CHAPTER II

EXPERIMENTAL

The details of the piezo-electric and magnetostrictive transducers and the construction of an RF oscillator to excite them have been described in the beginning of this chapter. The measurement of the intensity and wavelength of the waves emitted by them has also been described. This is followed by the description of the assembly of the apparatus to obtain ultrasonographs and sonoluminiscence flux and of the construction of an optical densitometer to measure the blackening of the image. The chapter ends with the experimental details to measure the amount of water absorbed by the film and the effect of temperature and humidity on its blackening.
2.1 PRODUCTION OF ULTRASONIC WAVES.

Ultrasonic waves were produced using piezo-electric and magnetostrictive transducers. A brief description of these transducers is given below.

2.11 PIEZO-ELECTRIC CRYSTALS:

High frequency ultrasonic waves can be produced using piezo-electric transducers which transform electrical oscillations into mechanical oscillations. Piezo-electricity is a property possessed by certain crystals like quartz, tourmaline, cane-sugar etc., which when subjected to pressure or tension develop electric charges on definite surfaces. Conversely if an electric field is applied along a definite direction called the piezo-electric axis the crystal expands or contracts in certain other directions. Quartz found extensive applications since it is dense, hard, strong, resistant to chemical attack, impervious to moisture and can be easily worked out. But quartz has high impedance and in practice if high power levels are to be obtained, it will be necessary to use radio-frequency potentials of several thousand volts.

A major improvement in the field was the discovery of a new group of crystals known as the ceramic crystals. They are manufactured by heating a mixture of metallic oxides in the furnace and this results in a solid mass
that resembles earthenware. This has poor piezo-electric characteristics as it comes from the furnace but it can be improved upon by the polarisation process. This process consists of keeping the crystal in a powerful electro-static field. Suppose a flat disc is to be polarised, first its opposite faces are silvered to make them conducting. They are then provided with connecting wires and the crystal is kept immersed in oil. The temperature of the oil is raised above the Curie temperature for the material and then a steady voltage of the order of 200 volts for every millimeter thickness of the crystal, is applied. The temperature is then steadily reduced below the Curie point. The disc will now behave like a quartz disc of similar type. This polarisation is reasonably permanent but it rapidly deteriorates as soon as the crystal reaches the Curie point once more. It can however, be repolarised.

(A) Plane crystals: In the present investigation we have used X-cut (longitudinal) crystals with parallel faces made of lead zirconate ceramic, type PZT-2 and PZT-4 details of which are given in Table 2.1.
Table 2.1: Details of the piezo-electric transducers.

<table>
<thead>
<tr>
<th>Ceramic</th>
<th>PZT-2</th>
<th>PZT-4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency in MHz</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>Thickness in cm</td>
<td>0.15</td>
<td>0.080</td>
</tr>
<tr>
<td>Diameter in cm</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>Curie Temp. °C</td>
<td>300</td>
<td>325</td>
</tr>
<tr>
<td>Manufactured by</td>
<td>National Physical Lab., New Delhi</td>
<td>Valpey crystal Corp., Holliston, Mass, U.S.A.</td>
</tr>
</tbody>
</table>

A PZT-2 crystal of frequency 1 MHz and thickness, 0.3 cm, was manufactured and the PZT-4 crystals were chrome plated by sputtering as the N.P.L., New Delhi. These crystals were mounted in a holder made in our workshop. Holders with solid metal backing were used in the investigation. The crystal was hemmed between a metal ring of diameter, 1.4 cm, and a backing plate and both of them were separately connected to the output terminals of the oscillator.

(B) Focussing Bowl: Ultrasonic waves of large intensity were required to study sonoluminiscence and for this purpose a ceramic crystal which was spherical in shape was used as it could focus the ultrasonic waves to produce intense waves at its centre of curvature. The details about this crystal are in Table 2.2.
Table 2.2: Details of the focussing crystal.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ceramic</td>
<td>Ba TiC₃O₃</td>
</tr>
<tr>
<td>Internal diameter</td>
<td>11.62 cm</td>
</tr>
<tr>
<td>Internal aperture</td>
<td>9.06 cm</td>
</tr>
<tr>
<td>Thickness</td>
<td>0.62 cm</td>
</tr>
<tr>
<td>Internal height</td>
<td>2.18 cm</td>
</tr>
<tr>
<td>Density</td>
<td>5.5 g/cm³</td>
</tr>
<tr>
<td>Frequency</td>
<td>450 kC/S</td>
</tr>
<tr>
<td>Curie temperature</td>
<td>120°C</td>
</tr>
</tbody>
</table>

This crystal was manufactured by the Brush Clevite Corporation, Cleveland, U.S.A. It had two fired in leads on either side of it and could be readily used by connecting these leads to the output terminals of the oscillator.

2.12 Magnetostrictive Transducer.

If the intensity of the ultrasonic waves is large then it is possible to expose photographic films to ultrasonics without any liquid transmitting medium. Since the attenuation of the ultrasonic waves is proportional to the square of the frequency, low frequency waves obtained by a magnetostrictive oscillator were used.
The magnetostrictive effect occurs in ferromagnetic materials such as iron, nickel, cobalt and certain special alloys. When these materials are subjected to magnetic fields there will be a change in their physical dimensions. Similarly there will be a change in their magnetic properties if their dimensions are altered by external force. The magnetostrictive effect which involves the change in the length of the material along the direction of the magnetic field is called Joule effect and is utilised in the generation of ultrasonic waves.

Magnetostriction can be explained by consideration of the domain theory. In an atomic system the orbital electron produces a magnetic moment. Due to random orientation of atoms in a solid, the magnetic moments of separate atomic groups are generally cancelled out. However ferromagnetic materials are an exception to this. In these systems an exchange force exists that causes all the atomic magnetic fields within a volume of about $10^{-3}$ cm to lie parallel to each other. These domains of magnetic moments can be aligned by an external magnetic field applied along one of the many fixed directions called the directions of easy magnetisation which depend on the structure of the crystal.
When an external magnetic field is increased the domains originally magnetised in the direction of the field grow in size taking over the other differently oriented domains until the complete crystal becomes a large domain. As the field becomes more intense the domain in each crystal rotates until it is parallel to the field and in doing so the material expands or contracts externally.

By passing electric current through a solenoid a suitable magnetic field can be generated along its axis. A sine wave source that excites the solenoid will produce directional changes in the core at twice the exciting frequency. If a varying external magnetic field is produced by a solenoid whose core is formed by the magnetostrictive element, it will operate in two directions of easy magnetisations which are opposite to one another. Since the initial system consists of completely random domains their average movement will cancel out. But if an initial magnetisation is given to the core and it is then subjected to a varying magnetic field the domain movement will be considerably greater in one direction. This causes a change in length of the order of $10^{-5}$ cm per cm. Whereas iron expands in a weak field and contracts in a strong field, nickel contracts at all field strengths. All magnetostrictive
materials are temperature sensitive and the material loses its magnetostriction as the temperature rises until the Curie point is reached where all magnetic properties cease.

In the present investigation a magnetostrictive oscillator containing a nickel transducer of frequency 20 KC and manufactured by Dr. Lehnfeldt and Co., Heppenheim, Germany was used. The length of the transducer, \(d = 7 \text{ cm}\) and it possessed a node at the centre of its fundamental mode of vibration. It was fitted with a tapering coupling stub of length 3d cm.

2.2 Intensity, Wavelength and Frequency of Ultrasonic Waves:

**Intensity**: It is customary to express the intensity of the ultrasonic waves as the ratio of the total power output by the crystal to the effective area of the crystal. The effective area can be easily measured and the total power output is obtained by the calorimetric method using the following formulae. If waves are passed through a liquid for a known time \(t\), the total power output \(W = JH\) and \(H = (m_1 s_1 + m_2 s_2) (\theta_2 - \theta_1) t^{-1}\). 

\[\text{(2.1)}\]
where $J$ is the mechanical equivalent of heat $\theta_2$ and $\theta_1$, are the initial and final temperatures, $m_1, m_2$ and $s_1, s_2$ and are the masses/specific heats of the liquid and calorimeter respectively.

In the near field or the Fresnel region, there will be positions of intensity maxima and minima on the beam axis (art. 1.31) and it is necessary to know the wavelength of sound in the liquid medium in order to calculate these positions.

**Wavelength**: The wavelength of sound $\lambda_l$ in the liquid medium was measured using Debye and Sears technique. The experimental arrangement is shown in Fig. 2.1. The whole assembly of the apparatus was built by Andhra Scientific Co., India. The ultrasonic waves passing through a liquid are elastic waves, in which compression and rarefaction travel one behind the other at regular distances apart. The density of the liquid is therefore changing periodically in the direction of the sound wave and hence also its index of refraction. This variation in refractive index is used to produce diffraction of light. This diffraction pattern is visible on a ground glass plate and is measured using a travelling microscope. $\lambda_s$ is calculated from the formula

$$\lambda_s \sin \theta_k = k \lambda_l \quad \cdots \cdots \cdots \cdots (2.2)$$
Diffraction of Light by Ultrasounds
where \( \theta_k \) is the angle of diffraction for the \( k \)th order and \( \lambda_l \) is wavelength of the monochromatic light. Since \( A \), the distance between the screen and the diffraction grating set up by ultrasonics is very large as compared to \( d_k \), the distance of the \( k \)th diffraction image from the central image, \( \sin \theta_k \) can be replaced by \( d_k/A \). A sodium vapour lamp was used to produce monochromatic light of \( \lambda_l = 5896\AA \). Ultrasonic waves were produced by a quartz crystal of frequency, \( n = 3.2 \) Mc. The velocity of sound in the liquid \( V \), can be readily obtained from the formula, \( V = n \lambda s \).

**Frequency:** The frequency of the crystal was quoted by the manufacturers. The crystal can vibrate in its fundamental or odd harmonic modes. The intensity of the waves of harmonic modes is very small and so the crystal was used to emit waves in its fundamental mode and its value was checked by connecting the oscillator to an oscilloscope.

2.3 **RF Oscillator:** The piezo-electric crystal was excited by an RF oscillator of output power 100 watts and frequency range 1 to 6 \( \text{MC} \). The DC voltage to the Oscillator was supplied by a stabilised power supply, operating in the range 0-500 Volts and 0-500 milliamperes.
built by the General Electric Company, U.S.A.
The oscillator was constructed in this laboratory. Its circuit diagram is given in Fig. 2.2 and its working is briefly described below.

An oscillator can sustain oscillations and will be useful in practice if the output power is sufficient to overcome the losses of the circuit and also to provide the power that is transferred to the external circuit. This can happen if the tube acts as a negative resistance.

The oscillator that has been constructed is essentially a plate coupled multi-vibrator oscillator with an L-C tank circuit connected between the plates of the two 829B tubes. It acts as a negative resistance oscillator. The two tubes have identical characteristics and if the voltage between the two points A and B is zero there is no flow of current between them. But if a small increment of voltage is applied between A and B, a small current will flow between them. This current will increase the plate voltage and decrease the grid voltage of one tube and decrease the plate voltage and increase the grid voltage of the other tube. If the resulting amplification is large then the change in the plate current exceeds the
current flowing between A and B and will be in a direction opposite to that of the applied voltage. So apparently a negative resistance will be produced between A and B. If an LC tank circuit is tuned between A and B, sustained oscillations can be obtained. Though the multivibrator by itself is a class C oscillator since there is only instantaneous flow of current the addition of the tank circuit makes it a class A oscillator because then the current will flow in the plate circuit throughout the cycle.

The two RF chokes of 50μH each, stop the AC component from getting back into the power supply. The two condensers of 2μH each cut off the DC components and feed back RF Voltage of proper phase to the grids. The 14 K resistance keeps the screen grid at a slightly lower potential than the plate. The 7K resistances act as grid bias resistors and supply proper voltage to the grids. The frequency of the oscillator \( f = \frac{1}{2\pi\sqrt{LC}} \) where \( L \) is the inductance of the coil and \( C \) is the capacitance. Induction coils were wound using the formula

\[
L = \frac{2\pi^2}{3a + 9b + 10c}
\]

where \( a = 2.5" \), the diameter of the coil; \( b = 1.5" \), the distance between the extreme turns of the coil, \( c = 0.4" \),
FIG 2.3  Experimental Arrangement to obtain Ultrasonographs
the diameter of the wire and \( n = 16 \), the number of turns of the coil. The value of \( L \) thus calculated was about 15 H and using a ganged condenser in the range 0-300 pf oscillations in the frequency range 2.5 MHz to 5 MHz could be obtained. Another coil with \( n = 50 \), was used to obtain oscillations in the frequency range 0.8 to 2.5 MHz.

The piezoelectric crystal was excited by connecting it across a secondary coil which would round the above inductance coil. One surface of the crystal was earthed. The RF voltage across the crystal in the range 50 to 110 volts was measured using a Philips vacuum tube voltmeter.

2.4 Assembly to obtain Ultrasonographs:

2.4 (a) Piezoelectric Oscillations: The assembly of the apparatus is shown in Fig. 2.3 and 2.4. The ultrasonic waves from the piezoelectric crystal were transmitted through transformer oil to a stainless-steel cell. This cell was 2 X 4 X 5 cm. in size and had a 2 X 2 cm. removable window of polythene sheet of thickness 5 microns. Stainless-steel does not react with the test liquid. The incident ultrasonic waves that were transmitted through the window propagated through the test liquid to irradiate the photographic film kept in the cell. The position of the cell could be accurately adjusted by a micrometer, and a gear...
FIG 2.4 Assembly to obtain Ultrasonographs.
arrangement enabled to keep the cell at different angles. The film was cut from a film-roll obtained from the local market, and its entire exposure and processing was done in total darkness. The piezo-electric crystal, the transmitting liquid and the test-cell were all kept in a 6X6X6 cm. copper-box with a removable lid. The box was light-proof when the lid was closed. This copper-box was kept in a big tray containing water which helped to control the temperature of the liquid inside the container and the cell. The temperature of the water in the tray was adjusted by adding ice, and was about $5^\circ$ C less than the temperature of the liquid inside the test-cell. The temperature of the test-liquid was measured before and after the exposure and it changed by about $2^\circ$ C during the exposure. The film was then removed and developed.

2.4 (B) **MAGNETOSTRICTIVE OSCILLATION**

The assembly of the apparatus to expose photographic films to ultrasonic waves obtained from a magnetostrictive oscillator is shown in Fig. 2.5. Electric oscillations of frequency 20 KC$\omega$ were obtained from an oscillator of frequency range 30 cycles$^{-1}$ to 30 KC$\omega$ built by Bharat Electronics, Bangalore. The output voltage of the oscillator was increased to 120 Volts by means of an amplifier manufactured
by Motwane Private Ltd., Bombay. This oscillatory current was fed to the solenoid of the magnetostrictive nickel transducer. The prepolarising current of 3.5 amperes and 24 volts, obtained by a battery was also fed to the solenoid. The 2µF condensers stopped any direct current voltage from entering the alternating current supply and the 0.5H induction coil stopped any alternating current voltage from entering the battery circuit. The oscillations produced were coupled to air by a connecting tapering stub. The entire transducer assembly was enclosed in a light proof wooden box. The rise in temperature of the transducer was checked by the continuous flow of water in the double walled vessel surrounding it. The photographic film was exposed to ultrasonic waves by keeping it in air directly in front of the coupling stub at a distance of about 2-3 millimeters.

2.5 BLACKENING OF THE FILM:

The latent image was visible on development and fixing of the film. The amount of blackening of the image was a measure of the interaction between ultrasonics and the emulsion.
A. Development: The exposed film was developed in D-19 developer for 4 minutes in total darkness. The chemical composition of this developer is given below:

D - 19 Developer

Water 1000 CC
Metol 2 gm
Sodium Sulphite 90 gm
Hydroquinine 8 gm
Sodium bicarbonate 52.5 gm
Potassium Bromide 5 gm.

B. Fixing: After development the film was dipped in water for half a minute and put in the fixer for five minutes. The fixer consisted of a saturated solution of sodium thiosulphite (hypo) in water. The film was carefully washed in water and allowed to dry in the air.

C. Density: The amount of blackening of the photographic film is measured in terms of density which was defined by Hurter and Driffield as the mass of silver per unit area. They observed that the mass of silver in the image is directly proportional to the absorption of light i.e., to its optical density. If $I_t$ and $I_o$ be the intensity of light incident on the sample and transmitted by the sample respectively then its optical density is defined as $\log I_o/I_t$. 
FIG 2.6 Optical Densitometer.
In practice the density invariably refers to optical density and is the logarithm of the ratio of the light intensity measurements by a photometer without and with the blackened film. The instrument used to measure the density of blackening is called a densitometer.

2.6 DENSITOMETER:

The details of the densitometer constructed in this laboratory are shown in Fig 2.6 and 2.7. An ordinary bulb of 200 watts operated at 230 volts and enclosed in a wooden-box having a small circular aperture in its front served as the source of intense light. The beam coming out from the aperture was converged by a system of lenses and an intense narrow beam of light fell on a slit. The slit had a circular opening of diameter less than a millimeter and the film whose density of blackening was to be measured was kept on the other side of the slit in immediate contact. The position of the film could be easily adjusted such that light could scan the full image on the film. The transmitted light liberated electrons from the cathode of the photocell RCA 923 to produce electric current of about 10 microamperes.
The current from the photocell was amplified by a 6SN7 double triode which acts as a differential amplifier as shown in Fig. 2.7. It was operated at 100 volts. The 6000 PF condenser allows the D.C. output from the photocell to be fed to the grid of only one tube and A.C. output if any, to the grids of both the tubes. The 500 KΩ resistances act as grid bias resistors. The variable 20 KΩ resistance acts as a cathode bias resistor and the used variable 2000KΩ and fixed 10 KΩ resistances were to adjust the potential difference so as to obtain zero current in the microammeter. The difference in current across the plates of the two tubes was read on a microammeter in the range 0 to 250 mA. The current thus measured was taken to be proportional to the intensity of light falling on the photocell. Deflections in the microammeter were noted when the narrow beam of light was made to pass through different regions of the film.

2.7. Assembly to study Sonoluminescence:

Sonoluminescence is the light emitted by liquids cavitated by ultrasound. Its intensity will depend on the intensity of ultrasonics, nature of the liquid, temperature of the liquid, etc. But its origin is not clearly understood and the results on its intensity
measurements are contradictory. So its intensity at
different temperatures and in different liquids have
been independently measured. The experimental arrangement
is shown in Fig. 2.8, and its details are given below.

The ceramic bowl shaped crystal (2.1 B) of
frequency 450 Kc/ was used to obtain intense ultrasonic
waves to study sonoluminescence. The crystal when
excited by an RF oscillator, focussed the waves at its
centre of curvature. "Ultrasonic Generator" model E-600,
manufactured by Fico-Products company, Framingham, Mass.,
was used to excite the crystal. The oscillator had a
frequency range of 120 Kc/ to 1 Kc/ with an input power in
the voltage range of 0-1500 and current range 0-200
milliamperes. The RF voltage across the crystal in the
range 0 to 30 volts was measured by Hewlett-Packard Vacuum
tube voltmeter. The ultrasonic waves were transmitted
through water and then through a thin polythene window
of 30 c. of test-liquid contained in a glass beaker.
The top surface of the liquid was just above the centre
of curvature of the ceramic bowl. The sonoluminescence
produced in the liquid was guided by a perspex rectangular
block 10 x 4 x 3 cm in volume to a photomultiplier tube,
RCA 931, kept enclosed in a metallic box with a 3 x 2 cm.
opening. This tube was operated by electric power, 1250 volts
and one milliamper current, supplied by a High voltage Unit
FIG 2.8 Experimental Arrangement to Measure Conoluminiscence Flux (1) Curved transucer (2) Transmitting liquid (3) Window (4) Beaker containing test liquid (5) Light-guide (6) Photomultiplier tube (7) Coolen box (8) Cooling system.
HV56 constructed by Atomic Energy Establishment, Bombay. The whole assembly was enclosed in a light-proof wooden-box. On passage of ultrasonic waves the temperature of the temperature of the test liquid rose rapidly. The rise in temperature was controlled by circulating water in a double walled outer vessel. The current produced by the photo-multiplier tube was read on a spot galvanometer, manufactured by Cambridge Instrument Co., Ltd., England. The sensitivity of the galvanometer was determined experimentally and was $5 \times 10^{-8}$ amperes per millimeter. The anode dark current of the galvanometer was $0.16 \times 10^{-6}$ amperes. Deflections in the range 0-100 mm. were read on the galvanometer for different applied voltages in the range 0-30 across the crystal. It took less than a minute for each set of readings and the temperature of the test liquid changed by about 1 °C during this interval.

By removing the perspex guide and keeping a photographic film in the test-beaker, the above set-up can also be used to expose photographic films immersed in different liquids to intense ultrasonic waves.

2.8 APPARATUS TO STUDY THE EFFECT OF MOISTURE AND TEMPERATURE ON THE FILM

In order to study the variation of the density of blackening due to ultrasonic irradiation on a moist emulsion
with temperature, distance, nature of the liquid, intensity of sonoluminescence, etc., it will be useful to know the effect of these parameters on the photo-film in the absence of ultrasonic irradiation. The experimental details to study these are given below:

**Change in Weight:** When the photographic film was immersed in water it absorbs water. The amount of water absorbed depends on the temperature of the water and on time during which it was immersed. With increase in time of immersion the film absorbs more and more water and gets saturated in about one minute. Small strips of films were cut and immersed in water for different intervals of time in the range 5-60 seconds and at different temperatures, in the range 10-35°C. On removing them from the water bath they were quickly dried by filter papers and carefully weighted in a chemical balance correct to $10^{-4}$ gm.

**Change in Volume:** Besides a change in the weight of the emulsion on immersing in water there was a change in its volume too. On absorbing water the thickness of the emulsion increased and practically there was no change in its lateral dimensions. The change in thickness was measured correct to a micron by means of a Cooke's Research microscope fitted with an eye-piece of magnification 10.
Change in Density of Blackening: The density of blackening changes with the temperature of the exposure, humidity of the ambient atmosphere, distance between the source and the film, time of exposure etc. Faint light sources such as (A) Zero-watt bulb; (B) Sodium iodide crystals with traces of Thallium in it supplied by Harshaw Chemicals, Cleveland, Ohio and irradiated by γ-rays from Ra source were used. The light source was replaced by X-ray source, Pu-237 while studying the effect of temperature on the blackening. The change in humidity, in the range 0 to 100%, of the ambient atmosphere, was brought about by keeping the film in a polythene bag containing chemicals like silica gel, copper sulphate, etc.
REFERENCES


