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AEC RESEARCH AND DEVELOPMENT REPORT

FABRICATION OF SPHERICAL PARTICLES

E. S. Bomar H. Inouye
Metallurgy Division

Art Levy
Pratt and Whitney Aircraft Corporation

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FABRICATION OF SPHERICAL PARTICLES

E. S. Bomar H. Inouye
A. Levy

Abstract

Several potential methods for the preparation of spherical particles of alloys containing uranium have been examined. In each instance, surface tension in the liquid state has been employed as the force for formation of spheroids. Spraying from a metallizing gun or forced ejection of molten alloy from a hole in the bottom of a crucible gave the most positive results. Both these methods, however, produced a wide range of particle sizes.

Spherical particles have been included among the geometries suggested for the fuel-carrying components of reactors that use solid fuel. One design which has been considered for the Pratt and Whitney supercritical water reactor requires the preparation of 0.010-in.-dia spheres. The spheres would contain the fuel in the form of an alloy of uranium and would be nickel plated to minimize corrosion by the high-temperature water.

Three binary mixtures which may be classified into high- and low-melting-point alloys were examined – molybdenum plus 5% uranium for the high-melting-point alloy and copper or nickel plus 5% uranium for the low-melting-point alloy.

In the several methods tried, the force employed for forming spherical particles was the surface tension present in the liquid state. The means used to obtain molten alloy particles were varied and included the following:

1. suspension in a refractory powder followed by heating in hydrogen or vacuum,
2. momentary melting while passing through a carbon or a tungsten arc,
3. momentary melting while passing through an atomic-hydrogen arc,
4. momentary melting in a salt bath with the required temperature gradient,
5. ejection under pressure from a hole in the bottom of a graphite or an alumina crucible,
6. spraying from a metallizing gun,
7. ejection from molten electrode tips.

The first four methods are characterized by their potential ability to yield a consistent product size if the feed material is uniform in size. In the other methods, that is, spraying from a crucible or metallizing gun and the melting of electrodes, the size of the resulting particle cannot be accurately controlled.

SUSPENSION IN A REFRACTORY POWDER

Ingots of arc-cast uranium-nickel and uranium-copper alloys were swaged and drawn to 0.010-in.-dia wires. The nickel alloy was mixed with aluminum oxide powder and heated to 1450°C in hydrogen. A second sample of this mixture was heated to the same temperature in a vacuum of 3 microns. Clippings from pure copper and from uranium-copper alloy wires were spread on a refractory plate and heated to 1100°C in hydrogen. A surface film formed on each of the samples, except those of pure copper, under the conditions outlined above and prevented spheroidization, even though a liquid phase was present (Figs. 1 and 2).

A second high-temperature treatment of the uranium-nickel alloy was made for which better vacuum equipment was used. Segments of the uranium-nickel alloy were placed in an alumina boat and degassed in a vacuum of 10^{-5} mm Hg at 900°C. Examination of the particles after removal from the hot zone but while still under vacuum revealed bright surfaces. The furnace temperature was then increased to 1450°C, and the charge was reintroduced at such a rate that the pressure did not exceed 5×10^{-4} mm Hg. After being held at temperature for 5 min, the sample was cooled under vacuum. Examination showed that, as before, a film had formed on the particle surfaces and spheroidization failed to occur.

It is of interest with regard to surface film formation that both the uranium-nickel and the uranium-copper alloys were bright annealed in a hydrogen atmosphere between drawing operations. The maximum temperatures to which the alloys could be heated in hydrogen without formation of surface films appeared to be close to the eutectic temperatures. The eutectic temperatures are 950 and

1104°C for 5% uranium-copper and 5% uranium-nickel alloys, respectively.

MOMENTARY MELTING IN A CARBON OR A TUNGSTEN ARC

A cylindrical housing about 18 in. high by 5 in. in diameter that contained two ¼-in.-dia water-cooled electrodes was assembled. Provisions were made for introducing a tank-argon atmosphere, and a window for observing the arc was included. Particles of the uranium-nickel and uranium-copper alloys of the types used in the first method were dropped through the ⅛- to ¼-in. electrode spacing into a refractory cup. The particles that melted became spheroidal, but the alignment of the feed through the arc was very critical and yields were low. Particles dropped through the carbon arc were quite porous, while those dropped through the tungsten arc were not. Unfortunately, pictures of the alloy product are not available because the particles were inadvertently thrown away. Figure 3 shows a picture of segments of 0.005-in.-dia nickel wire that were dropped through the tungsten arc.

Work by a graduate student, M. Berg, at the Massachusetts Institute of Technology, with aluminum oxide indicated a greater conversion to spheroidal particles might be expected than that found. Berg worked with a three-electrode apparatus. However, samples taken from the wire stock used for the experiments described above were processed in his equipment without a marked improvement in yield. Pure copper gave fairly good yields, but uranium-nickel and uranium-copper alloys were not altered in the same proportion. A picture of spheroidal particles of alumina produced by Berg is included (Fig. 4) to show the behavior of refractory material.

Since the maximum current in Berg's apparatus was limited to 15 to 20 amp, the effect of higher arc current was investigated. A third ¼-in.-dia tungsten electrode was mounted in the housing used previously. Clippings from the 0.010-in.-dia uranium-nickel and uranium-copper wires were again dropped through the arc. With a current of 35 amp, only a few particles melted. With 40 to 45 amp, a fair percentage of the particles became molten, but alignment continued to be critical. The free-fall distance was not adequate, and consequently a large number of the particles did not solidify during the fall (Fig. 5). Metallographic examination of spheroids produced in this manner disclosed porosity and possible depletion of uranium, which may have been caused by the inadequacy of the protective atmosphere (Fig. 6). Tank argon was used as the protective atmosphere in all the electric-arc experiments.

MOMENTARY MELTING IN AN ATOMIC-HYDROGEN ARC

Uranium-nickel and uranium-copper clippings of the types used in the first and second methods were dropped through an atomic-hydrogen arc and collected in a refractory dish. The yield of melted particles was again low, and the particles were very porous. The particles fell through an air space after leaving the arc zone, and the contact with air while still hot may have caused the porosity shown in Fig. 7.

MOMENTARY MELTING IN A MOLTEN SALT BATH

Spheroidization of precut particles was investigated by using a temperature gradient in molten barium chloride. The barium chloride was contained in a graphite crucible which was, in turn, positioned in a standpipe that was continuously purged with argon. The upper half of the crucible was heated by induction to produce a temperature gradient in the molten salt that varied from 962°C (melting point of barium chloride) to 1200°C. Particles of uranium-copper alloy showed partial spheroidization under these conditions. When the maximum temperature of the salt was increased to 1300°C, most of the particles were spheroidized. The surfaces of the particles were slightly oxidized at both temperatures. The microstructure of the particles is shown in Fig. 8.

EJECTION UNDER PRESSURE FROM A HOLE IN A CRUCIBLE

The attempt to break small particles of molten uranium-copper alloy by means of gravity flow through a 0.030-in.-dia hole in a graphite crucible met with negligible success at temperatures up to 1450°C. A tendency for the alloy to wet the graphite was noted. For these tests, the crucible was surrounded by a tank-argon atmosphere.

On the basis of these tests, an alumina crucible with a 0.020-in.-dia hole and surrounded by a graphite sleeve was substituted for the graphite crucible. Arrangements were made for applying a

positive argon pressure in the alumina crucible. On heating to 1200°C, particles ranging in size from 0.010 to 0.070 in. were ejected from the crucible. The particle surfaces showed little oxidation. Microexamination revealed a cast dendritic structure of copper and UCu_5-Cu eutectic and a small amount of microshrinkage. The shrinkage resulted in small areas of surface indentation (Figs. 9 and 10).

SPRAYING FROM A METALLIZING GUN

Portions of the arc-melted uranium-nickel and uranium-copper alloys were drawn to 0.057-in.-dia wire to be used as feed for the Metco, type E, metallizing gun. The uranium-nickel wire was first sprayed in a conventional manner by using an air blast to break up the molten wire. The particles were sprayed down a 5-in.-dia by 3-ft-high tube and collected in water. The resulting particles approached spherical geometry, but they were again porous (Fig. 11). Substituting tank argon for the air blast resulted in sound particles, but there was a considerable range in particle size (Fig. 12).

Uranium-copper wire fed through the metallizing gun gave nonporous particles with both air and argon blasts. In both instances, the particles were badly distorted and covered with a surface film that was reducible at 900°C in hydrogen. Spraying into a larger receiver would, without doubt, improve the particle geometry.

EJECTION FROM MOLTEN ELECTRODE TIPS

During the process of dropping particles through a tungsten or a molybdenum arc, it was noted that at current values high enough to melt the electrode tips (100 amp through a 0.3-in. tungsten electrode), particles varying from 0.012 to 0.017 in. were ejected from the molten tips (Figs. 13 and 14).

Two ½-in.-long by ¼-in.-dia electrodes were pressed from a mixture of molybdenum and uranium powders (95 wt % Mo–5 wt % U). The compacts were sintered at 1200°C for 5 hr in a vacuum of 0.5 to 1.0 micron. These compacts were substituted for the tungsten electrodes previously used. The electrode tips were melted by passing a current of 140 to 175 amp through an arc between them. The ejected particles are shown in Fig. 15. These particles did not give a response on exposure to an alpha counter as did the parent electrodes and the uranium-nickel particles from a spraying experiment.

CONCLUSIONS

Failure of the attempts outlined above to give satisfactory products appears to have been due to the formation of refractory oxides of uranium. The purity required in the atmosphere surrounding the molten particles is dependent on the length of time necessary for heating into the liquidus region. This is illustrated by the inadequacy of vacuums as high as 0.01 micron in preventing extensive surface reaction during furnace heating as compared with the satisfactory protection offered by an argon blast coupled with comparatively rapid heating and cooling. It seems almost certain that the partial pressure of oxygen present in the latter case was a minimum of several millimeters of mercury.

Other methods for spheroidization of metal particles which might be applicable to this problem are:

1. dropping particles through the center of a length of resistance-heated tubing such as graphite or silicon carbide,
2. passing a high-velocity inert gas through an electric arc struck between electrodes of the desired alloy,[1]
3. sectioning wire to give small cylinders and then producing spherical particles by random impacts in a hammer mill,[2]
4. forming spheres in heading machines such as those used in the manufacture of electrical contacts.

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[1] This technique is described in *Atomization Method of Making Uranium Powder* by H. H. Hausner and H. Mansfield, NYO-1133, Aug. 7, 1950.

[2] This method was suggested by D. Rauch of the NYOO-AEC, who stated that it had been reduced to practice by companies that produce ballpoint pens.

Fig. 1. Uranium-Copper Particles (5 wt % U–95 wt % Cu) and Pure Copper (Spheres) After Heating to 1100°C in a Hydrogen Atmosphere on a Refractory Plate. 21X.

Fig. 2. Uranium-Nickel Particles (5 wt % U–95 wt % Ni) Suspended in Alumina and Heated to 1450°C in a Hydrogen Atmosphere. 31X.

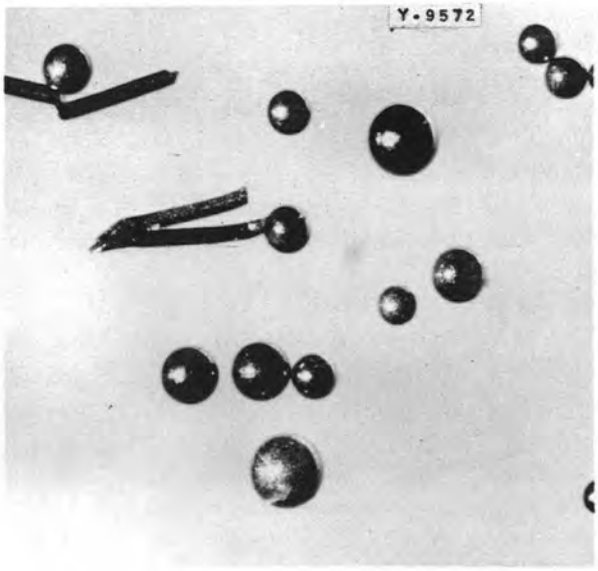

Fig. 3. Segments of 0.005-in.-dia Nickel Wire and Particles Dropped Through a Tungsten Arc. 27X.

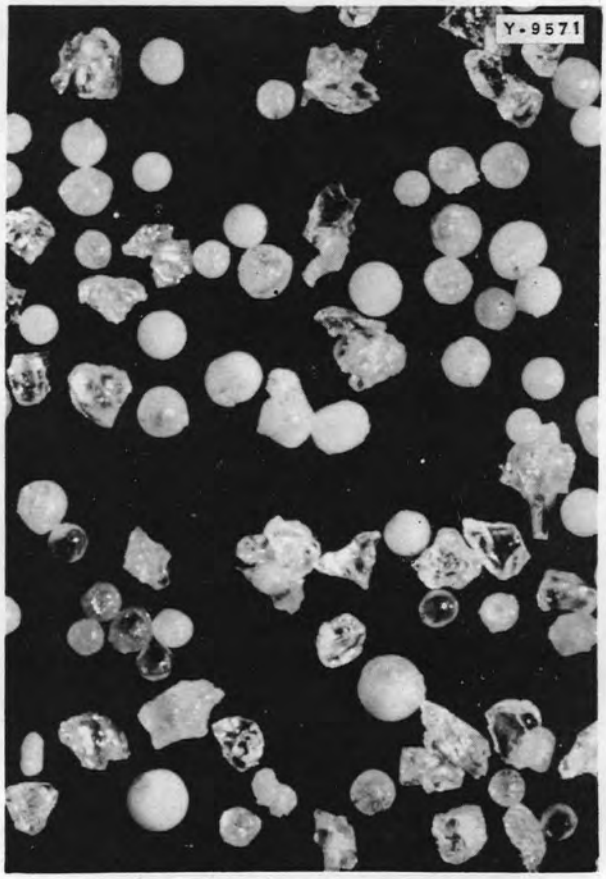

Fig. 4. Alumina Particles Dropped Through a Carbon Arc. 27X.

Fig. 5. Uranium-Copper Particles (5 wt % U–95 wt % Cu) Dropped Through a Three-Electrode Arc. 13X.

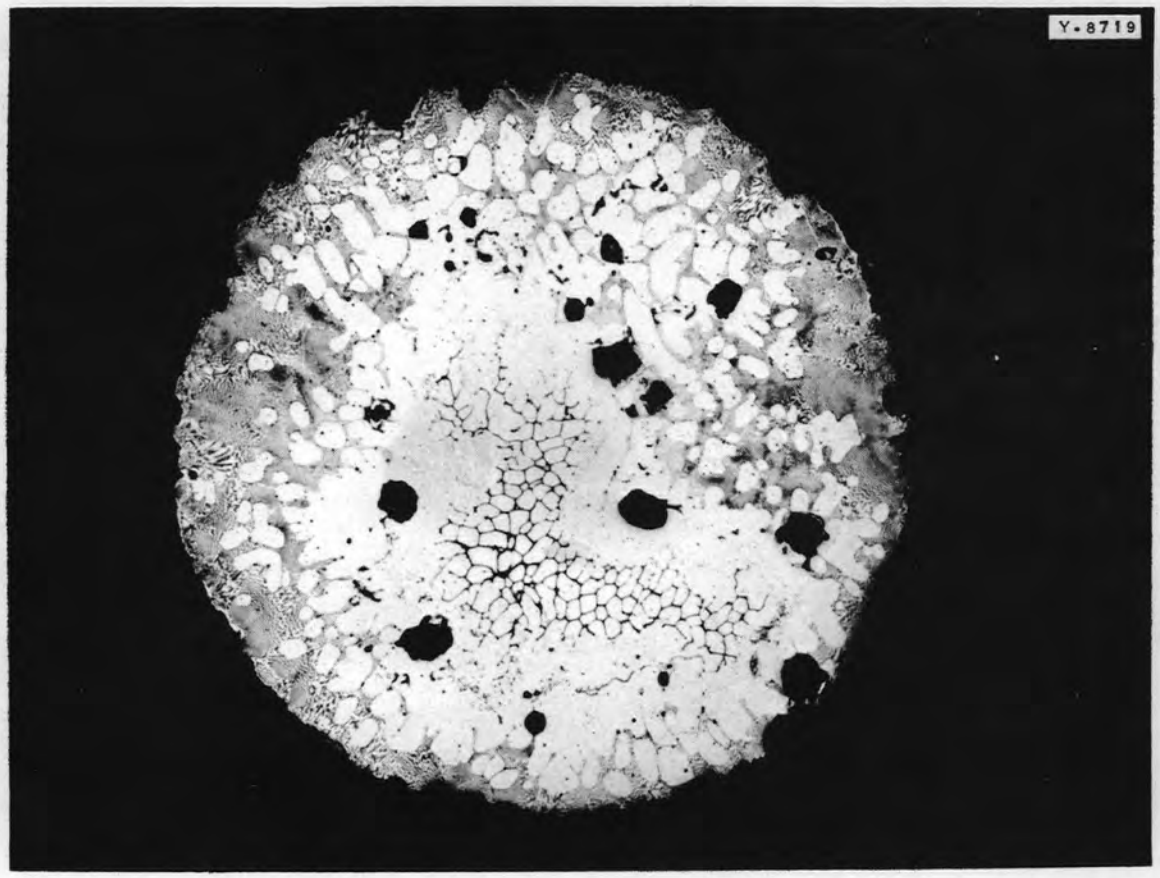

Fig. 6. Enlarged Cross Section of a Uranium-Copper Particle Shown in Fig. 5. 200X.

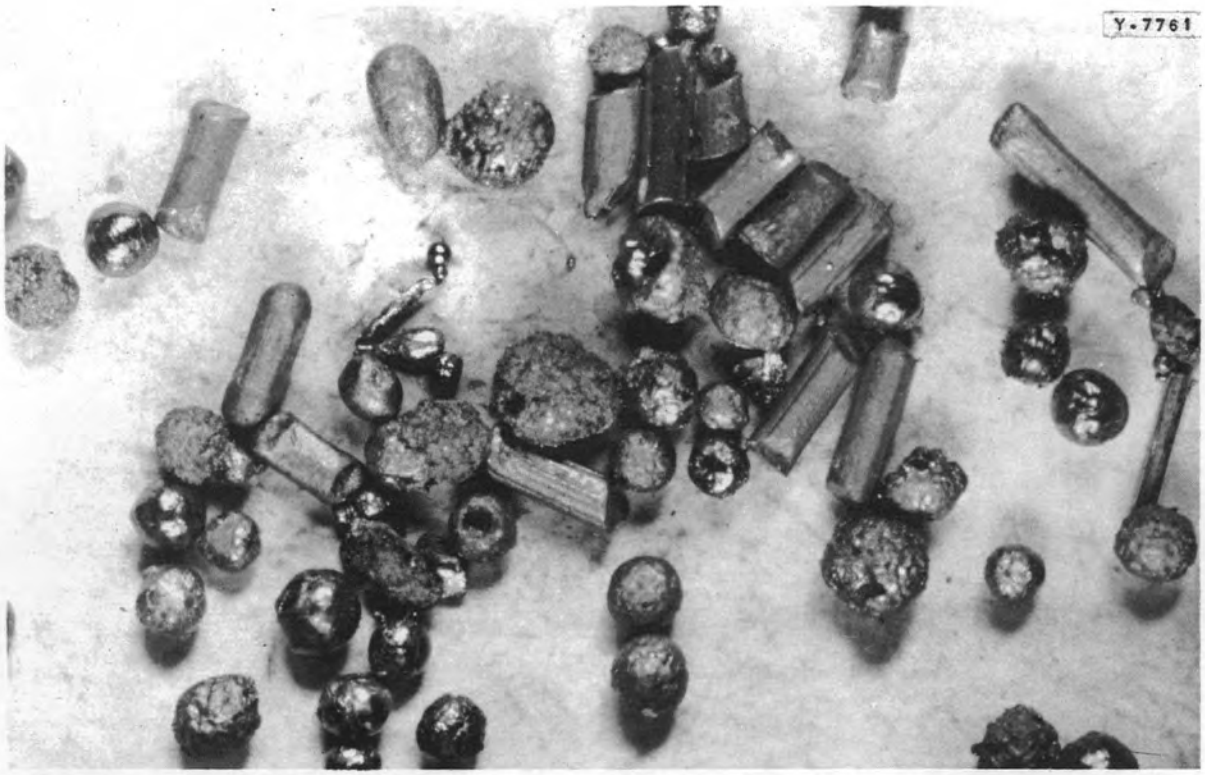

Fig. 7. Uranium-Nickel Particle (5 wt % U–95 wt % Ni) Dropped Throughan Atomic-Hydrogen Arc. 20X. Elongated particles are feed material.

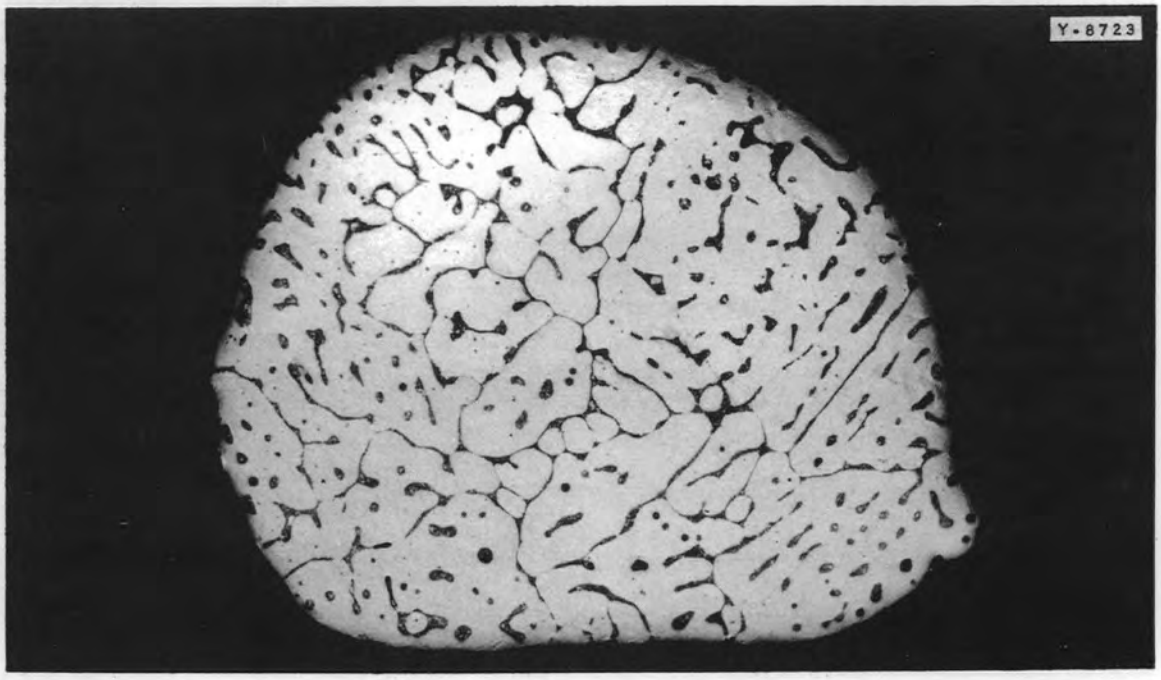

Fig. 8. Enlarged Cross Section of Uranium-Copper Particle Melted in a Salt Bath. 200X.

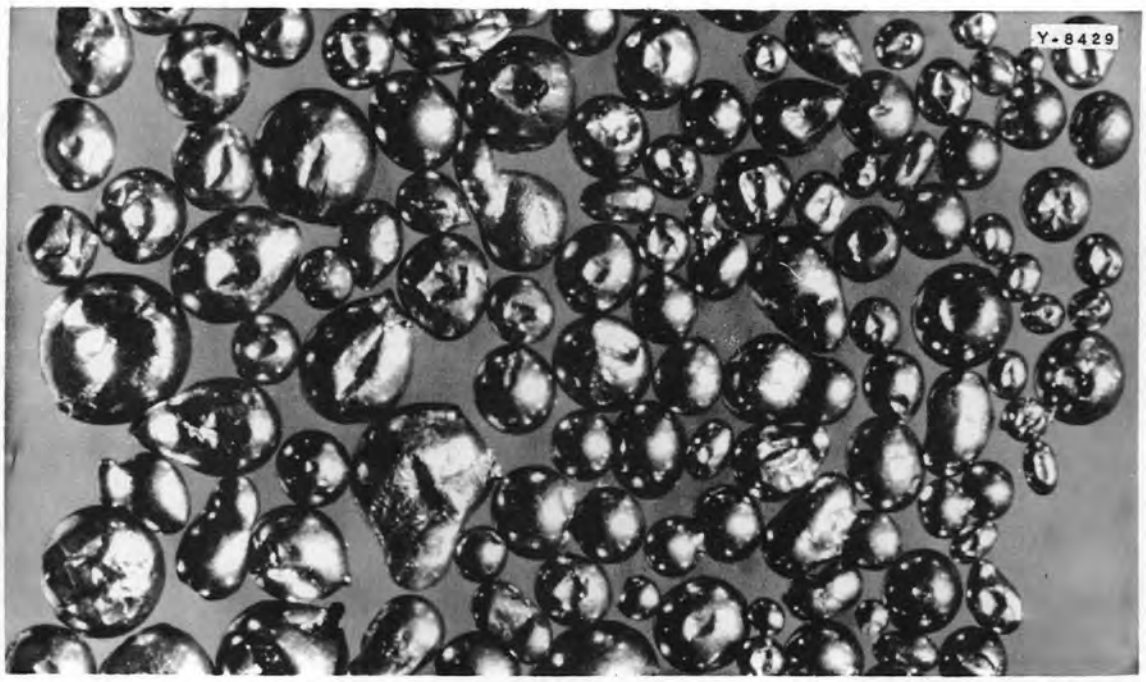

Fig. 9. Uranium-Copper Particles (5 wt % U–95 wt % Ni) Ejected Under Pressure from Hole in Bottom of Crucible. 11X.

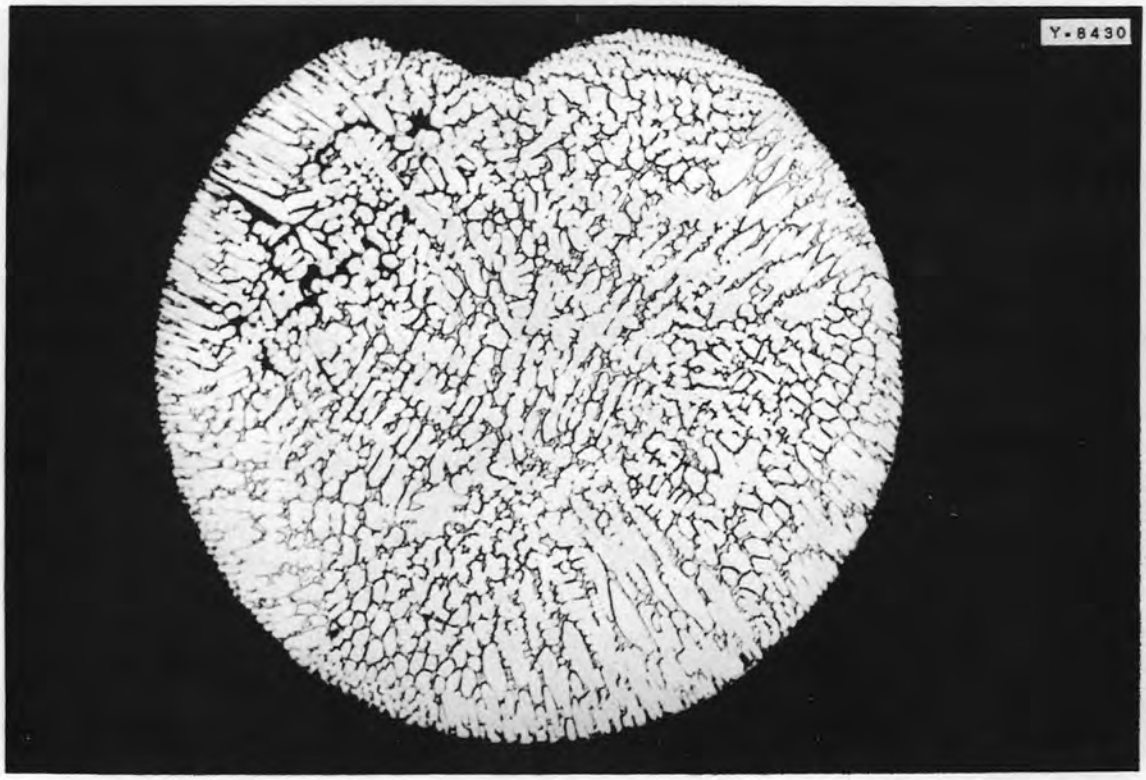

Fig. 10. Enlarged Cross Section of Uranium-Copper Particle Shown in Fig. 9. 60X.

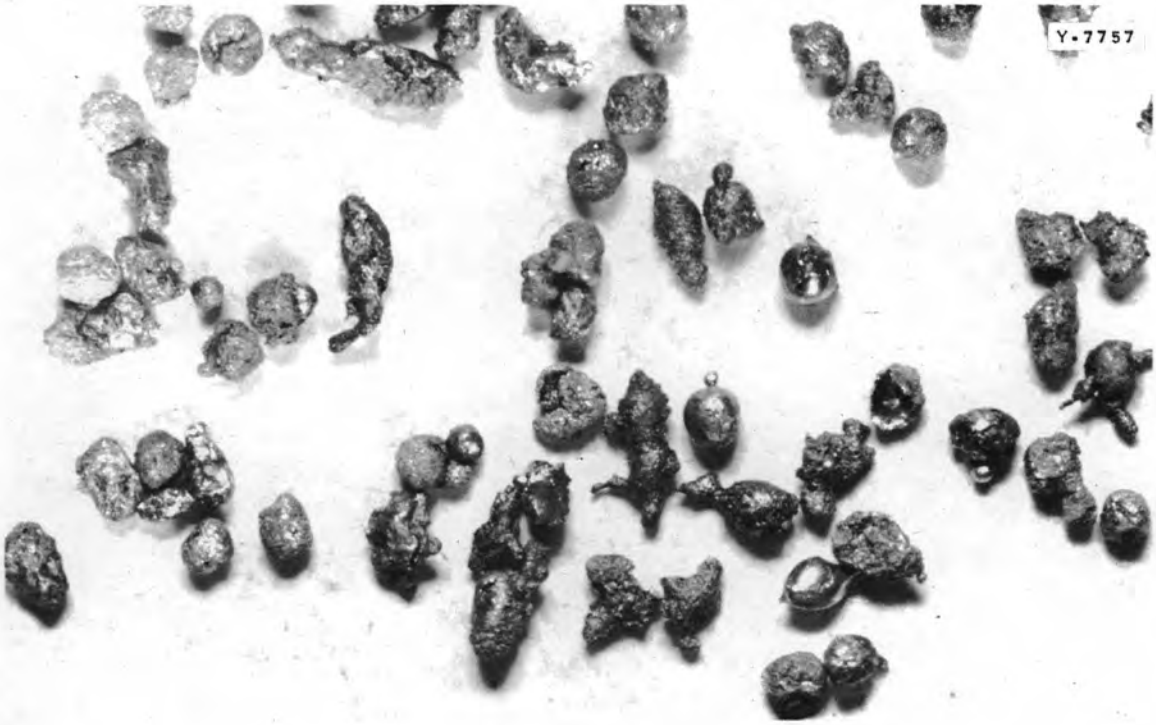

Fig. 11. Uranium-Nickel Particle (5 wt % U–95 wt % Ni) Sprayed from Metallizing Gun with an Air Blast. 33X.

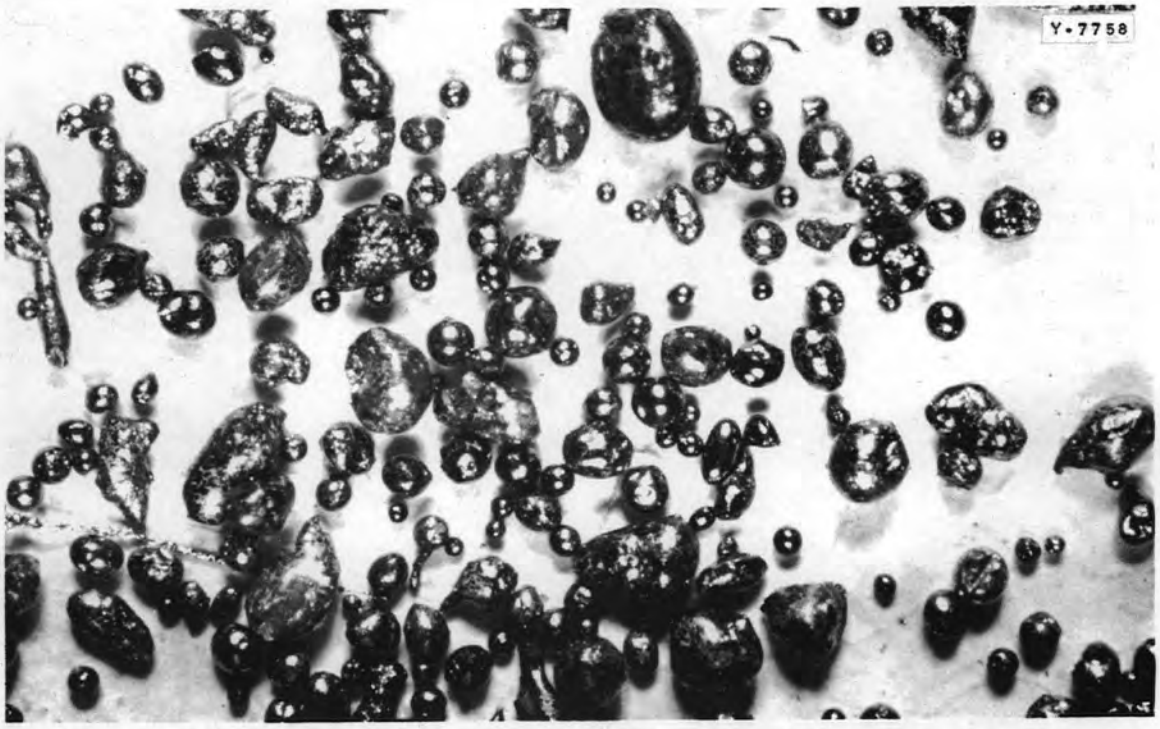

Fig. 12. Uranium-Nickel Particle (5 wt % U–95 wt % Ni) Sprayed from Metallizing Gun with an Argon Blast. 33X.

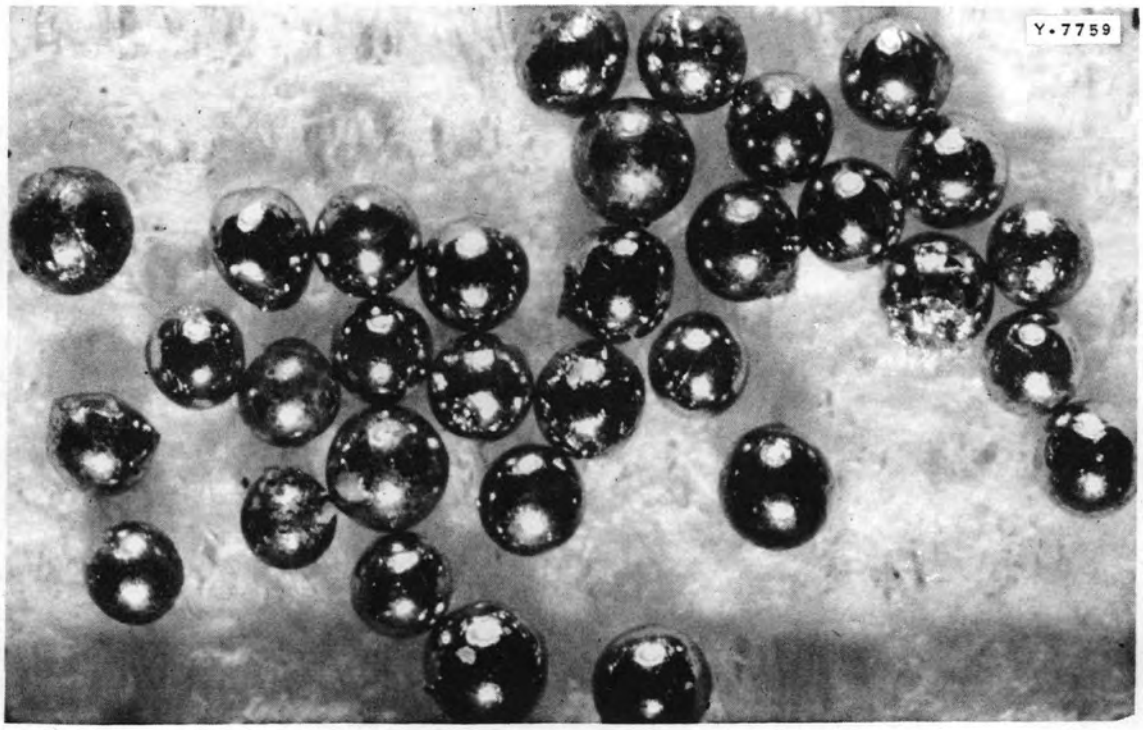

Fig. 13. Particles Ejected from a Tungsten Electrode. 20X.

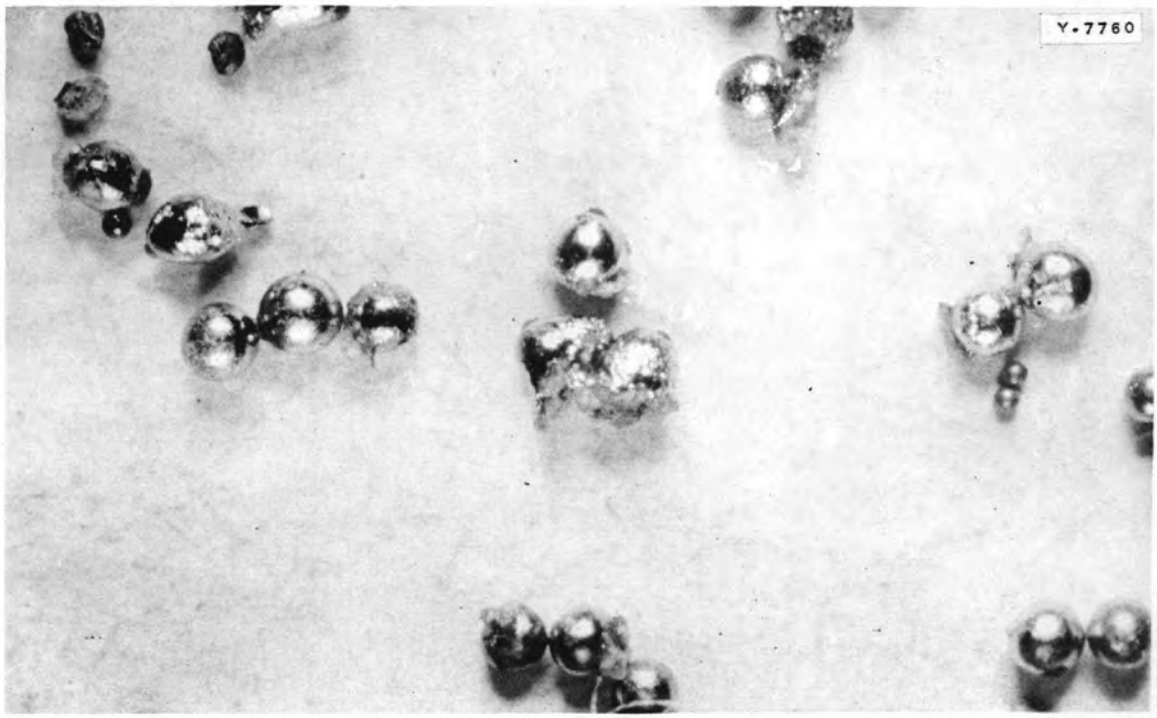

Fig. 14. Particles Ejected from a Molybdenum Electrode. 29X.

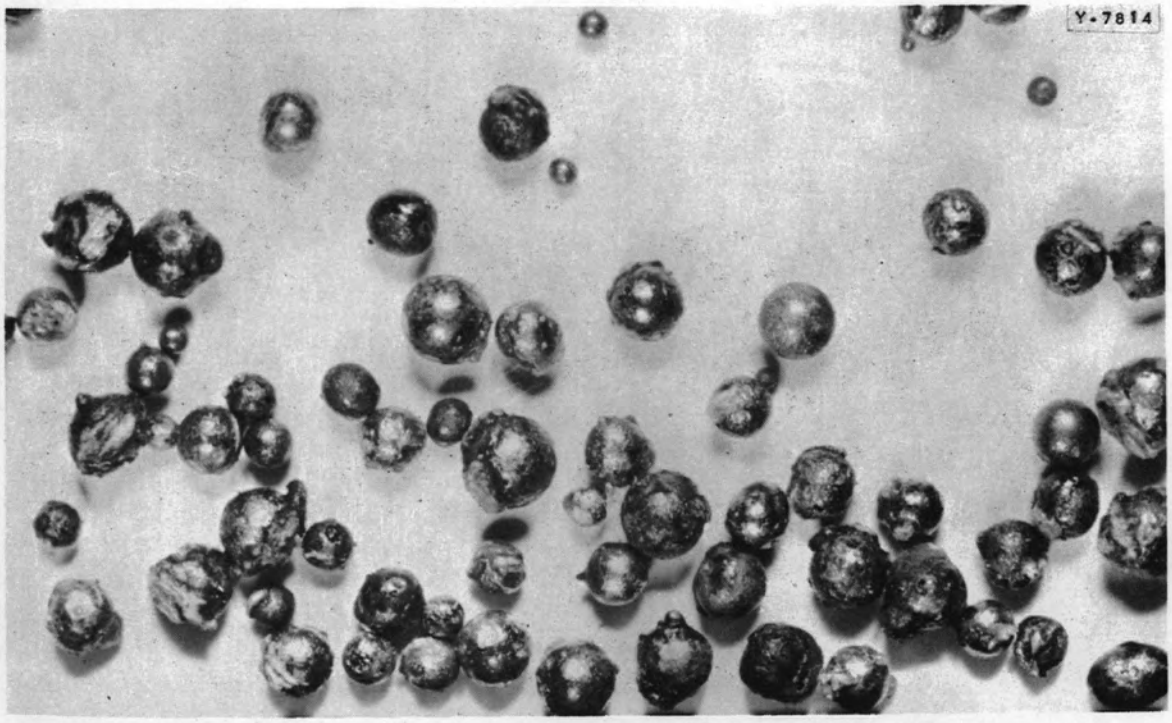

Fig. 15. Particles Ejected from a Uranium-Molybdenum Electrode (5 wt % U–95 wt % Mo). 22X.