PREPARATION OF SOFT (Th, U)O₂ MICROSPHERES AND ITS PELLETIZATION BY SGMP TECHNIQUE

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Abstract

A sol gel assembly based on internal gelation process has been set up in Fuel Chemistry Division, Bhabha Atomic Research Centre to explore the feasibility of employing sol gel microsphere pelletization (SGMP) process for the fabrication of thoria – urania fuel pellets for advanced heavy water reactor AHWR. This assembly has the capacity of producing gel microspheres of thorium - uranium at a rate of 1kg/day. The dried and calcined microspheres were characterized with respect to size, surface area, tap density and crush strength and were directly compacted into cylindrical pellets and sintered in air at 1600°C to high density. The mixed oxide pellets were characterized for their density and microstructure. The results of the initial experiments indicate that high density sintered pellets suitable for use in AHWR could be fabricated by sol gel microsphere pelletization process.

Introduction

Advanced heavy water reactor (AHWR) has primarily been designed for the utilization of thorium resources in India. The reactor is a vertical pressure tube type using D₂O as moderator and boiling H₂O as coolant in natural circulation mode at low pressure. The 54 pin core of AHWR will consist of composite fuel of 12 pins of (Th-²³³U)O₂ with ²³³U enrichment of 3.0% in inner most ring, 18 pins of (Th-²³³U)O₂ with ²³³U enrichment of 3.75% in intermediate ring and 24 pins of (Th-Pu)O₂ with plutonium enrichment of 3.25% in outermost ring. ²³³U produced from the irradiation of ²³²Th is always contaminated with some amount of ²³²U which in turn, decays to hard gamma emitting daughter products ²¹²Bi (½ = 0.7 -1.6 MeV) and ²⁰⁸Tl (½ = 2.6 MeV). This calls for heavy shielding and remote handling facility for fabrication of (Th-²³³U)O₂ fuel.

Fabrication of (Th-Pu) MOX can be done without much difficulty using the powder oxide pellet (POP) route and existing facility available for preparation of (U-Pu) MOX. (Th-²³³U) MOX fabrication will however call for development of an automated and remote fabrication technique. The technologies which are presently under consideration for fabrication of (Th-²³³U) MOX fuel are: 1) Powder Oxide Pellet route, 2) Sol-Gel Microsphere Pelletization (SGMP), 3) Pellet Impregnation or Microsphere impregnation and 4) Coated Agglomerate Pelletization (CAP).

The microsphere impregnation technique¹ and CAP process² have been demonstrated successfully on a laboratory scale. Sol-Gel Microsphere Pelletisation (SGMP) process consists of preparation of free flowable microspheres by sol – gel process and its suitability for pelletization and sintering.
The advantages of SGMP process includes handling of dust free microspheres, excellent micro-homogeneity and amenable to automation and remotization. The present work was aimed at exploring the feasibility of employing sol-gel microsphere pelletization (SGMP) process for the fabrication of Th–233U fuel for AHWR.

**Sol– Gel microsphere pelletization (SGMP)**

The assembly for sol-gel process (Fig. 1) consists of a feed tank, a silicone oil tank, a delivery tube, a wash tank and an oil separator belt and an oven, all housed inside a walk in fume hood. A closed circuit zoom camera has been installed to monitor the droplet formation. In the feed, total metal concentration was maintained at 1.14 M and HMTA, urea to metal mole ratio (R) was kept at 1.48. Six trial batch runs of each 200 ml volume were carries out to test the various equipment of the assembly and to optimize the heat treatment conditions to obtain soft (Th,U)O₂ microspheres containing 4 mole percent UO₂ suitable for pelletization.

After optimizing the process conditions, a demonstration run of three litre volume batch was carried out to yield about a kilogram of mixed oxide microspheres for pelletization studies. The gel particles obtained were given four washes of carbon tetrachloride to degrease the gel microspheres free of silicone oil and then washed with...
6% ammonium hydroxide solution for removal of unreacted gelation agents and unwanted gelation products. The hydroxide microspheres obtained were dried at 100 °C for 8 hrs and at 250°C for 4 hrs in the oven. They were then calcined in air at 500 °C for 1 h to achieve the required tap density.

Calcined thoria – urania microspheres were used as feed material for direct pelletization in a double acting hydraulic press at 375 MPa. Green pellets were sintered in air at 1600 °C to high density.

Results and discussion

Some of the important physical characteristics of the heat treated microspheres & pellets are shown in Table 1. The results indicate that the microspheres would form an ideal feed for compaction into pellets. The green pellets obtained from the microspheres were of good quality without any berry structure and had a density of 49% TD. Sintered pellets were polished and thermally etched for metallographic examination. Microstructure (Fig. 2) shows equiaxed grain structure (average grain size ~10μm) without microsphere boundaries indicating that the microspheres have completely lost their identities during compaction and sintering resulting in high density.

References
