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Simulation of alpha dose for predicting radiolytic species at the surface of spent nuclear fuel pellets

Abstract: In many countries, spent nuclear fuel is considered as a waste form to be disposed of in underground disposal. Under deep host rock conditions, a reducing environment prevails. In the case of water contact, long-term radionuclide release from the fuel depends on dissolution processes of the UO_2 matrix. The dissolution rate of irradiated UO_2 is controlled by oxidizing processes facilitated by dissolved species formed by alpha-radiolysis of water in contact with spent nuclear fuel. To understand the effect of the radiation, the information of the dose rate at the surface of the fuel and its proximity is needed. α particles contribute strongly due to their high linear energy transfer. However, their dose rate and the energy deposition at the fuel surface are difficult to measure. Cylindrical fuel pellets as used in fuel rods show specific features, such as the rim zone, where a higher Pu concentration and a different porosity of the fuel matrix is present. The α particle dose rate was determined by simulations with the code MCNPX with focus on the rim zone of a pellet. As a result a 40% increased dose level in the rim zone exists in comparison to the center of a pellet. The potential dominant and inhomogeneous α-dose distribution is supposed to have a strong impact on radiolysis phenomena and in turn on an inhomogeneous dissolution of elements over the surface.

Keywords: High burn-up spent nuclear fuel, RIM zone, MCNPX, dose distribution

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1 Introduction

In the case of groundwater directed to the waste in a spent nuclear fuel disposal, the environment is a dynamic redox system because of the time-dependent generation of radiolytic oxidants and reductants and the corrosion of Fe-bearing containers [1-4]. Due to the intense radiolytic field, radiation induced radiolysis products in the presence of water are considered to increase the waste form degradation rate and change radionuclide behavior. In an alpha-radiolytic field the local condition within a few tens of microns of the fuel/water interface layer may always be oxidizing even in reducing environments provided that H_2 is escaping from the water layer surface. Experiments with spent nuclear fuel are thought to be significantly influenced by their high beta-gamma-radiation field, which results in the generation of powerful oxidizing radiolytic species (e.g., $OH^{\cdot}$ and H_2O_2). McNamara et al. [5] found that studtite $[(UO_2)O_2(H_2O)_2]\cdot2(H_2O)$ and metastudtite $[UO_4\cdot2H_2O]$ were formed on the surface of spent nuclear fuel reacted at 298 K with deionized water for a period of 1.5 years and proposed that it would grow by incorporating peroxide created by the alpha radiolysis of water. This phase was investigated by P. C. Burns et al. [6] and Hughes-Kubatko et al. [7]. Efforts on modeling the radiation environment on the surface of fuel have been led by Sunder [8] and a conclusion of their works was that the beta field was the dominant field creating radiolytic oxidizing species. Different average dose rates for CANDU fuel with a burn-up of 685 GJ kgU^{-1} (8 MWd kgU^{-1}) are calculated in a water layer at the fuel/water interface. After one year of cooling time the dose rates amount to 70 Gy h^{-1} for alpha particles, the beta dose rate amounts to 10 kGy h^{-1}, and the gamma dose rate amounts to 400 Gy h^{-1}.

In light water reactors (LWR) uranium oxide and/or mixed oxide (MOX) fuel is used. The fuel consists of cylindrical UO_2 tablets (pellets). These pellets are inserted into a cladding tube, usually made of a zirconium alloy, forming the fuel rod. The fuel rods are bundled in structured arrangements forming the fuel elements which are inserted into the reactor core. During irradiation in the reactor, fission products and transuranic elements
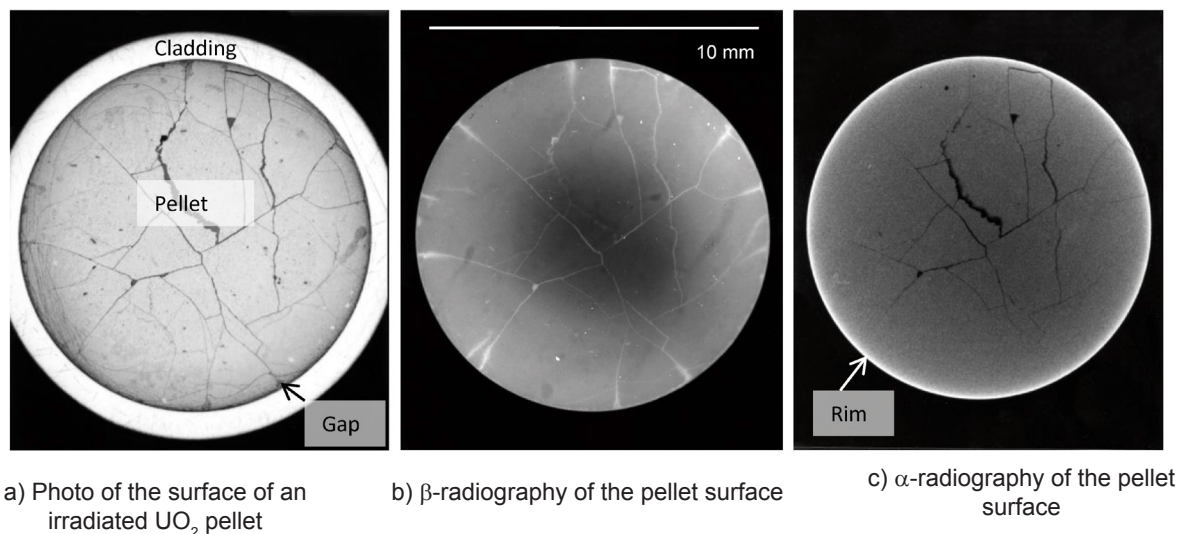
a) Photo of the surface of an irradiated UO₂ pellet   b) β-radiography of the pellet surface   c) α-radiography of the pellet surface

Figure 1: Photo and radiography pictures of the cross-sections through a Gösgen fuel rod. The UO_2 pellet, the gap between pellet and cladding, and the zircaloy cladding can be seen in a); the rim zone is visible in c).

(actinides) are formed. The fission products mainly cause the beta/gamma radioactivity whereas the actinides contribute to the alpha activity of the spent fuel. For the safety analysis of nuclear waste disposals, the potential radionuclide release (source term) is required. The source term depends on the dissolution of the UO_2 matrix of the spent nuclear fuel and on the related release of radionuclides. Unirradiated tetravalent UO_2 shows a low solubility as well as slow dissolution kinetics. However, investigations performed in recent decades revealed a different behavior of spent nuclear fuel: Due to the alpha-radiolysis directly on the surface of the fuel pellets in contact with aqueous solutions oxidizing species are formed causing oxidation of the tetravalent uranium to the hexavalent state which has significantly higher solubility [9,10]. For a better understanding of the processes, deeper insight in the dose rate distribution at the surface of the spent nuclear fuel pellets and the related energy deposition is needed. It is difficult to measure the α-particles dose rate directly. Therefore, in this paper, the dose rate induced by α-particles is calculated by simulations. For this, the code MCNPX [11] was employed.

2 Experimental Procedure

High burn-up (HBU) spent nuclear fuel refers to an average pellet burn-up above 45 MWd kg_{HM}^{-1}. Fig. 1 shows the surface of a pellet obtained by cutting a fuel rod between two pellets. It is a LWR-UO_2 fuel pellet (initial enrichment 3.8 wt% ²³⁵U) irradiated in the Gösgen pressurized water reactor [12] reaching an average burn-up of 50.4 GWd t_{HM}^{-1}. Table 1 shows the corresponding characteristic data of the fuel rod segment studied by KIT [13]. Properties of the pellet are: mass including cladding: 8.784 g, mass of irradiated fuel: 7.125 g (note that due to the cutting procedure the weight is less than for the unmachined pellet), diameter: 10.7 mm, height: 11.1 mm, β-dose rate at the shown surface ~100 Gy h^{-1}, and γ-dose rate ~5 Gy h^{-1} [14].

For the simulations, data for 48 GWd t_{HM}^{-1} [15] were employed, as they provide experimental data for the rim zone with a comparable burn-up. The composition of the investigated fuel pellets is summarized in Table 2; column "HBU center" is representative for the inner part of a fuel pellet, and "HBU Rim" shows the concentration in the outer rim zone which is relevant for a layer of ~200 μm depth from the cylinder surface. Note that the experimental data, obtained by inductively coupled plasma mass spectrometry and gamma spectroscopy, do not include elements such as the Xe (gas), I (volatile in acidic solutions), and oxygen (from UO_2). As shown in Table 2, the rim zone shows a significantly higher plutonium concentration and consequently a different fission product concentration. Furthermore, the rim zone of the pellet shows a porous structure [13].

3 Results and Discussion

Investigations of the rim microstructure in PWR fuels in the burn-up range 40 to 67 GW dt_{HM}^{-1} were performed by Spino *et al.* [16] and tabulated data on burn-up, radial position

Table 1: Characteristics of the fuel rod segment N0204 KKG-BS studied by KIT [13].

Property	Value
initial enrichment:	3.8% ^{235}U
pellet diameter:	9.3 mm
pellet density	10.41 g cm^{-3}
rod diameter:	10.75 ± 0.05 mm
zircaloy wall thickness:	0.725 mm
initial radial gap:	0.17 mm
number of cycles:	4
average burn-up:	50.4 GWd t$_{HM}$$^{-1}$
av. linear power:	260 W cm^{-1}
max. linear power:	340 W cm^{-1}
discharge date:	27. May 1989
duration of irradiation	1226 days
storage time	23 years

Table 2: Chemical inventory for the pellet in the center and rim region [15].

Element	HBU center (µg g^{-1})	HBU Rim (µg g^{-1})
Rb	320	490
Sr	700	700
Y	540	660
Zr	3800	4500
Mo	3600	4500
Tc	840	1090
Ru	2400	3200
Rh	510	700
Cs	2600	4200
La	2700	2800
Nd	4900	4000
U	790000	780000
Np	900	750
Pu	10400	15200
Am	500	800
Cm	70	110

and measured porosity, and pore sizes in 2D and 3D are provided. Also an equation allowing the calculations of the burn-up – porosity relation is given [13]:

$$\text{Porosity} = (\text{burn-up} - BU_o) \times d_1 + \exp(-d_2 + d_3 \times r/r_0)$$

where BU_o, d_1, d_2, d_3 are fit parameters, r the radius and r_o represents the maximum pellet radius. BU_o is the lowest burn-up for which the equation is valid. According to

Figure 2: Density and porosity in the rim zone of a pellet derived for spent fuel of a burn-up of ~50 GWd t$_{HM}$$^{-1}$.
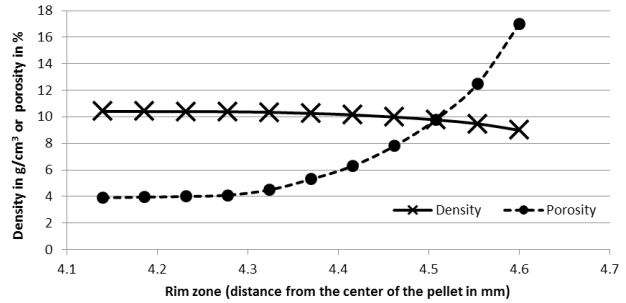
Kienzler *et al.* [13] the following data were obtained for ~50 GWd t$_{HM}$$^{-1}$ fuel: a rim porosity of 3.8% for r/r$_0$ = 0.9, 17% for r/r$_0$ = 1, showing an average of 6.2% between this two values. Based on fit parameters for a pellet with r$_0$ = 4.6 mm, Fig. 2 shows the porosity and corresponding density of the rim zone for the radius between 4.1 and 4.6 mm.

A pellet model for MCNPX was created consisting of a cylinder with a diameter of 9.2 mm and a height of 11.1 mm. The range of α particles with 6 MeV in water is approximately 0.05 mm. Therefore, the cylinder model was surrounded with a thin layer of water of 0.1 mm, to capture all of the deposited energy of α particles. To account for the rim zone on the surface of the pellet, as shown in Fig. 1, the rim zone was divided in 10 segments each containing a different chemical inventory and density. For the simulation the rim zone was modeled according to the position and density as given by the curve in Fig. 1. As only the radial region of the outer 500 µm is of concern, the corresponding inventory was simply assigned as a linear interpolation between the HBU center and HBU Rim as defined in Table 2. The specific activity of the fuel and element composition was calculated with the webKorigen software package [17]. α spectra of the decay of the nuclides, based on radiation decay data obtained with the software RadDecay [18], were calculated for 11 different inventories in the different regions of the pellet surface. They provided in turn the different source definitions for the input file of MCNPX. The *F8 energy deposition tally was used to obain the the total energy deposition in the water layer above each region. 10^8 „particle histories" were calculated with MCNPX in order to achieve a statistical accuracy better than 2%.

Fig. 3 shows the simulated dose distribution at the pellet surface with focus on the rim zone. According to the simulation results, in the rim zone the local dose per emitted α-particle in a 0.1 mm thick layer of water varies between 1.4×10^{-10} and 1.9×10^{-10} Gy. Multiplication with the activity (Bq) results in the dose rate which gives
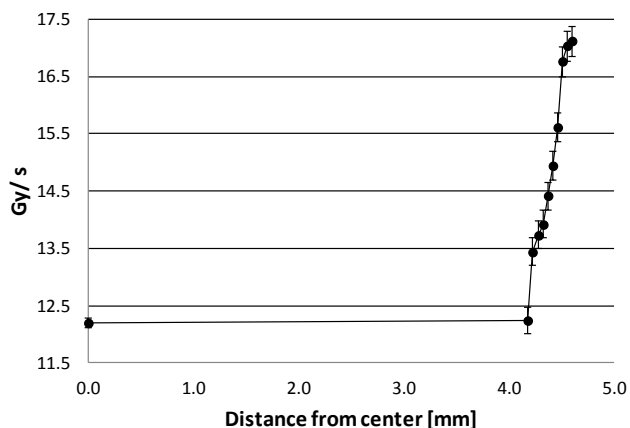
Figure 3: Simulated local α-dose rate on the surface of a pellet (the data points are connected by lines to guide the eye).

a corresponding local dose rate of 12 to 17 Gy s^{-1}, assuming an average specific α activity of 12 GBq g^{-1}. The result shows that in the rim zone the dose rate is increased by ~40% in comparison to the center of the fuel pellet. This effect enhances the yield of oxidizing species close to the rim zone leading to an incongruent oxidation of UO$_2$ over the fuel pellet surface. Previous results on a different pellet (α-activity 0,5 GBq g^{-1}) [14,19] revealed averaged measured and calculated γ-dose rates of 4 Gy h^{-1} and β-dose rates of 135 Gy h^{-1}, as well as calculated α-dose rates of 360 to 486 Gy h^{-1}. Here the α-radiation field is dominant for radiolytic oxidizing species. According to the results of this work the α-dose rate is supposed to increase by up to 40% in the rim zone towards the outer side i.e. the corresponding alpha-radiolytic field could have a high local dose rate of up to 680 Gy h^{-1}. Thus for the presented example the dose in the alpha-radiolytic field is half an order of magnitude higher than for the beta particles.

4 Conclusions

Radionuclide release from the fuel depends on dissolution processes of the UO$_2$ matrix, and the dissolution rate of irradiated UO$_2$ is determined by oxidizing species formed by radiolysis of water in contact with the fuel [20]. As the dose rate and energy deposition at the pellet surface is difficult to measure, we simulated the dose rate induced by α particles at the surface of the pellets deposited in a thin water layer. Furthermore, we took the rim zone of the fuel into consideration. Compared to the center of the pellet, the rim zone shows an increased dose rate up to a factor of 1.4. The potential dominant and inhomogenius α-dose distribution is supposed to have a strong impact on

radiolysis phenomena and in turn on an inhomogeneous dissolution of elements over the surface. As a conclusion the simulation data presented in this work provide the basis for subsequent determination of resulting chemical effects at the fuel/water interface.

Concerning the effect of the α-dose rate, it is worth pointing out that repository designs also incorporate large quantities of cast iron at depths of about 500 m in various host rocks. A significant H$_2$ pressure has been predicted to be generated due to the anoxic corrosion of the iron canisters. Dissolved hydrogen can significantly contribute to establishing reducing conditions in the near field [21-23] and effectively suppressing the UO$_2$ oxidation process even in the presence of a high alpha field.

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