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APPLICATIONS OF ULTRASONIC ENERGY

Progress Report No. 7

for December 1957-January 1958

by

Aeroprojects Incorporated

ABSTRACT

Ultrasonic precipitations of thorium oxalate, carried out by the addition of solid oxalic acid to thorium nitrate solution, yielded smaller and more regularly shaped particles than did non-ultrasonic precipitations involving slow addition of oxalic acid solutions. The importance of digesting the precipitate and of thorough washing to remove excess reactants was emphasized. Preliminary experiments were undertaken in the reaction of solid thorium nitrate and oxalic acid with concomitant water addition and ultrasonic application.

Studies were undertaken in the ultrasonic disintegration of composite fuel elements in mercury. Rapid erosion of Hastelloy C, stainless steel, mild steel, molybdenum, and graphite couplers indicate a problem in selection of an ultrasonic coupler material, although it may be possible to use a sacrificial coupler. Relatively short exposure resulted in extensive pitting and erosion of stainless steel sheet. The high erosion rates of the above materials indicate that such claddings may be effectively disrupted in mercury with applied ultrasonics.

Ultrasonic leaching of cesium-bearing alumina waste with 0.01 N nitric acid indicated a positive ultrasonic effect on rate of leaching in a continuous-flow or fixed-bed array. Undesirable breakup of the calcined waste pellets obtained under certain conditions suggests that non-cavitation leaching should be investigated.

Investigation was initiated on ultrasonic application to the magnesium extraction of plutonium from uranium-chromium alloys. Preliminary studies in a room-temperature simulant system indicated high efficiency in emulsifying mercury in water and a substantially increased rate of extraction of copper from mercury with aqueous reagents. The possible use of a tantalum ultrasonic coupler will be investigated prior to tests with the actual system of interest.

A theoretical evaluation and experimental program has been initiated on ultrasonic agglomeration of submicron radioactive particles in very

(over)

dilute gas streams. A new approach, adding an aerosol of particles in the 2-10 μ size range of a suitable wetting agent and superimposing ultrasonic vibration, appears promising both theoretically and practically. The production of additive aerosol particles from a viscous non-evaporating surfactant liquid in this size range with the Aeroprojects atomization techniques has been demonstrated.

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SECTION I

ULTRASONIC CONTROL OF CRYSTAL SIZE DURING AQUEOUS PRECIPITATION

PURPOSE AND SCOPE

Thorium oxide particulates of submicron size are desired for evaluation purposes and for use in HTR blanket slurries to avoid settling and plugging of the system. The thorium oxide is currently prepared by calcination of thorium oxalate precipitated from the reaction of thorium nitrate and oxalic acid, and the size of the thorium oxalate particles determines the final oxide particle size. However, difficulties are encountered in achieving suitably small, unagglomerated particles of thorium oxalate. This may be accomplished if precipitation is carried out at low temperatures, but the reaction rate is undesirably slow; elevated temperatures accelerate the reaction but produce large, agglomerated particles.

This work was undertaken to evaluate the effectiveness of ultrasonic energy applied during precipitation in producing submicron thorium oxalate particles at reasonable reaction rates. Initial ultrasonic studies with a simulant system of calcium nitrate and oxalic acid at room temperature produced small, unagglomerated particles and indicated that the desired submicron size might be achievable. Investigations are being carried out with the actual system of interest to evaluate significant chemical and ultrasonic parameters and to establish the conditions most favorable for producing fine particles of thorium oxalate.

HIGHLIGHT DEVELOPMENTS

It was previously reported (1)* that thorium oxalate particles of 1-2 microns diameter were produced by ultrasonic application during precipitation from thorium nitrate and oxalic acid, using a 20-kc, converging-coupler ultrasonic treatment vessel, under each of three conditions: (a) with fast addition of solid oxalic acid to thorium nitrate solution, (b) with fast addition of oxalic acid solution to thorium nitrate solution, and (c) with fast addition of thorium nitrate solution to oxalic acid solution. Further experiments have been made using other methods of preparation.

* Numbers in parentheses refer to references on pages 31-35.

Detailed information was obtained concerning the present technique of preparing thorium oxide at Oak Ridge National Laboratory. The oxalate is prepared by slowly adding 1 M oxalic acid solution to 1 M thorium nitrate solution at 10°C, with very rapid stirring. After the addition is complete, the suspension is allowed to stand at room temperature for 8-10 hours. The precipitate is then filtered through a sintered glass filter, washed, dried, and converted to the oxide. The resulting oxide particles are in the 1-micron size range.

A batch of thorium oxalate was prepared in this manner without ultrasonic application during precipitation. A second precipitation using solid oxalic acid with applied ultrasonics was run for comparison and allowed to stand for 8-10 hours. The resulting precipitates were filtered, washed, and dried, and particles were evaluated by microscopic examination and by a sedimentation analysis somewhat modified over that previously described (1). A 5% suspension of dried thorium oxalate was prepared in 0.005 M tetrasodium pyrophosphate solution, and 350 milliliters of this suspension was poured to a height of 42 inches in a glass tube 25 millimeters in diameter. A G-M tube (TGC-2) with a thin window connected to a utility scaler was placed 6 inches below the liquid level, with the window end contacting the wall of the tube. Activity counts were taken over a 24-hour period, and the rate of decrease in activity was related to the particle size of the suspended thorium oxalate. This method provided qualitative information on relative particle size distributions, but no quantitative data.

Figures 1 and 2 are typical photomicrographs of the thorium oxalate precipitates as they appeared in suspension in the tetrasodium pyrophosphate solution. In general, the material precipitated under ultrasonic influence (Figure 2) shows a smaller and more regular particle size than the non-ultrasonic precipitate. Figure 3 shows the ultrasonically prepared material smeared in the dried state on the slide.

A few larger particles, up to 5 microns diameter, were found in the ultrasonically treated oxalate after drying; these are evident in the resuspension shown in Figure 2. No particles of this size were noted in previous preparations, for which microscopic studies were made with the oxalate suspended in the mother liquor, i.e., before washing and drying. The size and general appearance of these large particles suggest that they may have been formed during the drying operation as a result of unreacted thorium nitrate or oxalic acid that was not removed by washing; such unreacted material may have either crystallized into large particles during drying or cemented some of the thorium oxalate particles together. If this is the cause, the large particles should be eliminated by calcination, which would decompose any excess thorium nitrate or destroy any excess oxalic acid. Efforts are being made to refine the washing procedure to eliminate such particles.

Another procedure for preparing thorium oxalate was investigated at the suggestion of personnel of Oak Ridge National Laboratory. Equivalent amounts of dried thorium nitrate and solid oxalic acid were premixed and

then added to water, ultrasonic energy being applied during the addition. The precipitate obtained in this manner was filtered, washed, dried, and evaluated in the same manner as before. Sedimentation analysis showed relatively rapid settling, and microscopic examination of the oxalate suspended in tetrasodium pyrophosphate solution revealed the large cubic crystals evident in Figure 4.

FUTURE WORK

Future work will involve the preparation of thorium oxalate particles washed completely free of excess reagents and the preparation of several samples by ultrasonic treatment for evaluation at ORNL.

SECTION II

ULTRASONIC PROCESSING INVOLVING MERCURY

PURPOSE AND SCOPE

Ultrasonic energy transfer to mercury systems appears to be an efficient process and one in which significant ultrasonic effects can be anticipated in a variety of processes. This part of the program therefore involves several subtasks relating to dissolution, dispersion, agglomeration, gel destruction, nucleation, and possibly others.

Dissolution or Disintegration of Fuel Elements in Mercury

The use of molten metals for the dissolution of spent fuel elements to eliminate fission products and recover fissile materials has been reported to possess several advantages over aqueous systems (3). Besides an economy of volume, certain molten-metal systems provide a radiation shielding power that obviates the necessity for long-time "cooling-off" of the fuel elements.

One of the more promising applications is the dissolution of fuel elements containing thorium or uranium in mercury. Both thorium and uranium have appreciable solubilities in mercury at elevated temperatures, and subsequent reclamation of these elements by volatilization of the mercury can be accomplished. However, the process is sometimes erratic with massive pieces of thorium or uranium, which become inert or dissolve slowly because of the build-up of films, presumably mercurides, on the surface of the elements. Furthermore certain fuel-element claddings and matrix materials used in dispersion-type fuel elements, such as Zircaloy and stainless steel, are normally resistant to attack by mercury and have low solubility in this metal even at elevated temperatures (4).

It has been demonstrated that appropriate application of ultrasonic energy accelerates erosion and dissolution processes (5,6,7,8,9), either by inducing high mechanical stresses in surface layers or by creating agitation in the liquid phase, promoting homogeneity and preventing supersaturation in the solution at the liquid-solid interface. In particular, it has been reported (4) that metals such as molybdenum and mild steel, which are resistant to attack by quiescent mercury at temperatures up to 500°C, are severely eroded in mercury even at room temperature under the influence of high-intensity vibratory energy.

Investigations are therefore being undertaken to establish suitable materials and methods for introducing ultrasonic energy into mercury systems and to determine the feasibility of accelerating the dissolution of fuel elements, rupture of fuel element claddings, and disintegration of dispersion-type fuel elements in mercury by this means.

HIGHLIGHT DEVELOPMENTS

Dissolution or Disintegration of Fuel Elements in Mercury

In view of the above-noted accelerated erosion of many metals in mercury under ultrasonic influence, a major problem in introducing vibratory energy into any mercury system is the selection of an ultrasonic-energy conductor (coupler) material that will be relatively resistant to such attack.

Scouting tests were conducted with several materials considered to be potentially suitable for couplers, including Hastelloy C, Type 304 stainless steel, mild steel, molybdenum, and dense graphite. Available couplers of each of these materials were attached to a 20-kc nickel-stack transducer. The couplers were then immersed in liquid mercury contained in a Type 302 stainless steel beaker, and vibratory energy was introduced through the coupler at power levels ranging from 300 to 1000 effective watts to the transducer for periods of 50-120 minutes. Some of the tests were made with continuous treatment and others involved pulsing at 30 seconds on and 30 seconds off. The tests are summarized in Table I.

All of the couplers showed marked cavitation erosion. Figure 5 shows the face and side of the Hastelloy coupler after a 60-minute exposure at 1000 watts power. The face has a peened appearance. The side appears to have been deeply etched, and microscopic examination revealed intergranular corrosion. The appearance of the molybdenum coupler face is evident in Figure 6.

The stainless steel beaker which contained the mercury in these experiments was also severely pitted and eroded, although it was located 2 inches from the radiating surface. The appearance of a section of the beaker is shown in Figure 7. The pitting in Section A resulted during one experiment in which the mercury level was higher than usual. Most of the pitting occurred in Section B below the surface of the mercury. The edge view of this section (Figure 7b) illustrates the gross erosion that resulted; the wall thickness below the mercury level was reduced from 0.030 to 0.021 inch. The rate of penetration can not yet be estimated. Local penetration in the stainless steel and Hastelloy couplers appeared to occur at a considerably higher rate; it was estimated that the average pit depth was 3 mils after 1 hour of pulsed ultrasonic treatment.

One test was conducted in which a polished Type 304 stainless steel rod of 1/2-inch diameter and 3 inches long was held in place vertically between a steel coupler face and the bottom of the mercury container. After application of ultrasonic energy at 800 watts for 30 minutes in 30 second on-30 second off pulses, the surface of the rod was pitted over part of its length. A short section of 1/4-inch-diameter stainless steel rod that had been allowed to float on the surface of the mercury during this test was wetted by the mercury and pitted over its entire surface.

An exploratory test was run using the direct-coupling technique that had been developed for the aqueous dissolution work (7). A strip of Type 316 stainless steel 1-1/2 by 10 by 1/8 inches, attached to the ultrasonic coupler, was immersed to a depth of 1 inch in mercury and flexurally vibrated for 1 hour at a power input to the transducer of 600 watts. Extensive pitting, producing a matte finish, was observed on the immersed surfaces of the steel strip. The average depth of the pits was estimated at approximately 2 mils.

Attention is directed to Section IV, which discusses water-mercury emulsification as an aid to extraction. This work also has implications for the mercury process flow sheet.

FUTURE WORK

Two approaches exist for the ultrasonic disintegration of fuel elements. One involving exposure at or near a vibrating face is most convenient from a process point of view, but introduces the coupler problem. Further efforts will be made to solve this problem.

A second approach, described in previous reports (7), involving direct activation by vibration of the fuel element rather than vibration of the mercury will also be studied. For this purpose, a sacrifice coupler of either mild or stainless steel could be used. The disintegration products of such a coupler will introduce no materials foreign to the process system.

After a suitable approach to the transducer-coupling array has been devised, tests will be made with thorium ingots to determine the extent to which ultrasonic application accelerates dissolution.

SECTION III

ULTRASONIC LEACHING IN AQUEOUS SOLUTIONS

PURPOSE AND SCOPE

After fuel materials are extracted from nitric acid solutions used in fuel-element dissolution, these solutions contain substantial amounts of fission products as well as aluminum. In processes under consideration for radioactive waste storage, such materials are calcinated to a form similar to sand or aluminum oxide. The material must be stored in thin layers with complex circulating systems in order to prevent melting of the alumina by the radioactive heat generated, according to calculations at Idaho Falls (10). Cesium and strontium isotopes are particularly troublesome. During the first few months of storage, these elements contribute only about 10 percent of the heat generated in the waste. But, since the other fission products decay much more rapidly, after the first year the strontium and cesium are responsible for about 90 percent of the heat. If these elements could be removed prior to storage, special cooling and storage techniques might not be required after the first 1-3 years. Furthermore, if substantial amounts of readily leachable fractions of strontium and cesium are removed so they cannot be subsequently leached by weathering, simple open-air or desert burial of the calcined and preleached solids could be accomplished, and material reduction in waste-storage cost could be anticipated.

With present methods (11,12), between 80 and 98 percent of the cesium-137 can be leached out in several stages, using distilled water at 80°C and several days' exposure time. The strontium-90 is more difficult to remove. Using 0.1 N nitric acid solution, only 30-70 percent of this element is removed (11), and use of the acid introduces the danger of dissolving unacceptable amounts of alumina (10). For safe storage, it is considered desirable that 99.9 percent of the strontium be removed.

Application of vibratory energy should facilitate the process by removing larger quantities of fission products and possibly reduce the amount of leaching liquid necessary or shorten the time required. Although little evidence exists of ultrasonic application directly to the leaching problem, significant results have been obtained in related fields. Cellular constituents have been released from a variety of biological organisms (13,14,15,16), proteins have been extracted from yeast (17), bitter materials from hops (18), and oil from peanuts (19) with greater efficiency than can be achieved by non-ultrasonic methods. Diffusion of salts through membranes has been accelerated 70-100 percent (20,21). The removal of sodium thiosulfate from photographic film during washing (22), as well as the rate and quantity of sensitizer-ion

transfer to the surfaces of silver bromide particles in photoemulsions (23), has been increased. Improved degassing of porous surfaces, promotion of wetting, increased diffusion in cracks and crevices, and disruption of diffusion gradient films at the surface of the particles have been postulated as the possible mechanisms.

These experimentally established effects appear to warrant investigation of ultrasonic application to the leaching of solid wastes. This subtask is therefore oriented to determining the feasibility of the process and establishing appropriate ultrasonic conditions for accelerated aqueous leaching to occur.

HIGHLIGHT DEVELOPMENTS

For a preliminary evaluation of ultrasonic leaching, three immediately available types of ultrasonic treatment vessels were considered:

1. A cylinder, resonant at 20 kc, surrounded by an outer jacket to form an annulus of 0.025 inch through which the solid waste and leaching liquid could be passed in continuous flow. This vessel is illustrated in the sketch of Figure 8.

2. A cylindrical vessel one end of which contains an O-ring-sealed ultrasonic coupler bonded to a 20-kc transducer. With a predetermined amount of solid waste in the vessel adjacent to the coupler face, leaching liquid could be continuously flowed through the waste (Figure 9).

3. A 20-kc converging coupler vessel providing high-intensity batch treatment for waste and leach liquid. This vessel can be suitably modified for continuous-flow treatment.

The first vessel as originally constructed proved unsatisfactory because of clogging of the inlet and exit tubes with the solid waste, producing irregularities in the feeding. Suitable modifications are being made to permit testing of this vessel. All tests to date have been made with the second vessel.

The material for the leaching experiments, obtained from Idaho Falls Operations Office, was a simulated aluminum-nitrate type solid waste to which had been added cold cesium and a small amount of cesium-137 as a tracer. The cesium content was not determined exactly but was no higher than 0.21 milligrams per gram. The leaching liquid used for these early tests was 0.01 N nitric acid solution.

For each test, 20 grams of the solid waste were placed in the treatment vessel and a total of 1 liter of the nitric acid solution was flowed through it while the ultrasonic coupler was activated with a power level of 300 watts to the transducer. Samples of 100 milliliters each were taken after the passage of 100, 300, 600, and 1000 milliliters. At the end of the test, the solution remaining in the vessel was drained off and the leached solid was recovered.

For comparison, a non-ultrasonic control run was made. Twenty grams of waste were placed in a flask and leached with ten successive 100-milliliter batches of 0.01 N nitric acid solution, using mechanical stirring and settling for each cycle.

Evaluation of cesium removal from the waste was carried out in the following manner: Each sample of leach liquor was centrifuged to remove suspended solids, evaporated to a small volume in a beaker, then placed on a stainless steel planchet and evaporated to dryness. The planchet was placed in a sample holder and the beta-emission radioactivity was measured on the basis of 0.1 gram of material and adjusted for background count, using a thin-window G-M tube with a utility scaler. The activity of the leached waste was determined in the same manner.

In the control run, the activity decreased from a count of 417 per minute for the first cycle of leach liquor to a count of 95 per minute for the tenth cycle. The recovered solid waste showed an activity of 129 counts per minute for 0.1 gram of material.

An ultrasonic test conducted at a leaching liquid flow rate of 20 milliliters per minute showed decreasing activity from a count of 420 per minute for the first 100 milliliters of leach liquor to 185 per minute for the last. The leached solid waste showed essentially no activity above background count.

In a similar ultrasonic test at a flow rate of 50 milliliters per minute, the activity of the leach liquor decreased from 297 to 96 per minute, and the solid waste showed a count of 103 per minute for 0.1 gram of material.

On the basis of the low activity in the residual solid waste, these preliminary tests indicate a positive ultrasonic effect on leaching on a continuous-flow basis and suggest that the flow rate may be significant in the results obtained.

A secondary and partially undesirable effect of the ultrasonic treatment was the breakup of the calcined waste pellets. Introduction of vibratory energy into the vessel created cavitation and considerable agitation of the material at the coupler face. The reduction in particle size is illustrated in Figure 10. The waste material as received had the rounded, opaque form of Figure 10a. After ultrasonic leaching, the particles were broken into irregular, partially transparent particles (Figure 10b).

FUTURE WORK

Further leaching studies will be made to determine the effects of flow rate, power level, and frequency. In an effort to avoid cavitation and thus particle breakup, lower-power runs will be emphasized. The use of frequencies in the range of 300-800 kc will be investigated, since cavitation thresholds in water are significantly higher at higher frequencies.

Tests will also be made using the other two types of treatment vessels, and the leaching of strontium-bearing waste simulants will be investigated as soon as samples are received.

SECTION IV

HIGH-TEMPERATURE MIXER-SETTLER

(ULTRASONIC EXTRACTION OF LIQUID-METAL SYSTEMS)

PURPOSE AND SCOPE

Certain actual or proposed methods for removing radioactive materials from fuels or breeder blankets involve extraction with molten metals or molten salts. In such processes the theoretical extraction may be high but equilibrium may be attained very slowly; conventional methods for accelerating the process are often impractical because of the inherent corrosive nature of the systems and the radiological hazards involved. The effect of ultrasonic energy in promoting the attainment of high concentration gradients has been suggested as a means for accelerating such extraction.

A process of current interest at Argonne National Laboratory is the extraction of bred plutonium from uranium with molten magnesium (24). The uranium, containing about 1 weight percent plutonium, is alloyed with 5 weight percent chromium to form a mixture that melts at about 860°C. The lower temperature imposes less severe operating conditions on the materials of construction and permits operation without serious vaporization of the magnesium. This alloy is contacted with molten magnesium until a reasonable concentration of plutonium in magnesium has been achieved. The phases are then separated and three more extractions are performed with fresh batches of magnesium. With this process, approximately 85 percent of the plutonium can be transferred to the magnesium phase, from which it is subsequently separated by vacuum distillation.

It has been determined that the distribution coefficient for extraction at 1150°C is approximately 0.2 mole fraction of plutonium in magnesium per mole fraction of plutonium in uranium. Under static conditions, this equilibrium distribution is approached very slowly and the process may require as long as 6 hours per batch of magnesium. Mechanical stirring has been found to reduce the required time to approximately 1/2 hour. However, the true equilibrium distribution is not achieved with any of the successive batches of magnesium. In a typical four-stage run (24) in which extraction times of 1/2 hour were used, the approach to equilibrium was 88 percent for the first stage and decreased through the other three stages to 45, 28, and 22 percent respectively.

It appears that the process could be accomplished more efficiently and rapidly with the use of ultrasonic energy in a suitable mixer-settler vessel, thus reducing the existent inventory of plutonium-contaminated uranium. The projected scale of 10 kilograms per day would be amenable to small-scale countercurrent batch extraction.

Previous experience with extraction in aqueous systems (25) indicated that the percentage approach to equilibrium in a given time could be increased by appropriate ultrasonic application in a mixer-settler vessel. Operating with molten metals obviously introduces new problems that are not present with aqueous systems at room temperature. If such problems can be solved, ultrasonic extraction with the system of interest should be advantageous.

This program will involve the investigation of ultrasonic mixer-settler vessel arrays suitable for use with the molten uranium-magnesium system and the selection of materials of construction resistant to attack by the molten metals under ultrasonic influence. Preliminary experimentation will be made with simulant systems, both at room temperature and at elevated temperature, prior to investigating the uranium-plutonium-magnesium system of interest at Argonne National Laboratory.

HIGHLIGHT DEVELOPMENTS

Preliminary experimentation has been carried out with a simulant system at room temperature to appraise certain design and operating problems prior to introducing the complexities of high-temperature operation. The mercury-water system was selected since these materials have properties approximating those of uranium and magnesium at 800°C (see Table II). The mercury-water system was first treated in an available ultrasonic vessel to determine mixing and settling characteristics. Simulant extractions were then carried out removing copper from mercury with aqueous reagents.

Ultrasonic Emulsification Studies

The experiments were carried out in the 20-kc converging-coupler vessel previously used in the thorium nitrate precipitation studies. Equal quantities by weight of mercury and water were introduced and ultrasonically treated for varying periods of time. The mixtures were kept cool by circulation of cold water through a steel coil immersed in the vessel. As soon as the vibratory energy was shut off, a sample of the aqueous phase was rapidly removed with a pipette and placed in a graduated tube to permit the dispersed mercury to settle. From determinations of the respective volumes of mercury and water in the sample, the weight distribution of the emulsified portion could be calculated.

The experimental results, listed in Table III, indicate a maximum of 38 weight percent of mercury dispersed in the water after 3 minutes of ultrasonic treatment. However, since settling was rapid, it is possible that a considerable amount of mercury dropped out during the brief

interval required to withdraw the sample, and the percentage of dispersed mercury may have been much higher. In all cases the dispersion was fine, which is desirable in that it provides a large surface area for mass transfer to take place. The emulsions were generally unstable, although a small amount of mercury (estimated at 0.1 percent by weight) remained suspended for as long as two days after treatment. This so-called stable dispersion could be broken by centrifugation.

Ultrasonic Extraction of Copper from Mercury

To establish the efficacy of this ultrasonic system in extracting a simulant solute from mercury into the aqueous phase, a mercury-copper alloy was prepared. Two types of extractant were used. One was a dilute (0.5 N or 1 N) nitric acid solution. The other extractant was 3 N ammonium hydroxide. After extraction, the nitric acid solution was made ammoniacal with ammonium hydroxide. The final concentration of copper in the extractant could then be determined by colorimetric evaluation of the copper-ammonia complex, using a Klett-Summerson colorimeter.

Ultrasonic tests were made in each of which a single batch of copper-mercury complex weighing approximately 200 grams was contacted with successive equal portions of aqueous extractant for periods of time ranging from 3 to 5 minutes. In an equivalent non-ultrasonic test, the two phases were mixed with a reciprocating stirrer for periods ranging from 3 to 12 minutes.

The results, summarized in Table IV, indicate that with ultrasonic treatment the transfer of copper to the aqueous phase was improved by a factor of about 15 over that obtained with mechanical stirring. It should be emphasized, however, that these tests do not actually simulate conditions existent in the extraction of plutonium from uranium by magnesium, since the extraction of copper from mercury by nitric acid or ammonium hydroxide solution is accompanied by chemical reactions.

Consideration of Materials of Construction for Molten-Metal System

In connection with other ultrasonic applications (see Section II of this report), it has been noted that selection of construction materials for equipment to be used with molten metals is critical because of possible erosion and dissolution which may not only reduce the efficiency of energy transmission but also contaminate the melt. Consideration was therefore given to potentially suitable materials for the molten uranium-magnesium system.

During prior studies at Argonne National Laboratory (24), only three materials were found to resist attack by this system under non-ultrasonic conditions; these were tantalum, graphite, and alumina. Tantalum is the best, but the other two materials will be considered. The behavior of

tantalum as an ultrasonic transmitter has been evaluated both theoretically and experimentally (26) and appears promising. However, the ultrasonically accelerated erosion reported for many materials in molten metals (discussed in Section II of this report) suggests that this aspect be investigated more extensively.

It should be noted that cavitation erosion of the tantalum, if it should occur with long-term exposure to the molten system, may not be an unsurmountable problem. If it can be established that good transfer of plutonium from uranium to magnesium can be achieved with brief periods of ultrasonic application, contamination of the system with tantalum for a particular batch may be negligible. In the event of continued erosion of the container and coupler with repetitive treatments, these units could be designed as expendable items.

A quantity of uranium-5% chromium alloy has been ordered for tests with tantalum.

FUTURE WORK

A tantalum ultrasonic coupler will be obtained and tests conducted to determine its compatibility with molten uranium-chromium alloy, molten magnesium, and a mixture of the two systems.

SECTION V

ULTRASONIC COALESCENCE OF PARTICULATE MATTER IN GAS STREAMS

PURPOSE AND SCOPE

Certain techniques in the processing of reactor materials are accompanied by the escape in off-gas streams of radioactive particles so fine that they cannot be collected by ordinary methods. Collection by cyclones, scrubbers, or filters would be possible if the particle size could be increased in some manner.

One such problem occurs in the calcining of waste products, wherein radioactive ruthenium is given off at temperatures above 250°C as a volatile oxidation product, presumably in the form of ruthenium tetraoxide. On cooling to below 250°C, this material condenses into submicron particles of a non-volatile lower-oxidation product, which are suspended in a gas that is largely air with oxides of nitrogen and water vapor. After passing through a cyclone, the radioactive dust concentration is of the order of 0.002-0.04 grams per cubic foot (0.07-1.4 milligrams per liter). It is desired that this concentration be reduced by a factor of 10^5.

A similar problem exists in the effluent from fluidized beds which have been proposed by Argonne National Laboratory both for feed-material preparation reactions and for fuel-element reprocessing. Many of the reactions carried out on these fluidized beds involve corrosive reagents or products, and dust collection therefrom is difficult to handle by filtration.

It appears that the agglomeration induced in an ultrasonic standing-wave field can be used to coalesce these particles to a size amenable to practical collection techniques. Such agglomeration has been discussed extensively in the literature. The principle was first enunciated in 1866 by Kundt (27), who generated standing sound waves in a gas within a glass tube containing dust particles and observed that collections of dust formed along the bottom of the tube at regularly spaced intervals corresponding to half wavelengths. This phenomenon was reproduced by Koenig in 1891 (28), and by Robinson in 1913 (29). During the past 30 years, extensive investigations of particle agglomeration in standing-wave systems in gases have been carried out (30,31,32,33,34,35,36). Particle size ranges include the 0.1-10 micron region, which encompasses such aerosols as magnesium oxide and zinc oxide smoke, ammonium chloride fumes, tobacco smoke, sulfuric acid and paraffin fogs, and carbon black. Sonic and ultrasonic agglomeration has been demonstrated with all these materials and with others.

Most of the above work involved the use of laboratory-type equipment of various types and of limited size. More recently high-powered siren generators have been used for certain industrial applications on a limited scale, but their economic feasibility has not yet been established. Hueter and Bolt (37) conclude that "Although powerful commercial sirens have been available for some time, the practical significance of sonic aerosol processing is still somewhat controversial. . . the complexity of variables, such as temperature, moisture, grain /dust/ loading, particle-size distribution, volume flow, has so far restricted the applications to the pilot stage."

The present program involves a study of the feasibility of using ultrasonic vibration to produce agglomeration of submicron ruthenium oxide particles in off-gas streams. The relationship of the aforementioned variables will be investigated and controlled. Earlier work with liquid coalescence conducted in this laboratory (25,38) has indicated the importance of minimizing circulatory effects, and the concept of an annular standing wave apparatus (38) will be extended to gaseous systems. It has also been proposed that an additive aerosol of surfactant be used to counteract the low dust loading in the ruthenium oxide case; such additive materials not only will increase the number of particles present and thus provide more points for agglomeration to occur but also, with appropriate surface-active characteristics, will improve adhesion between particles.

Initial work will involve the production of test aerosols of aqueous liquids in the 1-5 micron size range and the development of suitable apparatus for agglomeration of such particles. Successful accomplishment of this objective will be followed by experimentation with the more difficult submicron size range. Finally, equipment oriented specifically to the ruthenium oxide problem will be assembled and tested.

HIGHLIGHT DEVELOPMENTS

Study of Mechanism of Ultrasonic Agglomeration

Consideration of the fairly extensive theoretical, experimental, and patent background in the light of the basic physics of vibratory agglomeration phenomena is underway. Authorities are being contacted and possibly will be consulted as the work progresses.

The phenomena of agglomeration in gaseous and in liquid media involve many problems which are similar. The various forces acting on the particles in the standing wave causing the particles to be driven into contact have been considered in detail by Brandt, Freund, and Hiedemann (39), by St. Clair et al. (33), and summarized by Hueter and Bolt (40).

In their analysis of the theory, Greene and Lane (41) reached the conclusion that "A completely satisfactory theory of acoustic coagulation, which will account quantitatively for the manner in which vibration and acoustic forces increase the rate of collision of particles, has yet to be developed."

Briefly, the following forces appear to be involved in sonic agglomeration:

1. So-called orthokinetic forces which govern the relative velocity of the particles of various sizes and of the fluid medium, and which depend upon particle density and size, frequency of vibration, and the viscosity of the medium.

2. Radiation pressure forces, dependent upon particle size, wavelength, and energy density, which force the particles into close proximity in certain phases of the waves.

3. Bernoulli forces arising from hydrodynamic flow around the particles, through which particles are attracted to each other as a function of particle density and size and the density of the medium, and inversely as the fourth power of the distance between the particles.

4. Miscellaneous other effects, such as Brownian motion, which is significant with particles of submicron size; Oseen forces resulting from distortion of waves when harmonics are present; and average Stokes forces resulting from local variations in fluid viscosity caused by thermal gradients in the sound wave.

The orthokinetic coagulation theory is weak in that the centerlines of particles of different sizes vibrating at different amplitudes must be parallel to the direction of vibration of the medium in order for particles to contact; moreover, the range of effectiveness is only as great as the relatively small amplitude of the vibration. A reasonable rate of acceleration of flocculation could not be expected from such a mechanism (42). Radiation pressure forces do not appear to account for the rates of coagulation observed, and hydrodynamic forces can not explain the initial stages of flocculation because of the high-order inverse dependence of the forces on distance between particles in the initial aerosol.

While all of these mechanisms may be active, it appears that theoretical examination of the phenomena, either from a new point of view or from a combined mechanistic approach, should point the way to improvement in practical coagulation and should permit the use of lower-intensity and more economic sources, such as whistles or electromagnetic sonic generators, in substitution for the expensive and extremely complex maintenance problems of rapidly moving parts in ultrasonic sirens. Such highly complex equipment will be entirely unsuitable for corrosive and radioactive atmospheres.

Use of Additive Aerosol

In the cases of interest to the Atomic Energy Commission, particularly with the submicron ruthenium oxide particles suspended at low density (in the range of milligrams per cubic foot), it has been decided to investigate marked increase in particle loading by the addition of a liquid mist of a suitable inert material.

Foreign materials have been added to aerosols and the influence on coagulation rates evaluated; these materials are generally vapors of various types (42). It has been reported (35) that the addition of a "water fog" to the sonic agglomeration of carbon black resulted in a greater-than-fourfold increase in collection. Similarly a threefold increase in particle size was achieved by the addition of moisture to sulfuric acid mist.

More recently, Elton (43) and Pilpel (44) have employed appropriate surface-active agents to promote the coalescence of fog droplets through the modification of electric charges on the surface of the particles.

Combining this concept with ultrasonic coagulation seems to be a worthwhile and novel approach, since ultrasonic forces can only bring particles into contact with one another but apparently cannot cause the particles to adhere. It is therefore suggested that ultrasonic coagulation and collection efficiency will be improved by the addition to the low-concentration ruthenium oxide aerosol of a liquid mist of appropriate wetting agent, oil, or solution that will promote the coalescence and adhesion of the particles after the ultrasonic vibrations have forced them together.

While the orthokinetic theory of Brandt, Freund, and Hiedemann (39) does not explain coagulation completely, it may serve as a guide to choice of the appropriate additive aerosol properties. If the total concentration is raised to the point where the submicron ruthenium oxide particles, through their individual vibratory motion, will contact additive aerosol particles with augmented frequency, this controllable mechanism could possibly be made to predominate.

Examination of the vibratory frequency-vs.-particle size plot of Brandt et al. (39) reveals the following information on particle motion:

Frequency (kc)	Particle Size for Negligible Motion (μ)	Particle Size for Maximum Motion (μ)
1	10	3
5	5	1
20	2	0.5
50	1.5	0.3
100	1	0.2

Thus in a 20-kc sound field, all particles of 0.5 microns and smaller will follow the vibration of the gas, while all particles larger than about 2 microns will be relatively stationary. Hence additive aerosols need to be larger than 2 microns to maximize relative motion and smaller than 10 microns (45) to minimize fallout before sonic agglomeration can occur.

Using the work of Elton (43) as a guide, it has been decided to use a non-ionic surfactant as the additive aerosol. Since such materials are relatively viscous liquids, the need exists for efficient production of an additive aerosol of this material in the 2-10 micron size range. Aeroprojects' background in liquid atomization work for the Government (46) and subsequent commercial activities have resulted in the development of devices and techniques capable of producing over 95% of the particles in this specific size range (45). A typical aerosol produced with such a nozzle, using a non-ionic surfactant, polyoxyethylene sorbitan monolaurate,* was found to contain particles in the following size ranges:

% of Mass Atomized	Dispersed in Particles Smaller Than
65	8 μ
45	5 μ
35	4 μ

The desired size range for the additive aerosol is therefore readily achievable using the Aeroprojects ultrafine atomization nozzle.

Consideration of Ultrasonic Sources

The generation of sound waves in a gaseous medium is circumscribed by both frequency and intensity considerations (47). Practical use of airborne sound is limited to the lower frequencies because of increased absorption at high frequencies. It has been computed (48) that the intensity in air at 10 kc decreases to half its original value in 722 feet, while at 100 kc this decrease is effected in 7.22 feet, and at 1 mc in 0.072 feet. Thus frequency in the range of about 100 kc appears to be a practical upper limit for convenient-sized apparatus. Intensity is limited because a gas cannot support tension, and the fluctuating sound pressure cannot exceed ambient pressure; the upper limit is indicated to be about 1000 watts per square centimeter or 192 decibels (with reference to 10^{-16} w/cm^2) at the sound source. Neumann and Norton (35) conclude that "The threshold value causing noticeable agglomeration in a reasonable time is approximately 140 db."

* "Tween 20," Atlas Powder Co., Wilmington, Delaware.

Among the potential low-frequency sources, the usual electrically or electronically driven devices would have low efficiency because of impedance mismatch between a vibrating solid surface and the gas in which the vibratory energy is desired, although one apparently successful device for aerosol agglomeration consisted of a solid aluminum-alloy piston freely suspended at a nodal point and excited to vibration (10-20 kc) by an electromagnetic system (49); intensities of the order of 160 decibels were reported. Since it appears that a cylindrical configuration may ultimately be desired for the ruthenium oxide collection problem, there is possibility of using a radially vibrating cylinder made of magnesium (to minimize interfacial energy losses) enameled with a thin corrosion-resistant coating. Such a cylindrical radiator within a cylindrical treatment vessel would lead to a series of annular standing waves through which the aerosol-laden gas could be transported in an axial direction.

However, present knowledge appears to favor a gas-driven device for generating acoustic energy directly in the gaseous medium. Use has been made of sonic and ultrasonic sirens for this purpose (34,50). Another possibility exists with ultrasonic whistles such as the Hartmann air-jet generator (51,52), and the jet-edge devices (53), in which intensive sound is produced in air by the impingement of a high-velocity air jet on a resonant cavity. Several such whistles can be combined to provide more powerful equipment for industrial purposes (54,55). Jet-edge-resonator whistles have been reported to have very high outputs and were highly favored by the Penn State research group (53). Whistles are of simple construction with no moving parts and are highly suitable for the desired cylindrical geometry in corrosive atmospheres. Short distances between radiator and reflector could also possibly permit operation at materially higher frequencies.

Standing waves can be produced with suitable reflecting surfaces. Figure 11 illustrates a non-stable pattern of standing waves produced some years ago at Aeroprojects (56), in an aerosol of ice crystals; in this instance, the walls of the cold cabinet served as a reflector.

The whistles and sirens have certain disadvantages for this application, but these are not insurmountable. The wave pattern produced by all existing devices is not a cylindrical shell such as would be desirable. Difficulty also arises from the injected air used to operate these devices; enormous quantities are required for sirens. Such air will induce undesirable circulation effects, for previous experimentation with agglomeration in emulsions (25,38) established that phases are effectively separated when the wave pattern is sharp with minimum circulation in the fluid medium. However, it is possible that the air stream can be shielded from the standing-wave field by an acoustically transparent window.

Because of very extensive previous experience with whistles, the immediate availability of several such devices, and the ease of modification, this ultrasonic source is being used for initial work, aimed primarily toward exploring standing-wave patterns and determining the experimental parameters associated with coalescence of airborne particles.

Experimentation with Standing Waves

These range-finding studies were carried out with two types of available Hartmann whistles. One was a strut type in which the nozzle and resonant cavity were supported by an external frame. This had been designed to operate at a frequency of 27 kc and produced an energy output of 138 decibels* (ref. 10^{-18} w/cm^2) 1/2 inch from the source. After dimensional modification in accordance with developments by Palmé (57), the frequency dropped to 22.5 kc and intensity increased to 154 decibels (approximately 0.4 w/cm^2).

The second unit was a pin whistle, in which nozzle and resonator are connected by a central pin. The elimination of the external struts has the advantage of providing an uninterrupted sound field around the whistle. This unit operated at a frequency of 24 kc with an output of 138 decibels* 1/2 inch from the source.

These two units were tested with various types of reflectors and confining enclosures, as described below.

The presence of standing waves in the sound field of the whistles was determined by exploration of intensity variations with a precision probe-type microphone (58), by visualization of aerosols of solid or liquid particles suspended in the sound field, and by the pattern of fallout of such aerosols. The aerosols were produced from lycopodium powder, from a non-evaporating glycerine-water-blue dye solution, and from smoke from a smudge pot. Although no particle-size measurements were made in this preliminary work, it was estimated from aerosol stability that the glycerine-water-blue dye aerosol particles were in the range of 2-5 microns.

Exploratory work was directed toward the familiarization of laboratory personnel with the problems at hand through the development of standing-wave patterns and the coalescence of standard aerosols. Two basic arrays were used. In the first attempts to set up annular standing waves about the axially mounted pin whistle, an 8-inch-diameter reflector was used and lycopodium aerosol was introduced to establish the presence of standing waves. A concentric ring fallout pattern was observed at the base of the reflector but the air flow from the whistle prevented the observation of stratification of solids in the annular air space.

Plane standing waves were established in transparent 4-inch-diameter tubing about 4 feet long through locating the whistle at the focus of the parabolic reflector and directing the acoustic beam into the tube. The tube was closed at the end opposite the whistle with a plane reflector. While it was not possible to observe standing-wave agglomeration in air, the fallout pattern of smoke from the smudge pot (Figure 12) and blue dye-glycerine aerosol (Figure 13b) was established. Significant fallout patterns from the standing waves appear only on the bottom of the tube, indicating coalescence in the air above these points and fallout onto these portions of the surface.

* Approximately 0.007 w/cm^2.

For an evaluation of the extent of acoustic agglomeration which was actually occurring within the air and producing the banded fallout, semi-quantitative tests were made using the blue dye-glycerine aerosol dispersed in the test cell. The setup was essentially the same as before except that a blower was used to draw the aerosol into and through the tube (Figure 13a). Tests were made with the whistle operating at relatively low intensity and tuned to its resonant frequency and also with the whistle inoperative (detuned). The aerosol deposited on the walls of the tube in each test was washed out with water and the density of the washings evaluated colorimetrically. Approximately twice as much aerosol had agglomerated and fallen out in the ultrasonic tests run at relatively low intensity as in the non-ultrasonic tests.

One difficult factor in the above tests is the circulation in the confined area induced by the air used to drive the whistle. (As noted above, circulation had also inhibited the formation of stratification on standing waves in liquids.) A method was therefore devised for shielding the standing-wave field from this air stream. A thin Mylar membrane has been found in other work with liquids (59) to be acoustically transparent; theoretical considerations (60) indicated that it should be of the order of one-hundredth of the wave length in air or less to permit adequate transmission. At 24 kc, the wave length in air is approximately 0.5 inch. On this basis, 0.00025-inch Mylar was selected and placed over the opening of the tube adjacent to the whistle. Probe microphone measurements in the tube with and without the membrane and also using a 16-mesh screen to support the membrane gave no indication of reduction in the intensity of 154 decibels measured in the 4-inch tube 7 inches from the source. Minor losses occurred with a 0.00017-inch aluminum foil, which yielded readings of 151 decibels.

Using this air shield, many of the foregoing experiments were repeated with considerably greater success; not only were standing-wave formations produced in the fallout on the bottom of the tube, but also steady planes of coalescing aerosol were for the first time partially visible within the air confined in the tube. These were difficult to photograph, but are faintly evident at the arrow indications in Figure 14.

FUTURE WORK

Additional work projected for the immediate future will encompass the following:

1. Study of the propagation of intense sound through films of different materials and thicknesses, with the view to selecting a more corrosion-resistant shield for the generating air stream.

2. Investigation of other potential sources for generating airborne sound aimed at more practical, generally cylindrical geometries of higher intensities.

3. Study of frequency-particle size-intensity relationships in aerosol agglomeration.

4. Study of the influence of additive aerosols made from non-ionic surfactants.

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Figure 1

SUSPENSION OF THORIUM OXALATE PARTICLES PRECIPITATED WITHOUT ULTRASONICS
(Slow addition of oxalic acid solution)
Magnification: 1120X

Figure 2

SUSPENSION OF THORIUM OXALATE PARTICLES
PRODUCED BY ULTRASONIC TREATMENT DURING PRECIPITATION
(Fast addition of solid oxalic acid)
Magnification: 1120X

Figure 3

DRIED THORIUM OXALATE PARTICLES
PRODUCED WITH ULTRASONIC TREATMENT DURING PRECIPITATION
Magnification: 1120X

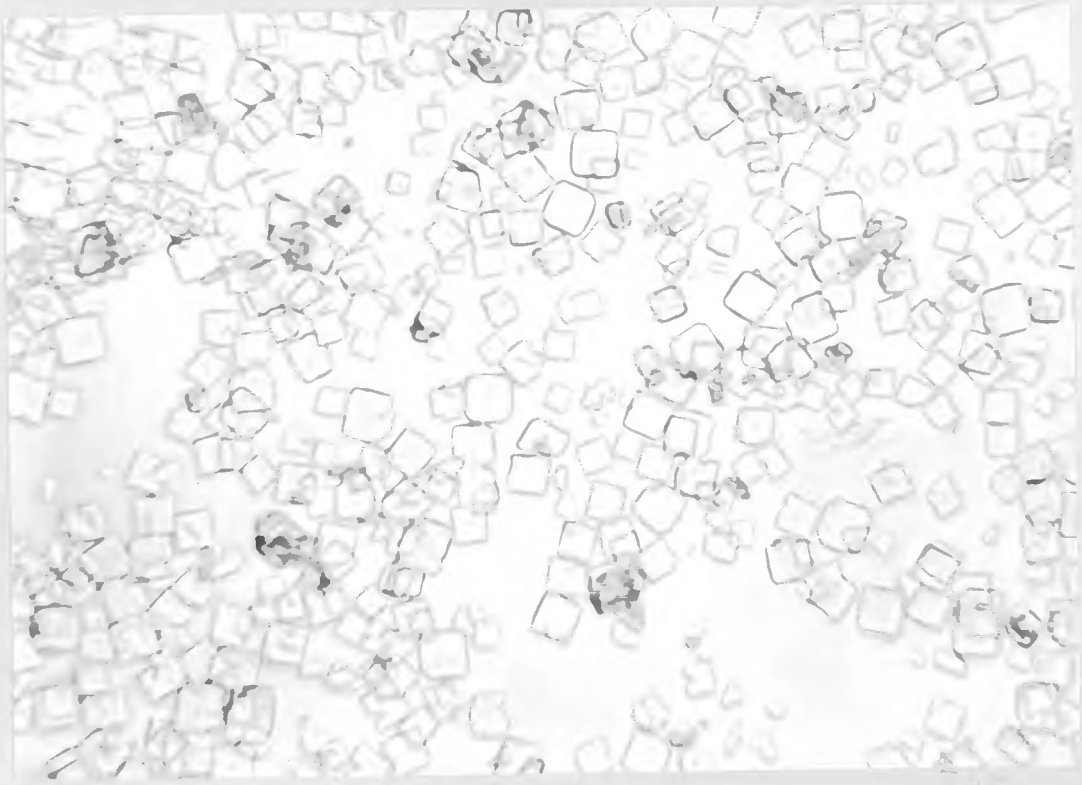


Figure 4

SUSPENSION OF THORIUM OXALATE PARTICLES
PREPARED BY ADDITION OF MIXED DRIED SALTS
TO WATER WITH APPLIED ULTRASONIC ENERGY
Magnification: 1120X

a. Face of Coupler

b. Side of Coupler

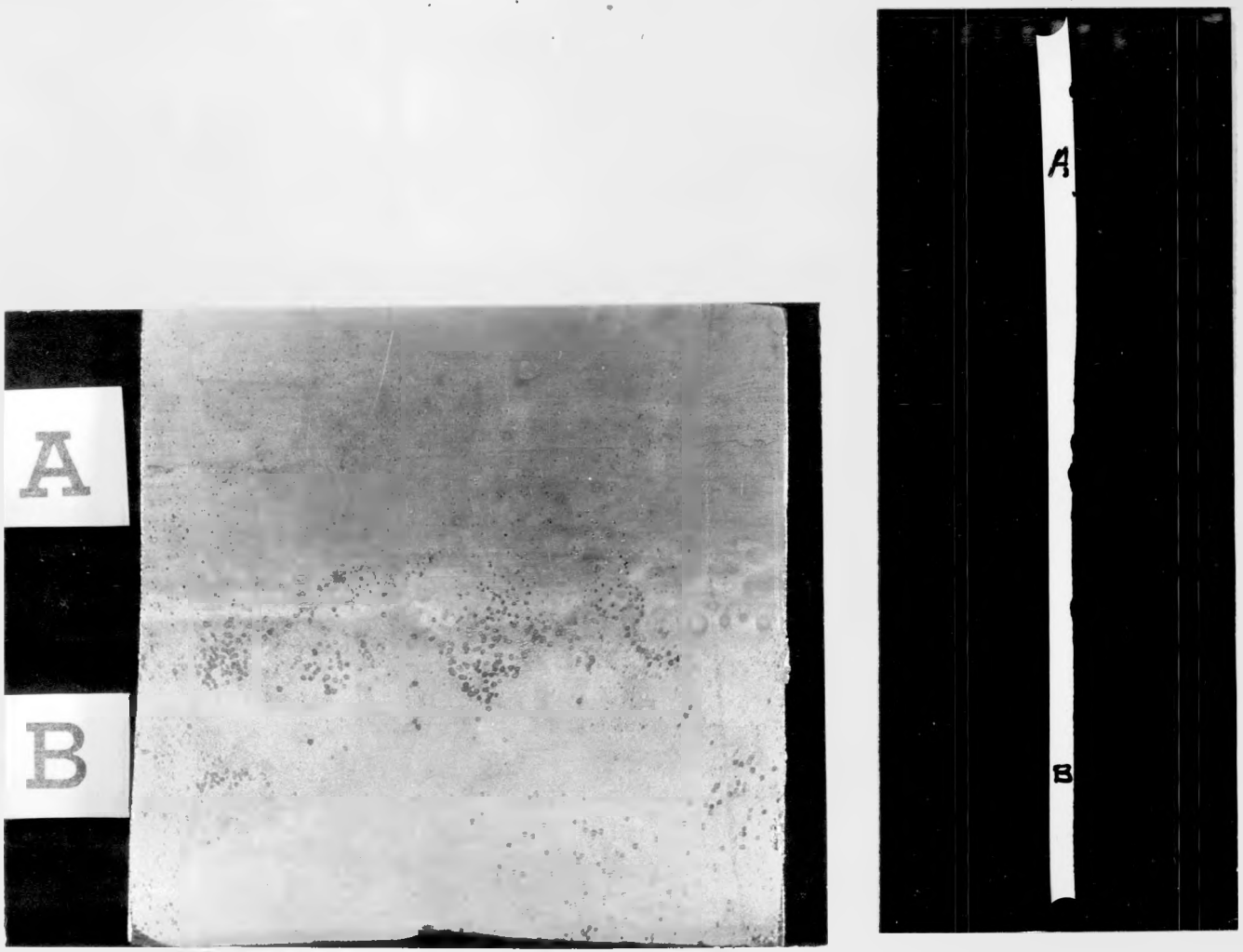
Figure 5

HASTELLOY C COUPLER AFTER 60 MINUTES
OF ULTRASONIC EXPOSURE IN LIQUID MERCURY
(22X)

Figure 6

FACE OF MOLYBDENUM COUPLER
AFTER 60 MINUTES OF ULTRASONIC EXPOSURE
IN LIQUID MERCURY

(22X)



a. Interior View

b. Edge View

Figure 7

SECTION OF TYPE 302 STAINLESS STEEL BEAKER
USED TO CONTAIN MERCURY IN ULTRASONIC EXPERIMENTS

Total Exposure: Approximately 16 hours

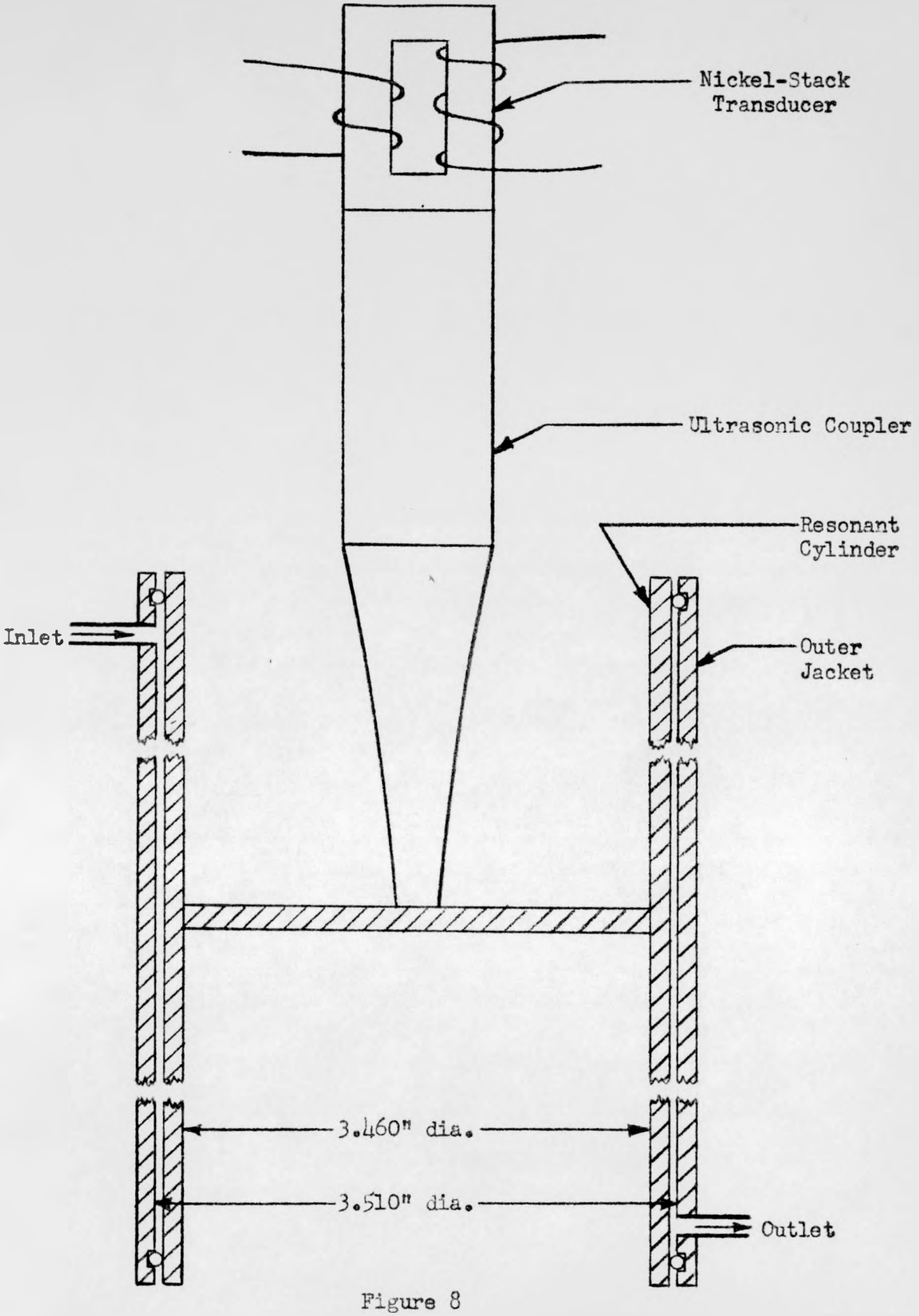


Figure 8

ULTRASONIC RESONANT-CYLINDER VESSEL
USED FOR EXPERIMENTAL LEACHING OF SOLID WASTES
(Vessel Type No. 1)

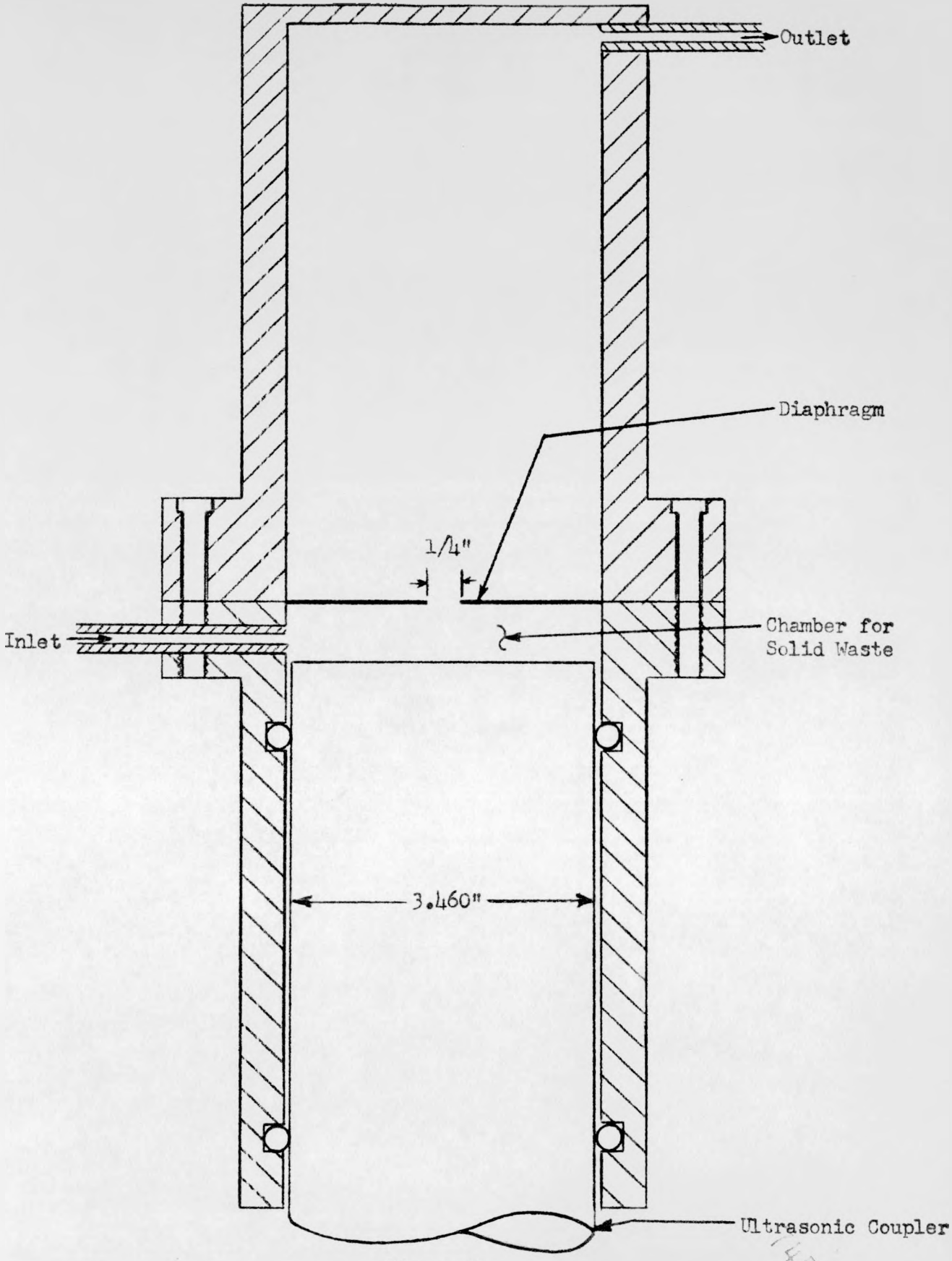
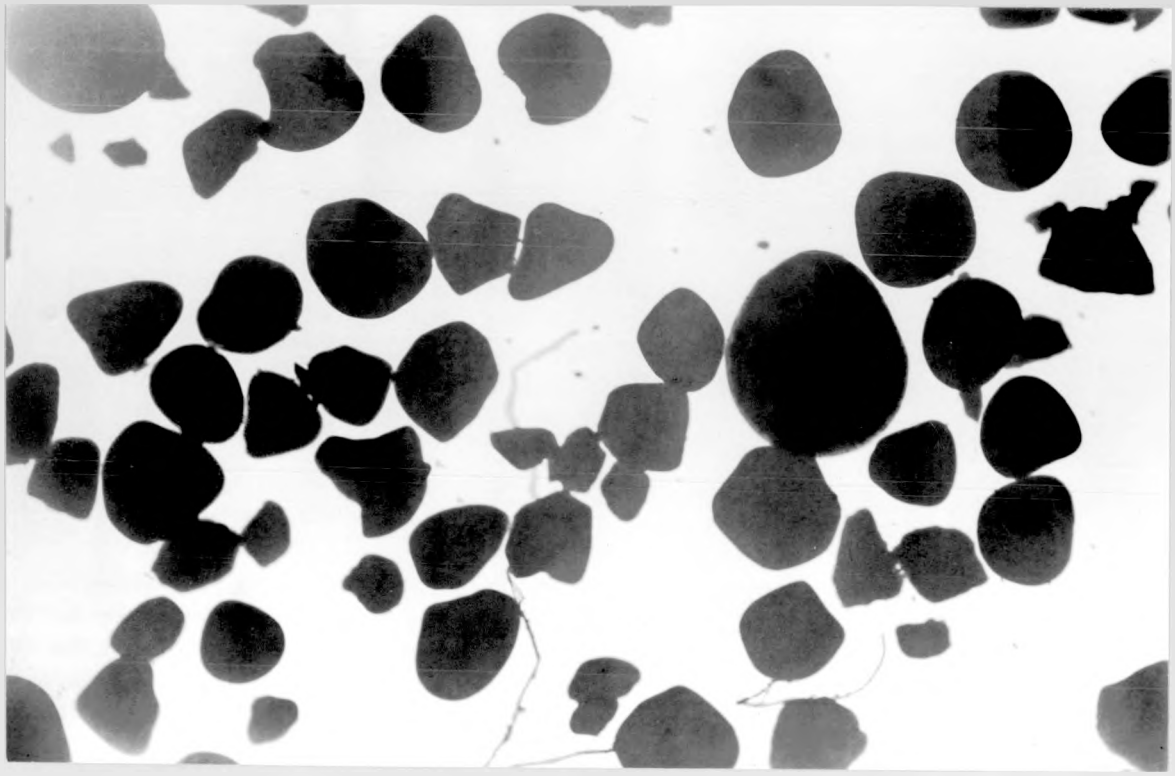
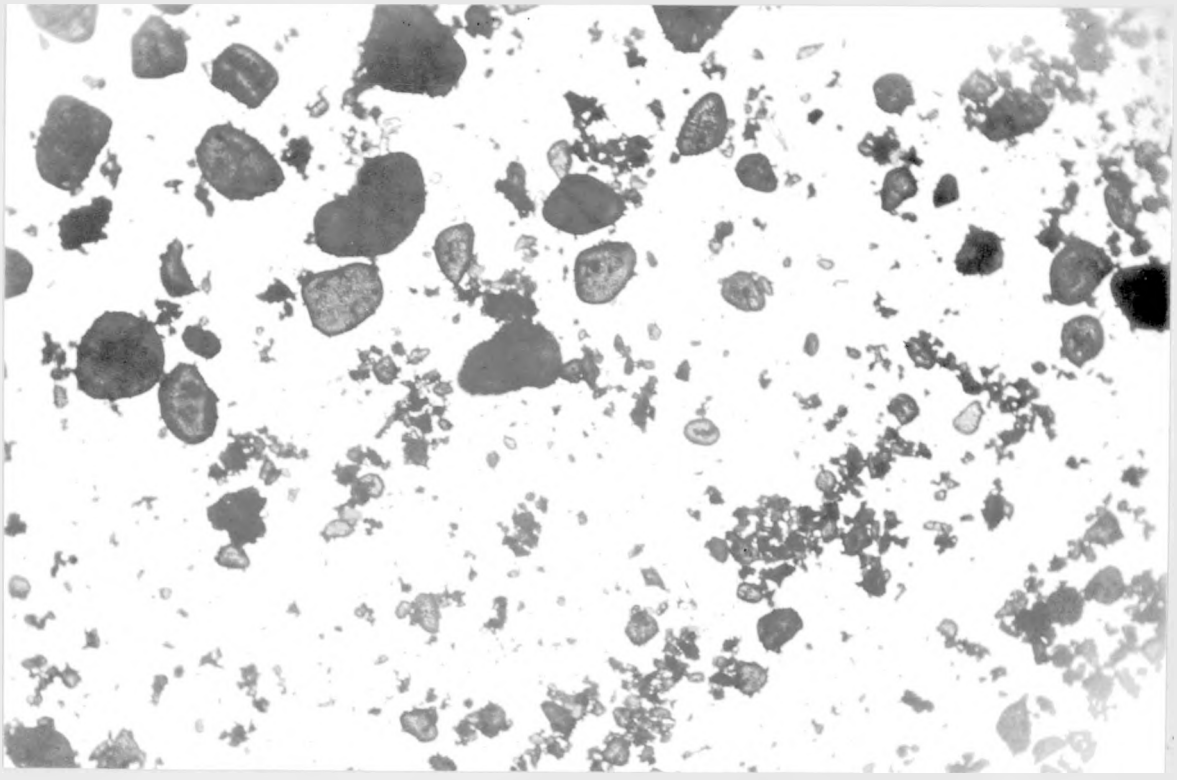


Figure 9

ULTRASONIC FIXED-BED CYLINDRICAL VESSEL
USED FOR EXPERIMENTAL LEACHING OF SOLID WASTES
(Vessel Type No. 2)



a. Before Leaching



b. After Ultrasonic Leaching

Figure 10

SIMULATED ALUMINUM NITRATE TYPE SOLID WASTE
USED IN ULTRASONIC LEACHING EXPERIMENTS
(44X)

Figure 11

NON-STABLE STANDING-WAVE PATTERN
DEVELOPED IN ICE CRYSTALS IN COLD BOX
USING HARTMANN WHISTLE

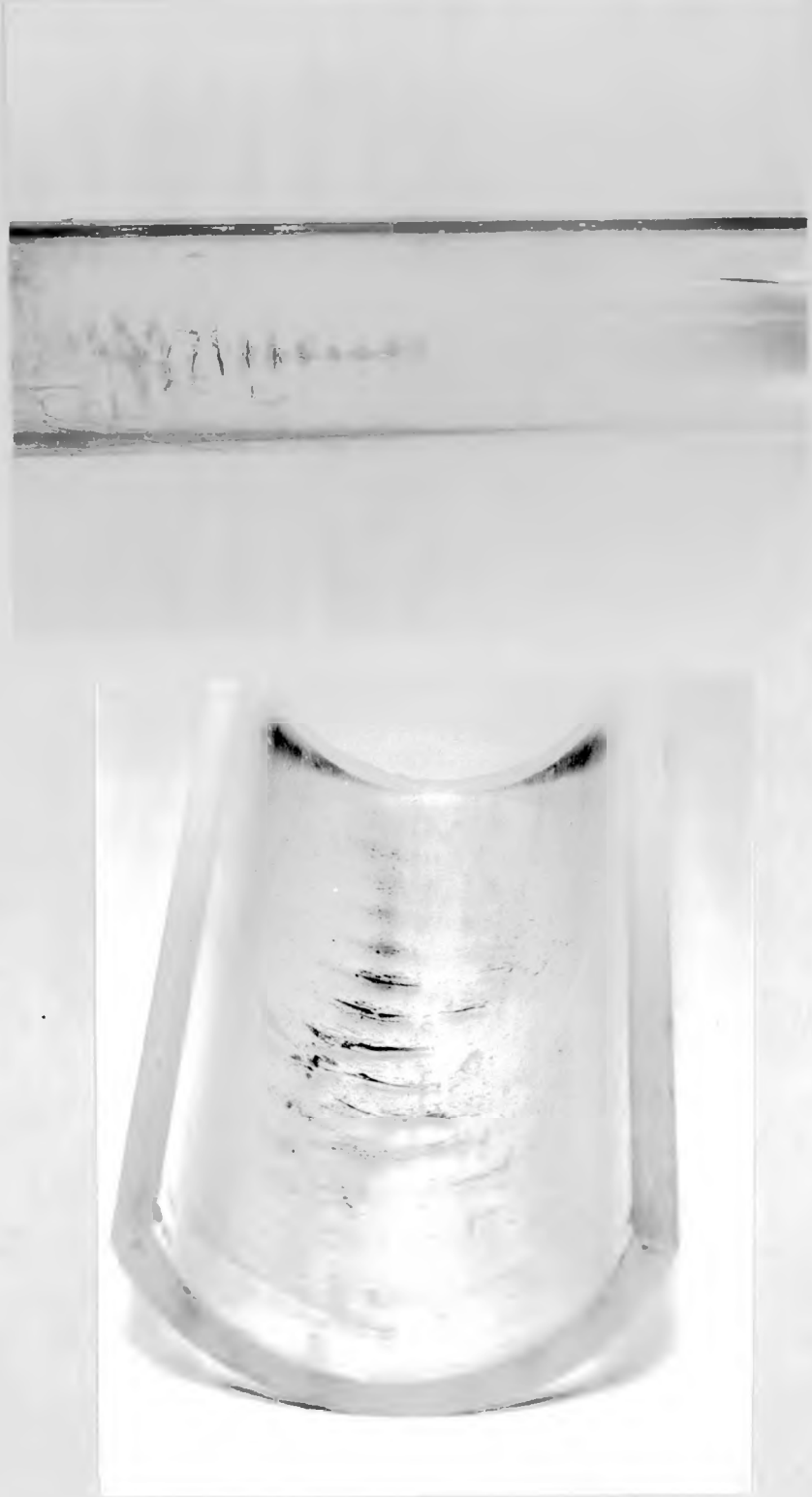


Figure 12

FALLOUT PATTERNS FROM SMOKE AEROSOLS
AGGLOMERATED WITH HARTMANN WHISTLE

Figure 13a

COLLECTION TUBE AND RECYCLE BLOWER
MOUNTED THROUGH WALLS OF TEST CELL
TO EVALUATE ULTRASONIC EFFECT ON AGGLOMERATION

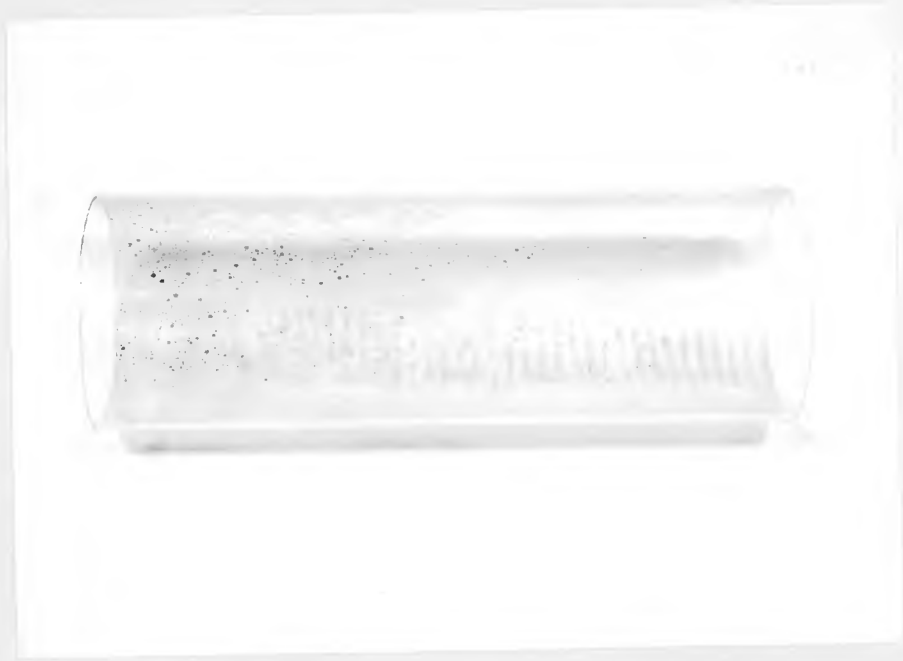


Figure 13b

TYPICAL STANDING-WAVE FALLOUT PATTERN IN TUBE
PRODUCED WITH ARRAY OF FIGURE 13a

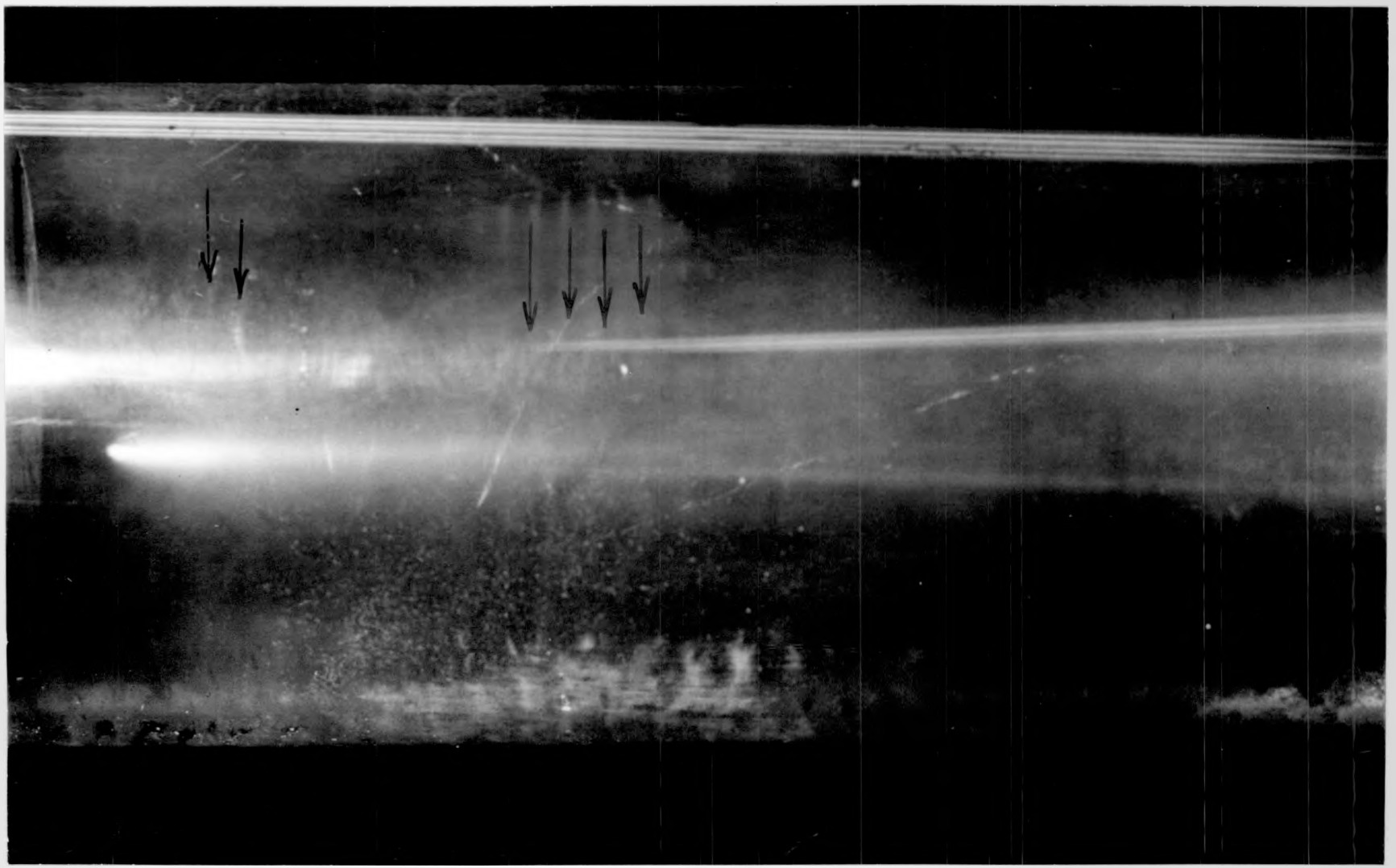


Figure 14

COALESCENCE OF AEROSOL IN STANDING-WAVE PLANES
PRODUCED WITH CONFINED AREA SHIELDED FROM WHISTLE AIR FLOW

(Oblique observation from right of photograph in regions indicated by arrows shows faint coalescence bands in air)

Table I

BEHAVIOR OF COUPLER MATERIALS IN LIQUID MERCURY
ULTRASONICALLY ACTIVATED AT 20 KC

Coupler Material	Treatment Time (min)	Temperature (°C) Start	End	Ultrasonic Power into Transducer (watts)	Estimated Acoustic Transmission Factor* (%)	Estimated Power at Coupler Face (watts)	Coupler Face Area (cm^2)	Estimated Unit Power at Coupler Face (w/cm^2)	Coupler Condition After Test	Remarks
Hastelloy C	50	21	149	400	88	350	25.6	13.6	Wetted by mercury, matte finish	---
Hastelloy C	60	30	178	1000	88	880	25.6	34.4	Severely pitted, see Fig. 5	---
Mild Steel	120	26	116	1000	88	880	25.6	34.4	Severely pitted	Ultrasonics pulsed
Mild Steel	60	18	32	900	88	790	25.6	30.8	Severely pitted	Coupler and mercury water-cooled
Graphite	60	14	24	1000	21	210	20.4	10.3	Severely eroded	Ultrasonics pulsed, mercury water-cooled
304 Stainless Steel	120	27	141	750	88	660	25.6	25.8	Severely pitted	Ultrasonics pulsed
Molybdenum	60	27	59	950	60	570	7.1		Severely eroded and pitted	Ultrasonics pulsed
Molybdenum	60	27	47	300	60	180	7.1		Severely pitted, see Fig. 6	Ultrasonics pulsed

* Theoretically calculated maximum percent of applied energy deliverable to mercury.

Table II

COMPARISON OF PROPERTIES OF MERCURY-WATER SYSTEM
WITH THOSE OF MOLTEN URANIUM-MAGNESIUM SYSTEM

Material	Temperature (°C)	Viscosity (cp)	Density** (g/cc)	Surface Tension (dyne/cm)
Mercury	25	1.5	13.5	460
Water	25	1.0	1.0	80
Uranium	900	1-2*	18.0	300-500*
Magnesium	900	1.2	1.35	520

* Estimated values
**Density Hg/Density H_2O = 13.5
Density U/Density Mg = 13.4

Table III

ULTRASONIC DISPERSION OF MERCURY IN WATER

Charge: Equal parts by weight of mercury and water
Power Level: 2000 watts to transducer

Test No.	Treatment Time (min)	Volume % Mercury in Water	Weight % Mercury in Water*
1	1	0.5	7
2	2	1	13
3	3	4	38
4	4	4	38

* Calculated from volume measurements.
The theoretical maximum mercury content is 50 weight percent or approximately 7 volume percent.

Table IV

EXPERIMENTS IN EXTRACTION OF COPPER FROM MERCURY

Test No.	Weight of Mercury* (gram)	Aqueous Phase	Weight Ratio Hg: Aqueous	Method of Extraction	Extraction Time (min)	Weight of Copper Extracted (mg)	Remarks
1a	202	0.5N HNO_3	1.1	Ultrasonic	3	4.7	Extraction Stage 1
1b	202	0.5N HNO_3	1.1	Ultrasonic	3	1.0	Extraction Stage 2
1c	202	0.5N HNO_3	1.1	Ultrasonic	3	Nil	Extraction Stage 3
2a	202	3N NH_4OH	1.26	Ultrasonic	3	4.7	Extraction Stage 1
2b	202	3N NH_4OH	1.26	Ultrasonic	3	Nil	Extraction Stage 2
3a	202	3N NH_4OH	1.26	Ultrasonic	5	4.2	Extraction Stage 1
3b	202	1N HNO_3	1.26	Ultrasonic	5	Nil	Extraction Stage 2
4	202	3N NH_4OH	1.26	Mechanical Stirring	3	0.32	
5	202	3N NH_4OH	1.26	Mechanical Stirring	9	5.5	
6	202	3N NH_4OH	1.26	Mechanical Stirring	12	5.5	

* Mercury-copper complex