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An attempt to reproduce high burn-up structure by ion irradiation of SIMFUEL

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ABSTRACT

Experiments in IC-100 and U-400 cyclotrons were conducted with SIMFUEL pellets (11.47 wt.% of fission products simulators) to reproduce some aspects of the long-term irradiation conditions in epithermal reactors. Pellets were irradiated with Xe^{16+}, Xe^{24+} and He^{+} at energies ranging from 20 keV (He^{+}) to 320 keV (Xe^{16+}) and 1–90 MeV (Xe^{24+}). Some samples were subsequently annealed to obtain larger grain sizes and to study defects recovery. The major microstructural changes consisted in grain sub-division observed on SEM and AFM images and change in composition registered by EPMA (pellets irradiated with 1–90 MeV Xe^{24+} ions at fluence of 5×10^{15} cm^{-2}). Lattice distortion and increase in dislocation density is also noted according to X-ray data. At low energies and high fluences formation of bubbles (20 keV He^{+} at 5.5×10^{17} cm^{-2}) was observed. Grain sub-division exhibits full coverage of the grain body and preservation of former grain boundaries. The size of sub-grains depends on local dislocation density and changes from 200 nm to 400 nm along the irradiated surface. Beneath it the size ranges from 150 to 600 nm. Sub-grains are not observed in samples irradiated by low-energy ions even at high dislocation densities.

1. Introduction

There are a number of unresolved material issues with uranium dioxide when it is used as fuel for epithermal-spectrum reactors analogous to PWR. Most relate to the change of structural and phase state of nuclear fuel during burn-up. The process is most intense on the periphery of fuel pellets where ^{238}U reacts with resonant neutrons producing fissile ^{239}Pu. At pellet average burn-up of 40–60 GWd/tU local burn-up in the outer rim region may reach 120 GWd/tU [1]. This leads to the fuel having a structure similar to a composite, the outer layer of which exhibits highly refined grains and large gas bubbles while the inner is more like the as-sintered pellet [2]. These layers become discernible in PWR conditions only at burn-up higher than some threshold value. Due to the tendency of increasing burn-up (e.g., average discharge burn-up of fuel loaded in Russian WWER-1000 increased from 48 GWd/tU before 1998 up to 67 GWd/tU in 2003, and is still increasing), the influence of HBS formation on the fuel performance may no longer be neglected and demands special attention. The mechanisms of this transient process are currently unclear. However, many guesses, unfounded or partly-proven assumptions on the reasons for this event exist [3]. Summarizing published data and excluding exotic and improbable theories, three scenarios of the onset of high burn-up structure (HBS) may be formulated:

- Dislocations scenario [4–7] (recent work – [8]): self-organization of chaotically distributed forest dislocations leads to grain refinement (polygonization). Small interstitial loops generated in cascade regions grow to create forest dislocations. The radiation-enhanced mobility allows re-arrangement of forest dislocations resulting in sub-grains. Possibly, further growth of sub-grains occurs [9].
- Bubbles scenario [10–12]: fission gas becomes trapped in vacancy pores and increases the pressure inside which becomes enough to punch dislocation loops out. Large density of forest dislocations formed as a result of continuous overpressure in gas bubbles induces polygonization.
- Fission fragments scenario [13,14]: non-uniform stress fields generated by the motion of fission fragments through the lattice with instable distribution of defects and growing density of fission products are subject to relaxation which possibly forms a new structure.

One of the reasons why there is no consensus on the problem of HBS formation is because post-irradiation examination of pellets irradiated in power reactors does not offer any chance in analyzing particular processes, e.g., the influence of bubbles and dislocations

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separately. Ion irradiation of samples with a chemical composition close to that of the spent fuel is an alternative to in-pile irradiation which may help. Formerly, ion irradiation was already used to study the mechanisms of HBS formation in UO_2 and important data was obtained [15–18]. Some irradiation experiments were conducted on SIMFUEL pellets [19]. However, true mechanisms behind high burn-up structure formation have not yet been clarified. This work is aimed at finding the major factors of microstructural changes in oxide fuel using pre-planned ion irradiation experiments with pellets of chemical composition close to that of high burn-up fuel.

2. Sample characteristics

Because the accumulation and heterogeneous distribution of fission products may influence the conditions of HBS transition, pellets of uranium dioxide already containing 11.47 wt.% of fission products simulators (FPS) were used in this work. This amount of FPS approximately corresponds to a burn-up of 120 GWd/tU.

Hydroxides of additives (Y, Zr, Sr, Ba, Ce, La) were mixed with the original ammonium polyuranate (2.4% ^{235}U) during co-precipitation, while Mo, Ru and special additives (Al_2O_3, SiO_2 and Cr_2O_3) were mechanically admixed to the molding material. Sintering was conducted at 1750 °C in hydrogen. The sintered pellets had a density of about 9.47 ± 0.03 g/cm^3 (89% of theoretical density) and a size of 10 mm (diameter) × 1.5 mm (height). Pellets were polished and cleaned in a heated ultra-sonic bath with ethanol. Samples were subject to grain etching in a mixture of HNO_3 and H_2O_2 until grain boundaries were clearly visible by means of optical microscopy at a magnification of × 1000. Mean grain size of the matrix was estimated to be 2–3 μm (Fig. 1).

The results of electron probe microanalysis (EPMA) show that uranium content in the matrix changes slightly from grain to grain whilst FPS additives are distributed less uniformly. Some elements as Ba and Mo which should precipitate in an individual phase are often found in the matrix phase. The dark phase (Fig. 1) is enriched by barium, uranium, zirconium and oxygen and shows a fluctuating composition. The ratio of elements present therein indicates it being the perovskite phase of $(Ba_{1-z}Sr_z)(Zr_{1-n}U_n)O_3$ type (plus small quantities of Mo and rare earths). Large particles almost free of uranium and oxygen and enriched by Mo, Ru and Zr are also found (Fig. 1).

3. Experimental conditions

3.1. Irradiation details

A fission fragment is a massively charged heavy ion with an initial kinetic energy of ~90 MeV. Its interaction with fuel material is characterized by such effects as thermal spikes, collision cascades, and ion implantation. If the path of a fission fragment was divided into segments, these could be selected on the basis of a predominant interaction effect. Then a measure of intensity could be introduced for quantitative characterization. The cumulative effect of many fission fragments produced with a given rate would simply be the integral over time and volume of these intensities multiplied by fission rate. However, if one required knowing the consequences of a particular interaction effect, one would choose to simulate the conditions in particular segments. Because each segment corresponds to a range of energies of the fission fragment, this would require using irradiation by ions whose energies fall into these ranges.

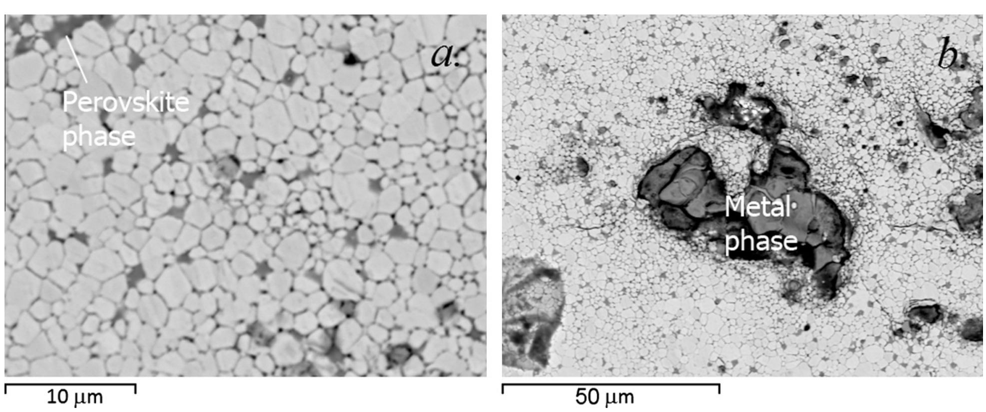

Fig. 1. Observation of as-sintered UO_2 + 11.47 wt.% FPS pellet: (a) matrix grains and perovskite phase $(Ba_{1-z}Sr_z)(Zr_{1-n}U_n)O_3$ precipitating on grain boundaries; (b) metallic phase particles enriched with Mo, Ru and Zr.

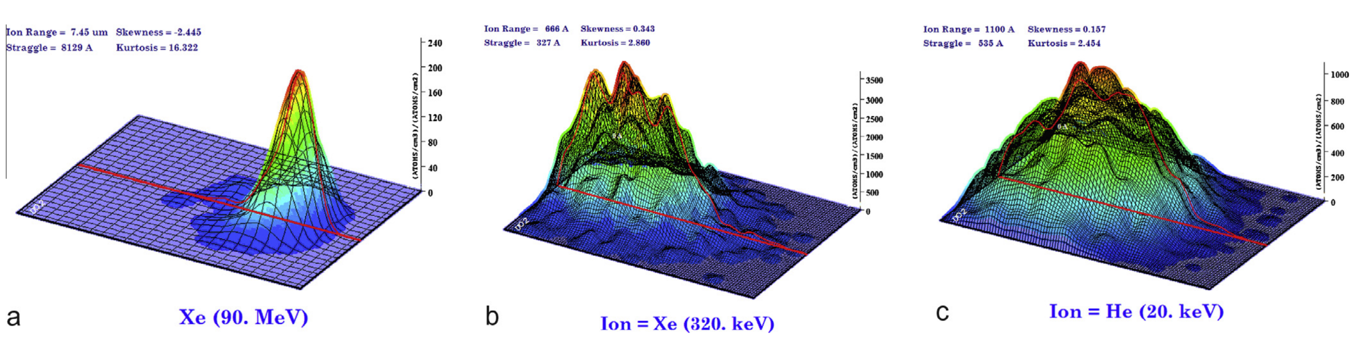

Fig. 2. Distribution of infiltrated ions in UO_2 for three possible irradiation schemes: (a) Xe^{24+} 90 MeV; (b) Xe^{16+} 320 keV; (c) He^+ 20 keV.