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Immobilization of inert TRISO-coated fuel in glass for geological disposal

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Abstract

Vitrification of TRISO-coated gas reactor fuel particles was achieved via two methods: glass melting and sintering. Inert TRISO-coated fuel particles and a borosilicate glass were used. With glass melting at 1200–1300 °C floatation and decomposition of carbon and silicon carbide occurred. Thermal pre-treatment of the particles for oxidation of pyrocarbon did not improve the coating properties of the glass. During cooling most of the particles floated and sorbed on the crucible or mold walls. The sintered glass at 700 °C showed better coating properties of the TRISO-coated fuel particles despite higher porosity compared to glass made by melting. Aqueous leaching properties of glass with particles are similar regardless the mode of fabrication, indicating the good chemical durability of the sintered glass. Sintered glasses may constitute a good technique for TRISO-coated fuel particles immobilization for an eventual deep geological disposal.

1. Introduction

High temperature gas cooled reactors (HTR) are receiving more interest around the world due to their advantages including high thermal efficiency and safety advantages due to low-power density and large thermal capacity of the core [1,2]. Development of these reactors is being conducted in Germany, China, Russia, South Africa, United States, United Kingdom and Japan [2]. HTRs use sub-mm sized coated fuel particles embedded in the graphite matrix of fuel pebbles or block type fuel. The small fuel particles (kernels) surrounded by buffer carbon (BC), inner pyrolytic carbon (IPyC), silicon carbide (SiC) and outer pyrolytic carbon (OPyC) [3,4] are called 'TRISO fuel particles'. All coatings are designed to better confine gaseous and metallic fission products formed during the operation of the reactor.

Up to today and over the last 30 years works on TRISO-fuel particles were conducted for fuel fabrication technologies, fuel testing and performance under irradiation conditions [1–12]. Hence, in Germany the fabrication process of the fuel is so strict that only about a 100 defects were found in 3.3 million particles produced [11]. For a HTR power plant (with a thermal output of 200 MJ s^{-1}) and below

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1620 °C the TRISO-particles with SiC were found to ensure a high confinement of fission products [11]. Higher temperatures treatments of TRISO-particles (up to 2100 °C) have shown a 4–5 order of magnitude increase in Xe-133, I-131 and Cs-137 release [13,14] due to SiC deterioration. In Japan, TRISO-particles with zirconium carbide (ZrC) showed better confinement properties at high temperature compared to particles with SiC [15].

In Germany and the United States reprocessing of fuel for ^{233}U extraction constituted a recycling option since the 1960s. In Germany reprocessing of fuel was successful at a semi-technical test facility to make ^{233}U fresh fuel [2,16]. However, for non-proliferation aspects the reprocessing option was terminated in early 1980s. Merz et al. [16] demonstrated that the fuel particles embedded in the graphite matrix of the fuel pebbles could be directly stored in a salt mine in Germany. However, the direct disposal of fuel pebbles requires a very large disposal volume and more compact disposal configurations seem to be beneficial.

In the present work, we report on the immobilization of TRISO-UO_2 particles in glass for an eventual geological disposal. The glass constitutes an additional barrier for particles protection against groundwater corrosion and radionuclides dispersion in environment. Industrial glasses were proven to be efficient matrixes for fission products and actinides immobilization in most of the countries reprocessing civilian and military spent nuclear fuels (France, Germany, Belgium, United Kingdom, United States, Japan, Russia, etc.) [17]. The main objective was to study the feasibility of embedding the TRISO particles in glass matrix. Optimization of process and determination of the maximum load of TRISO particles in the glass are not parts of this work.

2. Materials and methods

Inert TRISO-UO_2 fuel particles were supplied by Forschungszentrum of Jüllich, Germany within the framework of the European project 'Raphael: reactor for process heat, hydrogen and electricity production' (Fig. 1). The overall diameter of the particles is 895 ± 11 μm and density measured by helium picnometry is $3.59 \pm 0.1\%$ g cm^{-3}. The glass used for particles immobilization is a borosilicate glass provided by CEA and that is being used as glass matrix for long-lived high-level waste vitrification in France; the R7T7 glass (Table 1). The glass density is 2.48 g cm^{-3}, T_g is 528 ± 8 °C and the melting temperature is about 1200 °C. The glass was used in the form of two powders prepared by CEA: $\varnothing < 63$ μm and 40 μm $< \varnothing <$ 140 μm corresponding to a BET surface area of 0.49 and 0.27 m^2 g^{-1}, respectively. The procedure of glass powder preparation can be found in [18].

2.1. Vitrification by melting

The glass powder was melted under oxic conditions in an alumina crucible placed in a high temperature furnace at 1250–1300 °C during 30 min for complete degassing. The TRISO-particles were then added to the melt right before pouring into a stainless steel mold ($35 \times 22 \times 10$ mm^3). Melting of the glass powder in presence of TRISO-particles lead to a deterioration of the fuel coatings. For each batch we used 10–20 TRISO-particles together with 5 g of glass powder. Samples ($10 \times 10 \times 1$ mm^3) were cut from the 'glass + TRISO-particles' composite and were polished down to 3 μm for microscopy and leaching experiments. Detailed observation of the glass/TRISO particle interface was conducted.

2.2. Vitrification by sintering

Sintered glasses offer several advantages in particular low processing temperatures. Glass powder and TRISO-particles were mixed before pressing at 400 bars. The resulting pellets were placed on an alumina plate in a furnace and the temperature was raised above the T_g up to 700 °C for a few minutes. At this temperature the pellet begins to swell and then to collapse but the porosity was significantly decreased. The temperature was then decreased and maintained at 680 °C for 3 h where sintering took place. In these experiments inert and pretreated (without OPyC) TRISO-particles were used. Composite chips were prepared for microscopy and leaching experiments.

2.3. Solid analysis

Polished 'glass + TRISO-particles' composite were analyzed under a JSM 5800 LV JEOL scanning electron microscope (SEM) equipped with a Kevex Energy Dispersive X-ray spectrometry (EDS) system. Thin sections of standard phases were used to obtain quantitative analyses. Corroded glass and composite samples were also observed under the microscope. On polished thin section the error is about ±5%.