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ABSTRACT

This letter extends the work performed at Argonne National Laboratory on the QUADRISO fuel particles concept for High Temperature Reactors. Different configurations of the QUADRISO fuel particles concept are proposed and examined. The concept of QUADRISO fuel particles allocates an extra burnable poison layer next to the fuel kernel to reduce the initial excess of reactivity and enhance the reactor performance. The alternative proposed configurations introduced in this letter compare the performances of the previous configuration with two alternative configurations where the burnable poison is mixed in the fuel kernel for simplifying the manufacture process or in the outer pyrocarbon layer.

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1. Introduction

The QUADRISO conceptual design of coated fuel particles for High Temperature thermal Reactors (HTRs) has been developed at Argonne National Laboratory [1–4]. The new fuel design concept includes an extra layer of burnable poison next to the fuel kernel of the TRISO particle to better control the excess reactivity, consequently the new type of fuel can be referred as QUADRISO (Quadruple Isotropic) particles (Fig. 1). At beginning of life, when the core is loaded with both ordinary TRISO and new QUADRISO particles, the fuel in the QUADRISO particles is not 'seen' by neutrons because of the high absorption rate in the burnable poison. During irradiation, the burnable poison depletes and the fission reaction rate in the fuel of the new QUADRISO particles increases. This compensates the fuel depletion of the ordinary TRISO particles so that the reactivity curve is considerably flattened during the fuel cycle. The QUADRISO fuel particles concept has been inspired by the YALINA Booster configuration of Belarus, where a fast zone is coupled with a thermal one through an absorber layer [5, 6].

This work addresses some of the aspects of the QUADRISO fuel particles concept concerning the manufacture feasibility. For oxide or carbide fuel kernels the burnable poison is oxide or carbide form, respectively. Consequently, no additional transition layer is used between the kernel and the burnable poison. In order to coat

the europium oxide (Eu₂O₃) layer around the fuel kernel, a chemical vapor deposition (CVD) directly on the fuel matrix is suggested. The CVD technique for lanthanide oxide films is described by Shiokawa et al. [7]. In the present case, the CVD method uses β -diketonate metal chelates with 2,2,6,6-tetramethyl-3,5-heptanedione and some reactant gases as starting materials. The deposition is performed at atmospheric pressure and 843 K temperature. At present, the deposited Eu₂O₃ has a poorly crystalline film, which needs further improvement.

For the transmutation of neptunium and plutonium, the fuel is Np_{0.05}Pu_{0.95}O_{1.7}. The thermal expansion of the fuel is estimated to be 11.6×10^{-6} between room temperature and 1473 K (Ref. [8], for $Th_{1-x}Pu_xO_2$, where Ce surrogates Pu) and the thermal expansion of Eu_2O_3 it is reported to be 10.3×10^{-6} between room temperature and 1273 K (Ref. [9]). Consequently, the linear expansion of the burnable poison layer is 11% lower than the linear expansion of the fuel. This difference in linear expansion is small, therefore an expansion induced separation (spall off) of the coating due to thermal stresses is unlikely. Moreover, the poison layer is surrounded by the porous carbon layer, which would eventually hold up a separation of the coating. Besides the spall off effect, the whole restructuring of the kernel can influence the burnable poison layer. The article of Miller [10] exemplifies how important such a restructuring of the fuel kernel can be since it can introduce some cracks into the porous carbon layer and the inner pyrocarbon coating. Irradiation experiments are necessary to accurately predict the effect of the fuel kernel restructuring. Another important material aspect is the oxygen diffusion into the carbon layer, and the carbon diffusion in the oxide layer, both leading to the formation of carbon monoxide (CO) and Carbon dioxide (CO₂). This has been extensively researched for UO₂ kernels in ordinary TRISO particles, [11] but data do not exist for Np_{0.05}Pu_{0.95}O_{1.7} fuel kernel in QUADR-ISO particles. Generally the interaction between LnOx and C is



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Fig. 1. QUADRISO particle. The fuel kernel (pale-green) is surrounded by layers of: burnable absorber (orange), porous carbon (light-blue), inner pyrocarbon (blue), silicon carbide (yellow) and outer pyrocarbon (blue). (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)

easier if an intermediate valence state forms between Ln(III) and Ln(0). This is the case for Eu that could be reduced in Eu(II), while the reduction of Ln(III) by C is known to yield thermochemically the carbides and consequently carbon dioxide. An alternative candidate to europium oxide/carbide is erbium oxide/carbide [1–4]. More important materials aspects of different absorber materials, including Eu₂O₃, can be found in reference [12].

2. Different configurations for reactivity control

This study proposes new burnable poison configurations using the QUADRISO fuel particles concept. Fig. 2 compares the multiplication factor for six different burnable poison configurations as



Fig. 2. Neutron multiplication factor as function of time for different burnable poison configurations. Standard deviation lower than 60 pcm.

function of time for a prismatic HTR [1–4]. The six different configurations have the following features:

- 1. No poison is applied in the core and all the fuel uses ordinary TRISO particles. The porous carbon layer thickness of the TRISO particles in the central ring is $160 \mu m$ and the porous carbon density is decreased to have the same carbon mass as the ordinary TRISO particles. The six corner pins of each hexagonal block in the central ring are filled with pure graphite.
- 2. Eu_2O_3 QUADRISO particles are used with 10 µm absorber layer in the fuel pins of the central ring. The porous carbon layer thickness of the TRISO particles in the central ring is 150 µm with no density adjustment. The six corner pins of each hexagonal block in the central ring are filled by pure graphite.
- 3. The Eu₂O₃ layer of the QUADRISO particles in the fuel pins of the central ring is replaced by porous carbon. The porous carbon layer thickness of the ordinary TRISO particles in the central ring is 160 μm and the porous carbon density is decreased to have the same carbon mass as the ordinary TRISO particles. The mass of Eu₂O₃ of configuration two is reallocated and homogenized with graphite in the six corner pins of each hexagonal block of the central ring.
- 4. The Eu_2O_3 layer of the QUADRISO particles in the fuel pins of the central ring is replaced by porous carbon. The porous carbon layer thickness of the TRISO particles in the central ring is 160 µm and the porous carbon density is decreased to have the same carbon mass as the ordinary TRISO particles. The mass of Eu_2O_3 of configuration two is reallocated in the form of microspheres in the six corner pins of each hexagonal block in the central ring and the corresponding graphite density of the six corner pins has been accordingly increased to maintain the same fuel to moderator ratio as the previous configurations.
- 5. The Eu₂O₃ layer of the QUADRISO particles in the fuel pins of the central ring is replaced by porous carbon. The porous carbon layer thickness of the TRISO particles in the central ring is 160 μ m and the porous carbon density is decreased to have the same carbon mass as the ordinary TRISO particles. The mass of Eu₂O₃ of configuration two is reallocated and homogenized with the fuel kernel. The six corner pins of each hexagonal block in the central ring are filled by pure graphite.
- 6. Eu_2O_3 of the QUADRISO particles in the fuel pins of the central ring is replaced by porous carbon. The porous carbon layer thickness of the TRISO particles in the central ring is 160 µm with a decreased density to have the same carbon mass as the ordinary TRISO particles. The mass of Eu_2O_3 of configuration two is reallocated and homogenized with carbon in the outer pyrocarbon layer. The six corner pins of each hexagonal block in the central ring are filled by pure graphite.

The results illustrated in Fig. 2 show that mixing the burnable poison in the kernel provides a similar reactivity as function of time relative to QUADRISO particles. Mixing the poison in the outer pyrocarbon layer offers a better performance from the reactivity point of view. However, this configuration might have some problems if the radiotoxic isotope ¹⁵²Eu migrates into the coolant channels. The actinides burnup for all the analyzed configurations is rather similar.

3. Quadriso particles utilization benefits

The use of burnable poison in the fuel particle results in several benefits for transmuting neptunium and plutonium in thermal HTRs [1–4]. First the reactivity of the reactor core is considerably flattened during the fuel cycle. Second, the reactor can operate with a very low fuel packing fraction (only 7%); consequently,



Fig. 3. Fast fluence in the kernels for the deep burn fuel cycle based on QUADRISO particles proposed in reference [3].



Fig. 4. Failure fraction of TRISO particles in an accident scenario at the end of irradiation when next generation burnable poison designs are applied to a one pass deep burn [3].

the neutron spectrum is more thermalized and the fuel residency time for a specific burnup rate is considerably shortened. Fig. 3 shows the fast fluence in the fuel kernels when the one pass deep burn takes advantage of the QUADRISO fuel particles concept. The fast fluence is approximately the same as for the fuel cycles based on ordinary TRISO particles, as it can be seen by comparing with Fig. 4 of reference [13]. However, the fuel residency time is reduced by about 35%. The utilization of the QUADRISO fuel particles concept, based on two irradiation periods of 500 and 200 days, transmutes 52-58% and 95-97% of actinides and ²³⁹Pu, respectively [3].

The temperature excursion during a hypothetical accident scenario is illustrated in the top plot of Fig. 4 and the failure fraction associated to this hypothetical scenario, calculated by the PANAMA code, [14] is illustrated in the bottom plot of Fig. 4. In the latter plot it has been assumed that the average irradiation temperature is 800 or 900 °C during the 700 irradiation days. The results of the PANAMA code have shown an excellent resistance of TRISO particles for 100 h during a hypothetical accident scenario with a maximum temperature of 1570 °C.

4. Conclusions

The QUADRISO fuel particles concept proposed and analyzed at Argonne National Laboratory can be implemented into different configurations that include: adding an extra burnable poison layer next to the fuel kernel, mixing the poison with any of the coated layers or with the fuel kernel. From the neutronic point of view this study has shown small differences between the alternative configurations and all of them perform much better than the traditional design of the burnable absorbers that allocate the poison in the six corner pins of the hexagonal blocks. Irradiation experiments can determine which burnable poison configuration is the best.

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