



Evolution of spent nuclear fuel in dry storage conditions for millennia and beyond



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ABSTRACT

Significant amounts of spent uranium dioxide nuclear fuel are accumulating worldwide from decades of commercial nuclear power production. While such spent fuel is intended to be reprocessed or disposed in geologic repositories, out-of-reactor radiation damage from alpha decay can be detrimental to its structural stability. Here we report on an experimental study in which radiation damage in plutonium dioxide, uranium dioxide samples doped with short-lived alpha-emitters and urano-thorianite minerals have been characterized by XRD, transmission electron microscopy, thermal desorption spectrometry and hardness measurements to assess the long-term stability of spent nuclear fuel to substantial alpha-decay doses. Defect accumulation is predicted to result in swelling of the atomic structure and decrease in fracture toughness; whereas, the accumulation of helium will produce bubbles that result in much larger gaseous-induced swelling that substantially increases the stresses in the constrained spent fuel. Based on these results, the radiation-ageing of highly-aged spent nuclear fuel over more than 10,000 years is predicted.

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1. Introduction

Plutonium and minor actinides (americium, curium) constitute a small but very radiotoxic fraction of the elements produced in today's widely used UO_2 fuel but also in the mixed oxide ($\text{U}_x\text{Pu}_{1-x}$) O_2 fuel (MOX) during the production of electricity from nuclear reactors. These elements pose the main problems for non-proliferation issues, spent fuel disposition (the spent fuel stored worldwide will amount $\sim 350,000$ t of heavy metals in 2020 [1]), or for high level waste disposal in the countries where spent fuel is reprocessed (the reprocessed spent fuel will amount 120,000 t of heavy metals worldwide in 2020 [1]). Public acceptance of nuclear energy is strongly affected by the proposed solutions for these sensitive issues. Today's strategy for the back end of the nuclear fuel cycle varies from one country to the other [2–4]. However, whatever option will be chosen, i.e. reprocessing and recycling or direct disposal, potentially following interim storage, the nuclear waste will have to be safely stored for long periods of time.

While nuclear fuel in reactor undergoes significant restructuring due to the radiation effects from fission and to high operating temperatures [5], the radiation and temperature environment for spent fuel out of reactor is substantially different. After decay of the predominantly beta-decaying short-lived fission products, the spent fuel will be subjected primarily to alpha-decay self-radiation damage and radiogenic helium accumulation at near ambient conditions, where completely different atomic level processes will dominate for millions of years. Fig. 1 shows as an example the alpha-dose evolution, hence helium production, during time for different types of nuclear fuels. Thus, the spent nuclear fuel available and characterized today is not representative of the structure and state of aged fuel after hundreds or thousands years of storage because it has not yet experienced a significantly long accumulation of microstructural defects and of helium due to alpha-decay [6].

Because of the handling, transport and storage of spent fuel, it is of critical importance to study the aging of these materials in order to predict their long term behaviour and risk to the biosphere. The goal is to understand and assess the processes responsible for the alteration of materials subject to radiation damage in view of their long term integrity. The foremost alteration process is associated with the build-up of radiogenic helium and alpha-damage that could cause early material degradation and enhanced corrosion

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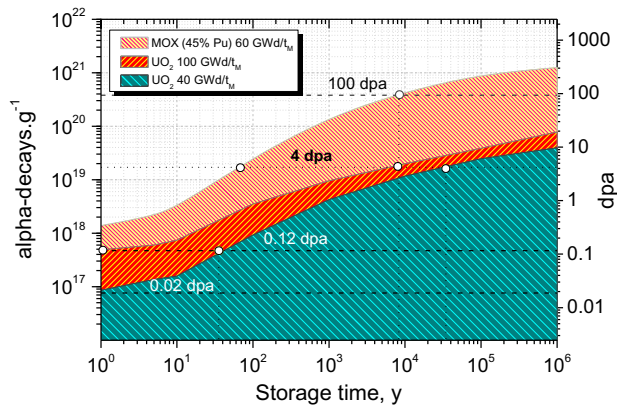


Fig. 1. Helium production and related alpha-dose as a function of time in different nuclear materials i.e. fuels of different composition and burnup. The equivalent damage is indicated as displacements per atoms (dpa) on the right ordinate. The green dashed lines indicate some examples of age correspondence between the studied samples and different types of aged nuclear fuels (see also Table 1 for a detailed sample description). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

[7–12]. Whether the fuel can or cannot retain the helium is of prime importance in assessing its long term mechanical stability. An increase of about 20 bars, in addition to the 25 bars of helium present at the beginning of irradiation, has to be accounted for in the fuel rod after a typical irradiation of a UO_2 fuel at a standard burnup. About 55% of this pressure increase is due to swelling of the fuel and consequent decrease of the free volume. If helium is generated during storage and not retained in the fuel, it will increase further the pressure. If disintegration of the fuel pellet occurs, additional gas will be released to the free volume from the opening of pressurized bubbles with a small compensation from the pores to the overall new free volume. This aspect would be more critical for fuel containing more shorter living actinides, as is the case for example for MOX fuel which in turn will produce more helium during storage (see Fig. 1). As a matter of fact, the cladding, which constitutes in some operational approaches the first confinement barrier after the fuel pellet in case of interim or long term storage, and should withstand over-pressurisation due to the production of helium in case it is released because of fuel embrittlement. For example, the production of $10^{20} \text{ He g}^{-1}$ in a MOX (burn-up 40 GWd t^{-1}) fuel after 10,000 years would result in an increase of 50 bar of total pressure if one assumes a free volume of $\sim 20 \text{ cm}^3$ in a cladding tube and a total release of He from the spent fuel pellet.

Increases in lattice parameter induced by the accumulation of gas and of defects from alpha-decay might cause swelling of the

spent fuel and its early failure. Interactions between the radiogenic helium formation and the defects produced by alpha-particles and recoil nuclei need to be understood to predict the evolution of the system. Aged spent fuel is not available today it can only be simulated by accelerated aging of materials with a structure comparable to fuel or by studying natural analogues having cumulated high alpha-dose [7,13–16].

After irradiation in a nuclear reactor the fuel has experienced about 4–5% of fissions and the formation of fission products (FP) that modify the chemical composition and microstructure of the fuel. However, part of the inventory of the FP is segregated from the UO_2 matrix. The gases are typically precipitated into bubbles with sizes between nm and μm as function of their radial position (temperature during irradiation). The five metal particles mainly composed of Pd, Ru, Rh, Tc and Mo and have sizes ranging between nm and μm also depending on the irradiation temperature [17]. The residual activity dominated by beta and gamma decay generates a decay heat of 1 kW/t ten years after discharge while after 10,000 years the activity will be predominantly caused by alpha-decay generating a decay heat of about 20 W/t [18]. At discharge, in a LWR fuel with a burn-up of 40 GWd/t having a residual temperature of 500 K, the thermal conductivity has decreased by $\sim 50\%$, from $5.8 \text{ W m}^{-1} \text{ K}^{-1}$ as in fresh fuel to $3.0 \text{ W m}^{-1} \text{ K}^{-1}$ [19]. The lower the temperature of the fuel the higher is the degradation of the thermal conductivity. However, since the total thermal power decreases with time, in wet storage conditions the fuel temperature will not exceed 473 K, which would even be the limit in dry storage conditions after a few decades [20]. For comparison our $^{238}\text{PuO}_2$ sample (see Table 1) has a decay heat of 350 mW/g. The oxygen potential of the fuel is almost close to the one of stoichiometric fresh fuel even if locally there can be slightly sub-stoichiometric domains.

The study here will therefore have some limitations but no other systems as those studied here could help to assess the basic and dominant effects responsible for the spent fuel structural degradation due to damage formation. In their paper, Janeczek et al. conclude that the natural analogues can exhibit similar properties as spent nuclear fuel like identical structure, high resistance to radiation damage, similar behaviour during oxidation, etc. [15]. This paper will also evidence a similarity of the microstructure of alpha-doped UO_2 , spent fuel and natural analogues.

This study has been mostly performed on UO_2 based samples doped with different concentration of strong alpha-emitters (e.g. ^{238}Pu) enabling to cumulate on a laboratory time scale alpha-dose representative of spent fuels aged up to millions of years. To take into account the kinetic aspects for the build-up of high damage levels and large quantities of helium we have also investigated natural analogues as described Table 1. The multiple microstructural aspects having a direct or indirect link with the formation or

Table 1
Samples* used in this study and measured properties.

Sample	Original composition	Age (y)	Damage (dpa)	Alpha-decay events ($\alpha \text{ g}^{-1}$)	Bubbles		Swelling (%)		Thermal desorption	
					Mean radius (nm)	Conc. (m^{-3})	Lattice	From bubbles	Release temp. onset (K)	Fraction of the initial helium still present
UO233	$(\text{U}_{0.9}^{233}\text{U}_{0.1})\text{O}_2$	5	10^{-5}	3.8×10^{14}			0.09			
UO-01	$(\text{U}_{0.999}^{238}\text{Pu}_{0.001})\text{O}_2$	9	0.02	7.6×10^{16}			0.5			100
MOX40	$(\text{U}_{0.6}^{239}\text{Pu}_{0.4})\text{O}_2$	12	0.12	4.7×10^{17}			0.7			
UO-10	$(\text{U}_{0.9}^{238}\text{Pu}_{0.1})\text{O}_2$	16	4	1.7×10^{19}	1.2	1.5×10^{22}	1.3	0.01	1000	100
RTG1	$(^{238}\text{Pu}_{0.9}, \text{Pu}_{0.1})\text{O}_2$	30	100	3.6×10^{20}	2.5	5×10^{23}	2.2	3	900	70
T4 ([73])	$(\text{U}_{0.33}, \text{Th}_{0.67})\text{O}_{2+y}$	550×10^6	170	7.2×10^{20}	3	8×10^{23}	1.5 [73]	9	800	
U2 ([73])	$(\text{U}_{0.92}, \text{Th}_{0.08})\text{O}_{2+y}$	220×10^6	130	5.8×10^{20}			1.5 [73]			

* For T4 and U2 the lattice swelling values are taken from the indicated references.

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