New Catalog on $(U,Pu)O_2$ Properties for Fast Reactors and First Measurements on Irradiated and Non-irradiated Fuels Within the ESNII+ Project

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Abstract. The thermal conductivity is an important parameter for the assessment of the FBR fuel thermal behavior during irradiation. In this study, the effect of burn-up on the thermal conductivity of FBR MOX fuel is investigated experimentally. The thermal diffusivity of FBR fuel with a burn-up of 13 at.% was measured by the laser flash technique and the thermal conductivity was deduced. It is shown that the thermal conductivity is higher than could be predicted with correlations issued from LWR UO_2 and MOX results. This effect is interpreted as being due to the diffusion of fission products out of the fuel matrix, leading to the formation of the JOG.

Key Words: mixed oxide, fuel, properties

1. Introduction

In order to develop the fast neutron systems, three prototypes of the Sodium Fast Reactor, the Gas-cooled Fast Reactor and a heavy liquid metal cooled Accelerator Driven System are studied in Europe: ASTRID (SFR prototype), ALLEGRO (GFR prototype) and MYRRHA (an LBE-cooled ADS system related to the ALFRED LFR-demonstrator). The ESNII+ project with its workpackage *7-FUEL SAFETY* aims to provide a set of oxide fuel properties needed for the fuel element design of each prototype. The improvement of fuel properties will also reduce uncertainties in safety behavior evaluations, in nominal conditions as well as during transients and will be achieved by the update of the European catalog on the MOX fuel properties, used for the fuel design of the ESNII prototypes. While the properties of fresh fuel will be updated with new measurements, properties on irradiated mixed oxide fuel will be integrated and a tentative correlation with burn-up will be proposed.

The associated uncertainties have to be rigorously determined; the two main driver criteria for fuel element evaluation are the margin to melt for the fuel and the risk of clad failure. It is commonly assumed that the properties of the fuel yield the dominant contribution to the uncertainties in thermal and mechanical evaluation. The measurements of the main thermal and mechanical properties on fresh and irradiated fuel should enhance the reliability of fuel behavior calculations and contribute to the design of safer fuel elements for ALLEGRO, ASTRID and MYRRHA. Property measurements are done on existing fresh and irradiated fuel samples, identified to cover the fuel characteristics for ESNII prototypes.

The review of the state-of-the art has shown that the knowledge on the thermal conductivity of irradiated FBR MOX is currently very limited. Only one publication is available providing an experimental result which is surprising: no degradation of thermal conductivity with burn-up was observed.

In this work, fresh and irradiated fast reactor fuel was characterised and its thermal diffusivity was measured. The considered FBR irradiated fuel has an average burn-up of 13 at %. First, the characteristics of the fuel and the measurement method are presented. Then the thermal diffusivity results are given and the thermal conductivity is calculated. Finally, the data and models available in the literature are reviewed and compared to the new experimental results in order to develop an updated recommendation.

2. Fuel characteristics

The samples are short segments of irradiated FBR MOX fuel $(U,Pu)O_2$ transported to JRC Karlsruhe within the ESNII+ European project for measurements of thermal diffusivity and high temperature stability. The origin of the fuel is the LECA/STAR facility in Cadarache. The MOX fuel corresponds to the NESTOR 3 irradiation and had a fresh fuel Pu content of 19.82 at. % (Pu/(U+Pu+Am)). The investigated fuel was irradiated up to the average burn-up of 13.3 at.% in the Phenix reactor. The irradiation temperatures were at the beginning of life: 1080K at pellet periphery and 2550K at the pellet centre, and at end of life: 1080K at pellet periphery and 2090K close to the central hole. The fuel density at end-of-life ranged between 95% TD at the pellet periphery and 97.1% TD close to the central hole. A fuel fragment covering the radial positions between approximatively the mid-radius to the central hole was analysed by SEM (*FIG. 2*). The microstructure with columnar grains can be clearly recognised.

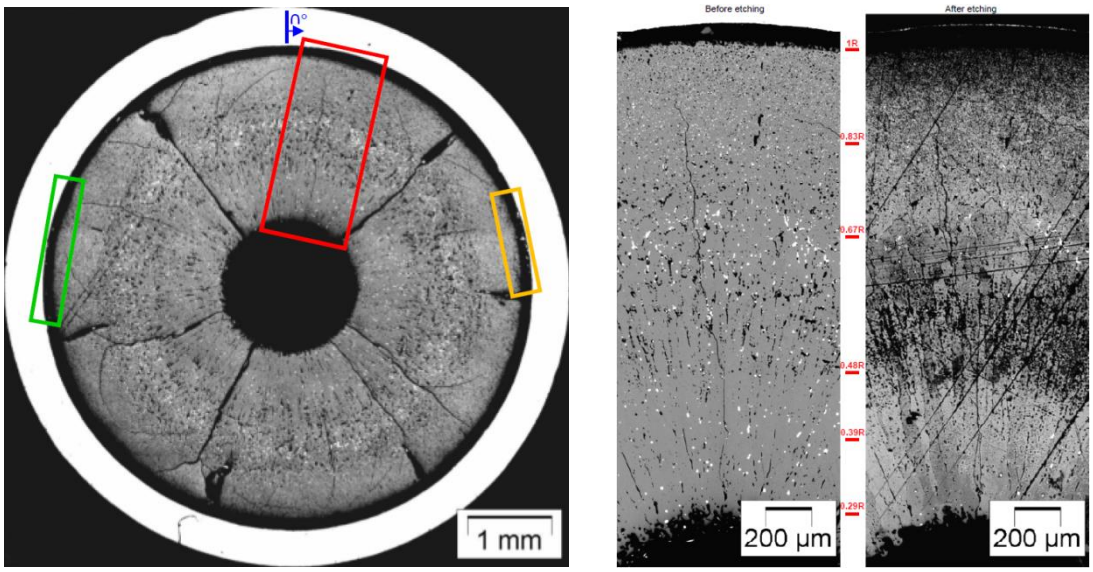

FIG. 1: Macrograph showing the complete fuel section and detailed view, obtained at the LECA star facility in CEA Cadarache.

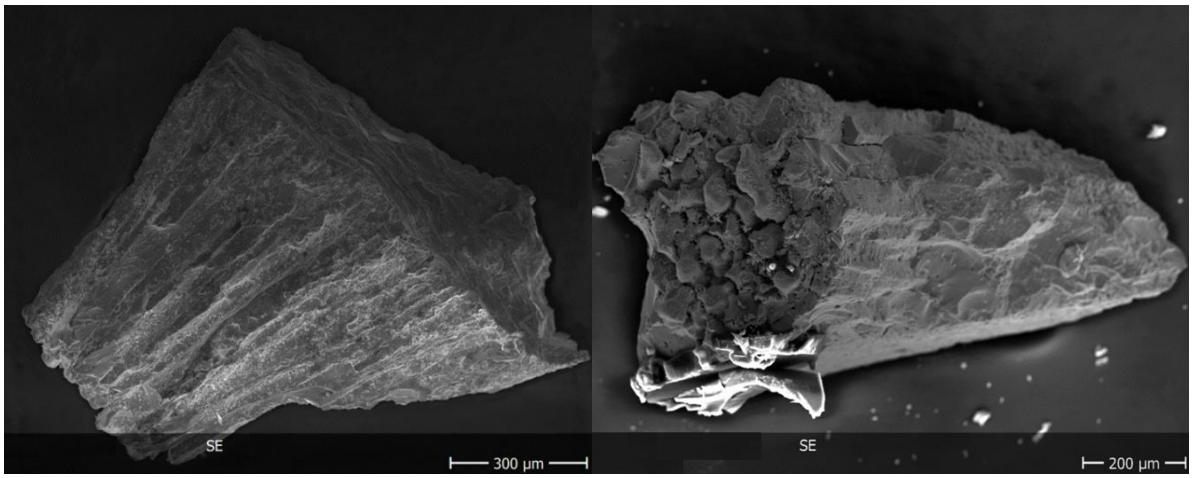

FIG. 2: SEM picture showing the microstructure of the fuel from the central hole to approximatively the mid-radius. Left: radial view. Right: view from the central hole position

3. Method for the thermal diffusivity determination by the laser flash set-up

The measurements of the thermal diffusivity were carried out in a laser-flash device (LAF.I) installed in a lead-shielded glove box provided with remote manipulators [1]. The sample is heated at the measurement temperature (between 500 and 1600K) in a high frequency furnace under a nitrogen atmosphere of 10^{-2} mbar. A laser pulse (10 ms duration in the case of irradiated fuel) is then applied to the front surface of the sample; the emerging temperature perturbation on the opposite surface is recorded by a photodiode pyrometer (0.05K sensitivity). The thermogram recorded by a 24 bit digitalizer, $T=T(t)$, is analysed by a realistic and accurate mathematical model of the pulse propagation in the sample: thermal diffusivity and various heat losses are calculated by a numerical fitting procedure. The local thermal diffusivity was measured in different radial positions by moving the pyrometer field-stop aperture (500 µm radius) through step motors across the image of the specimen surface produced by a water-cooled, high quality macro-objective placed in the vessel a few centimeters above the HF furnace coil. The aperture was drilled in an optical mirror that reflected the full image of the sample enabling an exact determination of the position of the measured field. The laser probe beam diameter was spatially homogeneous and slightly larger than the sample. Validation measurements were carried out with standard materials and fresh UO_2 pellets with known thermal diffusivity in order to check the quality of the results obtained for platelets of irregular shapes.

The precision of the individual measurements is better than 1% for the thermal diffusivity. The accuracy of the measured thermal diffusivity is, however, worse than the precision of the method, being principally determined by sample thickness variations which lead to a precision of 5% for the considered sample.

4. Thermal diffusivity at the burn-up of 13 at. %

The thermal diffusivity sample was a quarter of a disk, with clearly recognizable positions for the region in contact with the cladding and the central hole. The thermal diffusivity was measured in 3 radial positions: 0.6 mm from cladding, 1 mm from cladding and 1.4 mm from cladding. These positions were regularly distributed over the 2 mm width of the sample,

measured from the surface in contact with the cladding and the surface corresponding to the central hole.

The thermal diffusivity is shown in *FIG. 3*, the 3 radial positions are labelled as following:

-“cladding”: 0.6 mm from cladding, with estimated irradiation temperature of about 1600K

-“middle”: 1 mm from cladding with estimated irradiation temperature of about 1850K

-“center”: 1.4 mm from cladding with estimated irradiation temperature of about 2000K

Because of the limitation of the measurement technique, the extreme periphery of the sample corresponding to the JOG (the JOG having a thickness of about 150 micrometres for the considered sample) does not contribute significantly to the measurement results obtained at the “cladding” position, which is centered at a distance of 0.6 mm from the cladding.

Five annealing runs were done with the following maximum temperatures: 630 (*FIG. 3*), 710 (*FIG. 4*), 860 (*FIG. 5*), 1120 (*FIG. 6*), 1490 K (*FIG. 7*). The sample moved and lost some small fragments at a temperature of about 1500K, small sample portions detached on the cladding side corresponding to the low irradiation temperature region. The experiment was stopped, the sample was put back in position and the thermal diffusivity was measured again in order to quantify the recovery resulting from the high temperature annealing. It was decided not to increase temperature higher than 1500K (the maximum temperature for the set-up is 1600K) in order not to risk a more energetic fragmentation of the sample which could result in spreading sample fragments in the set-up. The fragmentation temperature corresponds relatively well to the average irradiation temperature of the sample region close to the cladding.

After the first measurement run (*FIG. 3*), recovery was observed, except on the position close to cladding. After the second measurement run (*FIG. 4*), recovery was only observed on the position close to cladding. After the third measurement run (*FIG. 5*), recovery was observed at all positions, with a particularly large amplitude for the position close to cladding. After the forth measurement run (*FIG. 6*), recovery was observed at all positions. The results at the position close to cladding show a behaviour that can deviate from a smooth behaviour, this could be due to some damage appearing in this region, even is no obvious sample damage was recognised visually. Some recovery was also observed after the annealing at the maximum temperature of 1490K (*FIG. 7*).

Because the recovery was observed at temperatures below the irradiation temperatures, the recovery is attributed to the annealing of irradiation damage produced out of pile by auto – irradiation. In the case of FBR fuel, irradiation point defects are annealed very efficiently in pile because of the very high irradiation temperatures, but auto-irradiation takes place out-of pile at the storage temperature and defects can accumulate. The results relevant for the in-pile fuel properties are therefore the thermal diffusivity obtained after annealing at the maximum temperature (fifth measurement run).

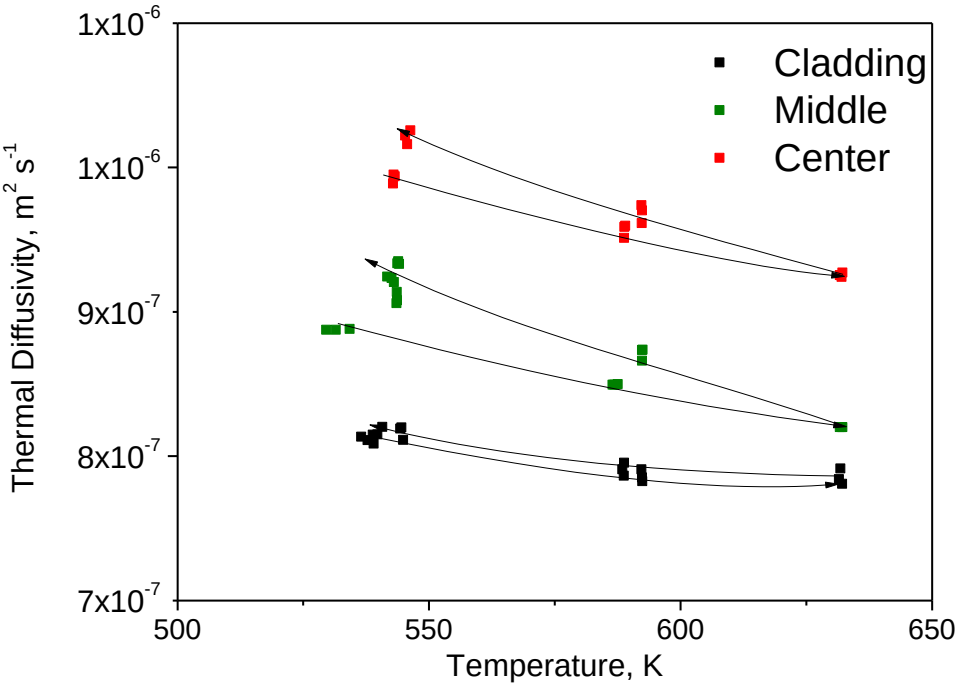

FIG. 3: Thermal diffusivity of the sample with 13 at% burn-up measured during the first run

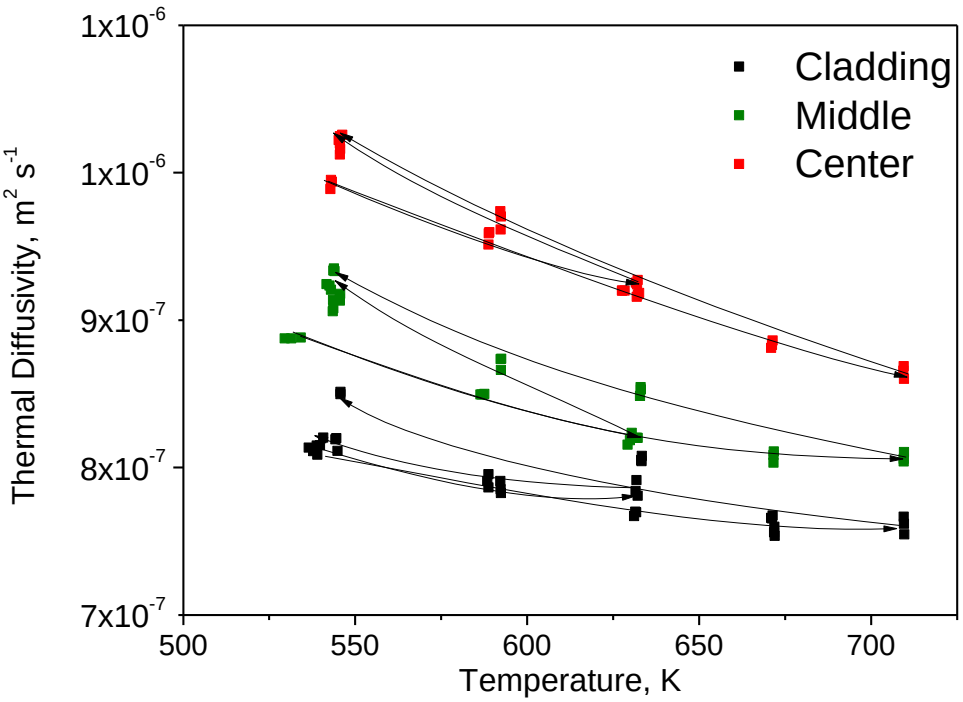

FIG. 4: Thermal diffusivity of the sample with 13 at% burn-up measured during the first and second runs

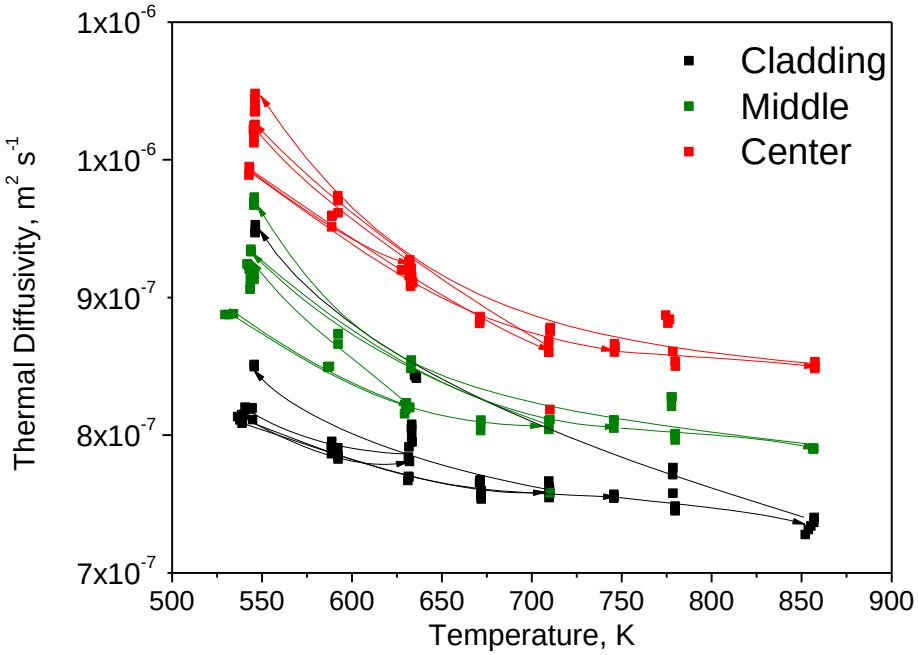

FIG. 5: Thermal diffusivity of the sample with 13 at% burn-up measured during the runs 1 to 3

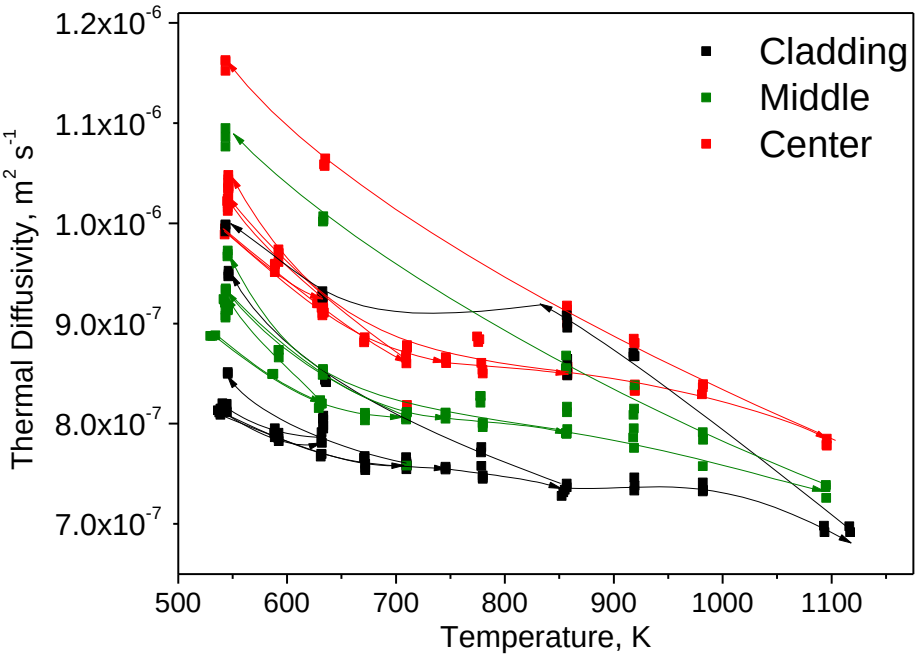

FIG. 6: Thermal diffusivity of the sample with 13 at% burn-up measured during the runs 1 to 4

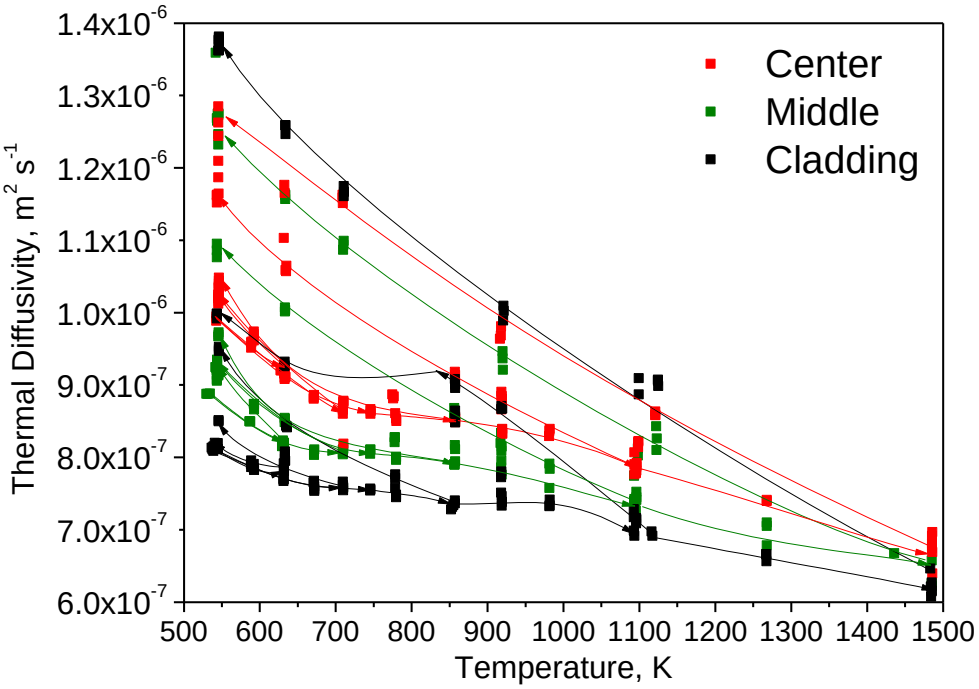

FIG. 7: Thermal diffusivity of the sample with 13 at% burn-up measured during the runs 1 to 5

5. Thermal conductivity

The thermal conductivity was deduced from the measured thermal diffusivity, the density predicted by a fuel performance code based the fresh fuel value and the specific heat of the fresh fuel. The results are shown in *FIG. 8*. For the last run, no large difference is observed between the three investigated radial positions, which is consistent with the relatively flat burn-up and density radial profiles. This shows that the differences in irradiation temperatures (1600 to 2000 K) did not induce significant differences in the local thermal conductivity. An effect of the increase of the plutonium concentration at the position close to the central hole region is also not observed, which is consistent with the observations on fresh fuels showing no significant effect of this parameter: the thermal conductivity of fresh fuels is almost identical at the stoichiometric state for Pu contents in the range 3 to 30 at.% [2] [3].

The knowledge on the thermal conductivity of irradiated FBR MOX is currently very limited. Only one publication is available [4] providing an experimental result which is surprising: no degradation of thermal conductivity with burn-up was observed, within the relative experimental uncertainties which were estimated to be up to 20 %. Philipponneau published a recommendation based on a review of fresh fuel results where the effect of burn-up was quantified by doping with simulated fission products [5-7]. The formula of Philipponneau includes the effect of solid fission products but no effect of irradiation damage.

At the same time, a large number of models is available for the thermal conductivity of irradiated LWR UO_2 and MOX. Some of these models provide a very detailed description of the different burn-up effects: soluble fission products, volatile fission products, radiation damage. The radiation damage effect is burn-up and irradiation temperature dependent. The concentration of irradiation point defects saturates early with burn-up, but the concentration of extended irradiation damage (dislocations, bubbles) increases with burn-up. Temperature has a strong impact on the concentration of irradiation damage point defects, with a recovery

taking place above about 1200 K. The fission gas atoms can either be considered as dispersed as atoms in the fuel matrix, where they have the highest impact on thermal conductivity, or released form the fuel matrix, in which case there is no impact on the fuel thermal conductivity. The case of fission gas atoms dispersed in the fuel matrix corresponds to the low irradiation temperature range in LWR fuel (for instance below about 1000K), which is not present in FBR fuels.

The thermal conductivity at End-Of-Life (EOL) of irradiated LWR UO_2 and MOX [8], evaluated for the low irradiation temperatures relevant to LWR and extended to the burn-up of 13 at % is shown in *FIG. 8* (line "LWR UO_2 and MOX, at EOL"). This thermal conductivity is well below the results obtained for the FBR MOX, which could be explained by the effect of radiation damage and fission gas atoms present in the LWR fuel. If the formula for irradiated LWR UO_2 and MOX [8] is evaluated for an annealed fuel (i.e. where the effects of radiation damage and dispersed fission gas atoms are suppressed, line "LWR UO_2 and MOX, Annealed" in *FIG. 8*), the obtained thermal conductivity is still well below the results obtained for the annealed FBR fuel. This shows that the very high irradiation temperatures in the FBR fuel have some supplementary beneficial effects, for instance the diffusion of part of the solid fission products from the fuel matrix to the JOG.

The measurements for the NESTOR 3 fuel at 13 at% are compared in *FIG. 8* with the thermal conductivity of the unirradiated MOX [2] and of irradiated fuels at the same burn-up: LWR UO_2 and MOX [8] and the recommendation of Philipponneau for FBR MOX [3]. The thermal conductivity of the irradiated FBR fuel is significantly below the fresh fuel value. The recommendation of Philipponneau for FBR MOX [3] is in good agreement with our experimental results. In the case of LWR fuels, the predictions from results obtained with fresh fuels doped with simulated fission products are usually underpredicting the effect of burn-up (i.e. predict a too high thermal conductivity for a given burn-up), as observed when the SIMFUEL results [9] are compared to measurements on real irradiated LWR fuel [10]. In the case of FBR fuels and supposing that a fraction of the FP have left the fuel matrix and are present in the JOG, the fraction of FP in the matrix of the real FBR fuel is smaller than the fraction of FP introduced and present in the simulated fuel with same burn-up.

Recent results obtained on fuel with lower burn-up (9.9 at. %) and lower irriadiation temperature have shown a thermal conductivity in the range of LWR fuel, i.e. much lower than in the case of the fuel with 13 at. % presented in this work. An analysis of the JOG is on-going in order to investigate if this result is due to a reduced diffusion of fission products out of the fuel matrix. The JOG was found to be twice more thick in the fuel with 13 at. % burn-up. Another significant difference is the presence of metallic precipitates in the fuel with 13 at%. burn-up, which are not observed in the fuel with 9.9 at. % burn-up. The extent of JOG and the presence of metallic precipitates are therefore proposed as major parameters for the estimation of the FBR fuel thermal conductivity.

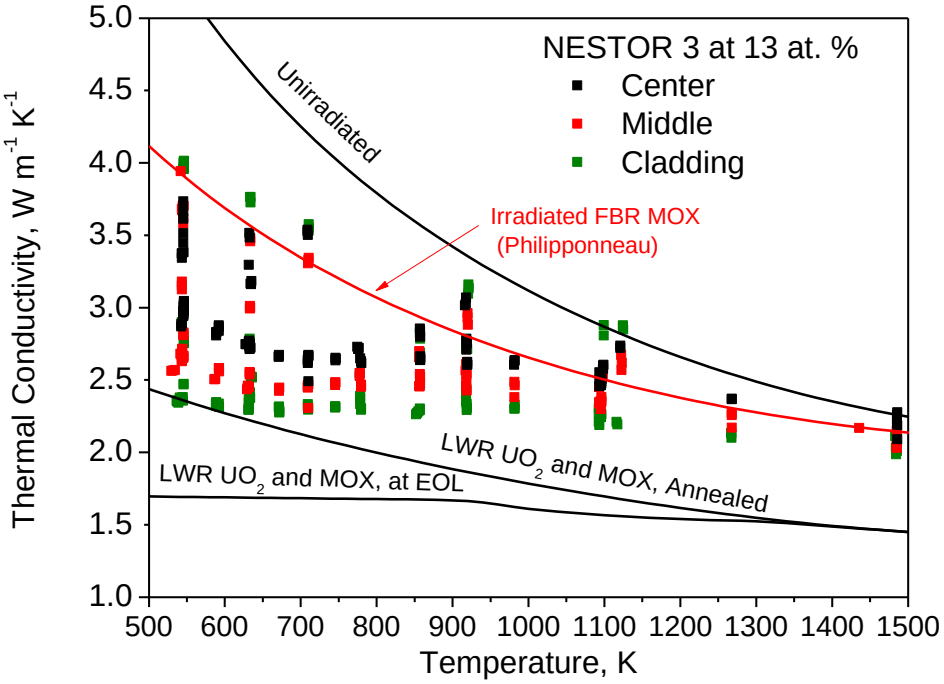

FIG. 8: Comparison of the measurements for NESTOR 3 fuel at 13 at.% with unirradiated MOX [2] and irradiated fuels at the same burn-up: LWR UO2 and MOX [8] and the recommendation of Philipponneau for FBR MOX [3]

6. Conclusions

The data and models available in the literature for the FBR MOX thermal conductivity were reviewed and new experimental results are obtained in order to develop an updated recommendation. Fresh and irradiated fast reactor fuel was characterised and its thermal diffusivity was measured. The irradiated fuel has an average burn-up of 13 at % and the thermal diffusivity was measured in 3 radial positions: 0.6 mm, 1 mm and 1.4 mm from the pellet cladding.

Auto irradiation damage is accumulated during storage after irradiation. This damage is annealed during out-of pile measurements, where a recovery of the thermal diffusivity is observed for temperatures below the irradiation temperature.

The thermal conductivity was found to be significantly higher than for LWR UO_2 or MOX fuels with similar burn-up. The impact of the irradiation temperature, radiation damage concentration, plutonium content, fission gas atoms and fission products was considered by comparison with the predictions of correlations available for irradiated LWR fuel. It was shown that the correlations for LWR fuels can not be adapted to FBR fuel even by adjusting the irradiation temperatures to levels allowing for a complete recovery of radiation damage and release of the fission gas atoms from the fuel matrix. The high thermal conductivity of the FBR fuel can only be achieved by considering a lower effect of the non volatile fission products, part of which are present in the JOG and therefore do not contribute anymore to the fuel thermal conductivity degradation.

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