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The effect of heat treatment on the microstructure and diffusion of silver in pyrolytic carbon coatings

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ABSTRACT

It is well accepted that TRISO (tristructural isotropic) coated nuclear fuel particles are capable of retaining fission products up to 1600 °C, however above this temperature fission products can diffuse through the pyrolytic carbon (PyC) and silicon carbide coatings that act as the containment barriers in this fuel. Despite decades of research and development, little is known on the origin of this fuel temperature limit. In order to understand the origin of this fuel temperature PyC coatings produced by fluidized bed chemical vapor deposition were heat treated at 1000 °C, 1400 °C and 1700 °C for 200 h in an innert atmosphere. We have observed that above 1400 °C the anisotropy, domain size and level of graphitization increases to twice its original value. Furthermore, at 1700 °C some samples exhibited the formation of nano-pores, which could be the origin of the maximum fuel temperature limit or at least contribute to it. The increased diffusivity of elements due to microstructural changes was corroborated by silver diffusion experiments. Furthermore, we have observed that not all the samples suffer the same level of graphitization, thus suggesting that some PyC coatings can maintain their capability to retain fission products even after temperature excursions above 1600 °C.

1. Introduction

The nuclear accident at Fukushima in March 2011 made evident the necessity of nuclear fuels with even higher tolerance to off-normal conditions. The TRISO (Tristructural Isotropic) coated fuel particles are among the options currently under study as accident tolerant fuels (ATF), and are an integral component contributing to the inherent safety of the High and Very High Temperature Reactors (HTR and VHTR). This type of fuel, first originated in the UK in the 1970s, is made of a uranium kernel coated with four layers of pyrolytic carbon (PyC) and one of silicon carbide (SiC) that operate as a miniature fission product (FP) containment vessel [1]. The kernel not only provides the energy in form of heat but also (partially) retains FP. The kernel is coated by a porous PyC coating layer, also described as the buffer, which attenuates fission recoil atoms and provides a void volume to accommodate gaseous fission products (e.g. krypton and xenon) and carbon monoxide. A dense internal PyC coating layer (IPyC) is deposited on top of the buffer layer, sealing this porous structure and acting as a containment of gaseous and metallic FP (e. g. Sr-90 and Cs-137). This dense PyC also prevents the chemical attack of the kernel during the production of the SiC layer, and gives a higher mechanical stability to SiC by introducing a compressive stress during irradiation. The SiC layer is the main diffusion barrier of metallic FP and provides most of the mechanical properties of the fuel. Finally, the outer PyC coating layer (OPyC) is the last diffusion barrier and ensures the SiC is under compression during irradiation. The purpose of these coatings is to stop the release of all fission products even under abnormal operating conditions [2–6].

In order to ensure the safe containment of all important radionuclides, the maximum operating temperature of this fuel has been set at 1600 °C. This temperature was established after experimental results showed that some FPs start to be released above this temperature. Although this value is well established, little is known on the origin of the release of FP at this temperature, especially if it is considered that the decomposition of the constituents of the fuel is expected to occur at temperatures above 2000 °C [7,8]. Therefore, to understand and even increase even further the safety of this type

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