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MEMORANDUM TO FILE

October 25, 1956

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Trip Report
Date CERAMIC INFORMATION MEETING, ORNL
October 1, 2, & 3, 1956

The meeting was opened by Leo Brewer, who spoke on a "Self-Purifying Reactor". This is a reactor with theoretically designed-in separations processes, involving calculations to show that it is possible to vaporize many fission products away from fuels. The reactor would be analogous to a graphite rod impregnated with a uranium or thorium compound, vapor-coated with zirconium, and cooled by liquid metal. The fission products will vaporize when the rod gets hot enough, and will be carried out of the pile in a flow of helium. The details of what will happen to the rest of the pile at these temperatures, and of the materials of construction required to handle the situation are yet to be worked out.

BMI and GE, Lockland, got very much higher vaporization and transport of uranium than Brewer's calculations called for. In the presence of H_2 , GE observed high volatility of UO_2 at $2000^\circ K$. BMI pointed out that U in graphite at these temperatures rapidly becomes UC_2 , and that when UO_2 is placed in a graphite thimble and heated, they detect 5 micrograms per sq. cm. per hour outside the thimble at $1600^\circ K$.

Ackermann and Thorn, ANL, talked on "The Thermodynamic Stability of Refractory Oxides", covering Bureau of Mines Bulletin 542 and remarks on Al_2O_3 , MgO , UO_2 , ThO_2 , and ZrO_2 . They vaporize UO_2 gas out of W or Ta crucibles above $1600^\circ K$; the volatility from Ta crucibles is 500 times more than from W crucibles. They say that the melting point of UO_2 is $400^\circ K$ below the published value, based on finding a negative meniscus in their crucibles.

A. G. Thorp, ORNL, "Uses of Thermodynamics in Ceramics", pointed out with examples how calculations based on thermodynamic data were often useful.

L. G. Wisnyi, KAPL, "Determination of the Melting Point of UO_2 and Other Refractories", reported $2750^\circ C$ as the melting point of UO_2 , using the Mendenhall wedge technique (optical pyrometric method employing a hollow wedge made by bending a Pt ribbon back on itself). Recrystallization of UO_2 is rapid at $2680^\circ C$.

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S. M. Lang, National Bureau of Standards, commented on the stages of oxidation of UO_2 . This compound oxidizes rapidly in air at 500° to $600^\circ C$ to form a series of quasi-compounds intermediate between UO_2 and U_3O_8 . The latter is thermally unstable, and dissociates above $1200^\circ C$ into UO_2 . C. Brassfield, GE, ANP, said that a material corresponding nearly to U_3O_8 volatilizes rapidly in air between $1200^\circ C$ and $1400^\circ C$, and can be condensed and collected. Lang was the only one at the meeting who proclaimed that relatively little is known about uranium oxides.

J. F. Quirk of BMI presented a resume of his Battelle report on sintering BeO .

G. L. Ploetz, KAPL, discussed samarium, europium, dysprosium, and gadolinium oxides. They are all cubic at room temperature, and hexagonal at high temperature. For fabrication, they are mixed with 1% plasticizer and fired at $1500^\circ C$.

C. Hyde, BMI, "Uranium Oxide Ceramics", said that UO_2 is relatively new in the ceramic field, and has been most used as crucible material for melting uranium. It is non-plastic, and unstable on firing in oxygen. UO_3 is reduced by hydrogen to form UO_2 ; the latter requires high forming pressure for fabrication and high firing temperature (30,000 psi and $1600^\circ C$).

Ed Epreman, AEC, made an appeal for a more thorough investigation of UO_2 , particularly of an alleged change in volume upon melting. He had no data, but referred to J. Belle of WAPD. Belle said that UO_2 shrinks from 5% to 35% in volume upon melting, which suggests a phase change. Upon questioning, Belle said that this information was based on the appearance of a central cavity in slugs irradiated to their melting point. There was no further discussion when the writer discounted the idea that Westinghouse slugs had melted at Chalk River.

C. Henderson, presently of MCW, formerly of Linde Air Products Co., discussed a method of making dense UO_2 . This method employed the Verneuil technique by which synthetic ruby and sapphire boules are made. MCW has made similar boules of UO_2 , 1/2-inch in diameter by 9 inches long, with densities up to 10.96, although some porosity was evident. UO_2 appeared to volatilize to some extent in the flame, and the base of the boule oxidized to U_3O_8 if the flame were not kept reducing.

C. A. Arenberg, ANL, presented a paper on "Fibrous Metallo-Ceramics". He dispersed filaments of Mo throughout ThO_2 , and made some W- ThO_2 and Zr- ThO_2 bodies. The latter cracked on firing because of a phase change in the zirconium.

M. J. Snyder, BMI, "Properties of Some Refractory Fuel Compounds", reviewed BMI-1124.

C. H. Hoenig, ANL, discussed some work on a boiling reactor. They are making fuel elements of ThO_2 containing 2.5% to 10% UO_2 , natural and enriched; the uranium is added as U_3O_8 , and fired to convert to UO_2 , thereby avoiding cracking. They are canned in aluminum or stainless steel, and exposed to 500°F water. They expect a burn-up of 0.95 atomic per cent, but have encountered cracking at 0.05 atomic per cent burn-up. They expect to have a MTR loading in three months. Norton Co. is making ThO_2 - UO_2 elements for the ANL-MTR loading.

W. E. Roake, HAPCO, spoke on "Compacting UO_2 by Swaging". They employ Norton Co. fused and ground UO_2 with an original pack density of 6 gm/cc in one inch stainless steel tubing. The initial swage at 600°C is a 40% reduction; this is followed by five passes with 15% reduction each, for an overall total of 75% reduction. The final density achieved with Norton UO_2 is 90% of theoretical; with MCW oxide the best they can do is 82% of theoretical density. They have also canned the oxide in aluminum tubes, supported on the outside by stainless steel, and swaged the combination. Roake claimed that the UO_2 showed grain growth as a result of swaging, but he had no data before and after swaging on particle size distribution.

D. J. Bowers, BMI, reviewed BMI-1117, on the effects of additions of ZrO_2 , SiO_2 , Be, BeO , CeO_2 , TiO_2 , SiN , to UO_2 ; the mixtures were fabricated into compacts by hydrostatic pressing. The additives did not improve the compacts.

E. M. Baroody, BMI, reviewed BMI-1102, a theoretical study on thermal fracture of hollow ceramic cylinders.

Miscellaneous

The writer discussed uranium-aluminum alloys with J. L. Cunningham, ORNL. He is casting flat slabs in vertical molds 5 inches wide by 1 inch thick by 10 inches high; the slabs are 4500 gm. Alloying and melting are done at 1150°C (vs. 1400°C at BMI). Compositions ranged from 20 w/o to 50 w/o uranium in aluminum.

The slabs are rolled at 600°C in 2S aluminum picture frame with an initial breakdown pass of 20% reduction, followed by reductions of 30% to 40% per pass. There is a small amount of edge cracking in the 40 w/o to 50 w/o compositions, which ORNL attributed to hot shortness. They had no trouble extruding rods at 500°C, but had trouble extruding flats, which they attributed to poor die design.

They have had trouble with gas porosity in their melts, but this has been remedied by vacuum melting followed by a remelting operation before pouring.

They have had some trouble also with growth of the cast billets of high uranium concentration. The riser seems to grow, and cracks off at its junction with the hot top. Upon remelting the scrap heads where an excess of U-Al_x compound may have risen to the top, there has been evidence of pyrophoricity.

They are assembling 48 w/o plates now for MTR. They went from 2S to 6061 aluminum picture frame in order to prevent dogboning during roll cladding. As the 6061 does not roll-bond to the core, they have had to roll a foil of 2S onto the core first to act as bonding layer.

The information pointed to less homogeneity in their alloys than BMI is getting, so they said they would discuss the situation with BMI.

Zirconium was specified as cladding for the homogeneous reactor, but in the presence of UOSO₄ and irradiation, its corrosion resistance dropped by a factor of 5, and it was severely radiation damaged.

Aluminum looks like a better container for UO₂ at low temperatures, but it reduces UO₂ at high temperatures (Pickelshimer, CF-56-8-135). ORNL is interested in 45 w/o to 60 w/o UO₂ in Al, all clad in Al, for operation at 500°F. For higher temperature, a stainless steel cladding seems best.

The instability, growth, and shrinkage of UO₂ is attributed to residual gas when UO₃ is reduced by H₂ to UO₂. ORNL thinks that a vacuum preparation would make a better product.

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