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LOCALIZING FISSION PRODUCTS IN IRRADIATED NUCLEAR FUEL PARTICLES WITH THE UNSHIELDED ELECTRON MICROPROBE

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Résumé - L'utilisation d'une microsonde non blindée pour examiner les produits de fission dans des petits échantillons de matériaux irradiés est montrée par quelques exemples d'examens de particules combustibles nucléaires enrobées.

Abstract - The application of an unshielded microprobe to examine fission products in small samples of irradiated material is demonstrated by some examples of investigations in coated nuclear fuel particles.

I - IRRADIATED MATERIAL IN THE UNSHIELDED MICROPROBE

The coated nuclear fuel particles - with diameter < 1 mm - which contain the fissile material of the high-temperature reactor in form of tiny fuel kernels, surrounded by pyrocarbon and silicon carbide coatings, are the object of extensive investigations. Irradiation tests are performed to verify the general integrity of coated particles, and especially to study the chemical behaviour of fission products and their transport properties.

X-ray microanalysis is an important method for the chemical examination after irradiation. Because only a small portion of nuclear material is concentrated in a single particle, after cooling-time periods of 1-2 years the radioactive background level can be decreased to practicable low values of some mCi (Fig. 1), thus giving us the unique possibility to work with an unshielded electron microprobe.

Fig. 1 - Effect of cooling-time on the activity and $\beta/\gamma$-ratio of fission products generated during 1 y irradiation of 1 mg U 235 assumed to be totally fissioned (100 % fifa).
It follows from Fig. 1 that for typical values of 0.1 mg U 235 per coated particle the activity can be reduced after 50 % fima burnup and 2 y cooling-time to about 2.5 mCi. This is in the order of magnitude which has been practically measured on specimens used for the microprobe examination. The residual activity does not influence the detection limits of various fission products very seriously i.e. by a factor of 2-3. The usefulness of the above considerations is demonstrated in the following examples.

II - FISSION PRODUCTS IN FUEL KERNELS

The primary object of the electron microprobe investigations is the kernel of the coated fuel particle, initially consisting of nuclear material of the oxide (UO₂) or carbide (UC₂) type, and then additionally enriched with fission products during the irradiation test. The fission products can be classified by their chemical behaviour in groups of elements which are soluble in the kernel matrix or reacting with the uranium containing fuel material and others which do not form stable compounds or precipitate in separate phases. From the results of microanalyses some characteristic distributions of typical fission products behaviour in this respect are shown in Fig. 2 and 3, also demonstrating the influence of the initial kernel composition (oxide or carbide).

![Fig. 2 - Typical fission product distributions of UO₂ coated particles after irradiation at 1300...1150°C and 50 % fima (= fissions per initial metal atoms). Scanning area of X-ray images: 400 x 400/um².](image_url)
1. Oxide Fuels (UO₂):

Cesium (Fig. 2a) does not form stable compounds and as a volatile element is deposited preferentially in the buffer layer surrounding the central kernel. Neodymium (Fig. 2b) like the other rare earth elements is soluble as oxide in the uranium oxide matrix. Barium (Fig. 2c) precipitates in a separated phase consisting predominantly of barium-strontium-zirconate. Molybdenum (Fig. 2d) is found in metallic inclusions containing also technetium and noble metals.

![Molybdenum-Mo Lα](image1)

![Neodymium-Nd Lα](image2)

**Fig. 3 - Typical fission product distributions of UC₂ coated particles after irradiation at 1030...940°C and 68 % fima burnup. Scanning area: 100 x 100/μm².**

2. Carbide Fuels (UC₂):

Molybdenum (Fig. 3a) forms ternary carbides of type UMC₂ (M = Metal) with uranium and technetium. Its distribution is also correlated with the noble metals M = Ru, Rh, Pd which are possibly forming another complex carbide phase of type U₂MC. According to the results of simulation experiments /1/ both phases can be mixed together in a fine dispersion unresolved by the electron microprobe. Neodymium (Fig. 3b) like the other rare earth elements is insoluble in the UC₂ matrix and precipitates in a separate carbide phase. The correlation of a low intensity level of Nd-Lα with the uranium distribution indicates the formation of a solid solution with UC containing small concentrations of rare earths as described in /1,2/.

III - FISSION PRODUCTS IN THE COATINGS

The moveable fission products must be retained within the fuel particles, for which purpose the coatings surrounding the kernel have been optimized to act as the main diffusion barrier. By quantitative measurement of concentration profiles of volatile solid fission products such as cesium with a stepscanning technique through the cross sections of interesting coatings a data basis can be obtained for retention properties of the fuel kernel and transport coefficients of coating material.

The method is demonstrated for cesium in coated particles with oxide kernels which have been modified by silica-alumina additives to improve the retention of fission products /3/. The amount released from the kernel during the irradiation can be obtained by integration of the concentration profiles of cesium (Fig. 4) preferentially found in the buffer layer neighbouring the kernel. After some recalculations based on the diffusion model of A.H. Booth /4/, an effective kernel transport coefficient for Cs 137 can be determined (Fig. 5).
Fig. 4 - Cesium concentration profiles measured by a stepscanning technique in the coatings of nuclear fuel particles irradiated at different temperatures to a burnup of 4-5 % fima.

Fig. 5 - Arrhenius plot of effective transport coefficients for cesium in oxide kernels with alumina-silica additives irradiated at different temperatures /5/. Data obtained from microprobe measurements are compared with a chemical etching technique and show excellent agreement.

REFERENCES
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4/ Booth, A.H., Report AECL-496 (1958)