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Instant release of fission products in leaching experiments with high burn-up nuclear fuels in the framework of the Euratom project FIRST- Nuclides

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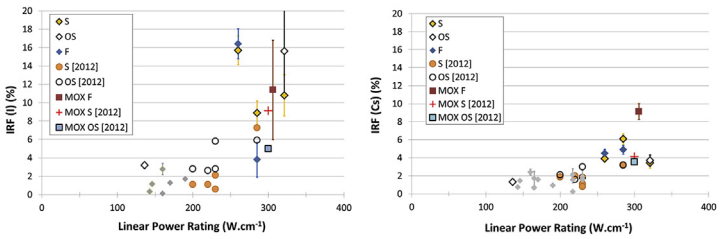
HIGHLIGHTS

- Leach tests were performed to study the instant release of fission products from high burn-up UO_2 fuels and one MOX fuel.
- In these tests, the fission gas release given by the operator was a pessimistic estimator of the iodine and cesium release.
- Iodine and cesium release is proportional to linear power rating beyond 200 W cm^{-1}.
- Closure of the fuel-cladding gap at high burn-up slows down the release.
- The release rate decreases following an exponential equation.

GRAPHICAL ABSTRACT



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ABSTRACT

The instant release of fission products from high burn-up UO_2 fuels and one MOX fuel was investigated by means of leach tests. The samples covered PWR and BWR fuels at average rod burn-up in the range of 45 −63 GWd/t_{HM} and included clad fuel segments, fuel segments with opened cladding, fuel fragments and fuel powder. The tests were performed with sodium chloride − bicarbonate solutions under oxidizing conditions and, for one test, in reducing Ar/H_2 atmosphere. The iodine and cesium release could be partially explained by the differences in sample preparation, leading to different sizes and properties of the exposed surface areas. Iodine and cesium releases tend to correlate with FGR and linear power rating, but the scatter of the data is significant. Although the gap between the fuel and the cladding was closed in some high burn-up samples, fissures still provide possible preferential transport pathways.

Acronyms: FGR, Fission gas release; IRF, Instant release fraction; LPR, Linear power rating; SNF, Spent nuclear fuel.

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1. Introduction

The Spent Nuclear Fuel (SNF) generated by thermal neutron irradiation of UO_2 or Mixed UO_2-PuO_2 oxide (MOX) fuels in nuclear power plants is a heterogeneous material, of which the structure, chemical composition and properties depend on the burn-up and in-pile irradiation history. The properties of irradiated fuels have been investigated since several decades, and comprehensive standard literature is available [1–3]. In an irradiated UO_2 fuel rod from Light Water Reactors (LWR), one can distinguish several structural-compositional elements: the zircaloy cladding, the gap between the cladding and the fuel (some tens of micrometers in thickness), and the fuel pellet itself, which is characterized by radial cracks formed as a result of the steep radial temperature gradient [4]. On a microscopic scale, the fuel consists of porous micro sized UO_2 grains containing sparse metal particles (known as “epsilon” particles). For the evaluation of leach experiments, structures representing potential pathways accessible to aqueous solutions are important. These include the fuel-cladding gap, the radial and longitudinal cracks, open pores in the matrix and fuel grain boundaries. In the case of SNF with an average burn-up higher than 40 GWd/t_{HM}, the formation of a rim structure [5–7] is observed, showing a distinctly smaller average grain size and increased porosity in comparison to the inner parts of the fuel. Chemically, the fission products in the SNF can be classified into four main groups [8]: (1) fission gases (mainly Kr, Xe); (2) fission products forming metallic particles (e.g. Tc, Ru, Pd); (3) fission products forming discrete oxides (e.g. Ba, Zr); (4) fission products dissolved as solid solution in the UO_2 matrix (e.g. REE, Sr). Some elements may partition in different phases, e.g. molybdenum plays a role in buffering the oxygen potential of the fuel and can coexist in metallic and oxide phases. The behavior of iodine and cesium, which are major contributors of fast release in leach tests, cannot be entirely rationalized in terms of the above scheme. During reactor operation, these elements may behave like fission gases due to their low boiling points, but they may also form a soluble phase (CsI), which is rapidly dissolved upon contact with water. Because the cesium concentration (in mole) surmounts the iodine concentration, cesium may also be associated to UO_2 and iodine may partition significantly in the fission gas phase.

When the SNF is disposed of in an underground repository, the radionuclides may gradually be released after failure of the canister and subsequent water ingress. The release rate of radionuclides differs depending on their chemical properties, their chemical speciation in the fuel, as well as the location where they are segregated within the SNF. Elements such as cesium and iodine are mainly volatile during irradiation of the fuel and undergo diffusion processes in the thermal gradient of the fuel rods. For fuel operated in high-power conditions (typically above 1100–1200 °C) these elements tend to segregate in the cooler regions at the pellet periphery and in the fuel/cladding gap. Typically, an operational distinction is made between the release from the fuel grains, from the grain boundaries and from the gap and cracks [9]. The release of fission products incorporated in the UO_2 grains (matrix dissolution) is very slow, especially under reducing conditions, as radiolytic fuel oxidation is suppressed [10,11]. The release of soluble segregated elements from the accessible gap, cracks and grain boundaries is fast. Most of their inventory is released within a few months or even days. The quantity of these rapidly released inventories normalized to the total nuclide inventories is commonly called the Instant Release Fraction (IRF) in safety assessment studies. Because some of the involved radionuclides are long-lived, and geochemically mobile (especially ^{129}I, ^{135}Cs, ^{36}Cl), they can significantly contribute to or even dominate the calculated dose exposure [12,13]. First measurements of the fast release were performed on low burn-up CANada Deuterium Uranium (CANDU) fuel [9,14,15]. Later, several European projects investigated the fast release mainly from Pressurized Water Reactor (PWR) UO_2 fuel [16–20].

Recently, experimentally determined fast release results were correlated with Fission Gas Release (FGR) [21]. The FGR can be measured directly by different methods [22,23] or calculated with relatively good precision [24,25]. It depends strongly on the operating conditions, especially on the burn-up, ramping and power rating. However, the correlation between burn-up, fast release and FGR proved to be weak due to the considerable scatter of the data and has therefore a low predicting power. Johnson et al. [21] suggested that linear power rating could be a more appropriate parameter to predict fast release. Up to date, the experimental database supporting this suggestion was small, partly due to the limited knowledge of power rating values for the investigated samples. Moreover, because the utilities tend nowadays to increase the burn-up in order to optimize energy production, it is particularly important to extend the experimental IRF database to such high burn-up fuels and to test correlations with well-known reactor operational parameters.

For this purpose, a common experimental program was set up by a group of laboratories, in the framework of the FP7 Collaborative Project FIRST-Nuclides (Fast/Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel, 2012–2014), carried out during the 7th European Union's Research and Innovation funding program (FP7) [26]. The experimental investigations included the gas release, rim and grain boundary diffusion processes, and leach tests, for quantification of the fast release of activation and fission products into the aqueous phase, with – to the extent possible – determination of their chemical speciation. The leach tests were performed with well characterized spent nuclear fuel samples by Karlsruhe Institute of Technology (KIT) Germany, Joint Research Centre – Directorate G (by the formerly called Institute for Transuranium Elements (ITU)), Paul Scherrer Institut (PSI) Switzerland, Studiecentrum voor Kernenergie - Centre d’Étude de l’énergie Nucléaire (SCK•CEN) Belgium, Fundació Centre Tecnològic de Manresa (CTM) Spain, and Studsvik Nuclear AB (STUDSVIK) Sweden. The leach tests were performed with fuels from pressurized water reactors or Boiling Water Reactors (BWR) with average rod burn-ups ranging from 42 to 63 GWd/t_{HM}. The leach tests were complemented by analysis of release rates of relevant radioisotopes from damaged and leaking VVER (Vodo-Vodyanoi Energetichesky Reaktor) fuel elements during their storage in a water pool. These fuels had been previously irradiated in a Hungarian pressurized water reactor. This paper summarizes the results of the leach experiments and attempts to establish correlations to improve the IRF estimations for high burn-up UO_2 fuel.

2. Experimental

Leaching tests were done with samples from various UO_2 fuels and one type of MOX fuel, irradiated either in PWR's or BWR's. The main characteristics of the investigated SNF samples are shown in Tables 1 and 2. In these tables, the average rod burn-up is listed as reported by the utilities, based on core physics calculations and the plant thermal output (‘rod ave-cal’ in Tables 1 and 2). The local burn-up of the tested sample can be different, and is given when this information is available (‘loc’ in Tables 1 and 2). The local burn-up values are either calculated by core physics calculations (‘loc-cal’ in Tables 1 and 2), measured by gamma scanning (‘loc-gam’ in Tables 1 and 2), or by dissolution of the sample and radiochemical analysis (‘loc-diss’ in Table 1). The linear power rating (LPR) distinguishes the rod average LPR (‘rod ave’ in Tables 1 and 2),

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