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Review

Determination of fission gas release of spent nuclear fuel in puncturing test and in leaching experiments under anoxic conditions

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ABSTRACT

During reactor operation the fission gases Kr and Xe are formed within the UO_2 matrix of nuclear fuel. Their quantification is important to evaluate their impact on critical parameters regarding the fuel behaviour during irradiation and (long-term) interim storage, such as internal pressure of the fuel rod and fuel swelling. Moreover the content of Kr and Xe in the plenum of a fuel rod and their content in the UO_2 fuel itself are widely used as indicators for the release properties of ^{129}I, ^{137}Cs, and other safety relevant radionuclides with respect to final disposal of spent nuclear fuel. The present study deals with the fission gas release from spent nuclear fuel exposed to simulated groundwater in comparison with the fission gas previously released to the fuel rod plenum during irradiation in reactor. In a unique approach we determined both the Kr and Xe inventories in the plenum by means of a puncturing test and in leaching experiments with a cladded fuel pellet and fuel fragments in bicarbonate water under 3.2 bar H_2 overpressure. The fractional inventory of the fission gases released during irradiation into the plenum was (8.3 ± 0.9) %. The fraction of inventory of fission gases released during the leaching experiments was (17 ± 2) % after 333 days of leaching of the cladded pellet and (25 ± 2) % after 447 days of leaching of the fuel fragments, respectively. The relatively high release of fission gases in the experiment with fuel fragments was caused by the increased accessibility of water to the Kr and Xe occluded in the fuel.

Contents

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1. Introduction

Presently, final disposal of spent nuclear fuels (SNF) in deep geological repositories is considered as the preferred option for the management of irradiated UO_2 fuels in a number of countries, such as Belgium, Canada, Finland, Germany, South Korea, Spain, Sweden and Switzerland [1–5]. Though, geological or geo-technical barrier system of an underground repository may prevent to some extent groundwater contacting the fuel, intrusion of solutions into disposal rooms has to be taken into account within the long-term safety case of a SNF repository. Upon contact with water, the alteration of SNF and the consecutive release of radionuclides involve the combination of many different processes, which can be grouped into two stages: (a) A fast release of radionuclides, which segregated during irradiation in trapping sites of the fuel pin as the pellet/cladding gap, fractures within the fuel pellets and UO_2 grain boundaries; this fast released radionuclide fraction is generally referred to as the fast/instant release fraction (IRF). (b) A much slower, long-term radionuclide release that results from the alteration and dissolution of the UO_2 matrix. Among other fission products (e.g. ^{79}Se, ^{129}I, ^{135}Cs and ^{137}Cs), the fission gases krypton and xenon are released as well.

The physicochemical and structural transformations in nuclear fuel, which occur consecutive to fission and neutron capture reactions, radionuclide decay and thermal stress during irradiation, determine to a large extent the release behaviour of radionuclides in the fuel. In particular for light water reactor fuels with a burn-up higher than 40 $MWd\cdot(kg_U)^{-1}$ formation of radial cracks, high porosity in the rim zone of UO_2 pellets and the closure of the initial gap between the pellet and the Zircaloy cladding is observed after irradiation. With respect to the spatial distribution and release of fission gases, the formation of inter-granular bubbles within the fuel is of relevance. The size of these bubbles varies with the distance, r, from the central line: micrometric size in the central part of the pellet, $r/r_0 < 0.5$ [6]; about 10–100 nm diameter in the region $0.5 < r/r_0 < 0.96$ [7–9]; micrometric size in the rim region, $0.96 < r/r_0 < 1$, determining the porosity of the peripheral high burn-up structure [10–12].

Kr and Xe isotopes, formed during irradiation, are characterized by a low solubility within the UO_2 matrix. As a consequence a large part of their inventory tends to precipitate into bubbles, both intra- and inter-granular [13–16]. These fission gases are distributed dynamically between (i) a solution within the oxide lattice, (ii) closed intra-granular and inter-granular bubbles within the fuel, and (iii) a part of the gases which are released to open porosity of the fuel and consecutively to the plenum of the fuel rod [17–20]. Indeed, within a fuel rod the release of Kr and Xe is partly due to recoil and knockout of a fraction of the fission gases formed close to an external surface; this process is referred as athermal release [17,20]. However, the largest contribution of the fission gas release (FGR) is the thermal release due to diffusion of the gases from the UO_2 lattice to free surfaces or grain boundaries [17,20]. Intra-granular bubbles act as traps for the diffusing gas. Irradiation-induced re-solution can remove gas from the bubbles and put it back in solution. Generally, gas release from the grain faces to the grain edges occurs via interlinkage of grain face bubbles, formation of channels, and subsequent venting to the edges. Bubbles accumulating along the grain edges link up and form tunnels and, eventually, if the tunnels are open and stable the gas can be released to a free surface [17,20,21]. With increasing burn-up, grain boundaries and pores are preferred sinks for fission gas atoms. Diffusion from the inner grain regions to the grain boundaries and pores is enhanced with increasing fuel temperature. In contrast to these physical and concentration gradient driven transport processes, there is no chemical interaction between the noble gases Kr and Xe and the fuel [11].

The quantification of the FGR into the free volume of a fuel rod is important to evaluate the impact of fission gases on the internal pressure of the fuel rod and the fuel behaviour during normal operation and incidental or accidental events [17]. The amount of fission gas released to the plenum during irradiation can be directly determined by means of puncturing tests [10,17,22–25]. In order to quantify the amount of fission gases retained in bubbles within the UO_2 structures, some studies on polished section of irradiated fuel pellets have been carried out during the last four decades using: scanning electron microscope (SEM), electron probe micro analysis (EPMA), and secondary ion mass spectrometry (SIMS) [6,10,14,26–29]. SEM is used to study the variation of the microstructure as a function of the fuel pellet radius. EPMA measurements allow determination of the radial distribution of Xe and other fission products in the UO_2 fuel matrix. However, the Xe determination by means of EPMA is limited by the fact that only Xe located in the fuel matrix can be measured, whereas the amount of Xe retained inside the pores cannot be analysed [33]. Using SIMS it is possible to determine the total amount of Xe in the fuel [30–36].

The present study deals with the fission gas release from a pressurized water reactor (PWR) fuel rod segment exposed to simulated groundwater in relation to the fission gas accumulated during irradiation in the plenum of the rod. Kr and Xe inventories released in the plenum during irradiation were determined by means of a puncturing test, whereas inventories of Kr and Xe, which were released during contact with water, were measured in anoxic leaching experiments with a cladded pellet and fragments of the PWR fuel rod segment. The study was conducted within the collaborative project, “Fast/Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (CP FIRST-Nuclides)” [37–39].

2. Experimental

2.1. Material

The studied spent nuclear fuel consisted of pure UO_2, fabricated by Kraftwerk Union AG using the NIKUSI (from the german: Niedrigtemperatur-Kurzzeit-Sintern) sintering process [40]. NIKUSI is a low-temperature, short-time sintering process under controlled oxidizing condition at a temperature (<1300 °C) below the temperature range of conventional UO_2 pellet sintering processes [41,42]. The UO_2 stoichiometry of U/O was reported to be 2.002 [40]. The lattice geometry of the fuel assembly consisted of a 15×15 array of fuel rods, where 205 of the 225 positions per assembly were occupied with fuel rods and the 20 remaining positions were available for control rods [43]. The material investigated