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Cermet fuel for fast reactor – Fabrication and characterization

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ABSTRACT

(U, Pu)O_2 ceramic fuel is the well-established fuel for the fast reactors and (U, Pu, Zr) metallic fuel is the future fuel. Both the fuels have their own merits and demerits. Optimal solution may lie in opting for a fuel which combines the favorable features of both fuel systems. The choice may be the use of cermet fuel which can be either (U, PuO_2) or (Enriched U, UO_2). In the present study, attempt has been made to fabricate (Natural U, UO_2) cermet fuel by powder metallurgy route. Characterization of the fuel has been carried out using dilatometer, differential thermal analyzer, X-ray diffractometer, and Scanning Electron Microscope. The results show a high solidus temperature, high thermal expansion, presence of porosities, etc. in the fuel. The thermal conductivity of the fuel has also been measured. X-ray diffraction study on the fuel compact reveals presence of α U and UO_2 phases in the matrix of the fuel.

1. Introduction

UO_2 & (U, Pu)O_2 mixed oxide (MOX) ceramic fuels have been used in various fast reactors world over and metallic (U, Pu, Zr) fuel is considered as the promising high breeding proliferation resistant fuel for the future fast reactors. The fuel cycle of MOX is well established. The advantages of the oxide fuel are its easy fabricability, good performance in the reactor and a well-established reprocessing technology [1,2]. The drawbacks of MOX fuel are its low thermal conductivity and low heavy metal density resulting in a lower breeding ratio and higher doubling time. Metallic fuel has been recognized as one of the candidate driver fuel for liquid–metal cooled fast reactor because of its inherently safe in-reactor performance and fuel cycle economics [3–9]. In Fast Breeder Reactor (FBR), the basic thermal and neutronic performance of metallic fuels is better than oxide ceramic fuels. The harder spectrum in the metallic fueled core results in high breeding ratio [3,4]. Metallic fuel is considered for future FBRs due to its high thermal conductivity, high fissile and fertile atom densities, low doubling time, etc. However, a few shortcomings of metallic fuels such as, low solidus temperature, high swelling rate and susceptibility to chemical and mechanical interaction with cladding materials prevent it from achieving its full potential. Therefore, the optimal solution might be to design a novel U–UO_2/U–PuO_2 cermet fuel which combines favorable features of both metallic and ceramic fuels [10]. Cermet fuel is a type of dispersion fuel consisting of particles of ceramic fuel uniformly distributed in a metal matrix. Presence of high density U metal in the fuel matrix gives the advantage of high breeding ratio of the fuel.

A cermet fuel is ideally designed to have the optimal properties of a ceramic, such as high temperature resistance and high hardness, and those of a metal, which improves the composite's thermal conductivity and mechanical properties. Cermet nuclear fuels have significant potential to enhance fuel performance because of low internal fuel temperatures and low stored energy.

As a cermet fuel, UO_2 has been used primarily in stainless steel matrices and to a lesser extent in refractory metal. The higher oxides U_3O_8 has been tested exclusively in aluminum and has been used in various research reactors. Zirconium and Zircaloy based UO_2 cermet fuel is being used in naval reactors [3].

Savchenko et al. [10] in their publication discuss about a new concept of Pu and minor actinides (MA) containing fuel for fast reactor. The U–PuO_2 fuel proposed is based on dispersion of PuO_2 particles in U or U alloy matrix. The study has been done using high density U alloys like U–Mo, U–Zr, and U–Zr–Nb. In the new fuel element design, a frame work fuel element having porous uranium alloy meat is filled with PuO_2 powder of <50 μm size. The high Uranium content fuel meat is metallurgically bonded to cladding and forms a heat conducting frame work.

Haertling and Hanrahan [11] have reviewed UO_2 dispersed in refractory metals. The most studied cermet has been W–UO_2 system. Other cermet system includes Mo–UO_2, W/Mo/Re alloys–UO_2 and Re–UO_2. Data are available primarily for W–UO_2 fuel. Failure mechanism of the cermet is typically degradation of mechanical integrity and loss of fuel. Techniques found to aid in retaining fuel include the use of coating around UO_2 fuel particles, use of oxide stabilizers in UO_2, minimizing grain size in metal matrix, controlling the cermet sintering atmosphere, etc.

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Cermet fuel has been considered as advanced fuel for the light water reactors (LWRs). Vatulin et al. [12] from Russia have worked extensively for the fabrication of this type of fuel. UO_2–Zr and UO_2–Al cermet fuel fabricated by traditional powder metallurgy route have been successfully tested in MIR research reactor in Russia. Their work reveals that in order to increase the stability of above mentioned fuel, oxygen to metal ratio of the fuel should be in the range of 2.15–2.25. With such oxygen to metal ratio, intensity of the reaction between matrix and ceramic material reduces. X ray analysis shows that bi-phase of UO_2–U_4O_9 forms in the fuel granules. Presence of U_4O_9 gives thermodynamic stability to the cermet fuel. Two basic methods namely low temperature impregnation and capillary impregnation have been developed by them to provide metallurgical coupling between fuel particle and matrix material [13,14].

In order to dispose plutonium through inert matrix fuel (IMF), the development of cermet fuel with PuO_2–Zr composite was carried out in Russia. It was concluded that PuO_2–Zr cermet system could be considered as a possible variant of new cermet fuel for Pu burning in LWRs [15]. The sub critical accelerator driven system (ADS) is now being considered as a potential means to burn long lived transuranium nuclides. In European Commission, JRC, Germany, the proposed fuel for ADS is Mo-92 cermet where plutonium and minor actinides will be dispersed in molybdenum matrix. To improve the neutronic characteristics, enriched Mo-92 will be required [16].

Sinha et al. [17] have recently discussed fabrication of U–UO_2 cermet fuel system for fast reactor and their characterization in terms of phase analysis and lattice parameter determination by X-ray diffraction technique.

The main objective of this study is to develop a flow sheet for the fabrication of the U–UO_2 cermet fuel by powder pellet route and carry out its characterization to evaluate its different thermo-physical properties.

2. Experimental

2.1. Fuel pellet fabrication

Uranium metal powder taken for the study was in the size range of 1–15 μm and was spherical in morphology as shown in Fig. 1. UO_2 powder was prepared from UO_2 pellets which were sintered at 1650 °C. The pellets were crushed in a jaw crusher and milled in an attritor. Sieveing was done to obtain UO_2 particle less than 75 μm size. The UO_2 powder was of irregular shape as shown in Fig. 2. The O/M ratio of the UO_2 powder was in the range of 2–2.015. Tables 1 and 2 represent the chemical compositions of U and UO_2 powder, respectively. Two compositions, namely U–15wt%UO_2 and U–30wt%UO_2 were made out of these powders. Uniform mixing of the powders was ensured by using a blender. Compaction was done at different pressure varying from 450 MPa to 1050 MPa using a double action hydraulic press at room temperature. Fig. 3 shows the flow sheet developed for the fabrication of the cermet compacts. The plot of compression ratio (volume of loose powder/volume of the compact) vs. compaction pressure of the mixed powder is shown in Fig. 4. Sintering of the green pellets was carried out in argon atmosphere at 1348 K for 8 h. The variation in density of the green and sintered pellet with pressure is shown in Fig. 5. One important concern in the handling of U metal powder is its pyrophorocity and oxidation when in contact with air. The oxidation of the powder may change the sintering behavior of the compact. Hence handling of the powder needs dry boxes filled with protective atmosphere like argon or helium. Dynamic flow of the argon gas was maintained inside the glove-box during the entire process of fabrication. O_2 and H_2O level in the glove box was maintained below 10 ppm.

2.2. Differential Thermal Analysis (DTA)

The phase transition temperatures of U metal powder and U–15wt%UO_2 sintered cermet were determined using DTA (Model No. Setsys Evaluation 24, combined TG/DTA/DSC; Make: M/s SETARAM Instrumentation, France). For DTA experiments, a sample of about 200 mg was loaded into an alumina crucible. During DTA operation, the furnace was purged with high purity argon gas flowing at 2 $dm^3\,h^{-1}$. To avoid oxidation of the sample, commercially available oxygen trap was used. The heating and cooling rates were programmed at 5 K min^{-1}. The temperature calibration was done by measuring the melting points of high purity metals, such as Zn, Al and Ag. The resulting thermogram was recorded. The instrument was evacuated with a standard mechanical pump and backfilled with argon several times prior to the tests. The phase transformation temperatures were determined from the heating curve.

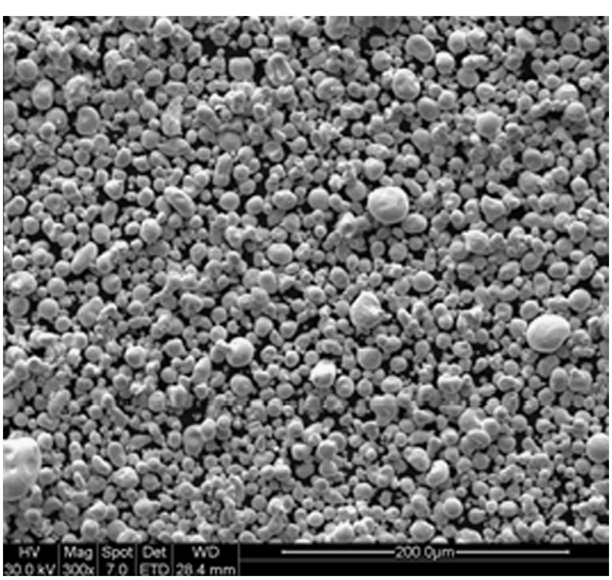

Fig. 1. SEM micrograph showing spherical morphology of U metal powder.

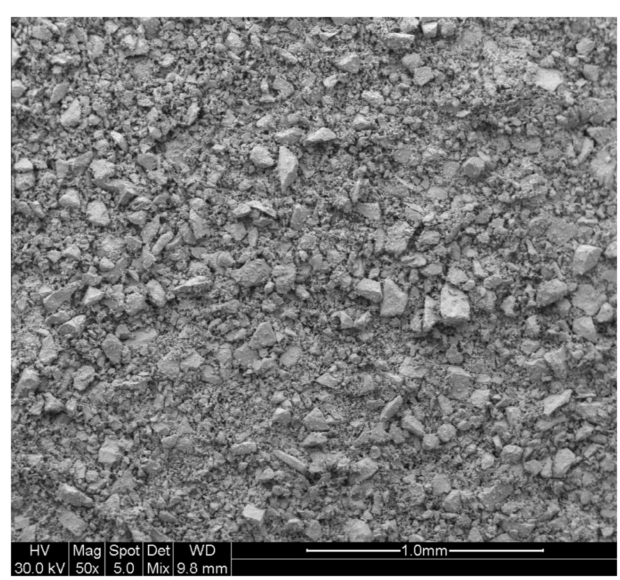

Fig. 2. SEM micrograph showing irregular morphology of UO_2 ceramic powder.