Equipment and Techniques for Studies on Irradiated Fuel and Fuel Rods Characteristics

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Abstract

Substantiation and prediction of reliability of the atomic reactors core elements require knowledge of different special properties of these elements and their components. To study such properties, SSC RF RIAR develops and uses different specialized measuring devices and techniques. In particular, the paper describes:

- The main technical features and methodic capabilities of the new X-ray diffractometer;
- The main technical features and examples of using the neutron autoradiography method based on application of $^{252}$Cf neutron source during material science investigations;
- The main technical features and examples of using the measuring technique of "fuel swelling stresses" based on application of special devices to standard mechanical test machines.

KEYWORDS: X-ray diffractometry, macro-stresses, amorphisation, neutron autoradiography, Californium-252, glass detector, metal fuel, restriction of swelling.

1. Equipment and methods of radiographic investigations.

1.1. DARD diffractometer.

The DARD type X-ray diffractometers (automatic remote X-ray diffractometers) have been used in hot cells of the Material Science Department since 1964. These diffractometers are manufactured by the "Burevestnik" firm in St.Petersburg. They are designed to conduct various radiographic investigations of specimens and products with high radioactivity (up to 15 Ku in $^{60}$Co).

In 2000 a new DARD diffractometer of the 5-th generation was installed in the hot cell. Fig.1 presents its schematic diagram. Its features are:
Figure 1 Diagram of the remote diffractometer: 1) counter; 2) lead shield; 3) monochromator; 4) specimen and 5) X-ray tube.

- Placement of the counter in the hot cell;
- Protection of the counter against background radiation by the lead layer, 320 mm thick, on the specimen-counter way;
- Use of the monochromator (pyrolytic graphite) to change the direction of X-rays;
- Critical angle of \( 2\theta = 164^\circ \);
- Possible radiography of long-length specimens (cylinders of \( \varnothing_{\text{max}} = 40 \) mm, \( L_{\text{max}} = 500 \) mm);
- Angles measurement accuracy of \( \pm 0.005^\circ \).

Thus, use of this type of the double spectrometer provides protection of the counter against direct radiation of the specimen. Therefore, the diffractometer can be equipped with different devices for investigating specimens with various forms and high activity.

Up to now the activity of any specimen has not been an obstacle for its study at the DARD type diffractometers. This refers even to 200 mm-pieces of regular fuel rods of the BOR-60 reactor irradiated up to burnup of 30 % h.a.

1.2. Investigations of large-size specimens.

The considerable part of articles in the atomic reactors core has a cylindrical form and large sizes (fuel elements, absorbing elements, wrappers). To handle such specimens, a special specimen holder was developed to radiography the surfaces of specimens, up to 0.5 m long: cylinders with diameter from 6 to 40 mm and the flat-surface specimens. When using this holder, no re-adjustment is required.

Use of the DARD diffractometer and holder of large-size specimens allowed study of macro-stress distribution on the surface of fuel element claddings, 6.9 mm in diameter and up to 250 mm long (Tab.1). These stresses had the maximum values near the core centre and were close to the elastic limit of stainless steel, from which the fuel rod claddings were made.

<table>
<thead>
<tr>
<th>Fuel rod No.</th>
<th>Distance from the core bottom mm</th>
<th>( \sigma ) MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>14 (non-irradiated)</td>
<td>150</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>150</td>
<td>340</td>
</tr>
<tr>
<td>4</td>
<td>150</td>
<td>460</td>
</tr>
<tr>
<td>21</td>
<td>30</td>
<td>130</td>
</tr>
</tbody>
</table>
The macro-stresses on the CPS sleeve (hexagonal guide tube) of the BOR-60 reactor after fluence of $1.6 \times 10^{-18}$ n/cm$^2$ were measured in the similar way. The measurements were performed in different sections and along the sides of the hexahedron with the across flats dimension of 44 mm (Fig. 2). These results were used for calculating the radiation creep and bending of the sleeve sides.

1.3. Investigations of small-size specimens.

The specimens with small sizes and various forms are used for standard investigations of the crystalline structure, phase analysis, texture analysis, etc.

Taking into account the specific character of the material science works, a device to the diffractometer was developed that allows radiography of specimens directly in the casing for metallographic investigations. Thus, amorphisation of the oxide fuel rim-layer was revealed in the VVER type fuel rods irradiated up to burnup of 56 MWkg U.

2. Equipment and techniques of neutron autoradiography

The methods of metallographic and radiographic investigations can be insufficient for study and description of the composition and state of heterogeneous type fuel – so-called dispersion fuel containing fuel particles in inert matrix. To a larger extent this insufficiency becomes apparent when studying the irradiated fuel rods with heterogeneous fuel types. Therefore, RIAR developed and successfully realized the neutron radiography technique of heterogeneous fuel and fuel rods with this fuel – in both initial and post-irradiation state. It is a technique of tracking autoradiography for metallographic specimens of irradiated materials containing atoms of fissionable elements. The surface (specimen section) is irradiated with neutrons inducing atom fission. The detector registers the fission-induced fragments, which is in
contact with the surface-flat metallographic specimen under study.

In terms of accessibility, simple handling in the hot cell, weak dependence on $\beta$- and $\gamma$- radiation the most preferable material of the detector is photoglass. After exposure the detector is etched in hydrofluoric acid resulting in revealing of dull areas due to damages that correspond to the locations of fissionable atoms on the metallographic specimen surface.

The layout of tracking autoradiography is presented in Fig.3.

The E-N/85 californium ($^{252}$Cf) source with neutron flux of $7.5 \times 10^6$ n/s 4π was applied as a neutron source. It was made in ORIP at RIAK (Department of Radionuclide and Isotopic Products). The cylinders of different height filled with water or plexiglass were used as a neutron moderator. The specimens under study were sealed to the standard casing for metallographic and radiographic investigations. This allows study of one and the same specimen by three independent methods: metallographic, radiographic, autoradiographic ones.

The efficiency of applying such a complex of techniques is demonstrated in Fig.4. They present the photos of one area of heterogeneous fuel metallographic specimen and the corresponding area of the glass detector after development of autoradiographic image produced at the same microscope with equal magnification.

Comparison of Fig.4a and 4b (left) shows their common quality conformance. The form of particles (or rather, their sections) coincides with their imprint on the glass (tracking image).

Under close examination, however, one can notice that the quantity of particles in the metallographic photo is larger than on the tracking detector. The particles not revealed on the detector (their locations are marked by icons $\oplus$) are inert (uranium-free) processing additions (third phase) in heterogeneous fuel.

The sizes of particles on the metallographic specimen (Fig.4a) and in autoradiographic image produced with the direct metallographic specimen-detector contact (Fig. 4b) were measured at the same microscope with equal magnification. These sizes coincide within the measurement error ($\pm 0.01$ mm).

After irradiation in the atomic reactor this heterogeneous fuel was investigated in the hot cell. Namely, each of the studied specimens sealed into its own casing was subject to investigations in this casing by metallography, radiography, tracking autoradiography methods. Only due to such a comprehensive investigation the effect of radiation-thermal homogenisation of the initially heterogeneous fuel was established and explained. Fig.4 (right) shows the results of comprehensive investigations of the metallographic specimen area of highly irradiated fuel, where homogenisation finished completely. Similar investigations conducted at intermediate irradiation stages allowed the homogenisation kinetics to be revealed in the particular heterogeneous fuel.
We suppose that such a complex of investigation methods will be most efficient during developments and investigations of heterogeneous compositions of cement type, cer-cer, etc. oriented on burning of accumulated actinides in special burner-reactors.

3. Equipment and techniques for studying the effects of mechanical restriction for fuel swelling

Development of new fuel types, fuel-containing compositions, designs of elements with these fuels and compositions assumes study of the swelling nature and parameters of these fuels and compositions.

One of the practically important characteristics of swelling can be "swelling stress", i.e. stress caused by mechanical pressure on the element cladding from the side of the swelling core.

The fuel rod cores operate under conditions of partial mechanical restriction of the fuel rod design by the cladding and other parts. Such conditions, through irradiation-free, can be reproduced in the hot cell.
To solve the set task, use was made of the MM-150D and 1797-1 type machines installed in hot cells. Both machines are designed for mechanical tensile tests (and compression tests) of specimens at temperatures up to 800 °C in inert gas medium and in vacuum not worse than $1.3 \times 10^{-2}$ Pa. The measurement error of loads does not exceed $\pm 2\%$. The trial scheme is presented in Fig. 5. The "dxh = 6x8 mm" specimens were subject to testing. The specimen being tested was placed in steel cladding. The compressive load of 200 - 300 N was applied to faces of the specimen being tested in the process of heating in vacuum. After temperature stabilization the load was reduced to 10 N. From that moment on the efforts of the swelling specimen in axial direction were recorded.

**Figure 5** Diagram of measuring the mechanical pressure of swelling specimen on cladding: 1) load recorder; 2) movable support; 3) irradiated specimen (being tested); 4) cladding; 5) irradiated specimen (reference specimen); 6) fixed support; 7) heating furnace.

![Diagram of measuring the mechanical pressure of swelling specimen on cladding](image)

**Figure 6** Swelling of U-0.5%BeO alloy under post-reactor annealing with different degree of mechanical restriction: "O" swelling of specimen in free state; "×" swelling of specimen in mechanical confinement state: A - uniform confinement state; B - without axial confinement; C - free swelling.

![Swelling of U-0.5%BeO alloy under post-reactor annealing](image)
The ratio of registered load to the specimen cross-section area can be considered as “gas swelling stress \( \sigma \)”. The experiments by the scheme in Fig.5 were performed on specimens of non-strengthened metallic uranium of technical purity and high-strength ceramic-metal composition U - BeO. These specimens were cut out from the cores of fuel rod irradiated at \( T_i = 400 \) °C up to burnup of 1 - 2 % h.a. It was established that at \( T_{\text{test}} = 600 \) °C the values for these fuels are different, but have small absolute values (2 and 6 MPa, respectively).

The measurements performed after a series of annealing showed no changes in sizes of the cladding in transverse direction or the specimen in axial direction (measurement error of ± 0.01 mm).

To study the effect of mechanical restriction degree on swelling, the experiment was conducted using the reference specimen that could swell freely. The swelling value was determined by density measurement method (measurement error of ± 1 %). Fig. 6 shows that swelling of the reference specimen during 6-hour annealing achieved 30% and remained almost unchanged during further annealing. The “restricted” specimen was slightly swollen for the first 6 hours of annealing. This swelling was caused by elimination of technological gaps between the specimen and casing. No additional swelling in area A was discovered. Swelling by 8 % occurred in area B corresponding to release of the specimen end surfaces from mechanical restriction. After decladding the additional swelling made up 20%. As the result, total swelling of the “restricted” specimen was equalized to swelling of the free specimen.

4. Conclusion

Replacement of the obsolete research equipment, development of new devices for the equipment in use, development and use of new investigation techniques, application of the complex of various techniques when investigating one specimen – all these components of research activities of employees in hot cells at RIAR, all and sundry, allow determination of new features and regularities of reactor irradiation effect on properties of reactor materials and in-reactor articles.

In its turn, knowledge of these features and regularities allows development of various improvements in fuel compositions, fuel element designs, operating conditions of large-size elements of atomic reactor cores.