SIMS – An Effective Addition to the Traditional SEM and EPMA Methods for Examination of Irradiated Oxide Fuel

Yu.D. Goncharenko, L.A. Evseev, V.A. Kazakov, F.N. Kryukov

Federal State Unitary Enterprise
State Scientific Center of the Russian Federation
Research Institute of Atomic Reactors
433510, Dimitrovgrad, Ulyanovsk region, Russia

The radial distribution of the fissionable element isotopes and fission product isotopes in the fuel pellet is closely related to such performance characteristics as swelling, local burnup, gas release from the fuel matrix, density and dimensions of gas bubbles etc. Moreover the experimental determination of radial distribution of uranium, plutonium and fission product isotopes is needed to verify the code efficiency.

The quantitative characteristics of the radial distribution of various elements in irradiated fuel are commonly determined by the EPMA method.

The quantitative isotope composition of spent fuel is generally determined by mass-spectrometric isotopic dilution analysis. This provides the averaged isotope composition of the fuel pellet and good confidence of the results.

In our case the SIMS method was applied to determine the radial isotope distribution and the SEM method to study the fuel pellet structure.

SIMS method provides not only the averaged quantitative isotope composition of the fuel pellet but the radial isotope distribution and the local isotope-elemental composition too.

The purpose of this presentation is to demonstrate the SIMS and SEM methods possibilities taking, as an example, the experimental determination of the radial uranium, plutonium and fission product isotope profiles and study of the irradiated oxide fuel structure.

Subject of inquiry:

The subject of inquiry was the VVER-440 type reactor fuel rod fragment with a local burnup of 69.1 MWd/kgU that was additionally tested in the power ramp conditions in the MIR reactor.

The initial fuel enrichment in ^{235}U was 4.4%.

A polished metallographic section of the fuel rod fragment was employed for examination.

The radial distribution of the various element isotopes was determined by the following procedure:

The distance between neighboring experimental points varies from 100 to 500 ìm;

In every point 10 measurements were made;

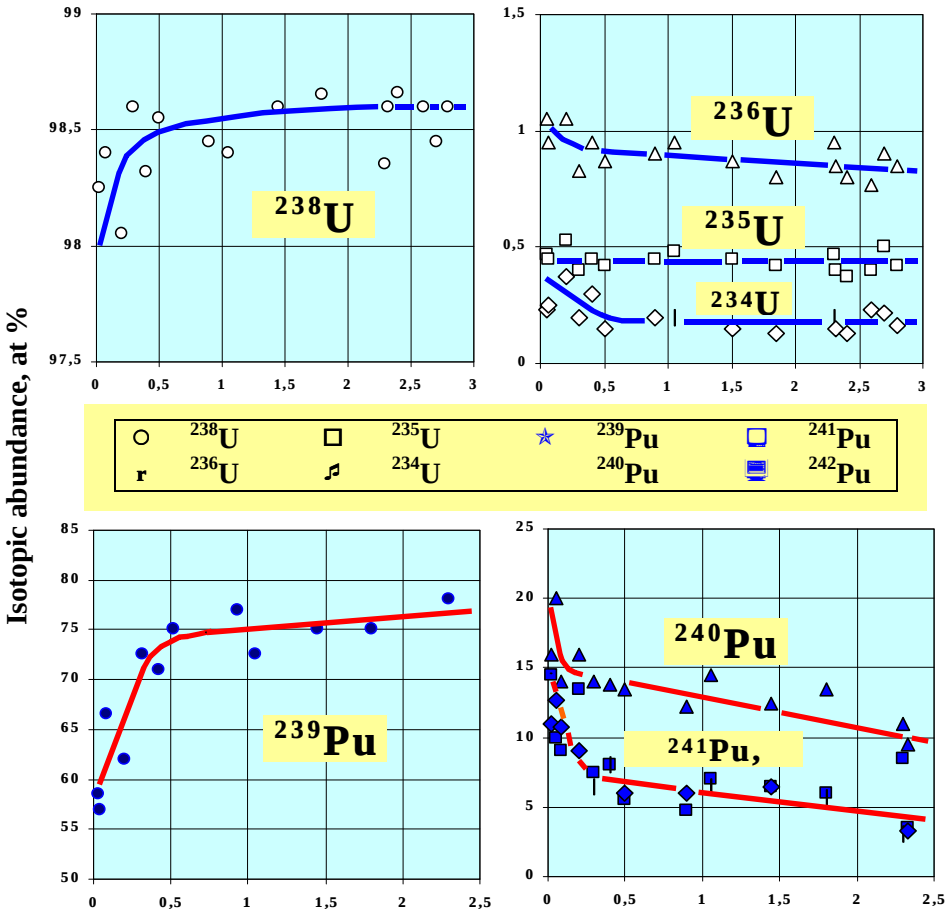
Diameter of the ion probe – 10 μm; Ar+ ions with the energy of 10 keV were used as primary ions;

The procedure implies recording of the selected mass-spectrum region containing mass-peaks with the mass to charge ratio from 250 to 259 for uranium and plutonium isotopes; thus the clusters from $^{234}U^{16}O$ to $^{242}Pu^{16}0$ or the mass-spectrum region containing mass-peaks with the mass to charge ratio ranging from 132 to 151 for cesium, neodymium and xenon isotopes were recorded.

The calculation of the isotope ratio in each point did not take into account possible superposition the mass-peaks with the same mass to charge ratio. For instance, possible superposition of ^{238}U-^{238}Pu, ^{134}Cs-^{134}Xe-^{134}Ba, ^{242}Pu-^{242}Cm etc., was not taken into account.

Total volume of the removed material from one measurement point was about 5000 cubic micrometers that corresponds to about 0.05 ì g of uranium dioxide.

The experimental radial distribution of plutonium and uranium isotopes on our sample is presented on pic.1.



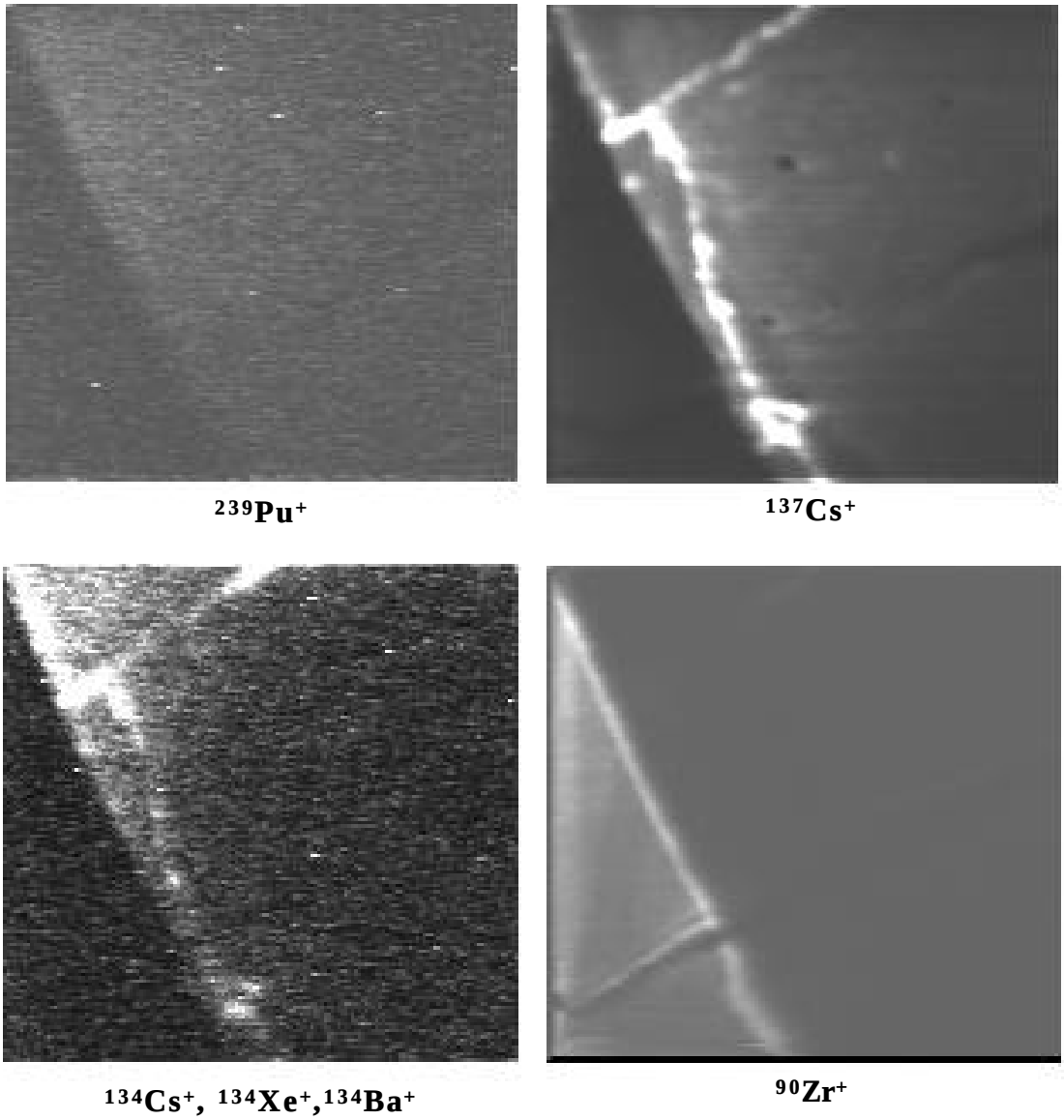
Pic.1. Experimental radial distribution of uranium and plutonium isotopes on the fuel pellet.

As is evident from the plots, the relative isotope content of ^{239} Pu noticeably decreases at the fuel pellet periphery in the region of about 500 ìm. The same region has slightly less ^{238}U and rather more other U and Pu isotopes.

In addition to the mass-spectrum record and calculation of the isotope abundance, the isotope maps of some elements from the fuel pellet surface were registered too.

The maps of some element isotopes were obtained in the following way:

- cleaning the selected surface area by ion beam for three minutes;
- recording a small mass-spectrum region containing the mass-peak isotope selected to obtain the ion image by the primary ion beam scanning in the same raster;
- recording the ion image when the exact position of the isotope mass-peak maximum is determined.

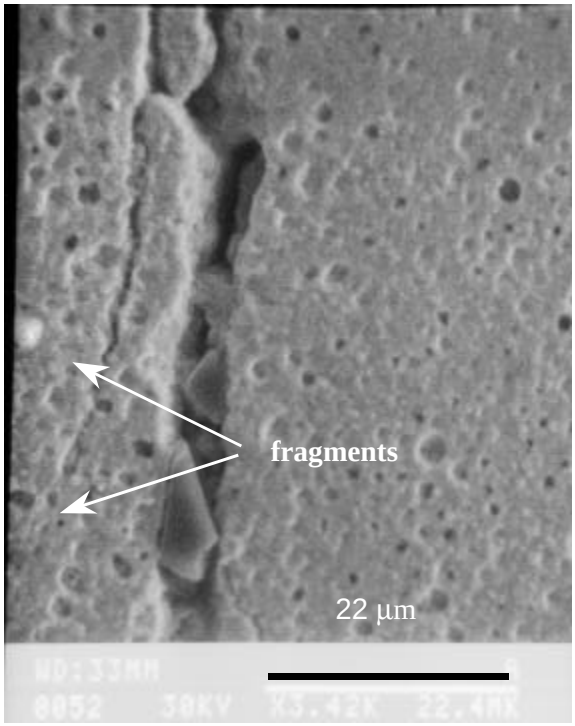


Pic.2. Isotopic maps.

The selection of the investigation place was caused by the desire to receive information on the radial isotopes distribution and on the isotope distribution features in the fuel pellet cracks. Pic.2 shows Cs, Pu, Ba, Zr isotope maps of the fuel pellet fragment. The maps resolution is 150x150 dots. On the maps the light regions correspond to the regions with relative high concentration of the corresponding isotope, the dark areas - to lower concentration.

What we could observe on the images:

- distribution of ^{239}Pu isotope is rather uniform but its concentration is slightly increased in the peripheral pellet regions;
- increase of ^{137}Cs concentration is observed in the fuel pellet cracks and it mostly increases in the discrete form;
- it is obvious that the fuel pellet and cladding are separated by the ZrO_2 layer.

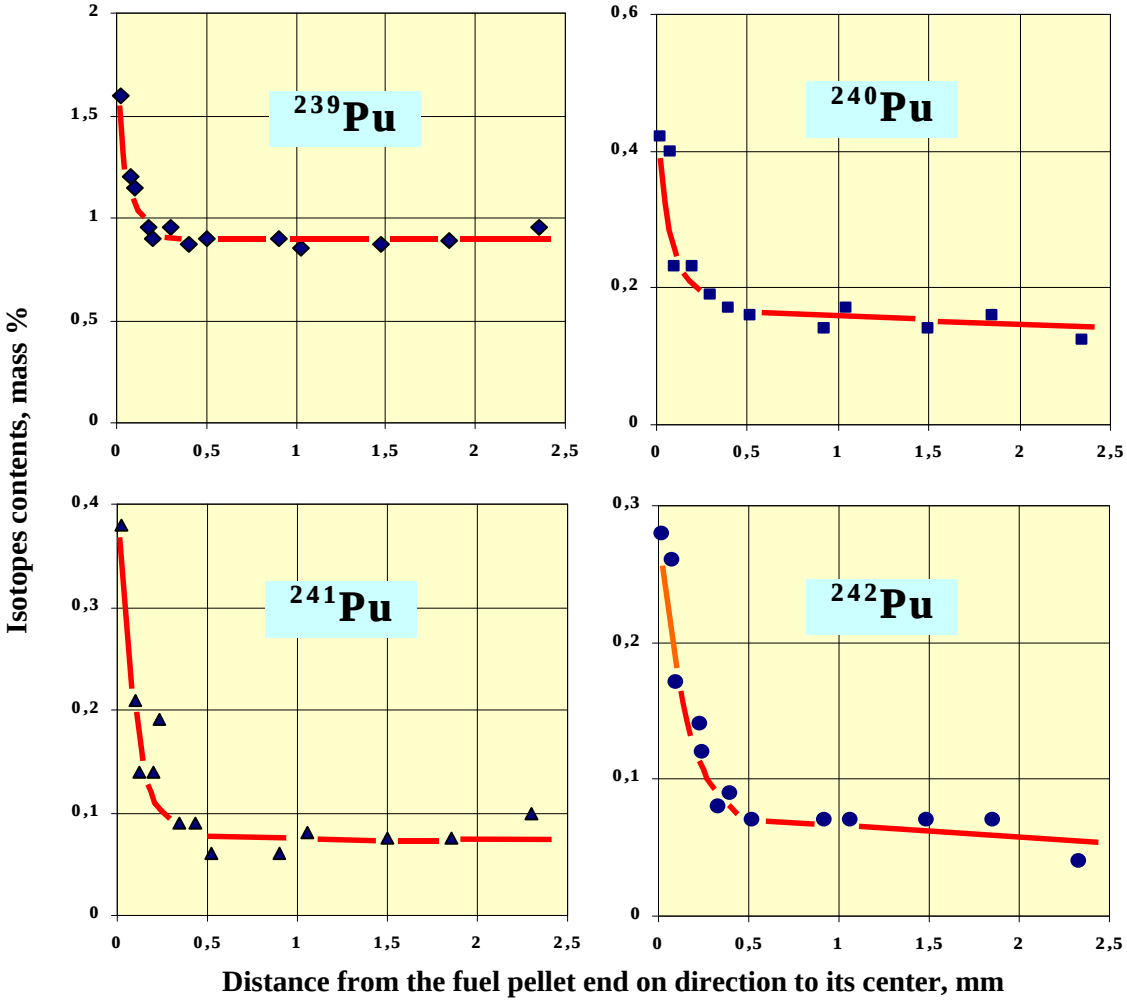


Pic.3. Fragments in the fuel pellet cracks of 3-7 μm width.

An increase of cesium concentration in cracks observed in the discrete form corresponds to the microstructure provided in pic.3. In the crack you can see the material fragments the structure of which differs from the fuel pellet structure.

The quantitative analysis of the radial distribution of fissionable element isotopes (as the elements) and fission product isotopes by the SIMS method is possible with the use of the SIMS facilities providing the quantitative elemental analysis or, as in our case, using the results obtained by the EPMA method.

So, due to the results of the quantitative radial distribution of elements in the same fuel pellet obtained with the EPMA method together with the quantitative radial distribution of the isotope abundance we were able to calculate the quantitative radial distribution of the fissionable element isotopes and fission product isotopes (as the elements), pic.4.



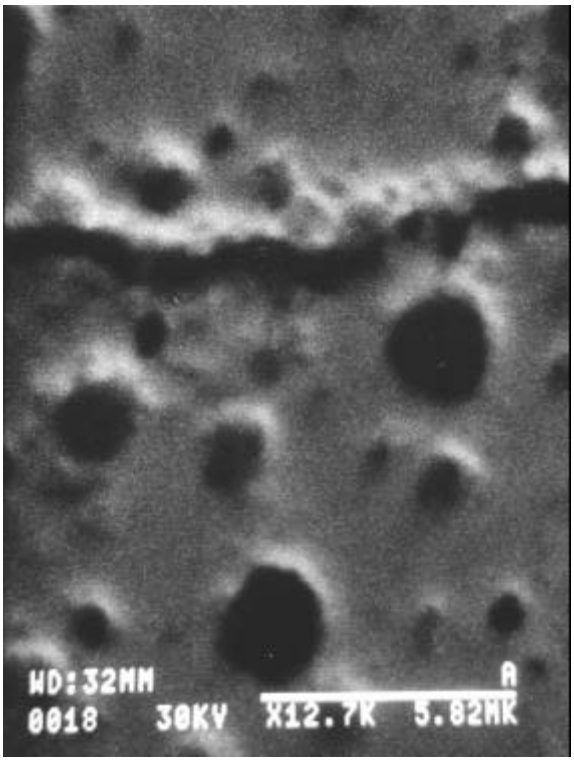
Pic.4. Quantitative radial distribution of plutonium isotopes in the investigated fuel rod fragment. Joint data of SIMS + EPMA.

It is obvious that the content of ^{239}Pu is increased on the fuel pellet periphery by a factor of 1.8 (that actually corresponds to the given ^{239}Pu isotope map), . ^{240}Pu – by a factor of 2.9, ^{241}Pu and ^{242}Pu – by a factor of 5.

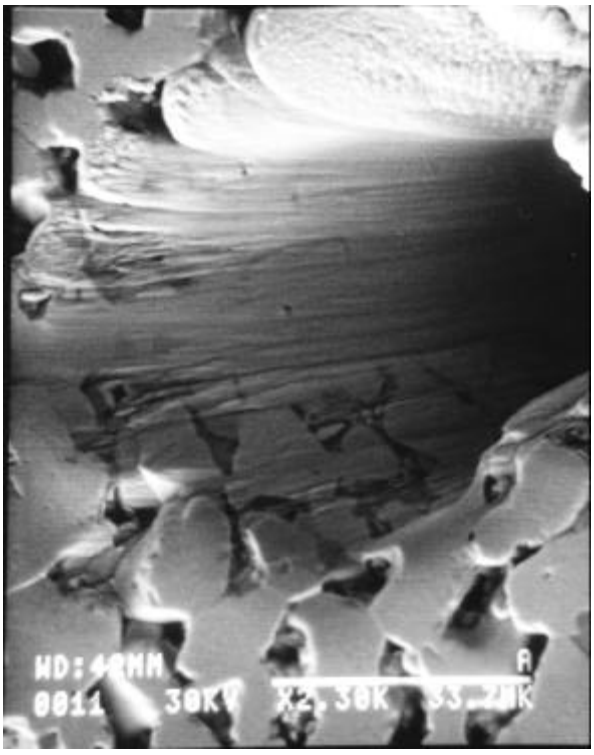
And last illustration. Because of insignificant volume of the received experimental results I shall pay your attention to one more SIMS capability for receiving additional information about irradiated fuel status, namely: possibility for the information receiving about distribution of gas fission products.

Structure investigation indicates that fuel contains high density of gas bubbles filled, probably, with xenon, krypton and helium. Basically bubbles have the round form, pic.5.

Pressure in the bubbles, having size from several hundred nanometers up to several microns, is estimated from several tens MPa up to 100-150 MPa [1].



Pic.5. Gas bubbles in surface layer.



Pic.6. Typical microstructure of irradiated fuel in the ion etching area (etching crater).

Interesting observation has been made for geometrical sizes definition of the ionic etching crater. In our case its size is approximately 20×35 microns, depth – about 7 microns (Pic.6). In the crater area there is rather a lot of bubbles which blow up as far as UO_2 evaporation. Probably, in such a way it is possible to estimate gas pressure in bubbles if to know a speed of layers removal and to use serial etching with small speed and intermediate non-expendable viewing of the crater area in a scanning electron microscope.

Conclusion

Analysis of the given results suggests that the new and more comprehensive information on the element-isotope composition and fuel structure can be obtained only in combination of the traditional methods such as the EPMA method, SEM method and the secondary ion mass-spectrometry method.

References.

1. K.Une, K.Nogita, Y.Suzawa, K.K.Hayashi, K.Ito, and Y.Etoh. “Effects of grain size and PCI restraint on the RIM structure formation of UO_2 fuels”. ANS Int. Topical Meeting on LWR Fuel Performance, Avignon, 10-13 April, 2000, Oral Session, pp. 615-625.