Melted and Granulated Depleted Uranium Dioxide for Use in Containers for Spent Nuclear Fuel

Vitaly T. Gotovchikov[a], Victor A. Seredenko[a], Valentin V. Shatalov[a], Vladimir N. Kaplenkov[a], Alexander S. Shulgin[a], Vladimir K. Saranchin[a], Michail A. Borik[a*], Charles W. Forsberg[b],

All-Russian Research Institute of Chemical Technology (ARRICT)
33, Kashirskoe ave., Moscow, Russia, 115409, E-mail: chem.conv@ru.net
Oak Ridge National Laboratory (ORNL)
Bethel Wall Road, P.O. Box 2008, MS-6165, Oak Ridge, TN, USA, 37831

Abstract – *Induction cold crucible melters (ICCM) have the potential to be a very-low-cost high-throughput method for the production of DUO_2 for SNF casks. The proposed work would develop these melters for this specific application. If a practical system can be developed, it has a major impact on the economic viability of DUO_2 for a variety of applications, but especially for DU shielded spent nuclear fuel storage and transport casks. Experiments on depleted melting uranium dioxide (DUO_2) were carried out in ICCM of diameter 120 mm, power supply 60 kWt and current frequency 1.76 MHz. The samples of melted DUO_2 of density near to theoretical one were produced. There were experimentally proved an opportunity to produce melted DUO_2 from mixed oxides of depleted uranium (DU_3O_8) by reduction melting in high frequency ICCM. This allows to make low-cost process of DUO_2 production. Experiments on the addition of gadolinium oxide as alloying components into DUO_2 melt while in crucible in order to improvement neutron and gamma radiation-shielding, physical and chemical properties of the final solids were conducted. Tests melting with DUO_2 melt discharge from cold crucible were conducted and proved principal opportunity for process realization at continuous conditions that allows to develop high-throughput process. Thermal and electric calculation of ICCM parameters are performed and there is carried out design of the apparatus for DUO_2 granules production by dispersion method of the melt drained from ICCM.*

I. INTRODUCTION

Research and development is underway to build spent nuclear fuel (SNF) storage, transport, and disposal casks using shielding made with depleted uranium dioxide (DUO_2) in a DUO_2-steel cermet or a DUCRETE with DUAGG (DUO_2 aggregate) with selective additives in cementious matrix. Such casks would have superior performance (higher capacity for a given weight, resistance against assault, improved repository performance) and the potential to use the entire inventory of excess depleted uranium (DU) in the United States and Russia. The production of dense DUO_2 particles and aggregates with the appropriate properties is a significant cost in the production of these casks.

II. CONCEPT

The preparation of DUO_2 particles and aggregates for shielding could be produced from technologies that are extrapolated from the costly multi-step nuclear fuel pellet technologies. However, the DUO_2 product requirements for shielding applications are different than for nuclear fuel. Shielding applications require only modest purity and dimensional control. The DUO_2 particles need to approach the theoretical density of DUO_2 in order to maximize shielding effectiveness. The DUO_2 must not produce significant volatile gases when heated. Note that conventional cermet and DUAGG production methods involve high-temperature steps where gas generation could create major problems.

Melting the DUO_2 and allowing it to freeze will produce a product near 100% theoretical density and assure that the product produces no volatile materials upon subsequent heating. Melting is a one step process that provides an opportunity to include additives in the DUO_2 to modify its chemical or nuclear properties (e.g. neutron absorbers) to improve cask performance.

The technical barrier to high-throughput melting is about the 2900 ^{0}C melting point of DUO_2. Russia has developed cold-wall induction heated melters that have operated above 3000 ^{0}C. The proposed work would develop these melters for this specific application. A cold crucible melter is an induction heated crucible where the furnace sides are actively cooled to create a frozen wall of the material being processed that provides insulation between the cooling coils and the melt. This allows operation at temperatures that exceed the capability of existing high-temperature materials. Solid DUO_2 and other additives can be added to the melter while liquid DUO_2 is withdrawn from the melter. The liquid can be sent to a shot tower where the liquid stream is broken up and the droplets solidify as they drop to the bottom of the tower. Most perspective process for production DUO_2 granules from the melt will be process in which source material is DU_3O_8 that is a product of conversion of depleted uranium hexafluoride. It is potentially a high-throughput low-cost process [1].

III. EXPERIMENTAL STUDY

III.A. Tests of Melting Uranium Dioxide in ICMM

For development of technology was decided to conduct experimental study with zirconium dioxide (ZrO_2) as DUO_2 simulator at existing ICCM of cold crucible diameter 120 mm (Fig.1), power supply 60 kWt and current frequency 1.76 MHz.

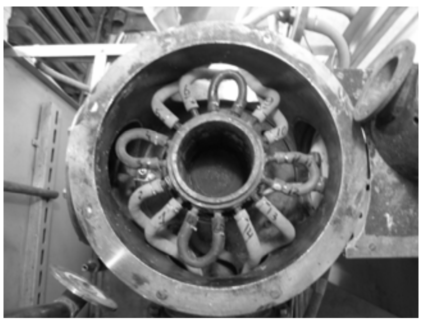

Fig.1. Induction cold crucible melter (from the top)

Process was conducted as follows: powder of zirconium dioxide by mass 1.5-2 kg. was charged in cold crucible on a graphite bottom. A start-up of heating was realized using zirconium wire ring, which was placed into powder at top coil level of three-coil inductor. When power was supplied to inductor zirconium ring and neighboring powder particles were heated up to temperature when zirconium ring melted. Temperature of zirconium dioxide located around zirconium melt was sharply raised. At high temperature zirconium dioxide conducts currents with frequency 1.76 MHz, heated more and melted. During melting process zirconium dioxide was periodically poured into crucible by portion into the melt. The melt was solidified into cold crucible by cooling water circulation through section.

The sample of produced monolith zirconium dioxide by mass 6 kg. are shown in Fig.2.

Fig.2. Monolith zirconium dioxide by mass 6 kg.

In next experiment test on zirconium dioxide melt draining was carried out (see Fig.3.).

Fig.3. Melted and drained zirconium dioxide

With the aim of production of UO_2 melt alloyed with neutron absorber, gadolinium oxide was added to source charge.

Melted uranium dioxide was very fragile and cracked into separate segments at cooling in crucible (Fig.4.).

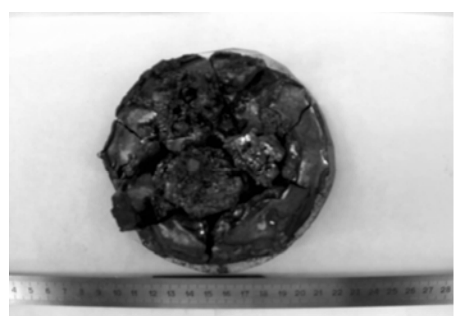

Fig.4. Melted depleted uranium dioxide (from the top)

Microstructure of melted uranium dioxide is shown in Fig.5.

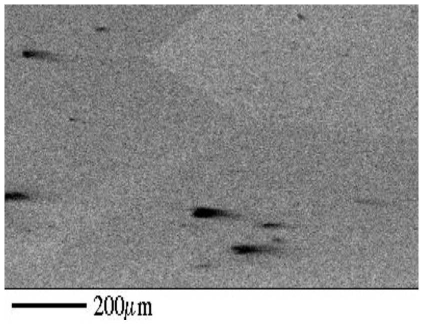

Fig.5. Microstructure and pores of melted uranium dioxide

The samples of melting products were subjected to X-ray analysis on difractometer by cobalt radiation with iron filter in range of 20-90°2θ.

It was found that powdered particles located on top and bottom parts of melted dioxide are mixed uranium dioxide (U_3O_8). Melted product was uranium dioxide with cubic lattice. Lattice constant *a* varied between 5.438 and 5.469. The last value corresponds to the theoretical one.

It is most likely that decrease in value *a* caused by generation of UO_2-U_4O_9 solution and inclusion of gadolinium oxide in structure [2].

From this it follows that during heating uranium dioxide is oxidized to mixed oxides and then uranium dioxide is reduced at high temperatures (above 1450^0C).

Uranium dioxide phase does not change during melting, solidification of the melt and the cooling of solidified product. It is accounted for dense monolith structure of UO_2 and probably protective layer of U_3O_8 powder.

From these facts it transpires that DU_3O_8 is able to use as source material for melted and granulated DUO_2 production. This allows exclude very expensive operation of DU_3O_8 conversion to DUO_2 and to make low-cost process of dense DUO_2 production.

Specific density of melted uranium dioxide measured by hydrostatic method was in the range from 10.15 g/cm^3, to 10.40 g/cm^3. This is attributable to presence of small gaseous pores (less then 1 mm) in uranium dioxide (see Fig.5, 6). Gaseous porosity will be excluded during technology improvement.

Analysis of gadolinium distribution in melted uranium dioxide was carried out in microsound analyzer "Camebax MS46" (Fig.6).

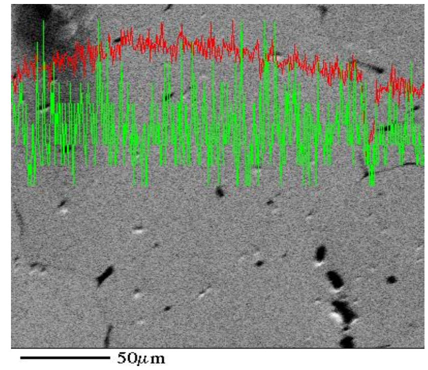

Fig.6. View in reflected electrons of melted uranium dioxide
Characteristics lines: upper - uranium
under - gadolinium

Metal gadolinium contents at five control points were (wt %): 1.26, 1.26, 1.20, 1.22, and 1.27.

It follows from this results that gadolinium oxide is uniformly distributed in melted uranium dioxide.

III.B. Tests of Cast Cermet DUO_2 – Steel by Impregnation Method Using of Melting Uranium Dioxide

System of uranium dioxide – stainless steel is system in which there are no wettability of solid surface of uranium dioxide by molten steel and solubility of components.

Interfacial tension of uranium dioxide – stainless steel melt is very high and surface forces prevent impregnation of liquid into pores and capillary.

In this connection melted uranium dioxide reduced to fine particles of 3-8 mm (bulk density 4.5 g/cm^3) was used in experiments (Fig.7). This range of size was determined by need of free space for molten steel penetration.

Fig. 7. Particles of melted uranium dioxide with diameter 3-8 mm.

Uranium dioxide and steel were alternately charged by portion of 15 grams into crucibles from aluminum oxide. Charge was heated up to 1600 ^{0}C in argon atmosphere. Uranium dioxide pores were impregnated with molten stainless steel. Then it was kept at temperature 1700-1800 ^{0}C during 5-10 minutes.

For the first time produced samples of real cast cermet were cast metal matrix with impregnated uranium dioxide particles (Fig.8). Experiments for choice of active doping agent improving wettability of uranium dioxide by steel were conducted. In present time optimal results were achieved by adding small quantities of cobalt and zirconium (1-3 wt %). Specific density of cast cermet samples was 9.20 g/cm^3. Experiments on searching metal active doping agent are continued now.

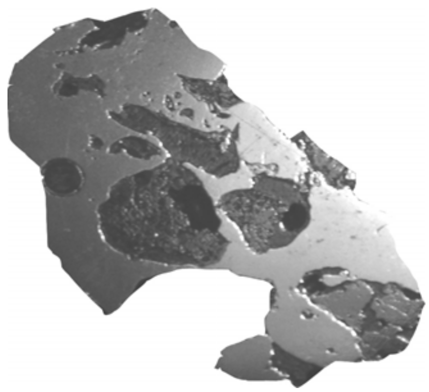

Fig.8. Cast cermet melted uranium dioxide – stainless steel (scale 2:1)

In our opinion technology for DUO_2 – steel cast cermet production is to include:

1. Production of granulated uranium dioxide by melting of uranium mixed oxides
2. Impregnation of uranium dioxide granules by molten steel.

It should be noted that properties of cermets produced by impregnation is much better then ones by powder metallurgy.

To improve wettability of uranium dioxide by molten steel surface, dioxide granules are needed to cover by copper and/or chromium oxides [3]. However, covering is very complete and expensive process. Besides, there are problems to achieve high adhesion of base and cover.

In this connection it is expedient to add doping agents in dioxide uranium at direct melting for wetting improvement. Metal active doping agents for wetting enhancement should be added to molten steel too. This complex approach allows to produce cermet with high physics-mechanics characteristics.

Based on conducted researches principal scheme of pilot apparatus for production of DUO_2 granules was developed (Fig.9). Design of apparatus is underway now.

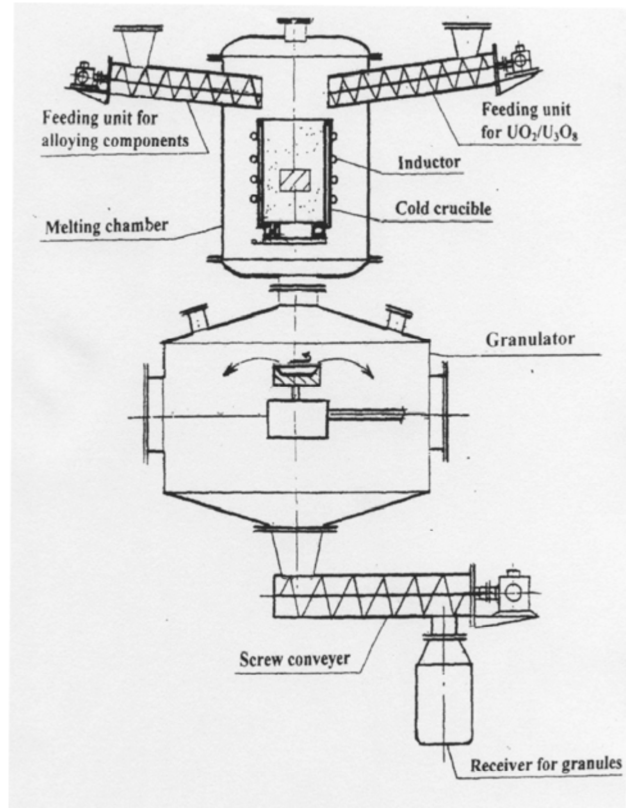

Fig.9. Principal scheme of pilot apparatus for production of DUO_2 granules

IV. CONCLUSIONS

There was experimentally tested melting process of DUO_2 in ICCM with frequency 1.76 MHz.

There were experimentally proved an opportunity to produce melted DUO_2 from mixed oxides of depleted uranium (DU_3O_8) by reduction melting in high frequency ICCM. Most promise process for production DUO_2 granules from the melt will be process in which source material is DU_3O_8 that is a product of conversion of depleted uranium hexafluoride.

It was experimentally proved that the addition of alloying components, (Gd_2O_3 and others) into cold crucible improves neutron and gamma radiation-shielding, physical and chemical properties of the final solid DUO_2

An opportunity of realization of uranium dioxide granulation process under continuous conditions was tested: charging source material into crucible and discharging the melt. Principal scheme of apparatus for the above process was developed.

Experiments on production of cast cermet using melted uranium dioxide were carried out. Conception for production of cast cermet using granulated uranium dioxide with active doping agents was developed.

REFERENCES

1. RUSSIAN FEDERATION PATENT # 2259903.
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* Michail Borik participated in experiments with zirconium dioxide only.