



The measurement of silver diffusivity in zirconium carbide to study the release behavior of ^{110m}Ag in the ZrC TRISO-coated nuclear fuel particle



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HIGHLIGHTS

- Developed new diffusion experimental method in lieu of problematic existing method.
- Measured concentration profiles of Ag in ZrC after diffusion annealing.
- Firstly determined diffusion coefficient of Ag in ZrC at 1500 °C.

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ABSTRACT

The tri-structural isotropic (TRISO) coated particle fuel has been developed and used for high temperature gas-cooled reactors (HTGRs). It provides a unique robustness of the first barrier for the fission products. The TRISO fuel particle has typically consisted of a UO_2 or UCO kernel, surrounded by successive layers of porous carbon, dense inner pyrocarbon, silicon carbide, and dense outer pyrocarbon. During operation, however, the SiC layer has been known to release radioactive silver ^{110m}Ag which makes maintenance more difficult and thus costly. Zirconium carbide has been considered as a promising alternative to the SiC fission product barrier. ZrC exhibits high temperature stability and possibly possesses superior Pd resistance, while the retention properties especially for silver have not been adequately studied.

To help elucidate the diffusive behavior of silver in the ZrC coating of the TRISO-coated particle, a new diffusion experimental technique, called the *encapsulating source method*, has been developed by constructing a constant source diffusion couple between ZrC and Ag gas originated from Zr–Ag solid solution. Scanning electron microscopy (SEM), wavelength-dispersive X-ray spectroscopy (WDS), electron backscatter diffraction (EBSD) and optical methods were used to analyze the diffusion couple annealed at 1500 °C. The resultant diffusion coefficient of Ag in single-crystalline $\text{ZrC}_{0.84}$ at 1500 °C was experimentally determined to be about $2.8 (\pm 1.2) \times 10^{-17} \text{ m}^2/\text{s}$.

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1. Introduction

1.1. TRISO-coated particle fuel

The very high temperature reactor (VHTR), or high-temperature gas-cooled reactor (HTGR), is one of the six classes of reactor designs in the Generation IV initiative [1]. The VHTR reference

concept is a helium-cooled, graphite moderated, thermal neutron spectrum reactor with target coolant outlet temperatures of 1000 °C. Such high temperatures offer higher efficiency electricity generation and new opportunities for a broad spectrum of process heat applications, including hydrogen production by thermochemical water splitting or high-temperature electrolysis [2,3].

The fuel of choice for the VHTR is the tri-structural isotropic (TRISO) coated particle fuel [4–10]. The VHTR aims to achieve enhanced levels of safety, which is in part based on the capability of coated fuel particles to confine the radiological fission products even at high temperatures. The TRISO-coated particle is designed to act as a miniature pressure vessel; it withstands high temperatures

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and prevents significant release of fission products from the reactor core under normal and postulated accident conditions [9–13]. In the current reference TRISO-coated particle design, the kernel (microsphere) is typically formed from enriched uranium dioxide (UO_2) or uranium oxycarbide (UCO) and is surrounded by successive layers of porous pyrocarbon buffer, dense inner pyrolytic carbon (IPyC), silicon carbide (SiC), and dense outer pyrolytic carbon (OPyC), as schematically illustrated in Fig. 1. Note that this figure is drawn to scale based on the design used in the AGR-1 experiment at the Idaho National Laboratory (INL), USA [14]. A lot of TRISO-coated particles are packed together into spherical or cylindrical fuel elements to be used in reactor cores.

The SiC layer in the TRISO particles, in particular, provides structural support to accommodate internal gas pressure and serves as the primary barrier to the release of fission products. It retains gases and most metallic fission products below 1600 °C. It has long been known, however, that silver exhibits relatively high release from intact SiC-coated TRISO fuel particles if they are maintained at sufficiently high temperatures for sufficiently long times [15–23]. The release of radioactive $^{110\text{m}}\text{Ag}$ in particular is of considerable concern since this strong γ -ray emitter (half-life of 249.8 days) can plate out on the various helium-wetted surfaces in the primary circuit of direct-cycle HTGRs and thus significantly complicate plant operations and maintenance [24,25]. Furthermore, SiC begins to gradually lose mechanical integrity especially above 1700 °C, by transformation of β -SiC to α -SiC and later thermal decomposition [26–28]. It is also widely noted that the SiC layer is subject to attack from palladium and rare earth elements [21,29–34], although some studies led to contradictory results, showing significant Pd corrosion of SiC has not been identified [16,35].

1.2. ZrC and Ag retention

Zirconium carbide has been considered as a promising alternative to the SiC fission product barrier of the TRISO-coated fuel particles since the 1970s, since the peak fuel temperature permissible under operating and accident conditions would increase while still retaining a very high degree of passive safety [7,8,36–42]. It has been demonstrated that the ZrC-coated TRISO fuel possesses much greater high temperature stability than that coated with SiC [38]. In one Japanese side-by-side test of TRISO particles of both SiC (36 μm thick) and ZrC (25 μm thick), irradiated to a burn up of 4.5% FIMA for about 100 effective full-power days at temperatures ranging from 1400 to 1650 °C, there was complete coating failure of only one of 2400 particles with ZrC, as opposed to 20 failures in the same number of particles with SiC [40]. In other Japanese studies [38,43], irradiated particles with ZrC survived at much higher rates when subsequently exposed to high-temperature heating tests. When

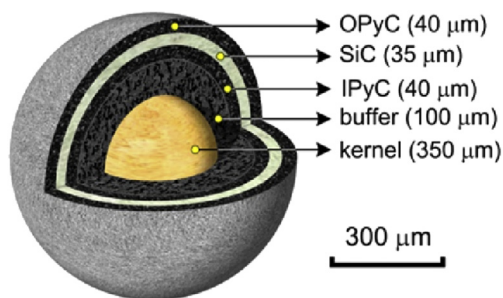


Fig. 1. Schematic cutaway drawing of a TRISO fuel particle showing the fuel kernel and coating layers. Note that the numbers in parentheses refer to the thicknesses of the coatings and the diameter of the fuel kernel.

heated to 2400 °C at 1 °C/min and held for more than 1.6 h, there was only a 1% failure of particles with ZrC, compared to 90% failure in particles with SiC by the time the temperature reached 2400 °C. The ZrC coating layer has also been known to exhibit immunity to Pd attack, although in a recent study Pd has not been found concentrated at the inner surface of ZrC layers, indicating possible migration through the coating rather than being retained [40,44].

With respect to Ag retention in the ZrC TRISO-coated particle, there are only a few studies that addressed this issue [44,45]. In order to assess the extent of silver retention in the ZrC layer under various operating conditions, it would be a good starting point to measure reliable silver diffusion coefficients in un-irradiated ZrC, although such a diffusion process is expected to be complicated further in actual service conditions by effects such as irradiation-enhanced trapping and adsorption.

Very few diffusion data on the transport of silver in zirconium carbide have been reported thus far, while there is a relatively large amount of research on that in silicon carbide (nevertheless, the Ag migration mechanism in the SiC coating layer has still not been well understood until now) [15,17,45–61]. This paucity of studies is not due to lack of interest but rather to the difficulty of carrying out experiments as discussed in the next section.

To the best of authors' knowledge, there is only one single study that explored Ag diffusivity in ZrC. In the former USSR, Chernikov et al. [45] investigated the diffusion behavior of Ag in ZrC (C/Zr unknown) coating layer over a wide range of temperatures (1730–2230 °C). $^{110\text{m}}\text{Ag}$ solution was deposited on the surface of what they called "simulators", which were unirradiated cylindrical ZrC pellets. After diffusion annealing, the distribution of the nuclide in the ZrC coating layer was determined by removing the ZrC stepwise until 10 μm thickness. Two linear regions with two different slopes were obtained in a diagram of the $^{110\text{m}}\text{Ag}$ activity versus the penetration depth squared. Chernikov et al. considered that the first region was due to the migration of silver in the grain volumes and the second region was due to that along the short circuit paths. However, they did not explain why the slope could vary depending on the diffusion depth.

Recall that it is well known that in polycrystalline metals the slope in an Arrhenius diffusion plot may vary with temperature (not diffusion depth) due to the change of the dominant diffusion mode, whether lattice or grain boundary diffusion. However the slope change with diffusion depth in likely homogeneous materials is not well known. We speculate that such a peculiar diffusion behavior is somehow associated with some of the experimental difficulties outlined in the next section. Therefore it seems worthwhile to revisit Ag diffusion in ZrC from a different experimental perspective.

1.3. Experimental difficulties measuring Ag diffusivity in ZrC

For Ag diffusion experiments in ZrC at high temperatures, such as 1500 °C, it is difficult to find an effective laboratory technique. Let us consider thin film method and the ion-implantation method as examples of experimental difficulties. Thin film method is used now almost exclusively for the measurement of diffusion coefficients, assuming that the entire deposited layer starts to dissolve instantaneously and diffuse into the matrix upon the initiation of diffusion annealing. To employ this type of method, a very thin section is deposited on the substrate applying various techniques. This thin layer contains atoms which are distinguishable from those in the substrate (or matrix). However, it is sometimes difficult to interpret diffusion profiles acquired using the thin film method mainly due to the following reasons [62]. First, deposited material tends to evaporate during the high temperature diffusion anneal. Secondly, deposited material lingers on the

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