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Enhanced diffusion under alpha self-irradiation in spent nuclear fuel: Theoretical approaches

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Abstract

Various theoretical approaches have been developed in order to estimate the enhanced diffusion coefficient of fission products under alpha self-irradiation in spent nuclear fuel. These simplified models calculate the effects of alpha particles and recoil atoms on mobility of uranium atoms in UO_2. They lead to a diffusion coefficient which is proportional to the volume alpha activity with a proportionality factor of about 10^{-44} (m^5). However, the same models applied for fission lead to a radiation-enhanced diffusion coefficient which is approximately two orders of magnitude lower than values reported in literature for U and Pu. Other models are based on an extrapolation of radiation-enhanced diffusion measured either in reactors or under heavy ion bombardment. These models lead to a proportionality factor between the alpha self-irradiation enhanced diffusion coefficient and the volume alpha activity of 2×10^{-41} (m^5).

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1. Introduction

Long-term interim dry storage and underground geological disposal are two of the options studied in France for the management of spent nuclear fuel. Studies are on-going in the framework of the PRECCI program (acronym in French for Research Program on Spent Fuel Long-term evolution) in CEA (Commissariat à l'Energie Atomique) and aims at predicting the long-term behaviour of spent nuclear fuel in the various environmental conditions encountered in dry storage or geological disposal. One of the final objectives is to propose radionuclide source term models for performance assessments of various types of spent nuclear fuel under repository conditions.

The release of radionuclides during geological disposal is classically divided into two terms [1]:

(i) an instantaneous release of radionuclides, often referred to as the instant release fraction (IRF); it corresponds to the fraction of the radionuclide inventory released rapidly as soon as water penetrates into the spent fuel rod;
(ii) a slow long-term release which corresponds to the dissolution of the uranium oxide matrix.

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The first term (IRF) is usually considered to be due to the radionuclides located within the regions of the rod presenting no confinement property in the long term (10 000 years). This fraction depends (i) on the location of the radionuclides inside the fuel rod on removal from the reactor, which is governed by the migration behaviour during irradiation in reactor, and (ii) on their mobility during the repository phase before breaching of the confinement. Therefore, to estimate the IRF it is necessary to determine the diffusion coefficients of radionuclides in conditions prevailing in geological disposal.

The mobility of heavy metal atoms (U,Pu), fission gases and volatiles in reactor has been extensively investigated for performance assessment and safety analysis during reactor operations because of its impact on thermo-mechanical behaviour of the fuel rod during irradiation. Extrapolating Arrhenius relationships given in the literature [2,3] shows that thermal diffusion of heavy metals and fissions products can be considered as negligible at the expected temperatures (<400 °C) over the time frame of both long-term dry storage (~100 years) and geological disposal (>10 000 years). However, studies of creep behaviour and swelling under irradiation have shown that creep and swelling rates of the pellet are markedly increased by irradiation under 1200 °C, where these phenomena are controlled by the low diffusion rate of uranium atoms [4]. Furthermore, direct measurements of uranium diffusion in UO_2 in reactor at 900 °C yielded a diffusion coefficient three to four orders of magnitude higher than the thermal diffusion coefficients [5]. These observations lead to the proposal of another mechanism of diffusion, which is not temperature controlled.

Enhanced diffusion of uranium and plutonium in UO_2 and $(U,Pu)O_2$ under irradiation was studied by Matzke [6]. The author showed that the diffusion coefficient was completely athermal below 1000 °C and was proportional to the fission rate. The same behaviour of the diffusion coefficient was observed by Turnbull et al. [7] for fission gases and volatile fission products.

After irradiation in reactor, the alpha activity is very high in the spent nuclear fuel (10^{15} Bq t_{iHM}^{-1}[1] during the first hundred years of cooling in a UO_2 fuel irradiated to 47.5 GWd t_{iHM}^{-1}). The effects of alpha decay in UO_2 are similar to the effects of fission. Therefore a mechanism of diffusion enhanced by alpha self-irradiation is expected to occur in the spent nuclear fuel out of reactor. This mechanism should be lower by some orders of magnitude than the one measured under fission in reactor. Thus direct measurements of the diffusion enhanced by alpha self-irradiation seem to be not possible within the time frame of laboratory experiments. Various theoretical approaches have been developed in order to propose rough estimates of the alpha self-irradiation enhanced diffusion coefficient. They are presented in this paper. The models are based on the effects of the recoil atom and of the alpha particle in the UO_2 lattice in comparison with the effects of fission fragments and also on the radiation-enhanced diffusion coefficients reported in literature.

Before extrapolating data measured in reactor to out-of-reactor conditions, it seems necessary to identify and quantify the mechanisms which cause the enhanced mobility of atoms observed under fission. Thus, the first part of the paper reviews the experimental data and mechanisms proposed in the literature to explain the enhanced mobility of atoms observed in reactor. The effects of alpha decays in UO_2 matrix are then described and their potential influence on atomic mobility is discussed. The diffusion coefficient corresponding to each particular mechanism is also proposed.

2. Review on enhanced diffusion under fission

2.1. Experimental data

Matzke described in [6] the complete results of the RADIF (*RAD*iation-enhanced *DIF*fusion) experiments performed in the 1980s by the Institute for Transuranium Elements in order to determine the enhanced diffusion coefficients of U and Pu atoms under irradiation. These experiments consisted of measuring diffusion coefficients of isotopic tracers of U and Pu in UO_2 and $(U,Pu)O_2$ during fission in a nuclear reactor. The influence of fission rates (between 7×10^{17} and 6.4×10^{19} fission m^{-3} s^{-1}) and temperature (between 130 and 1400 °C) was studied. The diffusion coefficient was found to be temperature independent below 1000 °C within the limit of measurement precision (see Fig. 3 in [6]).

The relationship between the radiation-enhanced diffusion coefficient of U and Pu in UO_2, D^* (m^2 s^{-1}) and the fission rate, F (fissions m^{-3} s^{-1}) derived from these observations was [6]:

$$D^* = AF \quad \text{with } A = 1.2\times10^{-39}\ \text{m}^5. \tag{1}$$

The radiation-enhanced diffusion coefficients in other nuclear ceramics ((U,Pu)C and (U,Pu)N) were also measured. The results showed that radiation-enhanced diffusion decreases in the order $(U,Pu)O_2 > (U,Pu)C > (U,Pu)N$ and is lower in (U,Pu)C than in $(U,Pu)O_2$ by approximately one order of magnitude [8].

Enhanced diffusion under irradiation was also studied for rare gases (Xe, Kr) and volatiles (I, Br). Experimental data and conditions reported in the literature for these elements are summarized in Table 1. In conclusion, experimental data have evidenced enhanced mobility of atoms due to fission. This mechanism is represented by a diffusion coefficient which is proportional to the fission

[1] t_{iHM} = ton of initial heavy metal.