Possibilities and Prospects of Investigation of Irradiated Structural and Fuel Materials Using Scanning Electron Microscope PHILLIPS XL 30 ESEM-TMP installed in the Hot Cell.

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Abstract

Scanning electron microscope Philips XL 30 ESEM – TMP with X-ray microanalysis system INCA has been installed at SSC RF RIAR. The microscope is placed in the hot cell. Monitoring and control system is installed in the operator's room. Irradiated specimens are supplied to the hot cell through the transport terminal and installed into the microscope by manipulators. Direct contact of the personnel with radioactive materials is impossible.

In addition it is developed the system of remote placement of the irradiated specimens into the specimen chamber of microscope. The system includes a stage with three seats, holders for different types of specimens and equipment for their remote loading in the holders.

Keywords: *Scanning electron microscope, EDX spectrometer, WDX spectrometer, hot cell, SM reactor fuel rods.*

Introduction

Scanning electron microscope Philips XL 30 ESEM–TMP of Philips Electron Optics production was installed in the hot cell of SSC RF RIAR in 2002. The microscope is equipped with the Energy Dispersive X-ray (EDX) and Wavelength Dispersive X-ray (WDX) spectrometer INCA of Oxford Instruments production. This paper presents the peculiarities of the microscope location in the hot cell and development of auxiliary equipment for its remote maintenance. The paper also presents the examples demonstrating the possibilities of the microscope in examination of irradiated materials.

Adaptation of the microscope

The microscope was adopted for operation in the hot cell. The adaptation included the following actions:

1. The plastic light-guide inside the Secondary Electron Detector was replaced with a glass one to make it less vulnerable to gamma radiation.
2. The opening distance of the stage door of the specimen chamber was increased to 30 cm. This allows for easier access to the stage when changing a specimen.
3. Extra shielding of the WDX spectrometer (between the specimen chamber and detector) was introduced to reduce the background signal in the wavelength spectrum.
4. An additional collimator for the EDX spectrometer was installed.
5. The length of the cables between the XL30 electronics console and the vacuum console was extended to a maximum length of 5 m. In this case the electronics console (with the user interface) can be placed outside of the hot cell.

Description of the area

The vacuum console is located in the hot cell (Fig. 1) on a separate base. The layout of the device is presented in Fig. 2. The vacuum console is remotely controlled through the cables of the electronics console located in the operators' room. A viewing system and two manipulators are installed. They allow the remote loading of samples into the microscope. The samples are transported to this hot cell from another one, intended for sample preparation, by means of conveyer. The hot cell is equipped with ventilation system allowing 20-fold air exchange per hour.

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Fig. 1 View of microscope through the hot cell window from the operator's room side

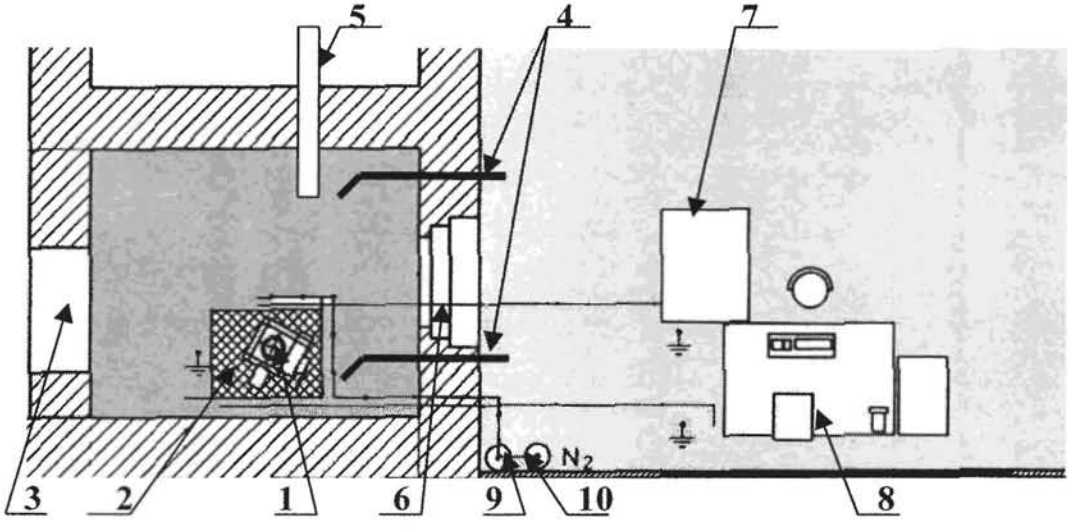


Fig. 2 Layout of the Philips XL 30 ESEM – TMP microscope. Hot cell is marked with red, operators' room is marked with yellow

1 – vacuum console of microscope, 2 – separate base, 3 – sealed door from the maintenance corridor side, 4 – manipulators, 5 – conveyer terminal, 6 – viewing system, 7 – control system for INCA spectrometers, 8 – electronics console of microscope, 9 - system for supply of argon-methane gas mixture to the WDX spectrometer, 10 – system for liquid argon supply.

An air-conditioner is installed in the hot cell to meet the requirement to temperature. Systems are installed for filling the Dewar of the EDX spectrometer with liquid nitrogen as well as for the WDX spectrometer supply with argon-methane gas mixture from the gas-cylinder located in the operators' room.
Before loading into the microscope, the sample surface can be cleaned in the ultrasonic cleaner, of which bath is located in the hot cell. Then, the samples are placed into special holders (Fig. 3). Each holder is

intended for a certain type of sample. Then, the holder with the sample is installed onto the microscope stage. This stage is equipped with two sets of standard elements for electron probe microanalysis (EPMA) of SPI Supplies, USA production. One set contains 59 metal and mineral standards and the other one – 15 rare earth standards.

The sample under examination and sets of element standards are located in the same horizontal plane that provides accurate quantitative EPMA.

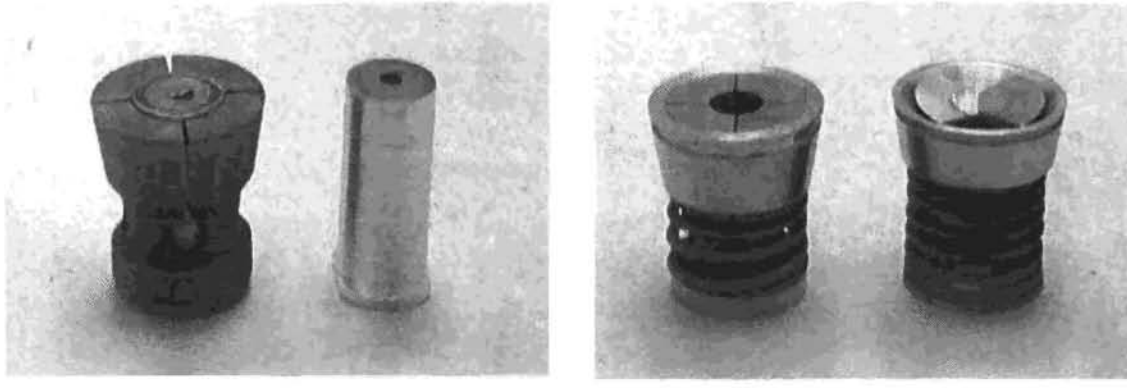


Fig. 3 Sample holders of different type

Examination of radioactive samples

In 2003 examinations of radioactive samples started. One of the activities performed within this period was study of the irradiation effect on the state of the SM reactor fuel rods with a fuel loading increased by 20% (from 5g to 6g of uranium-235). The work was aimed at upgrading of the SM reactor core so as to enlarge the experimental possibilities of accelerated high-dose irradiation of structural materials. The SM fuel rods are of dispersed type with a cruciform cross-section. The material of the fuel column is a composition containing uranium dioxide dispersed in the matrix consisting of mixture of copper and copper-beryllium bronze. The fuel rod cladding is made of steel C06Cr16Ni15Mo3B.

The SEM and EPMA methods were used to examine the SM experimental fuel rods, i.e. structure and elementary composition of fuel particles, copper-beryllium matrix and contact zones at the phase boundaries: "fuel particle-matrix" and "matrix-fuel rod cladding". The samples represented polished sections made from the full cross-section of a non-irradiated fuel rod (surveillance specimen) and irradiated one. The first one was used as an initial sample to determine the fuel rod state before irradiation. The second one, irradiated up to 41% of uranium-235 burn-up (accumulation of fission products achieved 1, g of fragments $*cm^{-3}$) was used to examine the fuel rod state after in-pile irradiation under operating conditions.

Examination results

Unlike the non-irradiated fuel rod, the matrix of the irradiated one contains large pores (Fig. 4), which are located mainly, at the boundary of two phases: "fuel particle-matrix". Large irradiated particles of uranium dioxide are characterized by very well developed porosity. A distribution of pores in the fuel particle volume is non-uniform; they are located, most probably, along the grain boundaries (crystallites). Fig. 5 shows one of the characteristic particles of uranium dioxide that was chosen to determine possible interaction between uranium dioxide and copper-beryllium matrix. Concentration curves of copper and uranium were plotted using X-ray spectrometers. No mutual penetration of copper and uranium can be seen. Copper is not available in the fuel particles. An interaction layer was revealed between the matrix and cladding. The difference in the thickness of the interaction layer was observed in the edges and cavities of the fuel rod cross-section. The irradiation did not effect the change in the layer thickness. A quantitative EPMA of the "matrix-cladding" interaction layer was performed using the non-irradiated sample. A re-distribution of elements was observed in this interaction area. Copper interacts with the cladding penetrating into steel in the form of separate "streamlet". In the course of the process evolution,

separate "streamlets" unite and form a system of "islands". At the next stage, mutual penetration of the cladding and matrix elements occurs in these "islands".

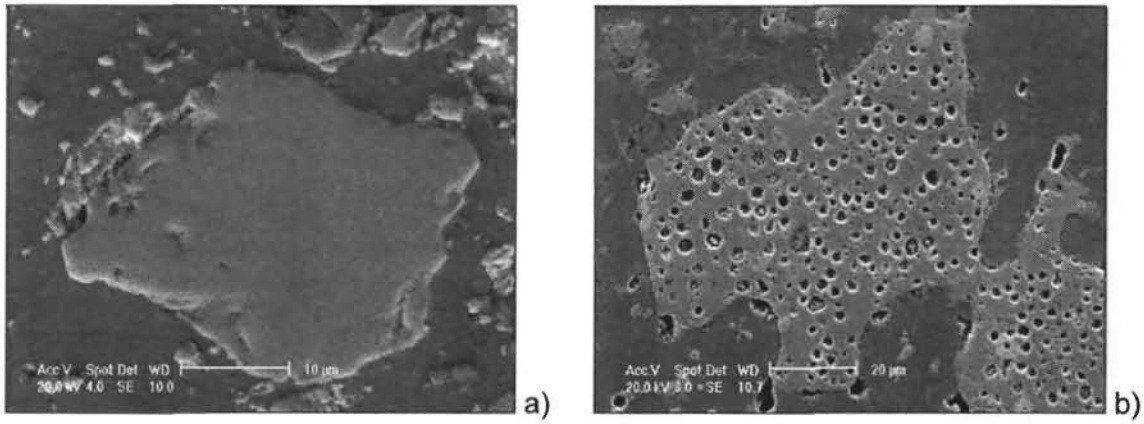


Fig. 4 Particles of uranium dioxide in the copper-beryllium matrix before (a) and after (b) irradiation

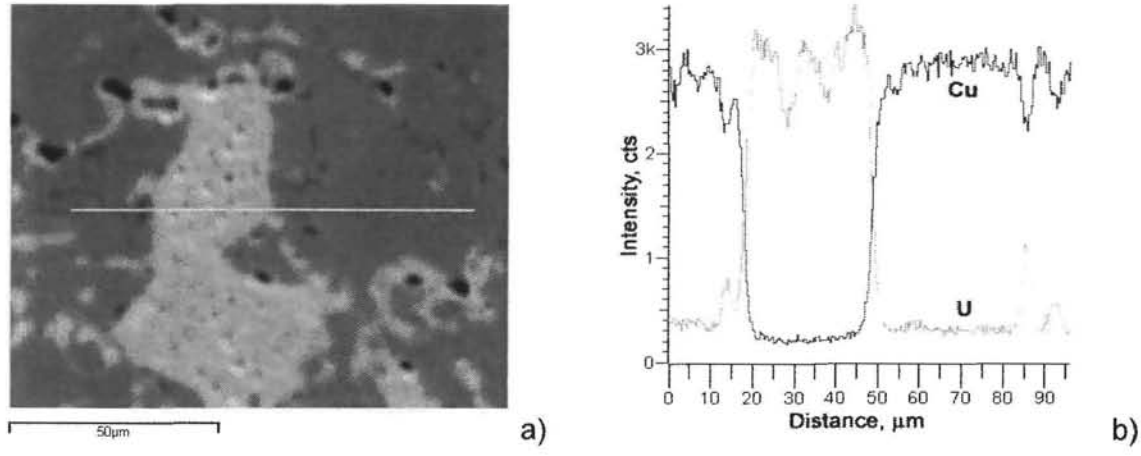


Fig. 5 Irradiated uranium dioxide in the copper-beryllium matrix (a). Distribution of copper and uranium (b) along the yellow line in the photo. The quality of the photo is worse than that in fig.4. It is related to the necessity of the increase of the probe current during the X-ray microanalysis that leads to degradation of the resolution of the microscope

It should be mentioned that during this process the elements contained in the cladding were re-distributed in different ways. Thus, in the "islands" the content of nickel and chromium decreased sharply and that of molybdenum increased. The iron concentration decreases only in the islands at the most distance from the cladding. A sharp increase in the copper content is also observed there.

The distribution maps of the irradiated fuel rod elements were plotted for the interaction area: "matrix-cladding" (Figs. 6, 7). They did not show any significant difference in the distribution of the above elements in the "matrix-cladding" interaction layer after irradiation.

a) b)

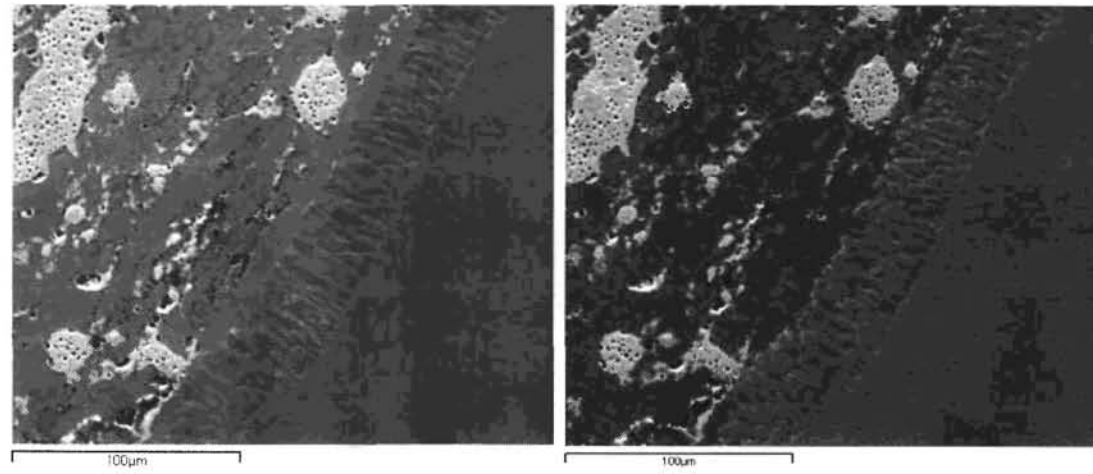
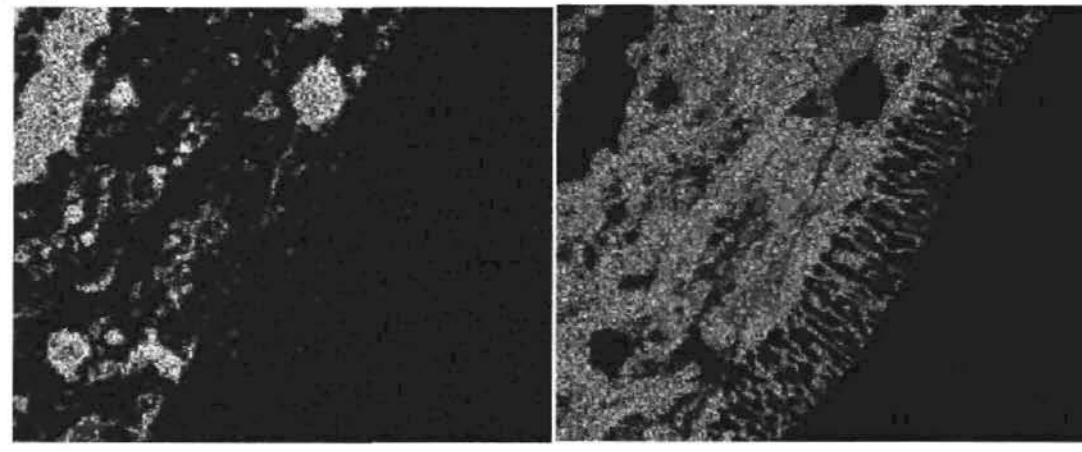
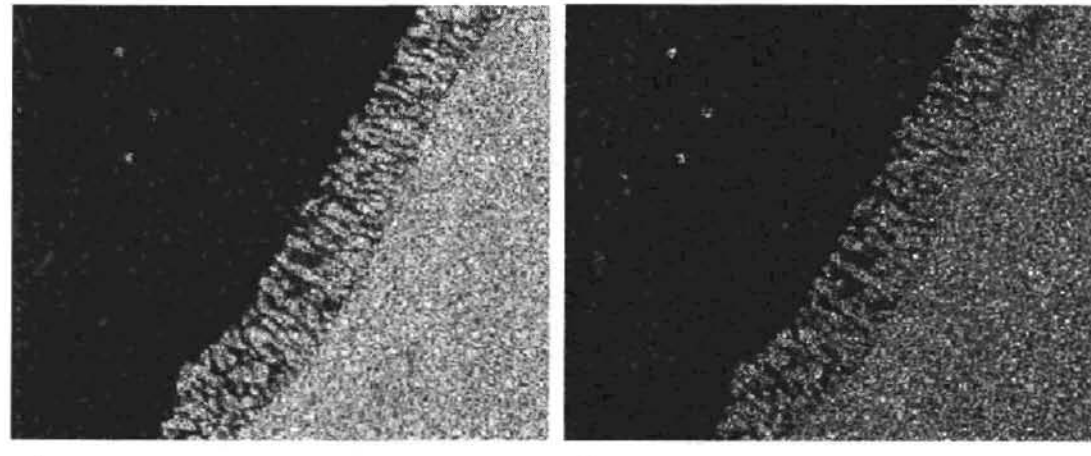


Fig. 6 The scanned area of the irradiated experimental fuel rod (a) Integrated SE image of the area with maps of elements distribution in the characteristic X-rays (b) Places of uranium concentration is marked in red, copper concentration – in blue and iron concentration – in green



a) b)



c) d)

Fig. 7 The distribution map of uranium (a), copper (b), iron (c) and chromium (d) over the area shown in Fig. 6. The EPMA image is obtained in characteristic X-rays

The device allows combining the SE image with maps of elements distribution. Places of concentration of elements in this case are well visible.

Conclusions

Microscope Philips XL 30 ESEM–TMP located in the hot cell allows the examination of radioactive materials with activity up to 10^{11} Bq (^{137}Cs source). It provides:

- obtaining of images of a sample structure with a resolution up to 3.5 nm.
- express-analysis of a sample composition.
- qualitative and quantitative EPMA of materials, including light elements.
- obtaining of maps of elements distribution over a sample.
- examination of dielectric materials without preliminary plating of conductive coating.

The above possibilities of the device allow the examination of materials used in nuclear power engineering to solve a vast spectrum of problems