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ABSTRACT

Metallic uranium alloys are candidate materials for use as the fuel phase in very-high-density LEU dispersion fuels. These ductile alloys cannot be converted to powder form by the processes routinely used for oxides or intermetallics. Three methods of powder production from uranium alloys have been investigated within the US-RERTR program. These processes are grinding, cryogenic milling, and hydride-dehydride. In addition, a gas atomization process was investigated using gold as a surrogate for uranium.

Grinding was found to be inefficient and introduced impurities into the fuel. Cryogenic milling of machine chips in a steel vial was found to have similar shortcomings.

The hydride-dehydride process has historically been used to produce very fine powder that may not be suitable for fuel fabrication. Uranium is made to form its hydride by heating in a hydrogen atmosphere. Subsequent heating under vacuum drives off hydrogen gas and returns the hydride to a metallic state. The volume change on hydride formation results in a fine powder upon dehydriding. The effects of alloying elements, partial hydriding, and subsequent milling treatments on particle size distribution are being explored.

Inert gas atomization is used on an industrial scale to produce metal powder. Current designs are not suitable for use with uranium. A system was specifically designed for uranium atomization. A prototype was built and tested using gold as a surrogate for uranium. The system operates efficiently and can produce powder in a variety of size ranges by changing the atomization nozzle.

INTRODUCTION

One feature of the RERTR project is the use of powder as the fuel form. This requires some means to produce and handle the fuel in the powder form. With the new higher densities that are required for future reactor conversions some changes in powder production and handling are

needed to meet program goals. In the past the uranium has been mixed with such elements as oxygen or silicon. The resulting alloy is typically friable to the point simple comminution is all that is necessary to reduce the bulk alloy to the proper powder condition [1].

The higher density requirements that the Advanced Fuel Development (AFD) group is addressing have dictated that the amount of alloying agent be a small enough addition and of the type that the metallic properties of the base uranium are essentially unaltered. Therefore, the alloys targeted for RERTR-AFD research are much less friable than those used in the past and require different powder processing techniques [2].

The metal alloy fuel also differs from its predecessors in that at a high free-energy state it is much more likely to pyrophorically react with atmospheric gasses. To avoid this type of incident and preserve quality control, inert atmosphere gloveboxes are required for production and processing of the powder.

Four methods of powder production have been targeted for the RERTR-AFD effort. These are mechanical grinding, cryogenic milling, hydride-dehydride and atomization.

MECHANICAL GRINDING

This method was performed to obtain the majority of the powder used in the first RERTR-AFD experiment currently being examined at Argonne's Alpha-Gamma hot cell facility [2]. Grinding is the most simple and crudest of the proposed powder production methods. This method was chosen for its low cost of equipment, simple operation, and rapid equipment procurement time.

Mechanical grinding is, in essence, using a file to grind the target alloy into the powder form. To aid the process, a small hobby lathe was used to provide a rotating bit into which an alloy slug was fed. The Scanning Electron Microscope (SEM) shows the resulting powder in the form of shavings (Figure 1A). The resulting shavings were sieved to obtain the necessary powder size. The size of the resulting alloy powder was larger than desired and resulted in a very high reject rate. With higher speeds on the lathe, the powder size was lowered but not enough to recommend this method on a production scale.

Other disadvantages of the mechanical grinding method were the high degree of contamination from the grinding bit, extremely slow processing times and the large amount of mechanical deformation imparted to the resulting powder. The uranium alloys used for the first experiment were similar to stainless steel in that they undergo a large amount of work hardening—hindering the process both by increased wear on the grinding bit and by slowing down the powder output. The cold work also imparts a high degree of strain into the ground powder. The resulting high density of dislocations could theoretically serve as nucleation sites for fission gas buildup during irradiation.

CRYOGENIC MILLING

Cryogenic milling relies upon the principle that most materials get more brittle at lower temperatures. For the initial production of the first RERTR-AFD experiment this processing method was tested as a possible stand-alone powder producing method and as a production step to be used in conjunction with mechanical grinding [2]. Tests were performed by placing the

bulk material or the mechanically ground powder into a steel vial with steel grinding balls, chilling the vial contents with liquid argon and agitating the vial in a high-energy ball mill.

The bulk material proved to be non-friable with only trace amounts of powder produced. The ground powders were reduced slightly in size but only after several cryogenic milling runs. SEM analysis shows the powder is in fact reduced in size and flaky in appearance (Figure 1B). The lack of promising results with this method and the high degree of contamination pickup from the grinding vial and balls caused early termination of this method as a candidate for RERTR powder production.

HYDRIDE-DEHYDRIDE

Hydride-dehydride has been used since the 1950's as a viable uranium powder production method [3-5]. It is considered a good method for producing very fine (often under 38 μm) powders. It is now being studied for possible use in future RERTR experiments. It is performed by heating a uranium alloy in a hydrogen atmosphere. At moderate temperatures (typically under 300°C) the uranium reacts with hydrogen. The uranium hydride is much lower density than the bulk uranium (10.9 Vs 19 g/cm^3) and it sloughs off as hydride powder. After the hydriding step is completed, the hydrogen is drawn off by heating the hydride powder under a vacuum. The hydrogen slowly dissociates from the powder leaving only the uranium alloy in powdered form. The process is expressed in the following, reversible equation [5]:



The RERTR-AFD main glovebox houses a tube furnace and an atmospheric control manifold in the powder processing area, which are used to perform the hydride-dehydride operation. The starting material is in the form of chunks or rods and is loaded into a stainless steel crucible and process tube that is connected to the atmospheric manifold. The hydriding step lasts approximately 30 minutes although actual times are unknown and will vary with alloy composition and surface area. The dehydride step is noted on a Pirani pressure gauge as a partial pressure which decreases as the hydrogen is driven out of the powder. This step may also be used to anneal the powder to provide larger particle size.

In initial tests pure uranium was processed with 29% of the resulting powder being in the target size range by sieve analysis. This percentage rose to 80% target size range after 2 minutes of high-energy ball milling. SEM analysis showed that the particle size was much smaller than the sieve analysis indicated (Figure 1C). This was due to agglomeration of the powder. SEM also showed that the powder is in a blocky form.

Disadvantages of the hydride-dehydride process are the size of the powders and safety concerns regarding containment and reactivity of heated hydrogen. In addition, powder produced by this process is extremely pyrophoric and can be handled only in an inert atmosphere.

ATOMIZATION

Atomization is a powder production method where molten metal is rendered into a fine spray, which is solidified forming powder. There are several different atomization methods that are currently commercially available. The most common atomization type is the two fluid method

where a falling stream of molten metal is impinged by a high pressure jet of gas or water. Centrifugal methods are also common with a rotating consumable electrode or a molten stream falling onto a rotating disk. For the RERTR project, an atomization method is required which contains the melt and allows it to homogenize prior to being dispersed into droplets. This is vital since the alloys will often preferentially melt leading to non-homogenous powder [4,6].

To avoid unwanted sticking and buildup, atomization methods require that the molten droplets solidify prior to contact with the atomization chamber walls. This is accomplished by inclusion of a quench medium or by ensuring that the chamber is large enough that the powder is fully solidified during its time of flight [7].

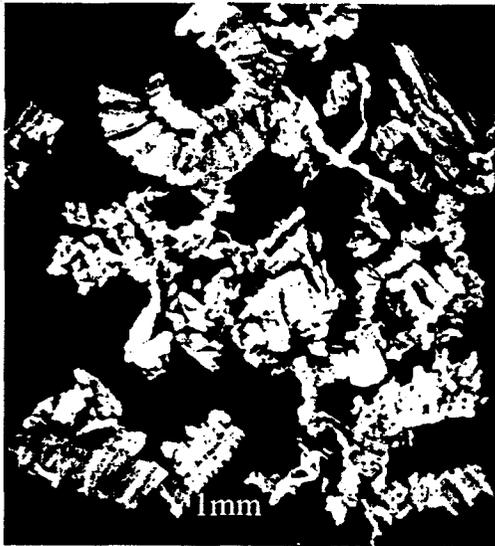
Atomization, while common for most production metals, is rare in the production of uranium or other radioactive elements. Typically the main goal of powder metallurgy is the production of extremely fine powder—for which the process of atomization is very well suited. The RERTR goals however require that the powder be of a size larger than typically fabricated using this process. The main disadvantage of atomization (aside from the cost) is the resulting spherical shape of the powders that easily segregate from the aluminum matrix material during and after blending (Figure 1D).

Of the commercially common atomization methods, inert gas atomization was chosen to be the most promising since its operating parameters are well understood as it has been used in the past to produce uranium powder. The centrifugal methods were judged to be unsatisfactory due to either high complexity of the machinery or segregation of alloy constituents which is common in some methods.

Since the startup cost of an atomization unit is high, a preliminary study was undertaken by the HJE Corporation to examine atomization's fitness for the specifics of the RERTR powder campaign. They were contracted to investigate the ability to produce powder in favorable and flexible sizes and distributions. Two target sizes of 40 and 180 μ m were chosen as a good representation of program needs. For the study, gold was used as a surrogate metal since its molten surface tension, viscosity and density are all similar to uranium. In addition, HJE was to establish a baseline hardware design and operating parameters based on the project dictated constraints. Specifically:

- Effluent gas flow below 1000 standard cubic feet per minute (SCFM)
- High efficiency (at least 99.8%) in material recovery
- Size constraints of the atomization chamber (research facility limitations)
- Operation at negative pressure (containment issues)
- Full density of produced powder

An existing commercially available gas atomization system was modified to meet project requirements. The size constraints dictated that a liquefied gas cooling system be employed to ensure that the powder be fully solidified before impacting the smaller atomization chamber's walls. The sub-ambient pressure operating requirement necessitated an evacuation pump and an atmosphere control system to maintain the pressure at desired levels (Figure 2). A specially designed atomization nozzle was employed to reach the larger target particle size.



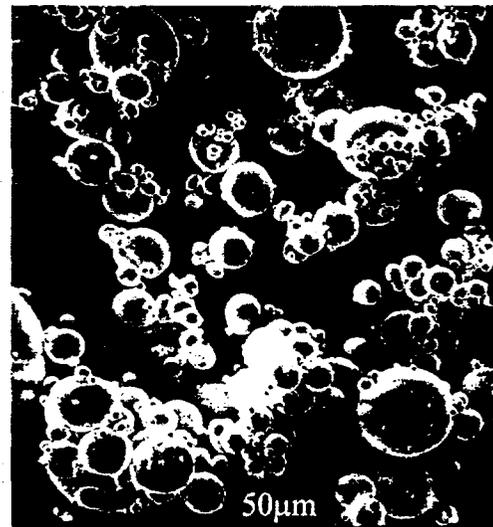
A



B



C



D

Figure 1. Representative Powder Samples Produced by A) Grinding (U-2Mo-1Nb-1Zr);
B) Cryogenic Milling (U-10Mo); C) Hydride-Dehydride (DU);
D) Gas Atomization (Gold Surrogate)

The HJE Corporation developmental system was found to meet or nearly meet all of the requirements listed above. Effluent gas use never exceeded 200 SCFM, the material recovery was above target and the size of the test equipment was well within the program requirements. The pressure of the system did have occasional excursions above ambient pressure but this could be overcome by enlarging the atomization chamber volume to that allowed by facility space and by rewriting the controller code to anticipate and counteract known high-pressure events. The atomized surrogate powders had a large number of satellites (Figure 1D) thought to be caused by turbulence in the atomization chamber. This would be corrected by modifications to the pressure

control system and the system layout. Density tests were somewhat unclear but seemed to indicate that the powders were nearly full density.

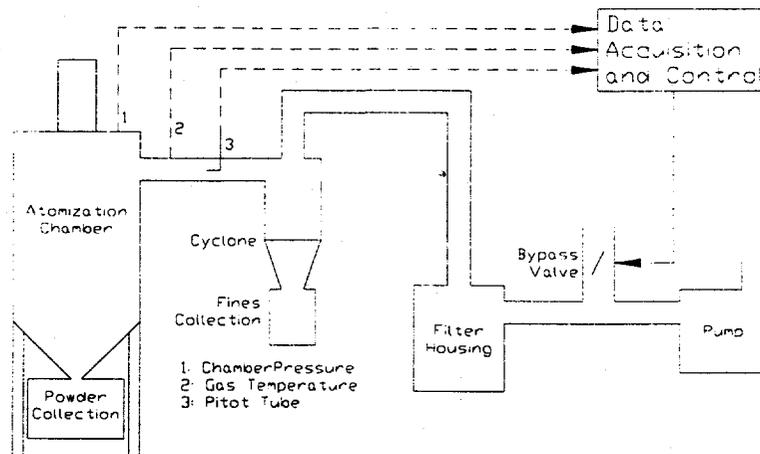


Figure 2. Schematic of Proposed Atomization Unit

The particle size was divided into two target sizes 40 and 180 μ . The 40 μ -target size goal was met using a standard atomization nozzle. For the 180 μ -target size test a special nozzle was designed and implemented. The designs met or nearly met both target powder sizes. See Table 1 and Figure 3.

Table 1. Particle Size Experimental Parameters and Results

Run #	Melt Orifice Size (in)	Melt Rate (g/s)	Mean Particle Size (μ)	Average Deviation (μ)
40 μ Target #1	0.073	69	50.2	2.18
40 μ Target #2	0.070	64	46.0	3.27
40 μ Target #3	0.073	68	51.4	3.51
180 μ Target #1	0.070	37.4	158.9	5.73
180 μ Target #2	0.076	45.0	173.5	6.41
180 μ Target #3	0.090	60.3	184.2	5.97
180 μ Target #4	0.081	49.3	185.1	8.11

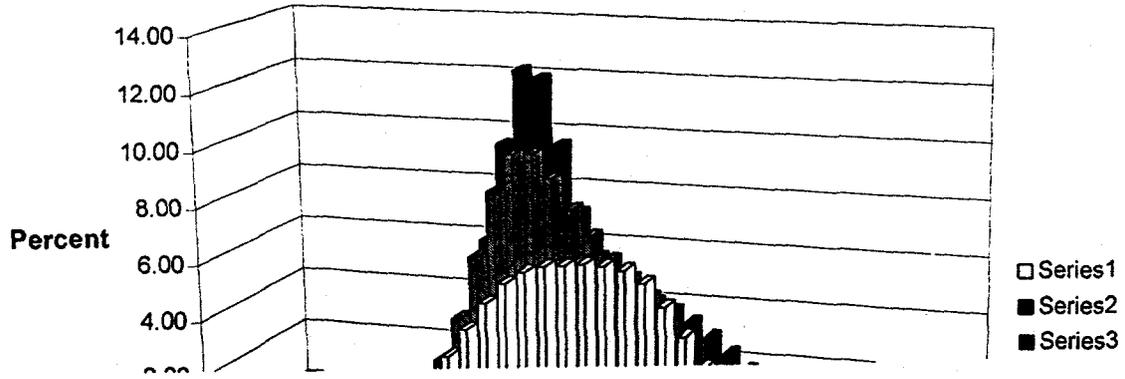
FUTURE EMPHASIS

The RERTR-AFD program will continue to examine several different powder production methods to increase the odds of overall success. The next RERTR experiment will consist primarily of atomized and hydride-dehydride powder.

The hydride dehydride process will be examined to find if it can be made into a viable powder production method. This may require additional annealing steps to coarsen the powder.

We also expect to look more at grinding as a production method specifically:

- Annealing of powder to relieve grinding-induced stresses
- Low temperature grinding to overcome work hardening
- As a known control



REFERENCES

- [1] R.W. Knight, "Observations in the Manufacture of Aluminum-Based Research Reactor Fuel Elements," ORNL/TM-11809, Oak Ridge National Laboratory, Tennessee, 1993.
- [2] C.L. Trybus, T.C. Wiencek, M.K. Meyer, D.J. McGann and C.R. Clark, "Design and Fabrication of High Density Uranium Dispersion Fuels," Proc. 20th International Meeting on Reduced Enrichment for Research and Test Reactors, 5-10 October 1997, Jackson Hole, Wyoming, USA (in press)
- [3] P. Chiotti and B.A. Rogers, "The Production of Uranium and Thorium in the Powder Form," AECD-2974, Ames Laboratory, Iowa, 1950.
- [4] J. Greenspan and T.R. Wright, "Powder Metallurgy Processing of Uranium Alloys," in "Physical Metallurgy of Uranium Alloys," Proceedings of the Third Army Materials Technology Conference, J.J. Burke Et Al. Editors, pp. 83-108, 1974.
- [5] W.M. Mueller, J.P. Blackledge and G.G. Libowitz Editors, "Metal Hydrides," Academic Press, 1968.
- [6] A. Lawley, "Atomization: The production of Metal Powders" MPIF, 1992.
- [7] J.T. Strauss, "Development of Uranium Atomization Process by the Use of a Surrogate Metal," HJE Corporation Report, 1998.