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Recovery and recycling of uranium from rejected coated particles for compact high temperature reactors

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HIGHLIGHTS

- The oxidation behaviour of coated particles was studied in air, O_2 and moist O_2.
- It was observed that coated layers cannot be completely removed by mere oxidation.
- Complete recovery of uranium from the rejected coated particles has been carried out using a combination of dry and wet recovery scheme.
- A crushing step prior to oxidation is needed for full recovery of uranium from the coated particles.

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ABSTRACT

UO_2 microspheres prepared by internal gelation technique were coated with pyrolytic carbon and silicon carbide using CVD technique. The particles which were not meeting the specifications were rejected. The rejected/failed UO_2 based coated particles prepared by CVD technique was used for oxidation and recovery and recycling. The oxidation behaviour of sintered UO_2 microspheres coated with different layers of carbon and SiC was studied by thermal techniques to develop a method for recycling and recovery of uranium from the failed/rejected coated particles. It was observed that the complete removal of outer carbon from the spheres is difficult. The crushing of microspheres enabled easier accessibility of oxygen and oxidation of carbon and uranium at 800–1000 °C. With the optimized process of multiple crushing using die & plunger and sieving the broken coated layers, we could recycle around fifty percent of the UO_2 microspheres which could be directly recoated. The rest of the particles were recycled using a wet recycling method.

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1. Introduction

A prototype Compact High Temperature Reactor (CHTR) is being developed by the Bhabha Atomic Research Centre as part of the Indian programme to produce hydrogen as a substitute for fossil fuels [1–3]. TRISO (TRI structural ISOtropic) coated particles (schematically shown in Fig. 1) will be used as the fuel in these reactors. A TRISO coated fuel particle considered here composed of a heavy metal (e.g., uranium, plutonium, thorium, etc.) oxide or mixed oxide/carbide fuel kernel (~500 μm diameter) coated with four layers of three isotropic materials. The four layers are: a porous carbon layer of around 90 μm thick (buffer), followed by a dense inner pyrolytic carbon layer (IPyC; 30 μm thick), a silicon carbide layer (30 μm thick), and a dense outer pyrolytic carbon layer (OPyC; 50 μm thick) in sequence. The buffer layer acts as a void volume for fission gas and CO. Its function is also to accommodate fuel kernel swelling and protects TRISO coating from fission recoil. The inner pyrolytic carbon layer retains the gaseous fission products and acts as a diffusion barrier for metallic fission products. It also acts as a mechanical substrate for SiC layer deposition. SiC layers are impervious to fission gases at normal operating temperatures and the SiC layer primarily acts as a structural material. The last layer, outer pyrolytic carbon layer retains gaseous fission products. This layer as well as IPyC layer, on irradiation, puts SiC layer into compression to limit stresses. Also it provides bonding layer with

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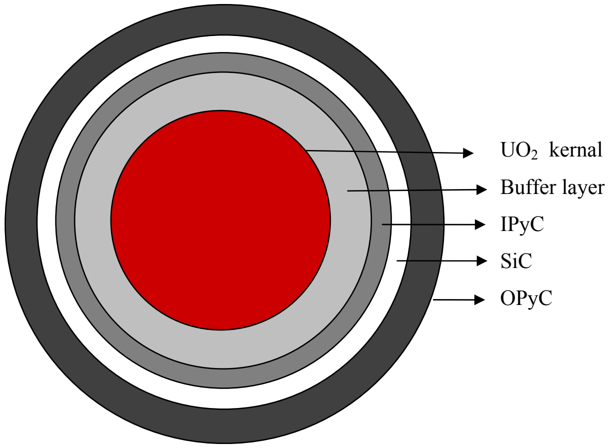

Fig. 1. Schematic of cross section of TRISO coated particle.

carbonaceous fuel element matrix. The TRISO coating fuel system acts as a pressure vessel which accommodates the internal gas pressures generated during fission of the fuel kernel material [4,5]. The TRISO coated particles to be used in these reactors should withstand high temperature of ~1500–1600 °C without any disintegration of the fuel particle [6]. The coated particle fuel is subjected to many forces that put stress on the TRISO coating. Potential failure mechanisms have been previously summarized [7]. These coatings of PyC and SiC on fuel kernel are formed by chemical vapour deposition (CVD) technique [8]. These coatings are obtained by pyrolytic decomposition of hydrocarbon gas or methyltrichlorosilane (CH_3SiCl_3) vapour in fluidised/spouted beds. A schematic of a spouted bed reactor is given in Fig. 2. Different coatings are prepared at different temperatures. All the layers are coated in an uninterrupted sequential process in the same fluidized bed reactor. The conditions under which layer deposition takes place are very important as these deposition parameters determine the properties and coating thickness of the coated particle formed. Parameters such as temperature, pressure, gas composition and gas ratios all play an important role in fixing the coated particle properties.

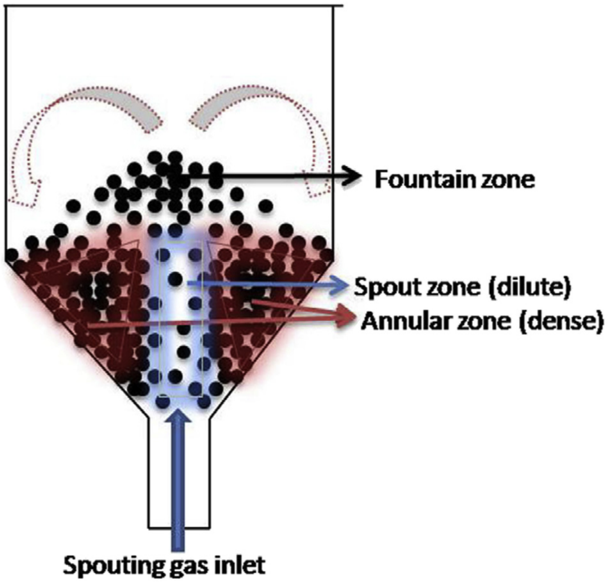

Fig. 2. Schematic of Spouted bed used for coating.

Since a very large number of TRISO particles (about 13.5 millions) [3] comprise the core, manufacturing of fuel kernels, deposition of multilayered coatings on the particles, and characterization of the coated as well as uncoated particles pose special challenges. It is very important to determine the fraction of defective coated fuel particle with respect to the specified coating thicknesses as the quality of the particles and, consequently, the acceptability of the fuel compacts and their irradiation performance is based on this parameter. The failure probability of a batch of fuel particles depends on statistical variations in the fuel design parameters as well as variation in the characteristic strengths of the coating layers in a batch. The fraction of defective or failed SiC layers is normally determined by burn-leach method and the gaseous acid leach method. In both methods the final step is extraction of kernel by leaching [9]. Recently, an integrated mechanistic coated particle fuel performance code known as “PARFUME” was developed at the Idaho National Laboratory which allows for statistical variations in the kernel diameter, the four layer thicknesses, the pyro carbon densities and several other parameters [10].

Since the fuel kernel contains ^{235}U or ^{233}U, recovery and recycling of these fissile materials from these failed/rejected coated particles are very much essential. The coated carbon layers should be removed by oxidation to recover the uranium from the failed/rejected coated particles. Several attempts have been made in the past to remove the coatings layer by layer but considered difficult regardless of the method employed. This is mainly due to the chemical nature and strength of the coatings, in particular the silicon carbide layer, which is rigid and non reactive with most of the acids and bases [11]. It has been reported that removal of the layers by ion sputtering technique does not result in uniform removal of the layers [12]. Removal of the silicon carbide layer by molten salts at 800 °C affects the distribution of fission products in the layer [13] for irradiated coated particle. In this paper, the oxidation behaviour of coated particles has been studied using Thermal Analyser and Temperature Programmed Desorption-Reduction-Oxidation (TPDRO) instrument. Based on these studies, a recovery procedure has been set up for the rejected coated particles (un irradiated). For these studies we have used sintered natural uranium oxide microspheres as a surrogate for $^{233}UO_2$ prepared by Internal Gelation Technique which were coated using CVD technique.

2. Material and experimental methods

2.1. Preparation of UO_2 microspheres

The UO_2 microspheres required for the coating experiments were prepared by Internal Gelation Process [14]. The dried UO_3 microspheres prepared by this method were heated at 1050 °C for 4 h in N_2+ 8% H_2 atmosphere. Further the spheres were sintered at vacuum/Ar +8% H_2 atmosphere at 1400 °C for 3 h. The density of these microspheres was determined by liquid displacement method [15]. The size (diameter) of the sintered microspheres was determined by optical microscope and specific surface area of the microspheres was determined by BET method [16].

2.2. Coating of carbon layers and SiC on UO_2 microspheres

The coating experiment was carried out in a spouted bed CVD furnace on natural UO_2 microspheres. Detailed reactor design had been reported elsewhere [17]. Spherical UO_2 microspheres of size range 500–600 μm and density 1055 ± 20 kg/m^3 were used for CVD coating. For coating porous buffer layer, acetylene (C_2H_2) gas was used. A mixture of acetylene and propylene (C_2H_2/C_3H_6) were used for coating IPyC and OPyC layers whereas, methyltrichlorosilane

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