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Irradiation-Induced Thermal Effects in Alloyed Metal Fuel of Fast Reactors

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Abstract. The paper presents the results of studying alloyed metal fuel after irradiation in a fast reactor. Determined is the mechanism of fuel irradiation swelling, mechanical interaction between fuel and cladding, and distribution of fission products. Experience gained in fuel properties and behavior under irradiation as well as in irradiation-induced thermal effects occurred in alloyed metal fuel provides for a fuel pin design to have a burnup not less than 20% h. a.

1. Introduction

Metal fuel remains the most attractive fuel to be used in fast reactors due to its simple manufacture process with the use of existing metallurgic techniques, high density of fission isotopes, high thermal conductivity, and compatibility with sodium coolant. Further electrochemical reprocessing of metal fuel and a metallurgical method of manufacturing new fuel slugs may be the decisive factors in minimizing the cost of the fuel cycle and generated electric power [1]. This fuel is considered to be one of the most promising fuels to be used in Generation IV Reactors. A large-scale program on studying metal fuel has been conducted in the U.S. [2-6]. The U.S. program focused on physical properties of uranium-based and uranium-plutonium-based alloys, their irradiation characteristics, optimal fuel composition and fuel design. Thorough work has been carried out in the feasibility of fuel performance including life tests and studying fuel behaviour under transient and abnormal conditions. Among the examined fuel compositions were U-5%Fs (uranium alloyed with metals – 5wt.% fission products), U-10%Zr, and U-Pu-Zr. The final choice of the fuel composition was made in favor of U-Pu-Zr. Zirconium was introduced into the alloy to increase the melting point and decrease chemical fuel-to-cladding interaction. The density of U-19%Pu-10%Zr in an extruded state is equal to 15.82 g/cm^3. The solidus temperature makes up approximately 1100°C, and the thermal conductivity ranges from 15 to 30 W/mK in the temperature interval from 400 to 1000°C. The alloy strength decreases rapidly as the temperature increases, e.g., the tensile strength of U-15%Pu-10%Zr ranges from 480 MPa at room temperature to 97 MPa at 400 °C. The yield strength at 400°C is equal to 69 MPa, and the relative elongation makes up 53 %. Low strength and high plasticity of U-Pu-Zr alloys favorably manifest themselves in swelling under irradiation exerting low pressure on the fuel cladding. In free swelling the U-Pu-Zr fuel density makes up approximately 75 % of the theoretical value at a burnup of 2-3 % h.a. By that time, a gap between fuel and cladding disappears, and fuel contacts the cladding.

Porous fuel appears to be of so high plasticity to provide weak mechanical fuel-to-cladding interaction. This effect became a decisive factor in maintaining the performance of fuel irradiated up to a high burnup. As the fuel swelling mechanisms were revealed, its quantitative characteristics were specified and the fuel pin design was improved, the burnup increased consistently from ~1 to ~18 % h.a. On single fuel pins the peak burnup of 19.9 % h.a. was achieved. At a fuel burnup of 16-18 % h.a. there was an 8% increase in the cladding diameter of fuel pins made of D9 austenitic steel that is likely to undergo irradiation swelling. As for fuel pins made of HT9 ferritic-martensitic steel that is not likely to undergo swelling under the same conditions, this value made up about 1%. Based on these data American scientists concluded that in fast reactors operated with metal fuel a burnup of 200 GW/t HM can be achieved, and the simple manufacture process and reprocessing technique of U-Pu-Zr fuel give benefits for its wide use.

In Russia the first experiments in the development of alloyed metal uranium- and plutonium-based fuel for fast reactors were launched in 1950-60 [7]. Later on, tenacious work related to these issues stopped. From 1988 to 2001 a range of activities has been completed including manufacture of four fuel assemblies (FAs) and their irradiation in the BOR-60 reactor. These FAs contained experimental fuel pins (U-Zr and U-Pu-Zr). Post-irradiation examination (PIE) data of three FAs are presented in papers [8-10]. This paper presents the results of studying FA VS-174e.

2. Fuel characteristics and irradiation conditions

Table 1 gives the main fuel parameters, design features and irradiation parameters.

Table 1. Parameters of experimental fuel pins and irradiation conditions.

Parameter	Value
FA shroud	EP-450
Fuel cladding	0Kh16N15M3B
Cladding wall diameter and thickness, mm	6.0×0.3 mm
Fuel column height, mm	450±3
Fuel composition	U-18%Pu-10%Zr
Fuel pin diameter, mm	4.7±0.1
Average relative effective fuel density, %	75±3
Heat-transfer fuel-cladding layer	Na
Peak burnup, % h.a.	9.7
Peak neutron fluence (E>0.1 MeV), cm^{-2}	1.28×10^{23}
Peak linear thermal power, W/cm	400
Peak temperature on the cladding inner surface,°C	600

3. PIE data

3.1. Irradiation swelling and fuel-cladding mechanical interaction

The cladding diameter was measured on ten fuel pins by profilometry. Figure 1 shows a profile diagram of one fuel pin.

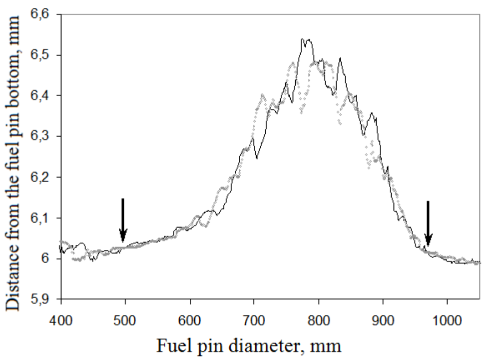

Figure 1. Typical profile diagram in two mutually perpendicular planes of a fuel pin with metal fuel after irradiation in BOR-60 to achieve the peak burnup of 9.7 % h.a. The arrows show the fuel slug boundaries.

This profile diagram was obtained by scanning along the fuel pin in two mutually perpendicular planes. The ovality of fuel cladding is seen in the area of the maximum diameter increase resulted from mechanical interaction with neighboring fuel pins through the spacer wire. According to the measurement data obtained on ten fuel pins in the deformation maximum areas, the average value of the relative diameter increase is equal to 7.1±0.1 %. The absence of the “steps” in the profile diagram on the boundaries of fuel slugs shows that there is insignificant pressure of fuel on the cladding. It agrees with the conclusions of American scientists about the results of metal fuel irradiation. The maximum swelling of steel 0Х16Н15М3Б in a solution treated state makes up 15% for the examined fuel pins under irradiation. Therefore, an increase in the cladding diameter due to irradiation swelling is 5%. About 2% of diameter growth results from irradiation-induced creep under stress due to fission gas pressure. Based on gamma scanning results the relative elongation of the fuel slugs makes up 4.2±0.7 %. The relative increase in the fuel volume under fuel-to-cladding interaction is equal to approximately 46 %, and the relative fission gas release from fuel into a free volume of the fuel pins makes up about 80 % according to the results of cladding puncture and gas phase analysis, which is close to the data obtained in the EBR-2 reactor [3].

3.2. Fuel microstructure

In examining cross-sections of the fuel pins of different levels relative to the fuel slug height with the use of a metallographic microscope, it was found that in all cross-sections the fuel slug diameter increased before fuel contacts the cladding. Sodium did not prevent swelling, and it was displaced from the gap. In most cross-sections fuel has a layered ring structure (Figure 2).

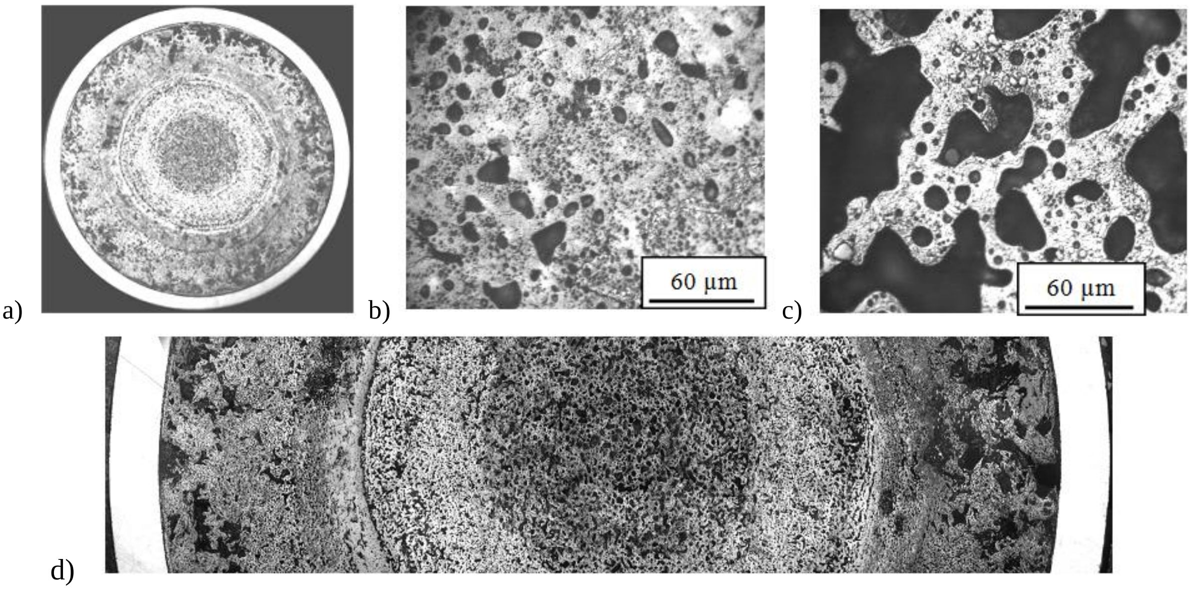

Figure 2. Fuel microstructure in the central cross-section of the fuel pin active core: a) – full cross-section macrostr d) b) – bubble area, c) – cavitation swelling area, d) – panoramic image in a diametrical direction.

The porosity nature depends on the fuel slug operating temperature. It changes both radially (in the fuel pin cross-section) and axially (fuel pin heightwise). When the fuel temperature is low, i.e. in the outer ring area of the fuel slug and in the lower parts of the fuel slug, cavitation swelling is observed with the presence of large-size pores with zigzag boundaries. At a higher temperature bubble swelling area is seen with pores of mainly smooth and round shape. A change in the porosity nature is clearly seen as ring layers on macrographs and panoramic photographs of the fuel pin cross-sections. This change arises from phase formation in fuel verses temperature. At a temperature lower ≈670 °C in alloy U-Pu-Zr an orthorhombic alpha phase is generated showing a tendency to strong anisotropy that manifests itself in the anisotropic irradiation-induced growth. This effect is based on anisotropic distribution of irradiation defects in the crystal lattice [15]. Under irradiation crystals grow in size in direction [010] and decrease in size in direction [100]. As a result, along with generation and growth of gas bubbles in the low-temperature outer ring area of the fuel slug there is redistribution of the material in the grain volume due to irradiation growth in a certain crystallographic direction. This leads to a break in the grain boundaries and formation of microscopic discontinuity flaws. Fission gas makes stable the generated discontinuity flaws that could further grow and change their shape. These processes result in a typical microstructure that is always observed in a low-temperature area of the fuel slug.

3.3. Distribution of fuel components and fission products

Distribution of the fuel components along the radius of the fuel slug, accumulation and state of fission products may affect the melting point and fuel thermal conductivity. Therefore, in the course of PIEs attention was paid on these issues. Figure 3 shows a typical microstructure obtained by scanning electron microscopy and distribution of the fuel components along the fuel slug radius in the fuel pin cross-section with the peak local burnup of 9.7 % h.a. obtained by electron probe microanalysis. The given results show layering of alloy U-Pu-Zr in composition and peculiarities of its structure formation affected by the radial temperature gradient. At a distance from 900 to 1400 μm from the inside of the cladding a layer was formed in which the zirconium weight fraction makes up less than 1 %, plutonium content is decreased and uranium content is increased. It is evident that zirconium migrated in the fuel central part. In addition, in distribution of zirconium the local maximum is observed due to the presence of zirconium alpha phase inclusions. On the periphery of the fuel slug zirconium migrated towards the surface with the generated alpha phase layer that is of high importance in terms of preventing fuel-to-cladding interaction.

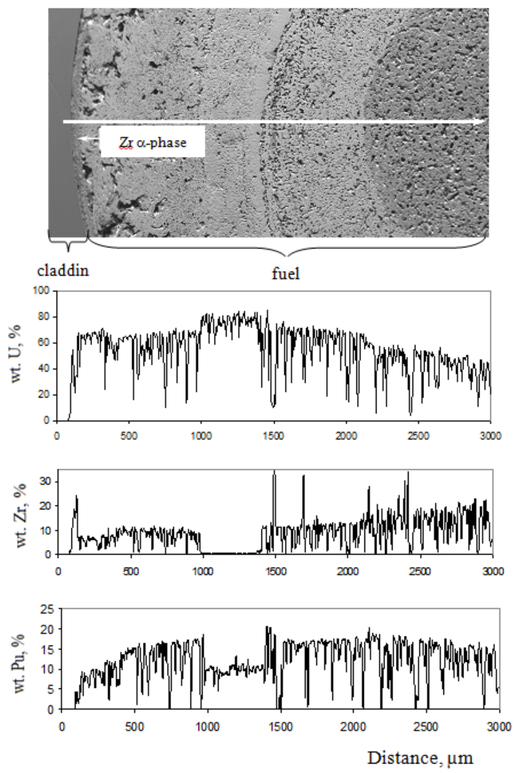

Figure 3. Electron microscope image of the fuel microstructure and distribution of the components along the fuel slug radius in the fuel pin cross-section with a burnup of 9.7 % h.a. The arrow shows scanning direction from the cladding to the fuel slug center.

The redistribution of components results from the diagram structure of the alloy U-Pu-Zr state. Despite the apparent complexity of structure formation the basic mechanisms are well-known, and their influence on fuel and fuel pin performance is determined. For instance, in studying chemical fuel-to-cladding interaction a favorable effect of zirconium alpha phase was observed on the fuel slug surface. There were some concerns about formation of a ring area of depleted zirconium in the fuel slug due to a possibility of a solidus temperature decrease in this area. However, it turned out that the main component in this area is uranium. Plutonium content was lower that the set value, and the correlation of components corresponds to the solidus temperature of about 1000 °C, which is significantly higher than the operating temperature in this fuel slug area.

Study of the fission product distribution shows that the content of the fission products in a solid solution is low except for molybdenum, and their main fraction presents in fuel as metal inclusions. Among all fission products molybdenum is described by the highest solubility in the alloy, and the nature of its distribution along the slug radius is similar to that of zirconium distribution.

4. Conclusion

At the early stage of irradiation under free swelling (without mechanical suppression) alloyed metal fuel increases rapidly in volume mainly due to formation and growth of gas pores. At a burnup of about 3 % h.a. the volumetric fuel swelling achieves 30-35 %, which corresponds to the effective density of about 75 %. The fuel state undergoes changes described by generated open porosity, high fission gas release, and as a result, a sharp slowdown in swelling. Further long-term irradiation does not change significantly the fuel state. Fuel swelling occurs due to solid fission products, and the swelling rate is low. Such fuel state that does not change much under irradiation was observed up to a burnup of about 20 % h.a. If the effective fuel density initially makes up no more than 75 % due to a slug-to-cladding gap, fuel contacts the cladding after the porous structure is formed. In the course of further irradiation this structure does not exert significant pressure on the cladding. No fuel cladding diameter increase was observed resulted from fuel-to-cladding mechanical interaction with the fuel density of 75 %. The major contribution to cladding loading was made by fission gas pressure that was compensated by an increase in the gas plenum volume. The PIE data fully confirm the described mechanism. Irradiation-induced thermal effects in alloyed metal fuel do not restrict fuel pin performance. Its operating life is determined by irradiation resistance of the cladding material.

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