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INNOVATIVE CONCEPTS FOR FUEL PLATE FABRICATION

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ABSTRACT

A number of fabrication concepts have been and are being explored at ANL. Although specific processes were addressed with silicide fuels in mind, most are applicable to fabrication with any fuel type. Processes include improved comminution procedures for converting U-Si alloy ingots to powder using a roll crusher and an impact mill. Aluminizing of core compacts by ion vapor deposition techniques in vacuum offers prospects for improved plate quality. Other items examined include the possible use of coatings on fuel particles, matrices different from pure Al, and ductile fuel alloys which might be used to produce fuel plates with uranium loadings higher than possible with conventional dispersed-phase powder metallurgy technology.

INTRODUCTION

As part of the ANL fuel development effort on the RERTR program, a number of processes were addressed to improve the quality of fuel plates and elements or to enhance manufacturing efficiency. Not all of these concepts proved successful; some have significant potential. It seemed in order to record the thoughts and activities of this phase of fuel development in a proceedings format so that the RERTR community might have the benefit of this work for any future application or consideration. As is true for any research activity, the time and effort devoted to any specific concept are dictated by budget considerations. Further, the availability (or lack of availability) of an irradiation environment to test some concepts precluded the possibility of bringing all studies to a logical conclusion.

DISCUSSION

Fuel Particle Coating

The irradiation performance of U_3Si/Al dispersion fuels at high burnup is considered to be relatively unsatisfactory. Coating fuel particles to preclude any Al (matrix and clad)-fuel reaction offered a potential solution to at least part of this behavior. Coated fuel particles are a reality in some applications but are not used in current research and test reactor elements. An assessment of the type and thicknesses of coatings required to serve as an effective diffusion barrier was made. Coatings of from 1 to 5 μm of Al_2O_3, Si, Ni or metal carbides are within the realm of existing technology and represented the final choices from an originally much longer list. Carbide coatings require temperatures in excess of the U_3Si peritectoid temperature while

Ni coatings require working with extremely hazardous materials. Both coatings were therefore dropped from further consideration. Si or Al_2O_3 at the thicknesses presumed to be required to serve as a diffusion barrier would diminish the maximum U loadings possible, assuming a 45 volume percent dispersed phase as a practical limit, to values comparable to those that could be achieved with U_3Si_2.

For these reasons, all considerations of coated fuel particles were discontinued and the concept has not been pursued further.

Ductile Alloys

Fuel plate fabrication procedures different from conventional dispersed-phase technology were considered with U-rich alloys fabricable to very thin sheet. Three compositions were chosen for study: U_3Si (96 wt.% U, density 15.2 g/cm^3), $U_{75}Ga_{15}Ge_{10}$ (91 wt.% U, density 15.8 g/cm^3), and $U_{75}Ga_{10}Si_{15}$ (94 wt.% U, density 15.6 g/cm^3). U_3Si may be reduced to sheet form but experiments demonstrated that hot rolling procedures are not simple and require protection from oxidation which renders the entire process technically feasible but very expensive and therefore commercially unattractive. No miniplates of any kind were produced with U_3Si sheet.

The Ga-containing alloys were selected from a total of ten alloys in this family on which experimental work was conducted. The preliminary screening included density and hardness measurements, metallographic and x-ray diffraction examinations and fabricability studies. The two Ga alloys chosen are quite soft and ductile and can be cold rolled, with intermediate anneals in a protective atmosphere, to sheet of virtually any thickness desired. With no irradiation performance data to support the potential value of continued experimentation with sheet fuel alloys in this category, it was decided that miniplates with dispersions of these alloys should be fabricated. Using a motorized flexible-shaft grinder and a carbide-tipped burr, particulate which can best be described as small chips was generated in a once-through nitrogen atmosphere glovebox. Because of the ductility of the Ga-containing alloys it was difficult to generate all particulate at -100 (<150μm) mesh. In order to proceed with experimental work in a timely fashion, miniplates were made with a range of particle size extending from ~250μm (60 mesh) to -325 mesh (<45μm).

As the miniplates containing the gallinide alloy "powders" were being completed, a decision was made to permanently shut down the ORR. Therefore, no irradiation experience has been generated with these fuels. Compatibility study plates with 20 volume percent gallinide fuel have been annealed at 400°C. The fuel zone volume growths of these unirradiated plates suggest a behavior poorer than that of U_3Si. Based on this very limited information and, at this time, no metallographic studies of the fuel zone in either the as-fabricated or annealed condition, it is suggested that neither the U-Ga-Si nor the U-Ga-Ge alloys are likely to be a panacea in looking for a high uranium density fuel for plate applications in an aluminum matrix.

Matrix Changes

In order to reduce or preclude fuel-aluminum reactions, several variations in the matrix powder used for the fuel cores were tested. Miniplates were made using prealloyed 6061 Al powder or ALCOA 718 Al powder. The 6061 powder is of the same composition as the clad used by ANL. There was a subjective perception that in irradiated plates the diffusion of Al into the fuel particles was less at the clad interfaces that at the pure Al/fuel interfaces. Limited irradiation experience indicated no apparent value to the use of the 6061 powder matrix. Further, the harder alloyed powder resulted in a less homogenous fuel distribution in the miniplates.

Several references (1,2) suggest that the diffusion of Al in U metal is reduced by a factor of from three to five if Al is replaced by an Al-Si alloy. To determine if an analogous reduction of matrix-fuel diffusion would occur with uranium silicides instead of U metal, ALCOA 718 powder (Al + 12 wt.% Si + 0.4 wt.% Fe) was used in the fabrication of a number of compatibility study plates with several U-Si alloy fuels. Thermal anneals at 400°C of unirradiated plates resulted in a fuel zone volume growth much more severe than that experienced with plates at similar fuel loadings and the conventional Al powder matrix. We have found in our work a correlation between unirradiated thermal compatibility and irradiation behavior (3). Therefore, no miniplates were ever fabricated for irradiation with the ALCOA 718 Al matrix.

Finally, Mg powder was used in two attempts to fabricate compatibility study plates. The use of Mg quite obviously eliminates any Al-fuel reaction except at the points of contact with the frame and cover plate. To prevent the formation of Al-Mg eutectic liquid, the hot rolling temperature for the first assembly which included six compacts, was reduced to 385°C. Except for this change, all regular cleaning, assembly, hot rolling, blister annealing and cold rolling procedures were followed. When the welded edges of the finished plate assembly were sheared away, there was no frame-to-cover bonding and the assembly came apart. A second attempt with Mg powder matrix and a hot rolling temperature of 415°C also resulted in a non-bonded product.

With further adjustments in the hot rolling parameters, this is still a viable procedure, in our opinion, for producing plates without an Al matrix. Other priorities required the abandonment of any further experimental work with Mg powder matrix plates.

Comminution

More efficient conversion of U-Si alloy ingots to powder has been of interest for some time. Procedures different from those employed by commercial fuel element vendors were examined; all experiments to date have been conducted with surrogate (non-U-bearing) materials.

A Retsch centrifugal grinding mill shown schematically in Fig. 1 was found to be quite unacceptable for this effort. Severe erosion of the components of the mill resulted in a gross contamination of the powder product and this type of equipment has been abandoned as a possible device for comminution.

Roll crushing with a commercially manufactured unit has been demonstrated to be effective for converting ingots (or ingot pieces) to granular material --10 mesh (<2 mm). The roll crusher is in effect a vertical rolling mill with corrugated rolls made from tool steel heated treated to a hardness of ~R_c58. Used with a speed control, the six-inch (15 cm) diameter rolls have worked efficiently to crush a Ni -10 weight percent Si alloy. Although no work has been done with U-Si alloys it seems safe to say that the roll crusher would be very effective for comminuting U_3Si_2. Experiments would still have to be conducted to demonstrate the applicability of this unit to U_3Si or to compositions between U_3Si and U_3Si_2 in either the as-cast or heat treated condition.

The Vortec impact mill shown schematically in Fig. 2 has worked quite well with surrogate fuel materials of a variety of types. The

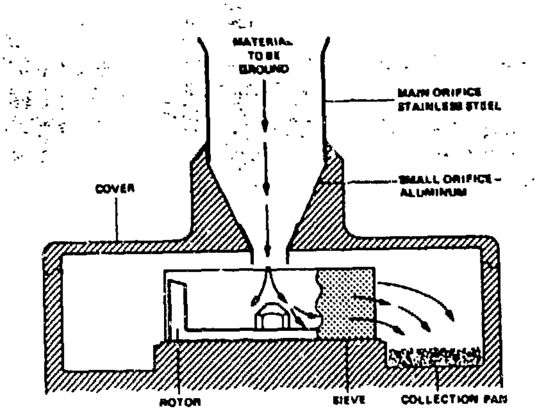

Fig. 1 Schematic representation of Retsch centrifugal grinding mill. For reference the overall diameter of the unit is ~9 in. (~23cm).

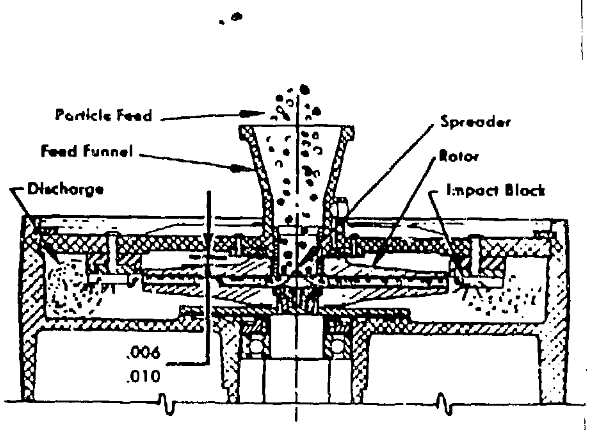

Fig. 2 Schematic representation of Vortec impact mill. For reference, the overall diameter of the unit is ~ 14 in. (~36 cm).

proof of the unit for silicide fuel remains a question that can only be answered by experiments in a neutral atmosphere glovebox. The Vortec does require as a charge material pieces no larger in any dimension than -0.38 in.(0.9 cm). Therefore, primary crushing with a roll crusher, or jaw crusher, if existing technology is to be used, to produce a product acceptable to the Vortec is necessary. The charge particulate for the Vortec should probably be considerably less than the maximum allowable by geometric considerations alone. The momentum of the particles must be reduced to a value which will allow efficient comminution without significant wear of the Vortec components.

Aluminum-Coating Compacts

Coating of core compacts with Al has been a consideration at ANL for several years. The reasons for conducting such a step include the following potential benefits:

- To prevent fuel particle oxidation during heating for hot rolling.
- To eliminate "white spots" (fuel-out-of-zone) in plates.
- To improve the integrity of the compact for easier handling and storage.
- To degas the compacts provided the process is conducted in a vacuum.

The general benefits of aluminizing compacts have been recognized and one vendor has already include such a step in their commercial fabrication procedure (4).

At ANL we have considered three processes:

1. The use of Al foil at the top and bottom of a compact charge before cold pressing.

2. The use of a small quantity of pure Al matrix powder beneath and on top of the core charge before cold pressing.

3. The use of an ion vapor deposition unit called the Ivadizer manufactured and marketed by the McDonnell Douglas Corporation (MDC) of St. Louis, MO, USA.

Of these three concepts no compacts were ever made at ANL using the Al-foil technique briefly described above. The procedure does seem to offer a high probability that stray fuel particles could not dislodge from the surfaces of the compact. The sides and edges of the compact, however, would probably not be protected. It is, in our opinion, worth trying.

Only two miniplates were fabricated implementing the second concept noted above. These plates, made with $\sim$50 volume percent fuel were neither better nor worse than conventionally produced compacts and plates. There was no time for further effort with this variation in compacting procedure and the sample size is too small for any definitive statement to be made regarding the value of such a step.

The treatment of compacts in the Ivadizer is being pursued. Ivadizer technology has been in commercial practice for over ten years and has been used to aluminize parts ranging in size from small machine screws to aircraft parts six or more feet long. Applications of the process have likewise covered a variety of materials including high strength steels, high strength Al alloys, depleted uranium penetrators, and composite materials. It is a well-established technology worthy of serious attention which, time and funds permitting, we plan to give it.

At the time of this presentation, twelve compacts, nominally of the size employed for ORR plates, were produced using W powder in lieu of silicide fuel. Prepared at 40 vol.% W, the compacts have been aluminized at the MDC plant in St. Louis. Characterization of the nature of the coating and its effectiveness in reaching the goals of an aluminizing process must still be addressed.

Ivadizer units operate in a vacuum of ~10^{-5} torr or lower, a level achievable with conventional vacuum systems. A 1 kV bias between the parts and the molten Al pool enhances the throwing power of the Al vapor. Compacts are easily supported in a rotating mechanism within the chamber to assist in ensuring a uniform Al deposit. Units usually are tailored to specific user-needs and have already been built, sold and used in sizes ranging from bench-top scale to those with chambers approximately six-feet in diameter by ten-feet long. Coating thicknesses of from 0.001 to 0.003 in. (0.02 to 0.8 mm) are possible in an operational cycle time of approximately two hours. Multiple compacts may be aluminized at one time in this batch-type procedure.

Concluding Remarks

The fuel plate fabrication concepts briefly presented here are to a large extent still unproven for silicide fuel element technology. The somewhat precipitous shut-down of the ORR has precluded generating any irradiation experience employing these ideas. As time, funds, and interest permit, however, methods for improved comminution and aluminizing of compacts which do not require any irradiation data to support the procedures if developed and will be pursued.

REFERENCES

1. L. S. DeLuca and H. T. Sumsion, "Rate of Growth of Diffusion Layers in U-Al and U-AlSi Couples," KAPL-1747, Knolls Atomic Power Laboratory, General Electric Co., May, 1957.

2. D. R. Green, "Diffusion of Uranium and Aluminum Silicon Eutectic Alloy," HW-49697, Pacific Northwest Laboratories, April, 1957.

3. T. C. Wiencek, R. F. Domagala and H. R. Thresh, "Thermal Compatibility Studies of Unirradiated Uranium Silicide Dispersed in Aluminum," Nuclear Technology, 71, Dec., 1985, p 606-616.

4. G. Thamm, "Proceedings of the 1986 International Meeting on Reduced Enrichment for Research and Test Reactors," ANL/RERTR/TM-9, Argonne National Laboratory, In Press. Meeting dates Nov. 3-6, 1986.