FUEL POWDER PRODUCTION FROM DUCTILE URANIUM ALLOYS

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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 research reactor fuel fabrication processes is the use of powder as the base fuel form. The higher fuel densities that are required for high-power reactor conversions will likely require the use of metal alloys. In previous experience, uranium oxides or intermetallics have

been used as the fuel phase. These compounds are typically friable to the extent that simple communition is all that is necessary to reduce the bulk alloy to the proper powder condition [1].

Higher density fuel requirements have dictated that the amount and the type of alloying elements be such that the uranium alloy fuel phase has metallic properties. Therefore, the alloys used for current Reduced Enrichment Research and Test Reactors (RERTR) fuel research are much less friable than those used in the past and require different powder processing techniques [2].

In powder metallurgy (PM), the powder size, shape and ductility dictate its ability to be fabricated into useful parts. Powder particle size distribution dictates the powder compact's ability to be pressed to a higher density with smaller sizes allowing greater compaction. The shape of the powder effects the strength of the pressed compact and the degree of segregation of blended powders. For example, spherical powders, with little or no frictional interaction do not hold together in a compact or resist segregation as well as other shapes. Ductile materials will readily deform giving good adhesion in a pressed compact. The uranium alloys currently under examination have higher ductility than other alloys that have been used, but the ductility is still much too low to play a significant role in adhesion of compacts pressed with aluminum. In these compacts, the volume fraction of aluminum serves as the binder for the uranium fuel.

Metal alloy fuel powder is likely to react with the aluminum matrix material during fabrication. It is possible that the high relative surface area of smaller powders will lead to a significant, unwanted U-Al reaction zone in the fuel plate. Powder size will be optimized based on the results from post-irradiation analysis and fabrication studies.

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

MECHANICAL GRINDING

Mechanical grinding was employed to obtain the majority of the powder used in the first US-RERTR irradiation experiment currently being examined at Argonne's Alpha-Gamma Hot Cell Facility [2]. Grinding is the simplest and most crude of the proposed powder production methods. This method was chosen for its low equipment cost, simple operation, and rapid equipment procurement time.

The mechanical grinding process makes use of a file to grind the target alloy into powder form. A small hobby lathe was used to power a rotary file into which an alloy slug was fed. Scanning Electron Microscopy (SEM) shows that the resulting powder is in the form of shavings (Figure 1A). The resulting shavings were sieved to obtain the powder size fraction required for fuel plate production. The size of the resulting alloy powder was larger than desired and resulted in a very high reject rate. The powder size decreased at higher file rotation speed, but not enough to recommend this method for production scale powder processing.

Other disadvantages of the mechanical grinding method were the high degree of contamination from the grinding bit, an extremely slow powder production rate and the large amount of mechanical deformation introduced into 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; this hinders the grinding both by increased wear on the grinding bit and by slowing powder output. The cold work introduced a high degree of strain into the ground powder. The resulting high dislocation density may lead to nucleation sites (sub-grain boundaries) for fission gas bubble formation during irradiation.

CRYOGENIC MILLING

Cryogenic milling relies upon the principle that most materials have a ductile-to-brittle transition temperature at some cryogenic temperature. At these temperatures the materials may become brittle enough that communition is sufficient to break the bulk ingot into powder. For the first US-RERTR experiment, this processing method was tested both 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 mechanically-ground powder into a hardened 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 that were cryogenically milled were reduced slightly in size but only after several milling runs. SEM analysis shows the cryogenically milled powder size is, in fact, reduced and it appears as flakes (Figure 1B). The lack of promising results with this method and the high degree of contamination from the grinding vial and balls caused this method to be rejected as a candidate for uranium alloy powder production.

HYDRIDE-DEHYDRIDE

The hydride-dehydride process has been used since the 1950's as a uranium powder production method [3-5]. It is considered a good method for producing very fine (often under 38 μm) powders and is now being studied for possible use in future US-RERTR experiments. The process is initiated by heating a uranium alloy ingot in a hydrogen atmosphere. At moderate temperatures (typically under 300°C) the uranium reacts with hydrogen. The uranium hydride has a much lower density than the uranium metal (10.9 vs 19 g/cm^3) and it sloughs off the ingot surface as hydride powder. After the hydriding step is completed, the uranium hydride is reduced 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 by the following reversible equation [5]:

$$3H_2 + 2U \leftrightarrow 2UH_3$$

The US-RERTR powder glovebox houses a tube furnace and an atmospheric control manifold 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 gas manifold. The hydriding step lasts approximately 30 minutes, although actual times vary with alloy composition and surface area. The dehydriding step is monitored by a pressure gauge. A low and stable pressure indicate that the dehydriding phase is complete. An annealing step may also be used to sinter the powder and provide larger particle size.

In initial tests, pure uranium was processed with 29% of the resulting powder being in the target size range (45-150μm) by sieve analysis. This percentage rose to 82% within the target size

range after 2 minutes of high-energy ball milling (Table 1). SEM analysis showed that the particle size was much smaller than the sieve analysis indicated due to agglomeration of the powder. SEM also showed that the powder is in a blocky form (Figure 1C).

Table 1. Hydride-Dehydride Process Sieve Analysis Results

Milling Time	Sieve Analysis Particle Size (%)		
	>150μ (+100 mesh)	150-45μ (-100+325 mesh)	<45μ (-325 mesh)
0 min	54	29	17
1 min	38	55	7
2 min	6	82	12
3 min	5	81	14
8 min	3	82	15

Disadvantages of the hydride-dehydride process are the smaller-than-desirable production powder size and safety concerns regarding containment and reactivity of heated hydrogen. In addition, powder produced by this process is very fine and must be handled in an inert atmosphere.

ATOMIZATION

Atomization is a powder production method where a fine spray of molten metal is produced, then solidified, forming powder. Several different atomization methods are commercially available. The most common atomization process 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 US-RERTR program, an atomization method is required which contains the melt and allows it to homogenize prior to being dispersed into droplets. This is vital since 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 so that the powder is fully solidified during its time of flight [7].

Atomization, while common for most production metals, is seldom used in the production of uranium or other radioactive elements. Typically, the main goal of PM is the production of extremely fine powder–for which the process of atomization is well suited. Dispersion fuel production however, requires that the powder be of a size larger than typically used for PM processes. The main disadvantage of atomization (aside from the capital cost) is the resulting spherical shape of the powders that can easily segregate from the aluminum matrix material during and after blending (Figure 1D).

Of the commercially common atomization methods, the rotating disk method for atomization of uranium has been developed by The Korean Atomic Energy Research Institute [8]. Inert gas atomization was evaluated as an alternative to the rotating disk method. Rotating electrode

methods were judged to be unsatisfactory due to either high complexity of the machinery or segregation of alloy constituents, which is common in such methods.

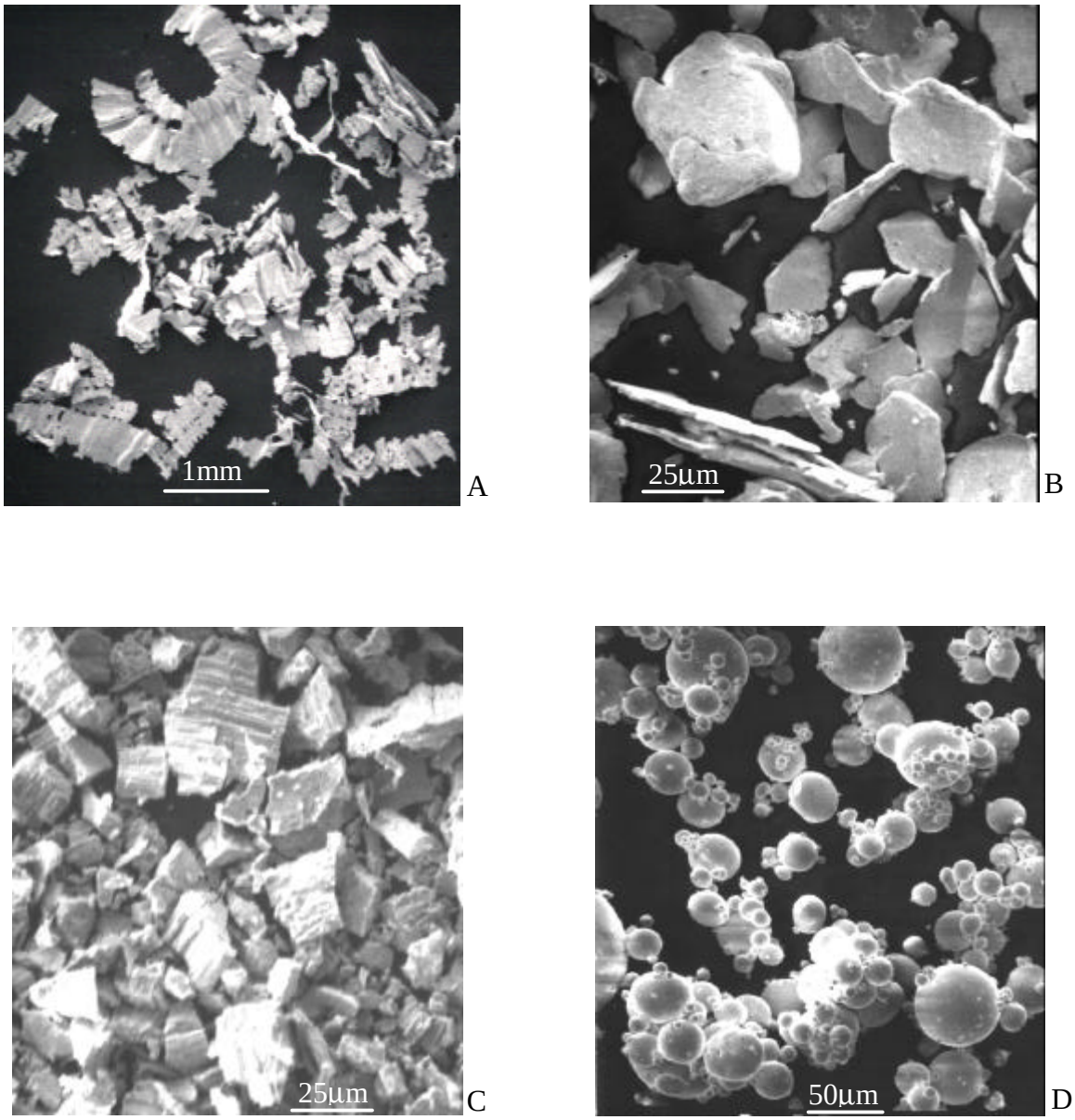

Figure 1. Representative Powder Samples Produced by: A) Grinding (U-2Mo-1Nb-1Zr); B) Cryogenic Milling (U-10Mo); C) Hydride-Dehydride (DU); D) Gas Atomization (Gold Surrogate)

Because the startup cost of an atomization unit is high, a preliminary study was performed by the HJE Corporation to examine the fitness of atomization for the specifics of uranium powder production. The focus of the investigation was the possibility of producing powder in favorable and flexible sizes and distributions. Two target sizes of 40 and 180µm were chosen as good representations of possible program needs for large and small powder. Gold was used as a surrogate metal for this study since its surface tension, viscosity, melting point and density are all

similar to uranium. An additional goal was to establish a baseline hardware design and operating parameters based on constraints dictated by facility limitations and concerns about contamination control. Specifically these constraints were:

- 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)
- Production of powder at full density
- No water (criticality)

An existing, commercially available gas atomization system was modified to meet these requirements. The size constraints and desire to exclude water from the system dictated that a liquefied gas cooling system be employed to ensure that the powder was solidified before impacting the walls of the small atomization chamber. 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 produce the larger target particle size.

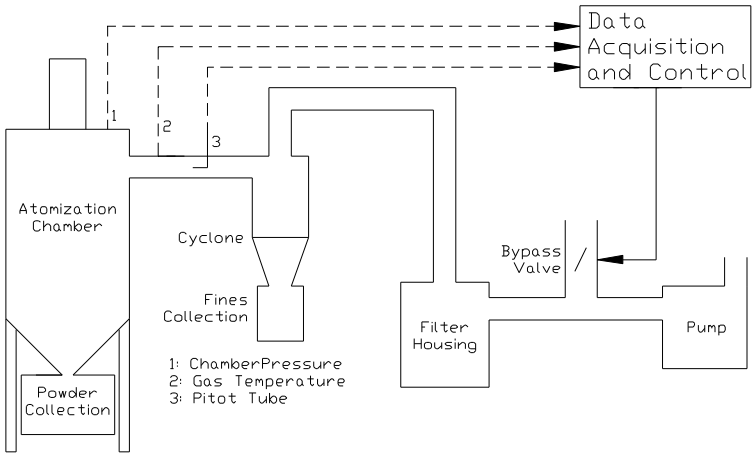

Figure 2. Schematic of Proposed Atomization Unit

The developmental system was found to perform within the requirements listed above. Effluent gas flow never exceeded 200 SCFM, the material recovery was above target and the physical size of the test equipment was within program requirements. The atomized surrogate powders contained a large number of satellites (Figure 1D) thought to be caused by turbulence in the atomization chamber. Satellite formation could be controlled by modifications to the pressure control system and the system layout. Density tests using gas pycnometry were somewhat ambiguous but seemed to indicate that the powders were near full density.

Powder production in the two target size ranges, 40 and 180μ, required the use of two atomization nozzles. The 40μ-target size goal was met using a standard nozzle. In order to produce 180μ-powder, a special nozzle was developed. These two nozzles met or nearly met both target powder size requirements. Particle size data is given in Figure 3 and Table 2.

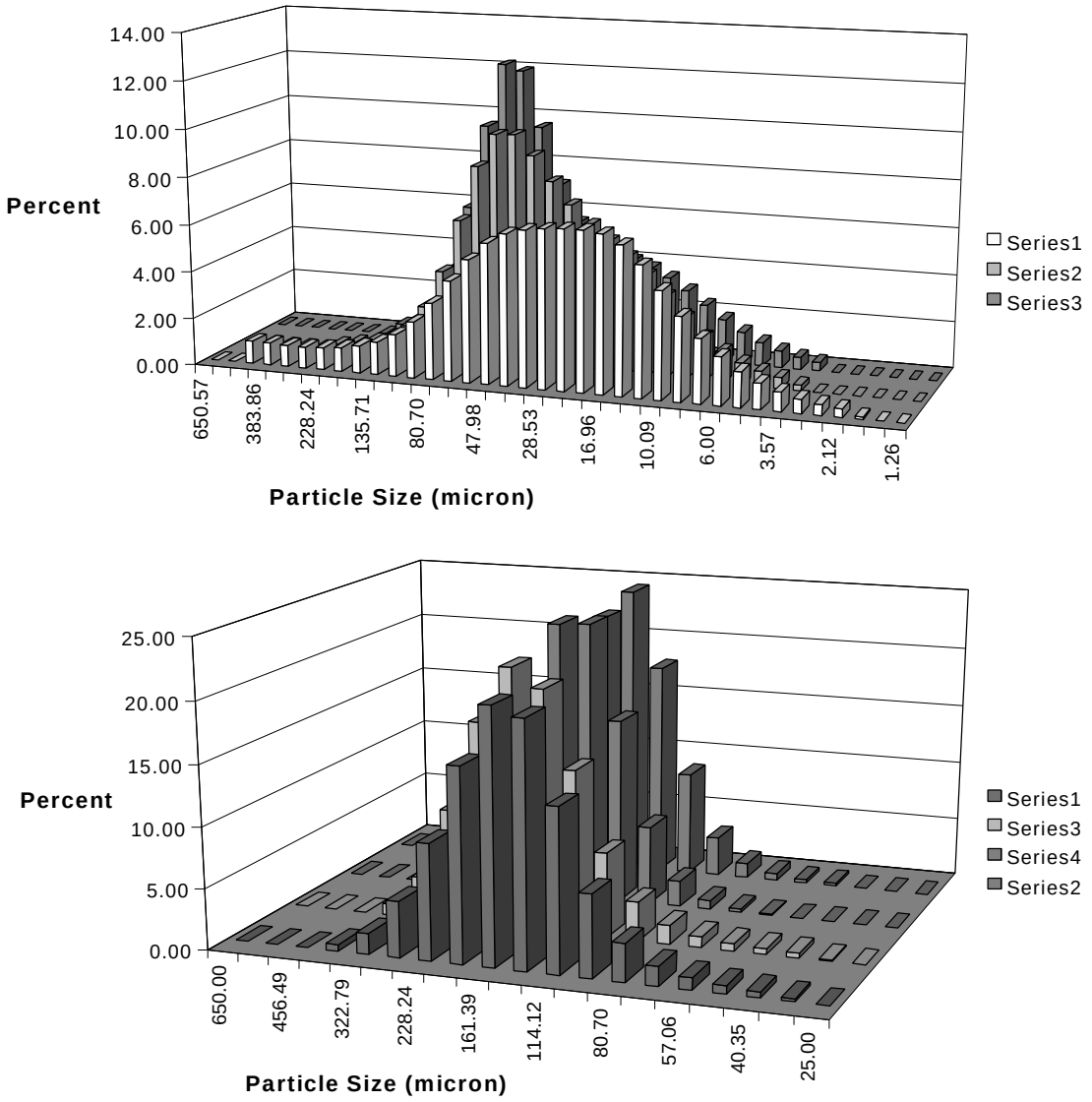

Figure 3. Plots of Particle Size for 40μ Target (top) and 180μ Target (bottom)

Table 2. Particle Size Experimental Parameters and Results

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

SUMMARY

Powder production by four different methods has been examined by the US-RERTR program. Cryogenic milling was found to be unsuitable due to ineffectiveness and high contamination. Grinding, while usable for near-term studies, is impractical for large-scale production and is also plagued by contamination. The hydride-dehydride process is a potential method for fuel powder production but may produce powder too fine for program needs. Atomization, a common commercial process, was examined in a surrogate study and could be made to meet program needs.

The US-RERTR program will continue to examine several different powder production methods to increase the prospects for success. The next US-RERTR irradiation experiment will consist primarily of powders produced by atomization and hydride-dehydride.

The hydride dehydride process will be examined to determine if it can be made into a viable powder production method. Production of powder in the desired size ranges may require an additional sintering step to coarsen the powder.

We also expect to experiment more with grinding as a production method. Specifically, powder produced by grinding will be used in future irradiation experiments to determine the effects of cold work on fuel performance.

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