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ULTRASONICALLY PRODUCED DISPERSIONS OF THORIUM BISMUTHIDE IN MOLTEN BISMUTH

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ABSTRACT

Ultrasonic energy was applied to the production of finely dispersed thorium bismuthide of potential use in the blanket preparation for the LMFR. A controlled-atmosphere ultrasonic-treatment furnace was assembled for operation at temperatures up to 1200°C, taking advantage of previously developed principles of ultrasonic furnace construction.

Using 5% thorium-95% bismuth systems, it was found that (1) when ultrasonic vibration was applied during cooling from complete solution temperature (1050°C) to 450°C, vibratory energy suppressed the normal precipitation of 1-centimeter-long thorium bismuthide platelets and produced more nearly equiaxed particles of a size generally less than 50 microns in diameter. (2) When applied to melts containing large platelets formed by cooling under the above conditions without applied ultrasonics, vibratory energy applied isothermally at 450°-500°C fractured the platelets into generally equiaxed particles smaller than 100 microns. Increased treatment time under these conditions appeared to increase the effectiveness of particle breakup.

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I. INTRODUCTION

A proposed breeder blanket for the Liquid Metal Fuel Reactor (LMFR) consists of a slurry of equiaxed thorium bismuthide particles in bismuth which is circulated around the core of the reactor (1)*; the most recent information indicates that this intermetallic corresponds to the compound $ThBi_2$ (2). The contemplated process for removal of bred uranium and protactinium from the slurry involves transferring a portion of the bred products from the solid $ThBi_2$ particles, in which they are primarily generated, into solution in the liquid bismuth. The liquid is subsequently separated from the solids and treated to remove the bred products (3, 4).

The Metallurgical Division of Brookhaven National Laboratory has considered at least two methods for achieving transfer of this bred uranium from the thorium bismuthide particles to liquid bismuth solution for subsequent processing, one involving complete solution of the thorium bismuthide in the molten bismuth, and the other involving thermal pulsing of the slurry. It appeared that both of these methods could be facilitated by appropriate ultrasonic application.

In the first method, the bred slurry is heated to a temperature at which complete solution of the thorium bismuthide will occur. For a thorium-bismuth mixture containing 5 weight percent thorium, the temperature should exceed 950°C (4). Cooling to 450°-500°C results in the precipitation of about 99 percent of the thorium as the bismuthide, uranium being retained in the liquid bismuth phase. The thorium bismuthide is subsequently filtered, reconstituted with liquid bismuth, and fed back into the process loop. During cooling, the solid $ThBi_2$ precipitates from solution in the form of platelets whose size varies with thorium content and with the rate of cooling from the temperature of complete solution through the temperature range within which most of the $ThBi_2$ precipitates from solution. Platelets up to 1 centimeter in diameter are produced in alloys containing 5-10 weight percent thorium cooled at moderate rates of the order of 30°C per minute. Increased cooling rate decreases the platelet size, and in order to precipitate platelets less than 50 microns in diameter, cooling rates of the order of 1000°C per second are necessary. Such rapid cooling would be difficult to achieve on a large scale; even if achieved, the resulting fine-platelet dispersion is highly viscous and must be subjected to subsequent heat-treatment to obtain the equiaxed particle shape necessary for high fluidity. If the solution were cooled in the normal manner, it is evident that the large platelets fed back into the process loop would increase pumping loads and initiate plugging (5).

* Numbers in parentheses indicate references in Bibliography, pp. 18-20.

The second method under consideration involves thermal pulsing of the bred slurry. If a bismuth-5 percent thorium slurry containing bred uranium is heated to 875°C, approximately 60 percent of the solids will dissolve. In this process, the slurry is held at 875° for a short time to permit the bred products to obtain an equilibrium distribution between the liquid and the undissolved solids; during this treatment some of the uranium is transferred to the liquid phase. If subsequent cooling is accompanied by agitation, platelet formation is prevented. However, a principal objection to this method is the eventual formation of large $ThBi_2$ particles which, even though equiaxed, may be difficult to maintain in suspension. The method must therefore be supplemented by some treatment to reduce the particle size.

Either of these processes might be more practical if the indicated problems could be solved. In the complete solution method, some means is required either to prevent large platelet formation during precipitation or to reduce the size of preformed platelets, so that the thorium bismuthide, when reconstituted with bismuth, may be reintroduced without difficulty into the breeder loop. The second process could be effectively converted to an isothermal treatment similar to leaching if it were possible to reduce the size of the equiaxed thorium bismuthide particles, thus creating more surface area and accelerating mass transfer of uranium to the liquid phase.

Based on reports for analogous systems, it would be expected that vibratory energy, properly applied, might accomplish the desired results by any of the following methods: (a) prevention of platelet formation by promoting bulk nucleation during cooling from complete solution temperature, (b) isothermal fracture of preformed platelets, or (c) isothermal reduction in size of equiaxed crystals.

Bulk nucleation by ultrasonic application has been repeatedly demonstrated on a laboratory basis, particularly in the grain refinement of such metals as aluminum alloys (6, 7, 8, 9), zinc (10, 11, 12), and copper (13). Furthermore, ultrasonic treatment has effectively reduced the size of various types of particles suspended in liquid media, such as silver bromide in aqueous gelatin solution (14), barium sulphate in water (15), and metal foils suspended in aqueous solutions (16). A particularly effective means of fine-particle production is to conduct precipitation reactions in an ultrasonic field, as exemplified by work at Aeroprojects with silver salts (14) and calcium oxalate (17). Recent work in this laboratory has demonstrated significant size reduction of unground uranium dioxide in sodium-potassium alloy without detectable coupler erosion (18).

Other experimental work has established ultrasonic dispersion of both solid and liquid metals within molten phases. Thus iron from a crucible or ultrasonic coupler has been dispersed in molten zinc (11, 19); lead and tin have been dispersed in zinc, the lead particles becoming

smaller with extended treatment (20); ultrasonic dispersions of zirconium in molten magnesium (19), of iron, lead, or titanium in molten aluminum (6, 21, 22), and of molybdenum in molten beryllium (13) have been demonstrated. Such ultrasonic effects suggested the possibility of utilizing similar methods to achieve the desired fine thorium bismuthide suspensions.

The above examples represent only a few of the many experimental efforts in applying ultrasonic energy to molten metal systems, which have been accomplished generally on a laboratory scale. However, such application in the continuous processing of substantial quantities of molten metal in the LMFR blanket system would seem reasonable only if it were possible to integrate furnace design with appropriate means for introducing substantial amounts of vibratory energy into a melt under vacuum or controlled-atmosphere conditions. The development of hermetically sealable pressure- and temperature-insensitive mounts for ultrasonic systems, together with experimental background in the design of transducer-couplings for use in unusual environments (6, 13), has clearly indicated the possibility of large-scale, controlled-atmosphere treatment. Critical appraisal of the ultrasonic, metallurgical, and chemical engineering requirements in our laboratory have indicated sufficient promise of an ultimate solution to justify the experimental feasibility program.

Utilizing this background, the experimental program was oriented to establishing the feasibility of accomplishing three specific end results:

1. The prevention of thorium bismuthide platelet formation by applying ultrasonic energy to a thorium-bismuth system during cooling from the solution temperature to 450°C.

2. The fracture of preformed thorium bismuthide platelets in molten bismuth by ultrasonic application at 400-500°C in order to achieve particle sizes no larger than 50-100 microns.

3. Reduction in the size of equiaxed thorium bismuthide crystals by isothermal ultrasonic treatment at 450°-500°C.

II. EXPERIMENTAL EQUIPMENT, MATERIALS, AND PROCEDURES

Ultrasonic Transducer-Coupling System

As noted above, one of the major problems in the assembly of an ultrasonic furnace for molten metal treatment is the development of a suitable transducer-coupling system for introducing vibratory energy into the melt at high temperatures and under controlled atmosphere conditions.

It has been generally agreed (23) that the vibratory frequency is of minor importance in achieving positive results in liquid metals and that intensity is a controlling parameter. Therefore, because of the several advantages of operating at frequencies in the lower ultrasonic range, the transducer-coupling was designed to incorporate a magnetostrictive nickel-stack transducer having a nominal frequency of 20 kc.

More careful consideration must be given to the coupler member used to transmit energy from the transducer into the melt. The requirements for satisfactory ultrasonic coupler materials for use in molten metal systems have been established in previous studies (24). Specifically, the coupler material must not seriously attenuate vibratory energy within the range of temperatures incident to the melt, must provide good impedance matching into the melt, must be susceptible to wetting by the molten metal without serious erosion of the coupler surface and without introducing undesirable contaminants into the melt, must have a melting point substantially above the maximum operating temperature, and must have a low heat conductivity to permit adequate cooling of the transducer end while the opposite end is maintained at the necessary elevated temperature.

Stainless steel has been found to provide satisfactory ultrasonic transmission into certain other molten metal systems. Its melting point is high and its thermal conductivity suitably low. Empirical evaluation of its behavior at elevated temperatures indicates fairly uniform acoustical transmission characteristics at temperatures up to 1000°C (6). Calculations indicate good impedance matching between stainless steel and molten bismuth, the interfacial reflection losses between these two media being only 15-20 percent, and approximately zero loss is obtained across a bond between a nickel-stack transducer and a stainless steel coupler.

No information was available concerning the degree of attack of molten bismuth on various types of ultrasonically activated stainless steel. Both iron and chromium are known to show good resistance to this metal under non-ultrasonic conditions at temperatures up to at least 600°C, while nickel is readily attacked even at 300°C (25). It therefore appeared advisable to use a stainless steel with high chromium and low nickel content, although its behavior under ultrasonic conditions

could not be predicted. Certain materials that are normally unaffected or only mildly attacked by molten metals demonstrate more or less rapid erosion under the same conditions when activated by ultrasonic energy (6, 11, 26).

The transducer-coupling eventually used consisted of a cylindrical coupler rod fabricated of Type 410 stainless steel bonded to a 20-kc nickel-stack transducer.

Ultrasonic Attenuation in Molten Bismuth

Further consideration in the design of a suitable ultrasonic furnace must be given to the characteristics of elastic energy transmission within the molten metal system itself.

High-intensity ultrasonic treatment of liquids is known to induce cavitation in the liquid, and this bubble formation and collapse is most violent adjacent the interface between the transmitting member and the liquid (27). Prior investigations (6) concerned with ultrasonic transmission into molten aluminum, tin, and zinc have shown that attenuation of vibratory energy within the cavitation zone is substantial, while attenuation beyond this zone is low, suggesting that high ultrasonic intensities can not be transmitted into a large bulk of the liquid metal.

Brief investigations were therefore made to determine the energy-transmission characteristics into molten bismuth, using the equipment array of Figure 1. Vibratory energy at a frequency of 20 kc was introduced through a horizontal coupler into the bismuth, which was maintained in the molten state (292°-298°C) by means of Bunsen burners. To insure effective energy transfer from the solid coupler to the melt, the coupler was prewetted with flux in molten bismuth. For measurements of relative energy levels, a high-temperature, probe-type microphone was inserted in the molten bismuth at varying distances from the coupler face. The immersed end of the microphone was also prewetted, using flux and 50-50 lead-tin solder, which alloys readily with bismuth at the temperatures used. Signals from the microphone were indicated on a vacuum tube voltmeter.

These relative energy measurements, obtained at input powers of 10, 20, and 30 watts to the transducer, are plotted as a function of coupler-to-microphone distance in Figure 2. The curve obtained from an earlier test with molten aluminum (6) is included for reference. The curves all show the same general configuration, i.e., attenuation is very high in the cavitation zone immediately adjacent to the coupler face, and maximum intensity was observed in the first 1/4-1/2 inch from the coupler. At distances beyond about 1 inch from the coupler, the energy level was relatively low.

On the basis of these data, it appeared that the ultrasonic furnace design should provide for the treatment of thin layers of the molten bismuth to insure maximum effects on the thorium bismuthide crystals.

Ultrasonic High-Temperature, Controlled-Atmosphere Furnace

In addition to the considerations discussed above, the chemical reactivity of thorium bismuthide and the necessity for operating at temperatures up to about 1000°C dictated the construction of an ultrasonic furnace array in which the ultrasonically active zone could be maintained in an inert atmosphere.

The apparatus devised for the treatment of thorium-bismuth slurries is illustrated in Figures 3 and 4. Vibratory energy was introduced through the transducer-coupling system into the molten metal container, which was sealed in a furnace tube to permit controlled-atmosphere operation. The melt container (C of Figure 3) was a recess 1-3/4 inches in diameter and 1 inch deep hollowed out of the free end of the coupler (E), thus providing the necessary shallow treatment zone. The 20-kc nickel-stack transducer (L) was driven by a 2-kilowatt electronic generator from which the transducer would accept approximately 1450 watts. All power measurements were made in terms of electrical watts to the transducer as indicated by a wattmeter with useful performance in the range of 0-200 kilocycles per second.

The transducer-coupling was mounted on a drill-press table which could be conveniently tilted for pouring the melt into a mold after treatment. The means of support was an all-metal pressure- and temperature-insensitive mount (K) sealed into the vacuum-atmosphere furnace wall, so that high-intensity vibration could be delivered through this wall without significant energy loss. The attached metal bellows (J) permitted positioning of the assembly within the furnace tube.

Flanged furnace tubes (B), made of either porcelain or stainless steel, could be placed over the coupler assembly and sealed to the bellows flange ring using an appropriate gasket. Both the flange and the joint between transducer and coupler were water-cooled (F). Provisions were incorporated for vacuum and inert-gas lines (H), and temperature control was accomplished with a chromel-alumel thermocouple (D) and controller. Heat was supplied to the upper end of the assembly by an electric resistance furnace (A) insulated with refractory brick. A rack and pinion permitted the furnace to be raised or lowered around the treatment array.

Experimental Materials

The thorium used in this investigation was electrolytic material supplied by Brookhaven National Laboratory. The bismuth was obtained in the form of C. P. Grade lump material. Argon for inert blanketing of the melts was used directly from cylinders without purification.

Except for the master melt described below, each of the melts consisted of 150 grams of bismuth and 8 grams of thorium, which provided a mixture containing 5 percent thorium by weight.

Control melts containing platelet formations were prepared by heating the mixture to approximately 1050°C for complete solution, soaking for a period of 1 hour under argon, then cooling to solidification of the bismuth in approximately 1 hour to permit the precipitation of the large thorium bismuthide platelets.

For the production of equiaxed thorium bismuthide crystals, a master dispersion of equiaxed crystals was prepared by the technique of Bryner et al. (5) by heating 750 grams of bismuth and 45 grams of thorium in a graphite crucible to 500°C under argon and soaking for 4 hours at this temperature, after which it was cooled and solidified.

Test Procedures

For tests to explore the effect of ultrasonic application on bulk nucleation (prevention of platelet formation), the charge of thorium and bismuth was placed in the molten metal container of the furnace array and the system evacuated. With the temperature on the controller set for 1050°C, the system was allowed to heat; as a temperature of between 400° and 500°C was reached, the system was backfilled with argon and retained under a slight positive argon pressure for the remainder of the run. After a 1-hour soak at 1050°C, the power to the furnace was cut off and ultrasonic energy at a power level of 1000 watts was applied in pulses of 30 seconds on and 30 seconds off for a total treatment time of approximately 1 hour while the melt cooled to approximately 500°C.

Experiments to investigate the ultrasonic fracture of preformed platelets were initiated by soaking the mixture of thorium and bismuth at approximately 1050°C for 1 hour as described above and allowing the system to cool over a period of about 1 hour to 450°-500°C, during which precipitation of large thorium bismuthide platelets occurred. The temperature was maintained at this level while ultrasonic energy was applied in pulses of 30 seconds on and 30 seconds off for varying periods of time. The first group of tests involved 30-minute ultrasonic treatment at 850 watts power. Subsequently, to investigate the effect of treatment time and power level, runs were made at an ultrasonic power level of 1400-1500 watts, with the pulsed energy being applied for respective treatment times of 10, 15, and 20 minutes.

In tests to determine the ultrasonic effect on equiaxed thorium bismuthide crystals, the requisite amount of the master equiaxed melt described above (prepared with a 4-hour soak period at 500°C) was placed in the ultrasonic treatment vessel, heated to approximately 500°C under an argon atmosphere, and pulsed ultrasonic energy at 1200 watts power was applied for varying lengths of time.

In each of the above tests, as soon as ultrasonic treatment had been discontinued, the molten charge was poured into a thick-walled graphite mold, and solidification was essentially instantaneous. The ingots thus produced were cylindrical in shape, approximately 1 inch in diameter and 1 inch long.

Evaluation Procedures

The solidified ingots were sectioned axially by cutting under kerosene, and the sections were prepared for metallographic study by the usual grinding and polishing methods. Exposure to air resulted in oxidation and darkening of the thorium bismuthide particles, so that no etching procedures were required.

Each specimen was examined microscopically, representative areas were photographed, and average particle sizes were estimated. No effort was made to establish particle-size distributions, since the primary intent was to determine only whether size reduction had been produced by the ultrasonic treatment. Typical samples were sent to the Metallurgical Division of Brookhaven National Laboratory.

III. RESULTS AND DISCUSSION

Prevention of Platelet Formation

The typical thorium bismuthide platelets formed during normal cooling from the temperature of complete solution (1050°C) are illustrated in Figure 5. Figure 6 illustrates the effect of pulsed vibratory energy at a power level of 1000 watts applied during cooling from approximately 1050°C to 500°C. The normal mode of crystallization has obviously been altered and no platelets are visible in either melt. The largest individual thorium bismuthide particles are estimated to have a maximum dimension of approximately 100 microns, and most of the particles are no larger than 25-50 microns. The photomicrographs at the highest magnification (500X) reveal the regular shape and rounded edges of the small particles, suggesting bulk nucleation rather than fracture of large particles.

Fracture of Preformed Platelets

Figure 7 illustrates the effect of vibratory energy applied isothermally at a temperature of approximately 500°C after the melt has been cooled normally from solution temperature and the usual thorium bismuthide platelets have been formed. In this case, the pulsed vibratory energy was applied for 30 minutes at a power level of 850 watts. Comparison with the control melt in Figure 5 at the same magnification illustrates the significant particle fracture that has occurred. Many of the particles are not as small as those produced by the bulk nucleation technique, although there is an abundance of very fine particles.

The effect of treatment time at the higher power level of 1400-1500 watts is illustrated in the photomicrographs in Figure 8. As before, these melts were cooled normally to about 500°C and the pulsed ultrasonic energy was applied isothermally for the indicated periods of time. These specimens again show significant platelet fracture, even with only 10-minute ultrasonic treatment time. The progressive size reduction with the treatment time increased to 15 and 20 minutes is evident. These fractured particles have an irregular shape that contrasts with the uniform shape of the particles obtained with ultrasonic treatment during cooling from solution temperature.

The effect of increased intensity of ultrasonic application is evident from a comparison of the ingots of Figures 7 and 8. More effective particle breakup appeared to be obtained with 10-minute treatment at high power (1400 watts) than with 30-minute treatment at low power (850 watts).

Size Reduction of Equiaxed Crystals

Figure 9 contrasts the master melt containing equiaxed particles with a portion of this melt that was ultrasonically treated at a power level of 1200 watts for a 5-minute period at 450°C.

The non-ultrasonically treated control melt shows agglomerates of small $ThBi_2$ particles in addition to larger equiaxed particles. The short ultrasonic treatment resulted in dispersion of many of the agglomerates and produced a melt which appears to have more clearly defined equiaxed particles and fewer large agglomerates.

Ultrasonic Coupler Erosion

Some of the ultrasonically treated melts exhibited inclusions such as the white areas in Figure 9. Although the source of these inclusions was not established, it is believed that they were introduced either from the coupler during ultrasonic treatment or formed during air exposure in the chill-casting procedure.

Experience with ultrasonic application to other molten metal systems has shown that such particles may be eroded from the ultrasonically activated coupler face in contact with liquid metal. However, the appearance of these inclusions was only sporadic, and the coupler itself exhibited little evidence of pitting or erosion, even after several hours of intermittent ultrasonic exposure. Further investigation is necessary to establish whether Type 410 stainless steel is a suitable material for treating thorium bismuthide slurries or whether other candidate refractory materials may be more satisfactory.

IV. CONCLUSIONS

These experiments in the ultrasonic treatment of liquid bismuth-5% thorium mixtures produced the following results:

1. When applied during cooling from complete solution temperature to 450°-500°C, vibratory energy suppressed the normal platelet precipitation of thorium bismuthide and produced much smaller, more nearly equiaxed particles of a size generally less than 50 microns in diameter.

2. When applied to melts containing large platelets formed by normal cooling from solution temperature, ultrasonic treatment at 450°-500°C fractured the platelets into generally equiaxed particles smaller than 100 microns; increased treatment time appeared to increase the effectiveness of particle breakup.

3. Ultrasonic energy applied to slurries containing equiaxed thorium bismuthide particles disrupted particle agglomerates, produced a more uniform dispersion, and may have effected a minor reduction in particle size.

4. The ultrasonic melts contained slight evidence of contamination perhaps from the Type 410 stainless steel coupler, but the source of the contamination was not established in this study.

It should be emphasized that this investigation was merely an exploratory study to determine feasibility, and insuffficient data are presently available for valid extrapolation of the results to estimate power requirements or equipment scale-up for plant operation.

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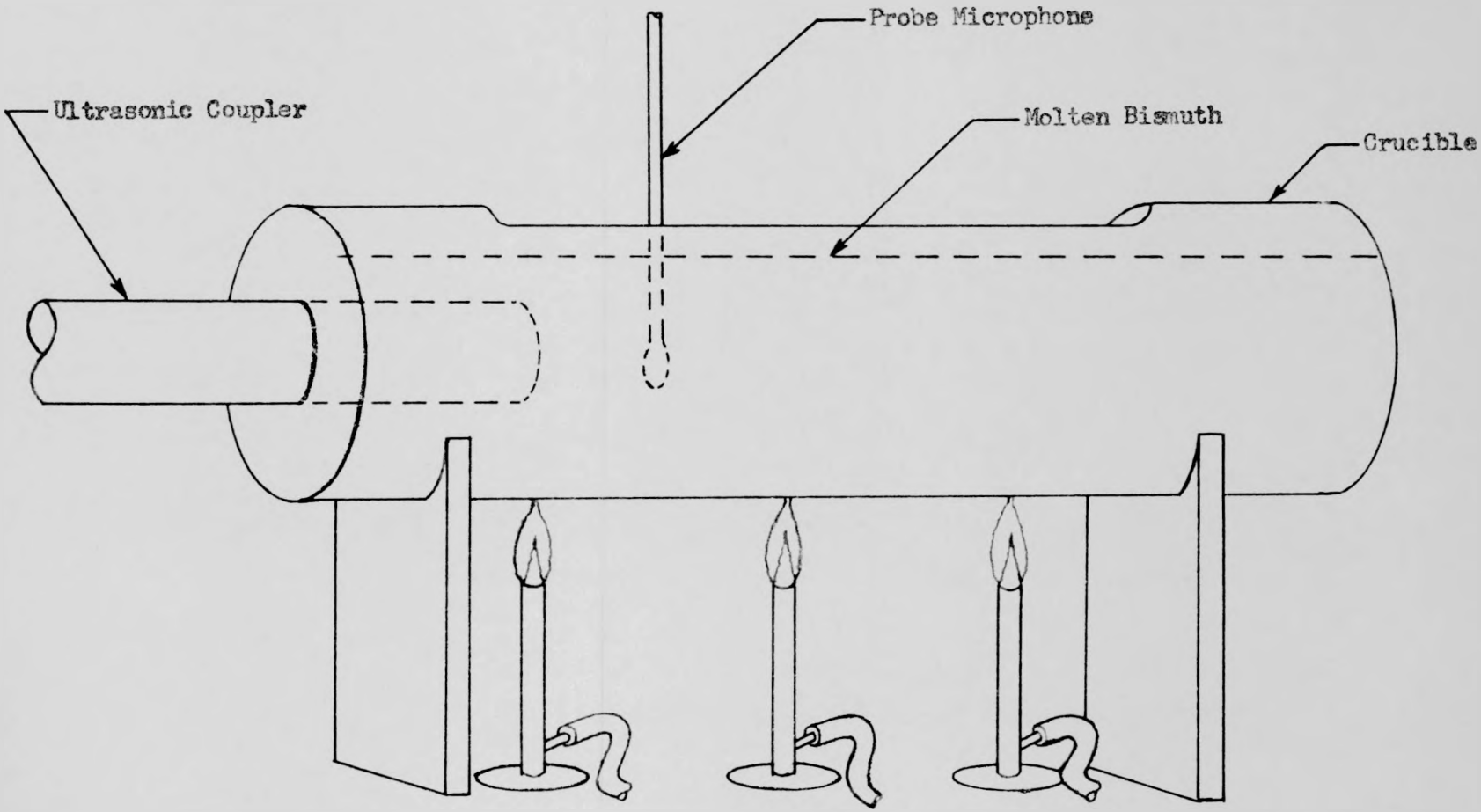


Figure 1

TEST SETUP FOR MEASURING ACOUSTIC ATTENUATION
IN MOLTEN BISMUTH

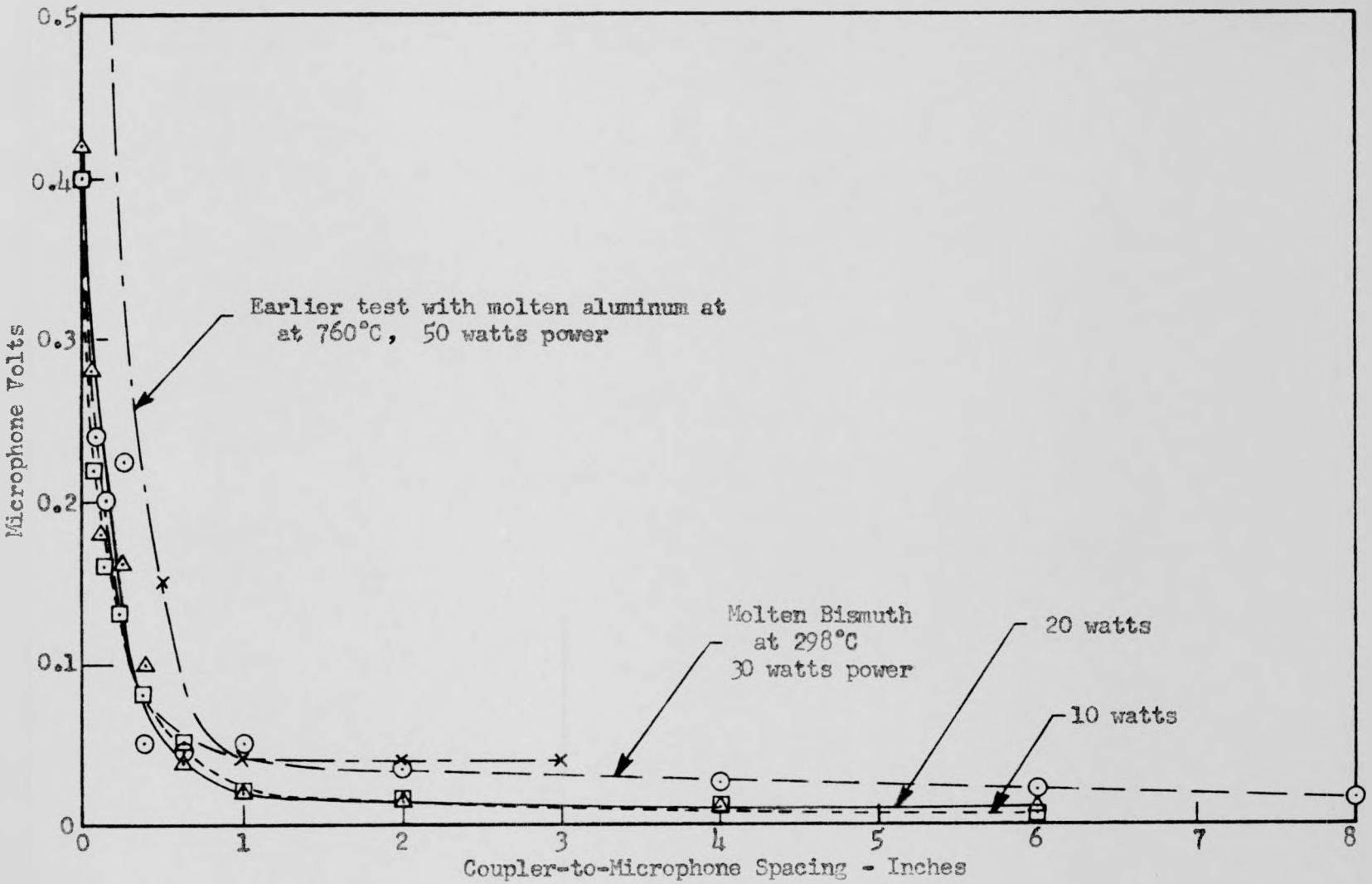


Figure 2

ULTRASONIC ATTENUATION IN MOLTEN BISMUTH
COMPARED WITH EARLIER DATA FOR MOLTEN ALUMINUM

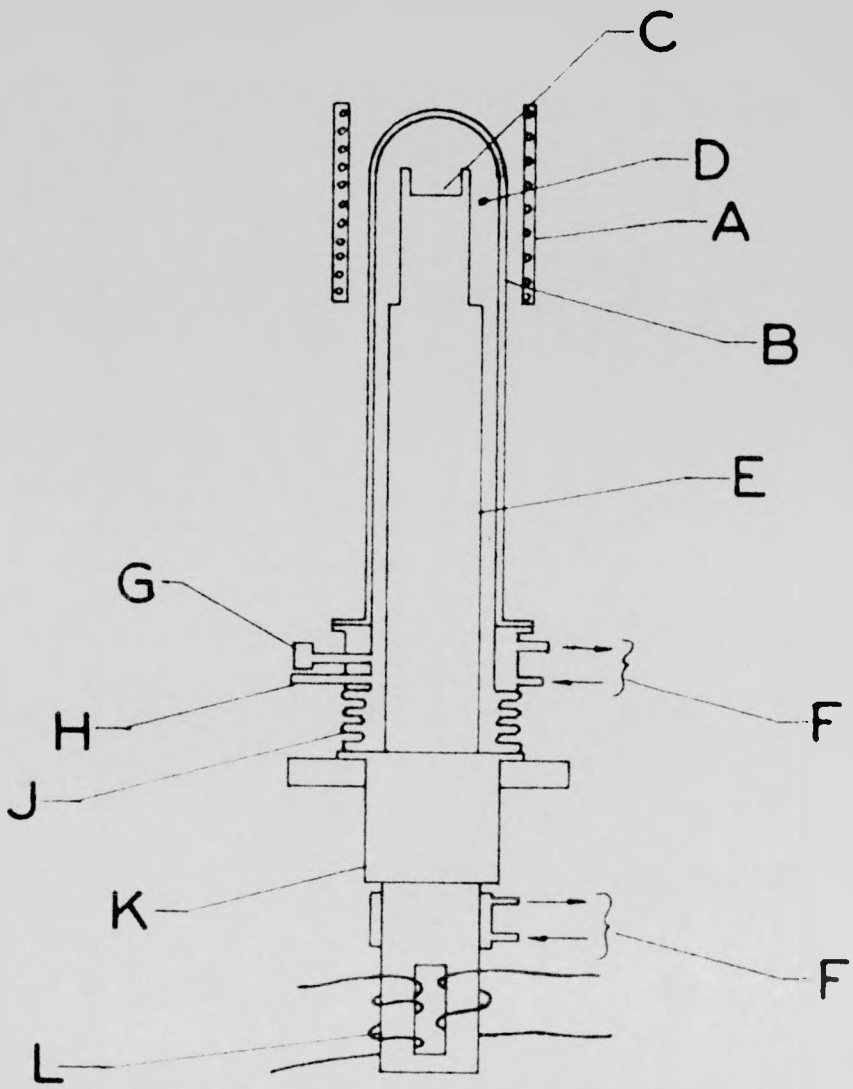


Figure 3

SCHEMATIC DIAGRAM
OF ULTRASONIC RESISTANCE-HEATED, CONTROLLED-ATMOSPHERE FURNACE

A. Resistance furnace
B. Furnace tube
C. Molten metal container
D. Thermocouple junction location
E. Ultrasonic coupler
F. Cooling water inlets and outlets
G. Thermocouple gland and port
H. Vacuum and back-fill inlet
J. Bellows assembly
K. Force-insensitive mount
L. Magnetostrictive transducer

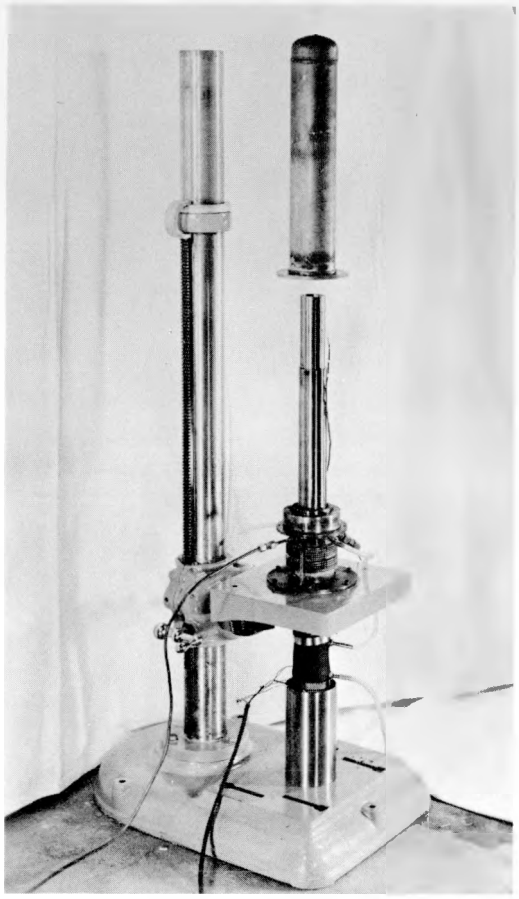


Figure 4

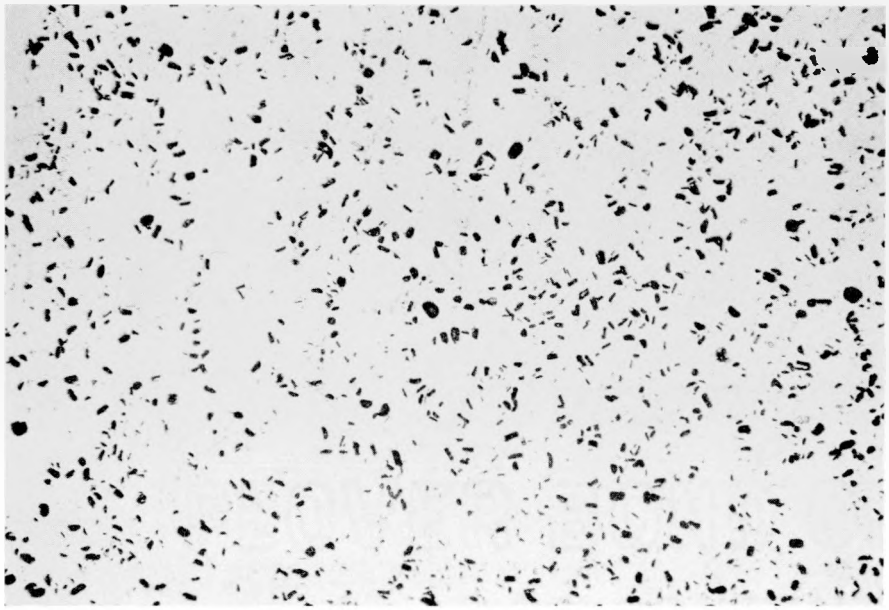
ULTRASONIC RESISTANCE-HEATED,
CONTROLLED-ATMOSPHERE FURNACE

Figure 5

THORIUM BISMUTHIDE PLATELET FORMATIONS
PRODUCED IN NON-ULTRASONIC CONTROL MELT

Bismuth-5% thorium mixture soaked for 1 hour at approximately 1050°C followed by slow cooling to room temperature

Magnification: 28X



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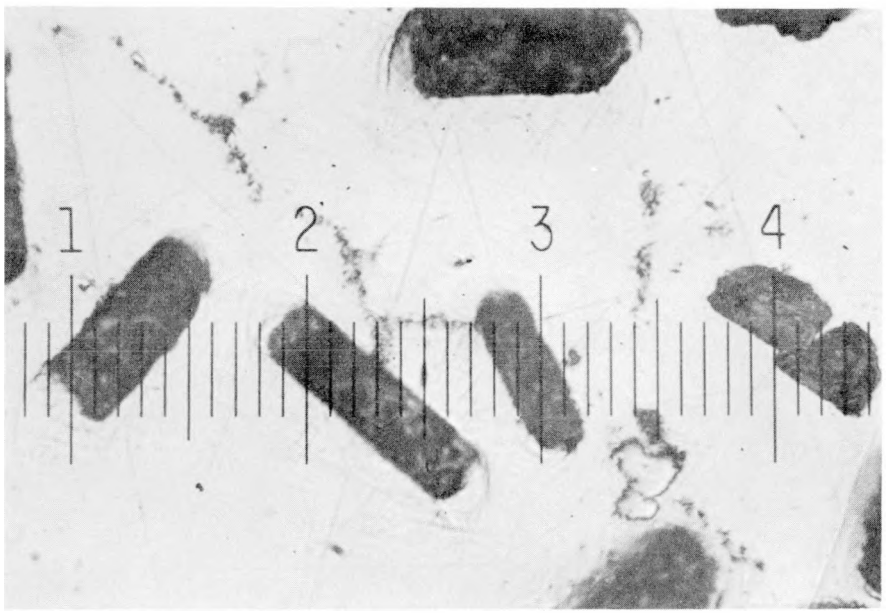
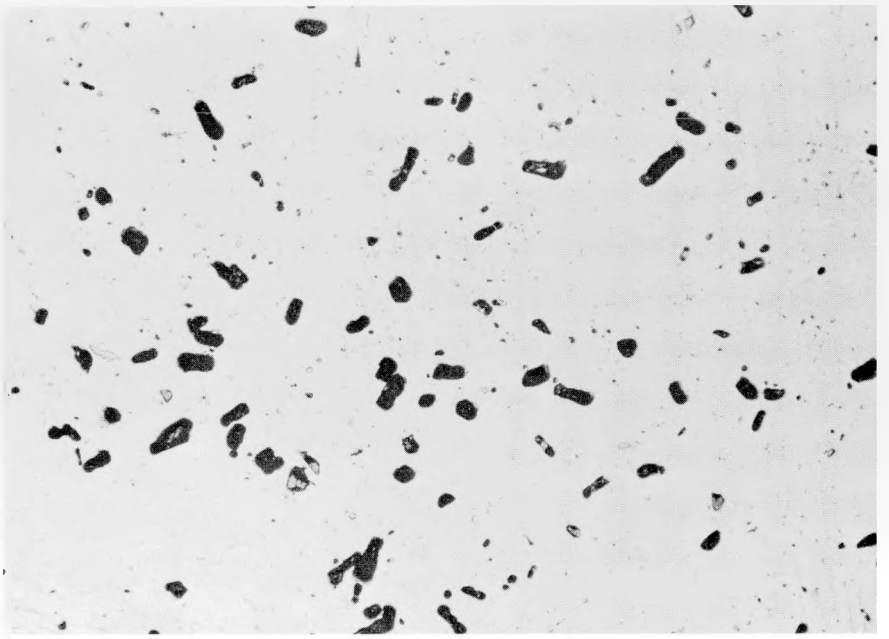


500X
(Each small division = 5μ)

Figure 6

THORIUM-BISMUTH MIXTURE ULTRASONICALLY TREATED
DURING COOLING FROM APPROXIMATELY 1050° TO 500°C

Duration of Treatment: Approximately 1 hour
Ultrasonic Power: 1000 watts



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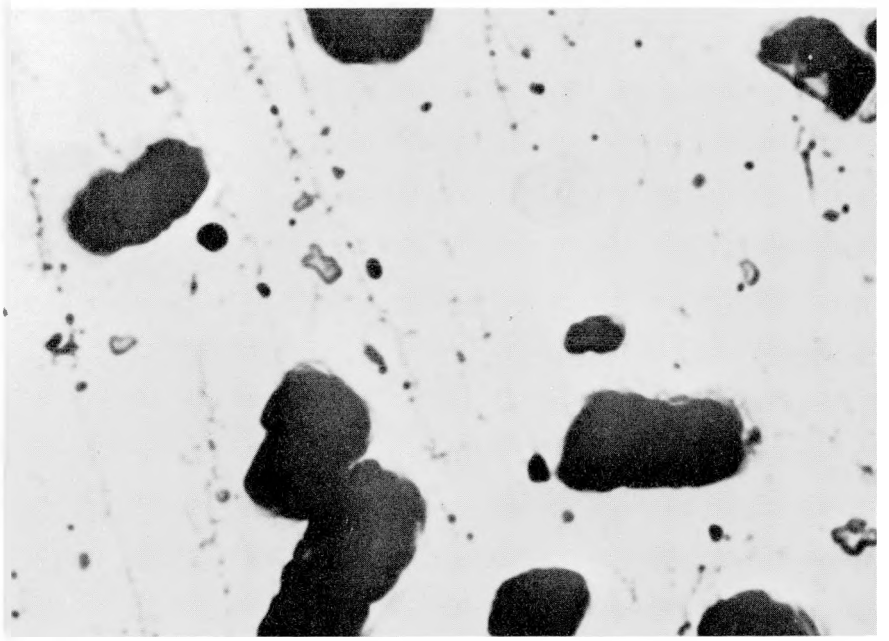
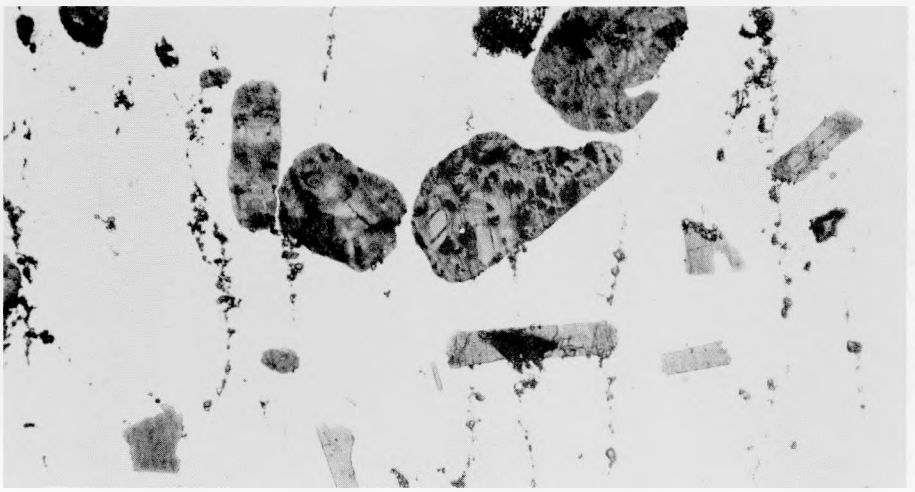


150X

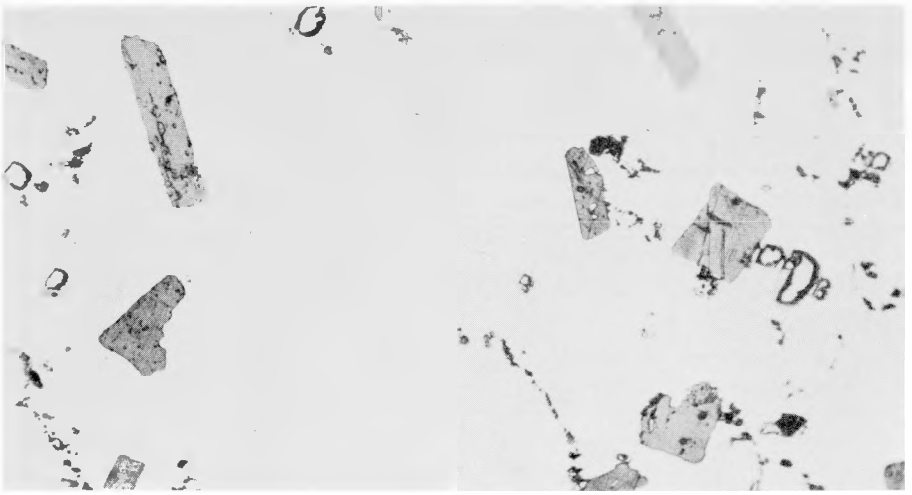
Figure 7

THORIUM-BISMUTH MIXTURE COOLED NORMALLY FROM APPROXIMATELY 1050° C AND ULTRASONICALLY TREATED AT 450° -500° C

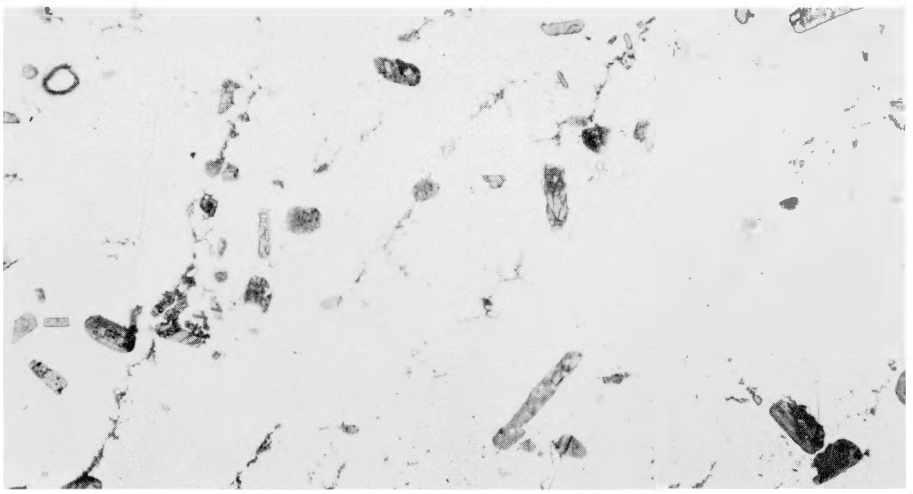
Duration of Treatment: 30 minutes
Ultrasonic Power: 850 watts



10-minute pulsed ultrasonic treatment



15-minute pulsed ultrasonic treatment

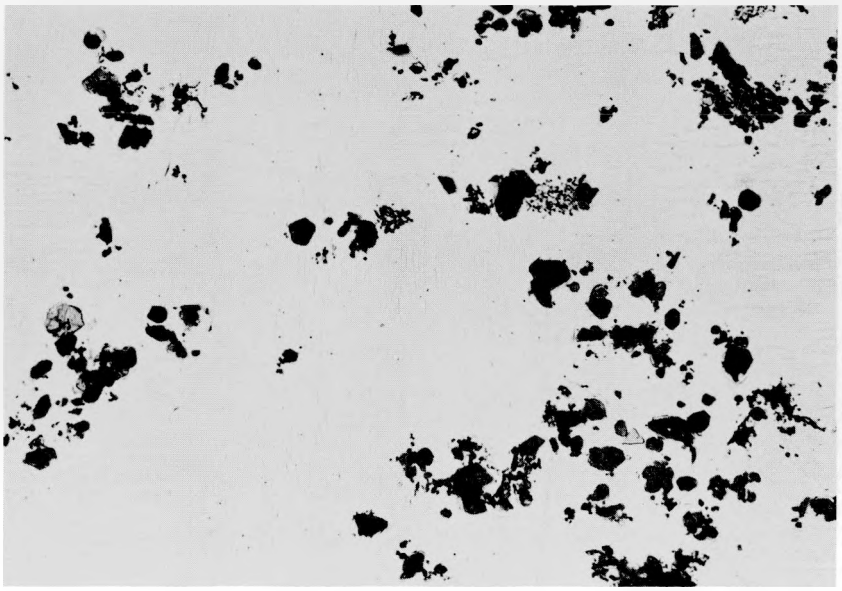


20-minute pulsed ultrasonic treatment

Figure 8

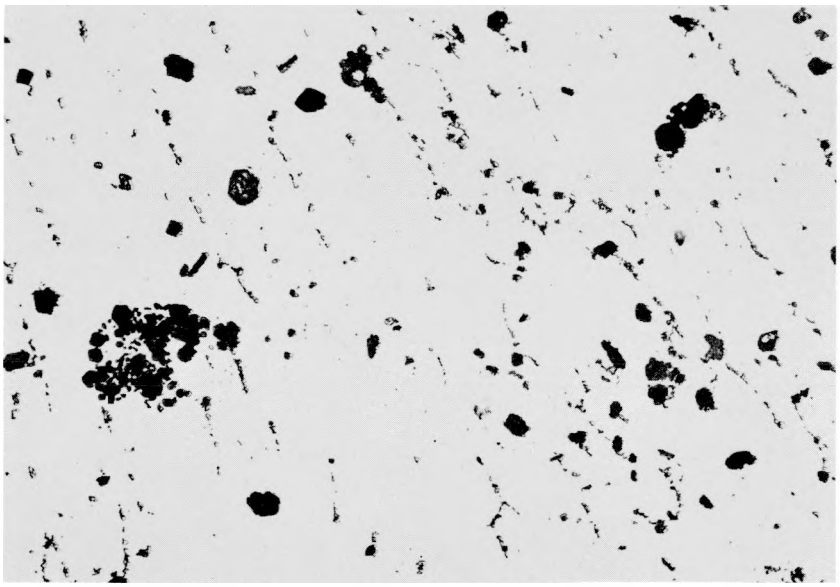
EFFECT OF ULTRASONIC TREATMENT TIME ON EXTENT OF RUPTURE OF PREFORMED THORIUM BISMUTHIDE PLATELETS

Treatment temperature: 470°C
Ultrasonic power: 1400-1500 watts
Magnification: 150X



150X

a. Non-ultrasonically treated control melt



150X

b. Portion of control melt remelted at 500°C and ultrasonically treated for 5 minutes at a power level of 1200 r-f watts

Figure 9

EFFECT OF 5-MINUTE ULTRASONIC TREATMENT AT 1200 R-F WATTS ON SIZE OF EQUIAXED THORIUM BISMUTHIDE CRYSTALS

(Crystals formed by soaking for 4 hours at 500°C in an argon atmosphere)