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Structure and properties of hafnium after a 9-year operation in the RBT-6 research reactor

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A R T I C L E   I N F O

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A B S T R A C T

Hafnium control rods in the form of plates 6 mm thick were examined after a 9-year operation in the RBT-6 research reactor in water coolant at 68–86 °C to a maximal fluence 7.5×10^{25} m^{-2} ($E < 0.625$ eV) and 2.4×10^{25} m^{-2} ($E > 0.8$ MeV). The Hf crystalline lattice volume increased by 0.4% and its density decreased. The oxide film thickness did not exceed 1 μm. Linear-type a-dislocations with a concentration of $(5 \pm 2) \times 10^{13}$ m^{-2} were found in the structure as well as fine precipitates, where iron and chromium were detected, in addition to hafnium. Hf strength changed insignificantly; the material preserved its plasticity at a level of 8–15%. The performed examinations allowed the prolongation of the control rod design operation for up to 20 years.

1. Introduction

Since the end of the 1950s, metal hafnium has been used as an absorbing material in control rods of nuclear reactors [1,2]. The available literature primarily provides its properties after operation in PWR and BWR reactors [3–6]. In 1997, hafnium was used for the first time in the VVER-1000 control rods (Zaporozhie NPP, Ukraine) [7]. Control rods of a complicated geometry are used in research reactors which are operated at relatively low coolant temperatures that allows hafnium to be used both as a structural and neutron absorbing material.

This paper presents the examination results of a Hf control rod after a 9-year operation in the RBT-6 research reactor. These examinations resulted in the justification of serviceability of control rods designed at RIAR for two Chinese nuclear reactors CARR and CMRR.

1. Specifics of the design and operating conditions of the control rods

The RBT-6 research reactor is a pool-type water-cooled reactor; its core comprises 54 fuel assemblies (FAs), 6 control rods that serve both as emergency protection and shim rods (EPSR), and one power automatic control rod (PACR). The core is assembled of FAs installed in a support grid. Gaps between FAs are used to move EPSR and PASR inside them. The reactor core is immersed into the pool filled with distilled water serving as a coolant. Parameters of coolant are presented in Table 1.

On opposite sides of the core, there are two blocks of EPSR located on the reactor vessel. Each block consists of three sections. Each section is comprised of two parallel absorbing plates of special profile (Fig. 1). One EPSR section has non-cladded Hf absorbing plates 6 mm thick. Table 2 presents the chemical composition of the metal hafnium used for plate fabrication.

The control rod under examination was operated in the RBT-6 reactor for 9 years, i.e. for 2047 effective days.

The results of neutron-physics calculations showed a significant non-uniformity in the distribution of the neutron fluence on the Hf plate surfaces. Fig. 1b presents the fluence distribution in a cross-section of one plate. The maximal neutron fluence made up 7.5×10^{25} m^{-2} ($E < 0.625$ eV) and 2.4×10^{25} m^{-2} ($E > 0.8$ MeV).

According to the thermo-physical calculations, the temperature of the Hf plates during operation was between 68 and 86 °C on the surface and 69 and 88 °C in the middle of the plates.

2. Examination of the Hf plate after operation

No failures of the hafnium EPSR were registered during its operation in the RBT-6 reactor; the physical efficiency remained essentially the same. All the components preserved their shape and integrity. The absorbing plates remained parallel, no deformation or bending of the surface was observed. There were neither sediments nor corrosion damage. In the area of the highest neutron fluence, the temper colors of the plate surfaces varied from yellow to blue. The plate thickness did not change during operation and varied between 5.73 and 6.20 mm, which corresponded to the plate fabrication tolerance.

To perform material testing, the control rod section was disassembled into components. Samples were cut off from one of the hafnium plates.

Examinations performed on optical and scanning microscopes using metallographic sections cut from the examined plate

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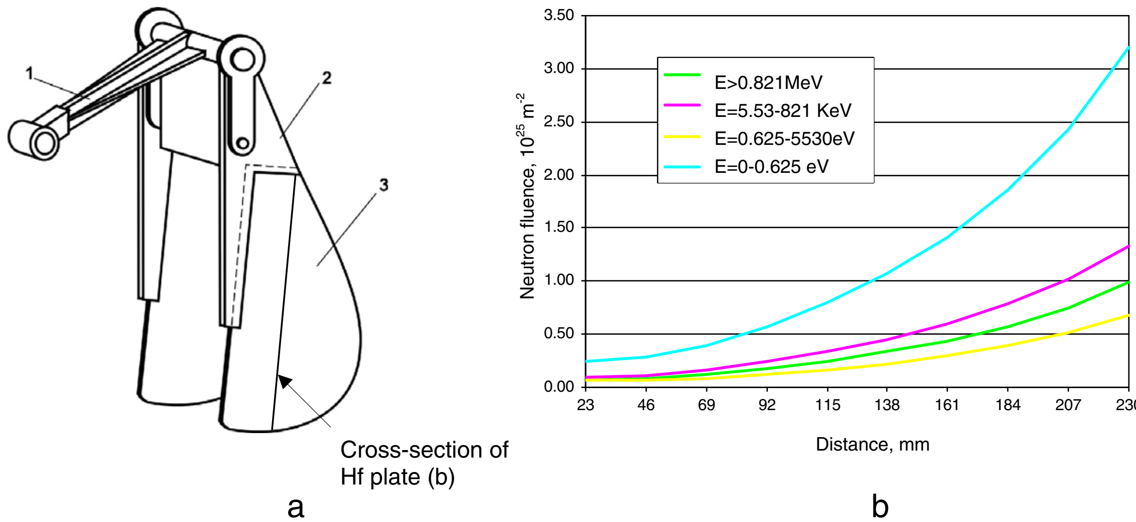

Fig. 1. Hafnium EPSR section (a): 1 – EPSR traction; 2 – shim plate 3 – hafnium plate; Calculated neutron fluence distribution in a cross-section of one Hf plate (b).

revealed no oxide film on the surface contacting the coolant. Consequently, the oxide film thickness was less than 1 μm (Fig. 2) in agreement with the temper film appearance described earlier.

The average equivalent grain diameter was 55 μm in the section transverse to the rolling direction and 69 μm in the longitudinal direction (Figs. 3 and 4).

The microhardness of the hafnium not changed after irradiation and its value was 2600 ± 300 MPa.

The structure of unirradiated hafnium is characterized by the presence of re-crystallized grains, inside which initial dislocations and their clusters are observed (Fig. 5a). There are no c-dislocations, as can be seen in the micrograph of a foil in a prismatic orientation obtained with an [0 0 2] reflection (Fig. 5b). A reflection of resulted in the appearance of linear-type a-dislocations (Fig. 5c), with a concentration of $(5 \pm 2) \times 10^{13}\ m^{-2}$. Precipitates were observed (Fig. 5d) that contained, in addition to hafnium, iron and chromium with an atomic concentration of (2.91 ± 0.46) and (2.80 ± 0.46)%, respectively.

Table 1
Parameters of coolant of the RBT-6.

Parameter	Value
pH	5.0–7.0
Electric conductivity	<4.0 μS/cm
Hardness	<3.0 μg/kg
Concentration of Cl^-	<50.0 μg/kg
Concentration of Fe	<50.0 μg/kg

Table 2
Hf chemical composition of the RBT-6 EPSR plate.

Element	Content mass portion, %
Hf + Zr	99.8
Zr	0.81
	Mass portion, ppm
Al	<15
Co	5
Cu	<15
Fe	240
H	10
Mg	<20
Mn	10
Mo	10
Nb	<60
Ni	<10
Si	<60
Sn	<30
Ti	10
Ta	<60
V	10
W	<20
Cr	<20
U	<0.5
Ca	<10

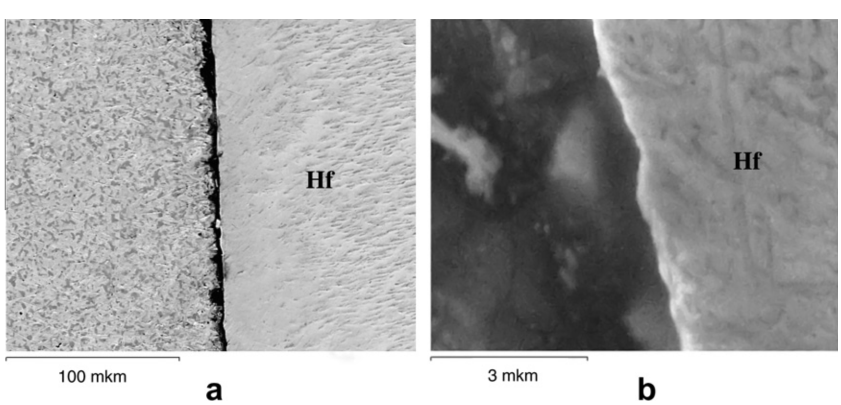

Fig. 2. Structure of hafnium plate surface at different magnifications: a – optic microscope; b – scanning microscope.

The structure of irradiated Hf sample cut off from the plate is shown in Fig. 6. The estimated fluence of the examined section was 0.25×10^{25} m^{-2} ($E < 0.625$ eV) and 0.06×10^{25} m^{-2} ($E > 0.8$ MeV). It resembles the structure of unirradiated hafnium. There are a-dislocations and no c-dislocations. The only difference is the presence of small radiation defects in the form of black-and-white dots 5 nm in size that appeared in the Hf matrix.

The Hf hexagonal close packed crystalline structure did not change under irradiation, but the parameters of the crystalline lattice and the volume of the elementary cell tended to increase by up to 0.45% as neutron fluence rose (Fig. 7). It is probably related to the accumulation of irradiation defects, transmutant elements, and hydrogenation of the hafnium.

Samples (10 mm × 10 mm × plate thickness) were cut from different parts of the plate to measure density by the hydrostatic method. The measurement results (Fig. 8) show a decrease of Hf density by about 1% at a thermal neutron fluence of 6.2×10^{25} m^{-2} ($E < 0.625$ eV).

To evaluate the mechanical characteristics of the irradiated Hf, tensile samples oriented perpendicular to the rolling direction (Fig. 9) were cut from the plate by electroerosion cutting.

The results of the mechanical testing performed in the temperature range 20–150 °C show that after operation in the RBT-6 reactor, the Hf ultimate strength increased by 1–5% and was 410–500 MPa (Fig. 10). The uniform elongation and total elongation remained higher than 3–5% and 8–15%, respectively after irradiation. These results demonstrate rather good Hf mechanical characteristics even after irradiation.

2. Conclusion

The Hf control rod (EPSR) operated for 9 years (or 2047 effective days) in the pool-type water-cooled research reactor RBT-6 up to the maximal fluence 7.5×10^{25} m^{-2} ($E < 0.625$ eV) and 2.4×10^{25} m^{-2} ($E > 0.8$ MeV) preserved completely its serviceability. The control rod and all its components preserved their shape and integ-

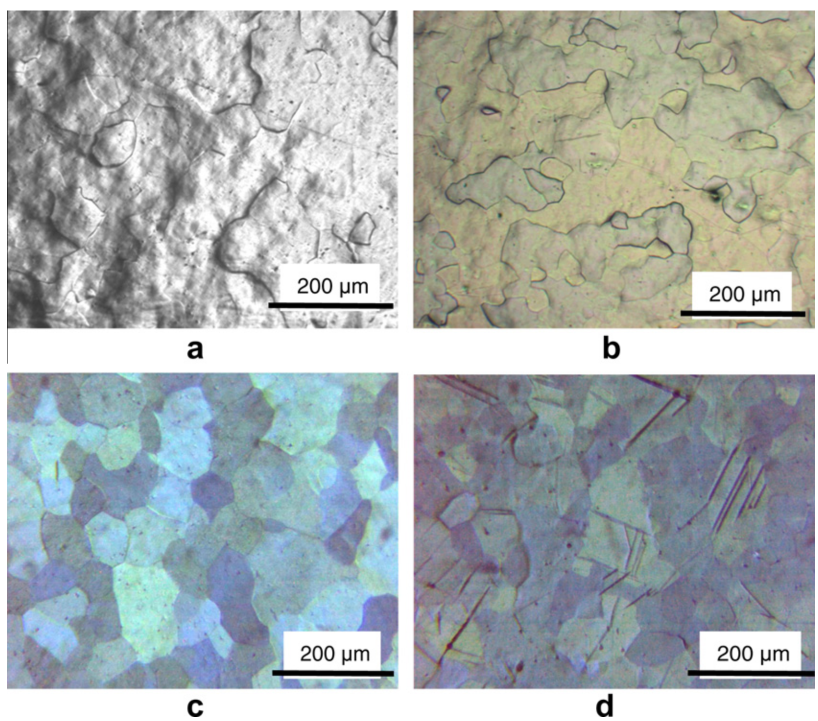

Fig. 3. Microstructure of hafnium irradiated in RBT-6: a – in indirect light after polishing; b – in direct light after etching; c – in polarized light after etching; d – in polarized light after etching.

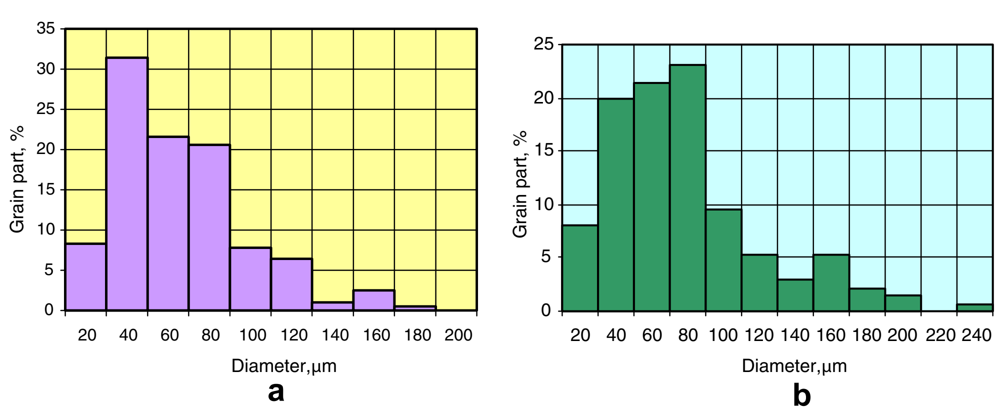

Fig. 4. Bar charts of the grain size distribution on sections in the transversal (a) and longitudinal (b) rolling direction.

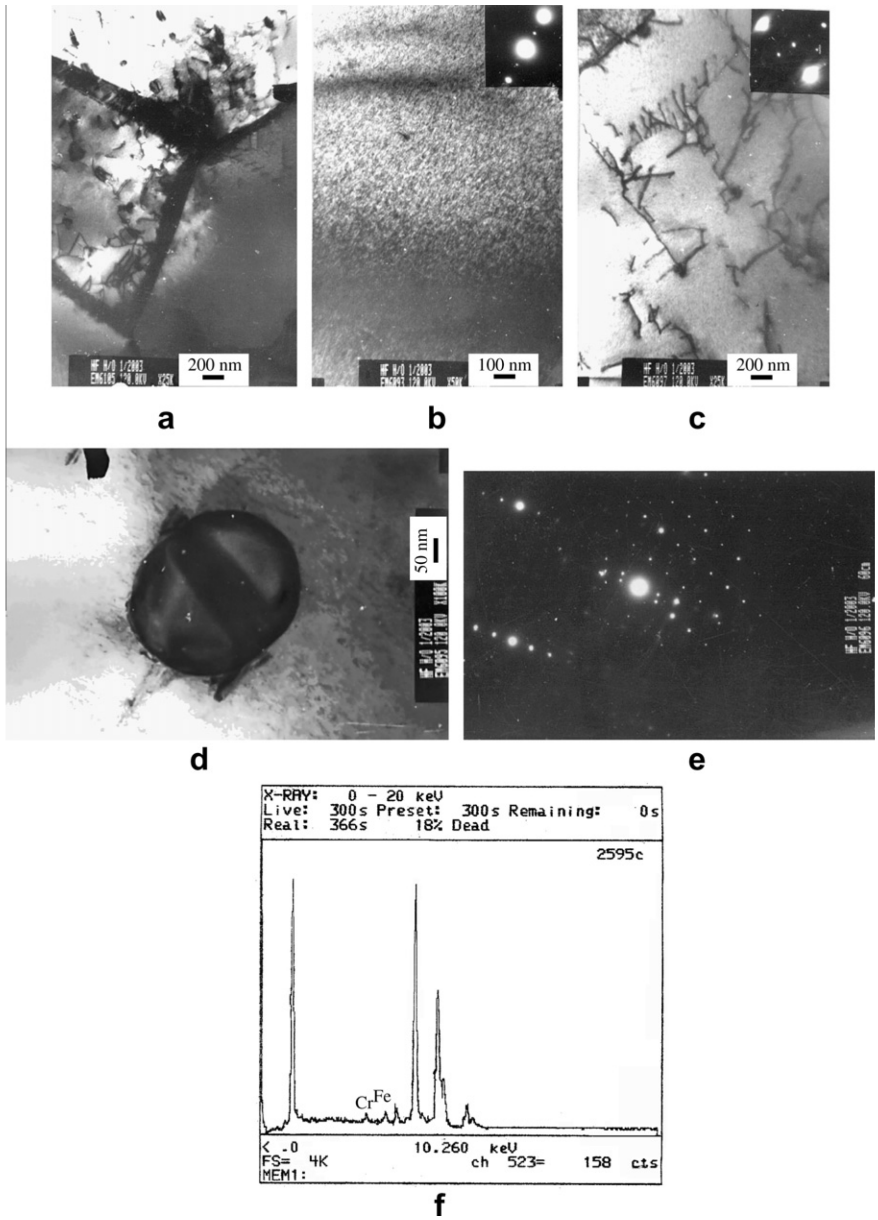

Fig. 5. Structure of unirradiated hafnium sample cut off from the RBT-6 control rod plate: a – general view; b – no c-dislocations; foil orientation – [1 0 0], reflection [0 0 2]; c – clusters of a-dislocations; foil orientation – [$\bar{1}$ 0 0], reflection [0 2 $\bar{1}$]; d – secondary phase precipitate; e – micro-diffraction picture from precipitate and matrix; f – energy-dispersed X-ray spectrum from extracted precipitate.

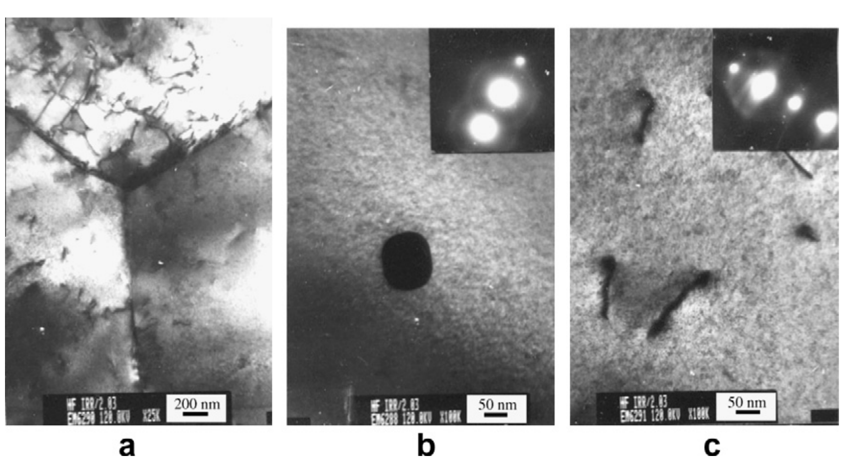

Fig. 6. Structure of irradiated hafnium sample cut off from RBT-6 control rod: a – general view; b – no c-dislocations, foil orientation – [1 0 0], reflection [0 0 2]; c – a-dislocations and radiation defects in the form of small black dots, foil orientation [1 0 0], reflection [0 2 0].

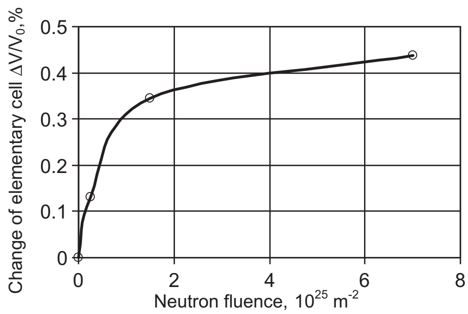

Fig. 7. Change of Hf elementary cell vs. neutron fluence ($E > 0.821$ MeV).

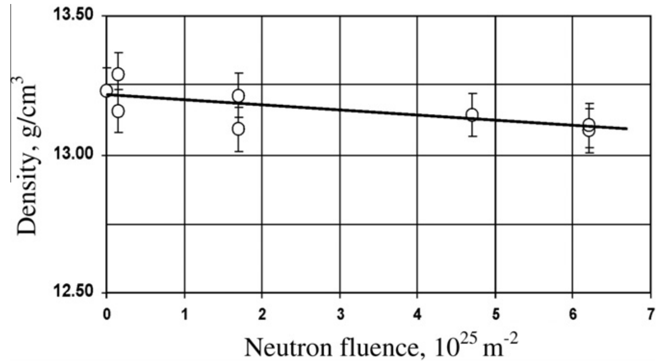

Fig. 8. Change of Hf density vs. neutron fluence.

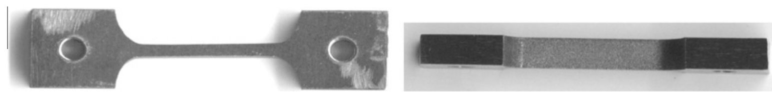

Fig. 9. Samples for tensile testing.

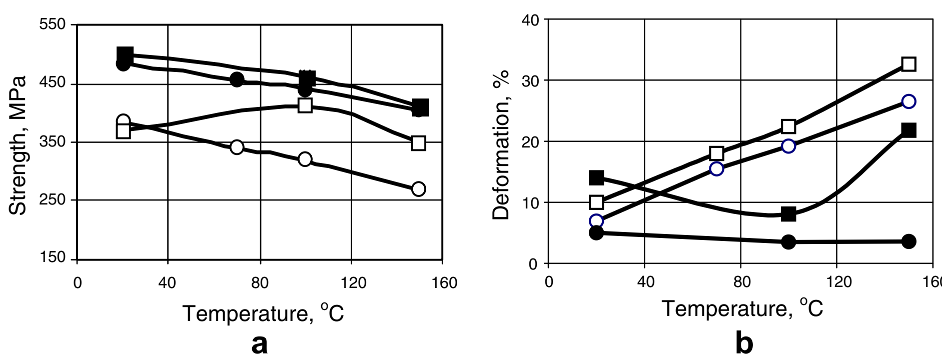

Fig. 10. Strength (a) and plasticity (b) characteristics of flat Hf samples vs. temperature: yield strength (a) unirradiated (○) irradiated (●), ultimate strength (a) unirradiated (□) irradiated (■); uniform elongation (b) unirradiated (○) irradiated (●), total elongation (b) unirradiated (□) irradiated (■).

rity. No mechanical defects were revealed. Rivet joints between the plate and section components were in good condition. No corrosion damage of the Hf plates was revealed.

Irradiated Hf preserved its initial hexagonal close packed crystalline structure and grain size distribution, the presence of linear a-dislocations and absence of c-dislocations but developed small radiation defects in the form of black-and-white dots up to 5 nm in size. Irradiation caused the increase of the elementary crystalline cell volume by 4.5% and a decrease of density of up to 1%. The mechanical properties of irradiated Hf are characterized a 1–5% increase of ultimate strength with plasticity in the temperature range of 20–150 °C at a level of $\delta_u = 3–5\%$ and $\delta_t = 8–15\%$.

The results of Hf operation in the RBT-6 reactor as an absorbing material as well as all examined physical and technical characteristics allow predicting its long-term (more than 20 years) operation under similar conditions.

There is every reason to consider Hf to be one of the most attractive absorbing and structural materials to fabricate control rods for water-cooled research reactors.

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