavitation does indeed occur immediately upon the incidence of radiation. It also appears that radiation can produce long-lived cavitation nuclei when

the field is non-existant. More study needs to be conducted in order to obtain sufficient data to state conclusively just what is occurring in the irradiated liquid.

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APPENDIX

The Glass Cylinder

In working with the glass cylinder resonator, an interesting technique was discovered. Though not related to the experimental work directly, the technique was considered important enough to include in the appendix.

Liquid height is of prime importance in the proper functioning of such a free surface system. The resonant frequency of the system is determined by the physical characteristics of the transducer and brass disc to which it is attached. Therefore, the water height must be the proper number of wavelengths along the tube before the liquid will be driven in resonance, sustaining high acoustic pressures.

Using the current probe, a simple method was devised to find the most efficient water height of the system; that is the height at which the Q of the liquid system is greatest. The system was first tuned with the variable capacitors. Then the frequency was set at the resonant frequency of the system. Water height was controlled by a valve at the exit tube. When the valve was cracked the liquid left the water column drop by drop. When the current measured on the

current probe had reached its maximum value, the proper water height of the system had been found.