Spent nuclear fuel/water interface behavior: Alpha dose rate profile determination for model surfaces and microcracks by using Monte-Carlo methods

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ABSTRACT

This work aims to better understand the nature and evolution of energy deposits at the UO_2/water reactional interface subjected to alpha irradiation, through an original approach based on Monte-Carlo-type simulations, using the MCNPX code. Such an approach has the advantage of describing the energy deposit profiles on both sides of the interface (UO_2 and water). The calculations have been performed on simple geometries, with data from an irradiated UOX fuel (burnup of 47 $GWd.t_{HM}^{-1}$ and 15 years of alpha decay). The influence of geometric parameters such as the diameter and the calculation steps at the reactional interface are discussed, and the exponential laws to be used in practice are suggested. The case of cracks with various different apertures (from 5 to 35 μm) has also been examined and these calculations have also enabled new information on the mean range of radiolytic species in cracks, and thus on the local chemistry.

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1. Introduction

For many years, studies to better understand the alteration process at the UO_2/water interface under alpha irradiation have been carried out internationally in order to be able to predict the long-term behavior of spent fuel in deep geological repositories [1—11]. The alpha irradiation from the actinides present in the fuel will dominate over the long term, and may thus lead to an oxidizing dissolution of the UO_2 matrix by the production of oxidizing species through alpha radiolysis of water. Alpha particles, whose energy is about 5 MeV, typically have a high linear energy transfer value (LET) and relatively short paths (a few microns) within the media they pass through, which can induce considerable fluctuations in local chemistry. Determining the energy deposits between the solid and the solution is therefore essential in order to better understand the physical and chemical evolution of the UO_2/water reactional interface. Several works have sought to describe the energy deposit profile in water at the reactional interface by using analytical

approaches which integrate the geometry of the system studied [12,13]. These have given decreasing exponential functions with a maximum energy deposit at the extreme surface of the fuel. The present work detailed here implemented another calculation approach, based on Monte Carlo-type simulations carried out using the MCNPX code, firstly for comparisons but also in order to add to and complete existing results. Thus the energy deposit profiles were calculated both in the water and in the UO_2 solid, which had never been done previously. Different geometries were taken into account, i.e. spheres, pellets, and different sizes of cracks, typical of those found in a fuel irradiated in a reactor. The complete set of results is given here, and first discussed as concerns the geometry and the calculation hypotheses ("refinement steps"). Then the general equations able to fit the profiles are proposed, and the consequences on the physics and the chemistry of the system are described.

2. Method and calculations

2.1. The MCNPX simulation code

To simulate the passage of alpha particles through the fuel from

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which they are emitted into water, the MCNPX calculation code [14] was selected because of its ergonomics in designing materials, geometrical shapes and radioactive sources as well as its ability to handle scales from centimeter to micron, necessary when focusing on alpha ranges.

MCNPX is an extension of MCNP (for Monte Carlo N-Particle Transport Code), a particle transport code based on the Boltzmann transport equation solver by the repeated random sampling Monte Carlo method [15]. Each particle has its own "history" and is followed from its emission to its disappearance. When the particle passes through matter, it leads to a large number of events (interactions) which are taken into account using physics equations and cross-sections representing the interaction probabilities, available in libraries. The results are obtained by tallying events (e.g. number of particles crossing a surface, or number of particles that have undergone a collision in a finite volume of the geometry …) and they are statistical. Thus the code gives results averaged over the events with the statistical error (standard deviation) [16]. In this study, an alpha particle was no longer considered once its energy decreased below a 1 keV cut-off energy.

2.2. Description of the modeled systems

First, the characteristics of the modeled spent fuel are described, followed by the three kinds of spent fuel/water interfaces of increasing complexity which were simulated.

2.2.1. Spent fuel features

This study chose to consider the features of a "fresh" spent fuel with a burnup of 47 GWd.t_{HM}^{-1}. The fuel was initially enriched to 3.1 wt%^{235}U and then irradiated in the French Fessenheim reactor for four cycles. The most important radionuclides contributing to alpha activity are detailed in Ref. [12]. They lead to an alpha activity of 4.73×10^8 Bq. g_{HM}^{-1} after 15 years of alpha decay. The average value of emitted alpha particles is 5.3 MeV. The simplified chemical composition considered in the calculations is $^{238}UO_2$, with a mass density of 10.8 g cm^{-3}.

2.2.2. Spherical and cylindrical geometries

Two different geometries were considered to describe the UO$_2$ fuel/water interface. First, to give a very simple approach to the spent fuel/water interface, a spherical spent fuel "fragment" was considered as a homogeneous material within which alpha particle emission was isotropic and homogeneous, and which was surrounded by water. Second, in order to be more representative of the real UO$_2$ fuel shape, cylindrical spent fuel pellets were designed.

To calculate the deposited energy as a function of the distance from the interface in each case, the system was virtually divided into several thin layers (also called "steps") on each side of the interface (i.e. the spent fuel and the water parts) over a distance corresponding to the alpha range in the material concerned, i.e. the last 12.6 μm in UO$_2$ fuel and the first 40 μm in water, as determined using the SRIM software [17]. Each layer's thickness was adapted to the accuracy necessary, up to 0.0625 μm very near the interface. The distance is referred to from the fuel/water interface: it is therefore negative for layers within the fuel and positive for layers within the water. A schematic layout of the two kinds of modeled systems is presented in Fig. 1. The main parameters taken into account in the MCNP calculations are given in Table 1.

Concerning the spherical geometry, UO$_2$ fuel fragment diameters ranging from 100 μm to 1 mm were considered in order to better understand possible changes in the energy deposition profiles. To better focus on the fuel/water interface of the flat part of the cylindrical geometry, 1 mm thick pellets were considered, and
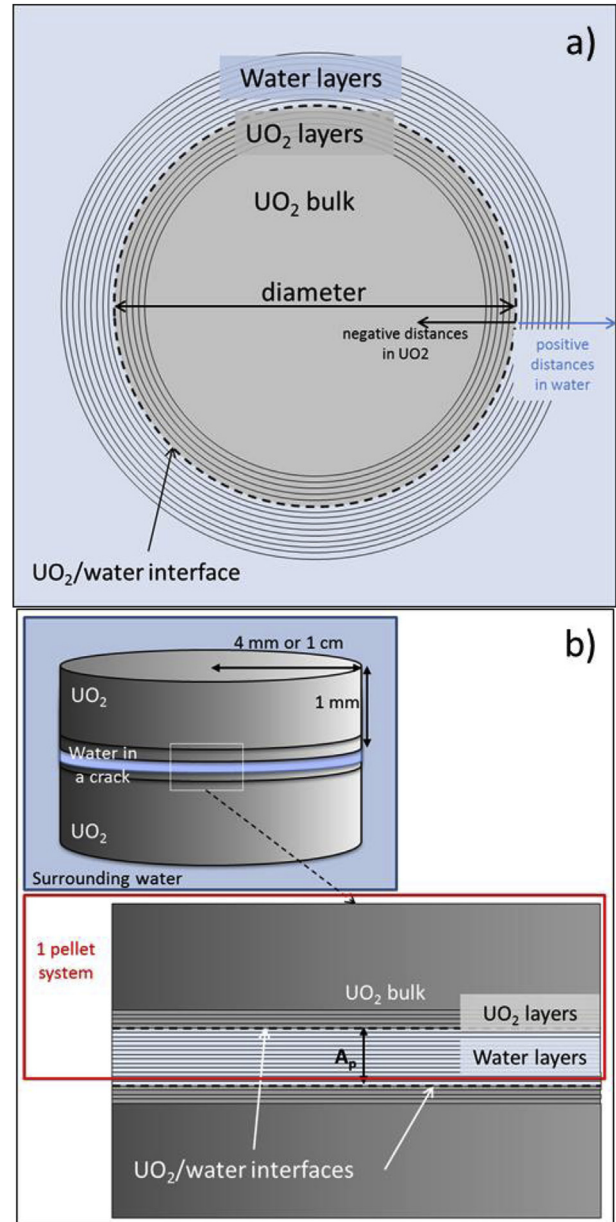

Fig. 1. Schematic view of the simulated geometries. (a) Spherical geometry with a UO$_2$ sphere surrounded by water. (b) Cylindrical geometry with UO$_2$ pellet(s) in water. In the latter case, the ideal crack filled by water is simulated by the water contained between two UO$_2$ pellets.

two diameters (8 mm and 2 cm) were studied in order to assess a possible edge effect on the interface description.

2.2.3. Cracks

During its residence time in the reactor, the temperature gradient between the center of the UO$_2$ fuel pellet and the periphery led to ceramic fracturing. This induced the formation of cracks with apertures ranging from 5 to 35 μm [18]. During storage in pools or under geological conditions, these cracks could be filled by water. In this work, the simplified geometry used for simulations is cracks with plane parallel faces. Fig. 1 b presents a schematic layout of an ideal crack as simulated here by 2 cylindrical UO$_2$ fuel specimens (2 cm diameter and 1 mm thick) spaced by the kind of water-filled aperture described above (from 5 μm to 35 μm aperture values).