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ZrN coating as diffusion barrier in U(Mo) dispersion fuel systems

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ABSTRACT

The control of the interaction between the U(Mo) fuel phase and the Al matrix is one of the challenges of dispersion fuel plate development for research reactors. Given the specific properties of this interaction layer, larger amounts of it in the meat could lead to a reduction of the plate mechanical integrity and thermal conductivity, eventually leading to pillowing. The SELENIUM project showed that by depositing a ZrN coating on the surface of the U(Mo) fuel particles, the amount of formed U(Mo)-Al interaction layer is limited but still present. Microstructural analysis performed on the as fabricated coating and fresh fuel plates containing ZrN coated U(Mo) dispersed in an Al matrix, revealed that the coating gets damaged during plate production. The post irradiation examinations (PIE) of the ZrN coated U(Mo) fuel plates, from the SELENIUM and SEMPER FIDELIS experiments, show how the U(Mo)-Al interaction layer is formed - only at those locations where the coating is missing or damaged - and the evolution of coating microstructure during irradiation. As a remedy, to further reduce the amount of interaction layer formed, the use of an Al-Si matrix was proposed based on the higher affinity of Si for U compared to the affinity of Al for U. PIE of a fuel plate consisting of ZrN coated U(Mo) dispersed in an Al-Si matrix irradiated in the SEMPER FIDELIS experiment, clearly demonstrates the benefit of adding Si to the matrix.

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1. Introduction

Low enriched U(Mo) has been selected as a viable candidate to replace the high enriched fuel currently still used in several high performance research reactors (HPRR). Two U(Mo) based fuel systems are being considered: a dispersion in which atomized U(Mo) particles are dispersed in an Al matrix or monolithic where a U(Mo) foil is clad with Al sheets. In the build up to qualification of this new fuel, several irradiation experiments have been performed. It was found that both dispersion and monolithic fuels suffer from detrimental interaction between the U(Mo) fuel and Al matrix or Al cladding that lead to poor in-pile behavior of the fuel systems. To limit or even eliminate the formation of such a deleterious U(Mo)/Al interaction layer, the application of a diffusion barrier was evaluated. The role of a diffusion barrier is to prevent or slow down the inter-diffusion of the two materials in contact. Therefore, to be effective, a good diffusion barrier requires inertness with respect to adjacent materials.

In the case of the monolithic fuel system, a pure zirconium layer is employed for the purpose of controlling the U(Mo)-Al interdiffusion at the foil/cladding interface during both plate fab-

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rication and irradiation. A barrier thickness of ~25 µm is usually selected to exceed the maximum fission fragment recoil range (~9 µm in Zr). For U(Mo) dispersion fuel the application of coatings directly on the powder particles surface by physical vapor deposition (PVD) [1], was expected to limit or possibly even avoid the interaction between the fuel and the Al matrix altogether. Even if the fission products are not stopped, the diffusion barrier does prevent the recoil atoms of the fuel phase and the matrix to reach each other, thereby preventing interaction. The SELENIUM (Surface Engineering of Low ENrIched Uranium-Molybdenum) fuel development project of SCK-CEN [2] consisted of the production, irradiation and post-irradiation examination of 2 flat, full size plates containing coated U(Mo) atomized powders dispersed in a pure Al matrix. Next to silicon, ZrN was selected as coating material as it is metallurgically inert towards U(Mo) and Al. The choice of ZrN was also based on the experience of the Russian MIR irradiated mini rods containing ZrN coated U(Mo) [3]. The post irradiation examination (PIE) clearly showed the positive effect of the coatings; for both Si and ZrN coated fuel a virtual absence of reaction between the U(Mo) and the Al is observed up to high fission densities after which a U(Mo)/Al interaction layer has formed. Even though both coatings have a clear effect on the kinetics of interaction layer formation, their behavior and evolution is completely different under irradiation [4]. Whereas the Si coating interacts with U(Mo) during plate production and the resulting U-Si rich interaction layer inter-

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