

New Possibilities of the Isotope Distribution Examination in Irradiated Absorbing Materials Using Secondary Ion Mass Spectrometry Method

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The SIMS technique (with using a linear analysis and 2D surface imaging) has been to measure the radial distribution of the boron isotope ratio in the boron carbide pellets irradiated in the fast reactor. It was revealed that a radial distribution of isotope ratio in the boron carbide pellets is significantly different after irradiation in fast and thermal reactors. It was showed the advisability of using ion images for such examinations.

Keywords: secondary ion mass spectrometry, absorbing elements of nuclear reactors, 2D analysis, isotopic charts

Introduction

Nowadays boron carbide (B₄C) is one of the main materials, which are used in the control rods of different neutron spectra reactors. The initial elemental and isotope compositions of absorbers are changed in the course of the reactor operation. These changes (they are of a radial character in the first approximation) are one of the factors affecting the efficiency of the B₄C absorbers, their radiation resistance and serviceability.

Currently these changes are usually analyzed with the use of calculation and volumetric analysis methods. Local methods of the isotope composition analysis are less common (nuclear reaction analysis using microprobe techniques [1, 2], secondary ion mass spectrometry method (SIMS) [3] and others).

The analytical potential of the secondary ion mass spectrometry method (sensitivity, lateral resolution and others) is appropriate for experimental isotope composition analysis of absorbing materials. But at the same time such research activities require much time and spending. Probably the use of secondary ion images (2D-analysis) will help in the local isotope composition analysis of absorbers. This paper demonstrates how the radial distribution of the boron isotope ratio in the enriched boron carbide pellets irradiated in the fast reactor can be examined using the SIMS technique.

Experimental

The isotope composition was examined using a MS7202M SIMS instrument. The Ar⁺ ions having the energy of 10 keV were used as primary ions. The beam current of primary ions was 0.05 μA. The chosen diameter of the probe was about 30 μm.

It has been known that in spite of the significant progress made due to the use of primary cesium ion beam in the SIMS technique and the MCs⁺ cluster registration [4], the quantitative analysis of the stored images has been presenting difficulties as before as a result of chemical, topographic and crystallographic effects [5]. The crosscut metallographic sections of irradiated boron carbide pellets (Fig. 1) were used for the topographic contrast effect reduction in the course of this work.

Sets of ion images (in the absorbed current and ¹⁰B⁺ and ¹¹B⁺ ions (Fig. 2)) were registered on the sample surface (nearly its edge) over an area of ~1x1 mm.

The resulted ion images were processed in the following way: the intensity of each image point in ¹⁰B was compared with the intensity of the corresponding image point in ¹¹B and then these relationships were averaged over lines (Fig. 3a). As the change of the isotope ratio in pellets under the reactor operation conditions is the radial character in the first approximation, such image processing procedure is well acceptable for similar examinations if a pellet size is rather big (it is 20 mm and 1 mm in our case) as opposed to the region of the ion image registration but the registration region itself should be located over the peripheral area of the pellet (Fig. 1). In the opposite case such averaging should be made with due

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consideration for the pellet geometry (that is over the imaginary radiuses starting from the pellet centre but not over the horizontal lines) (Fig. 3b).

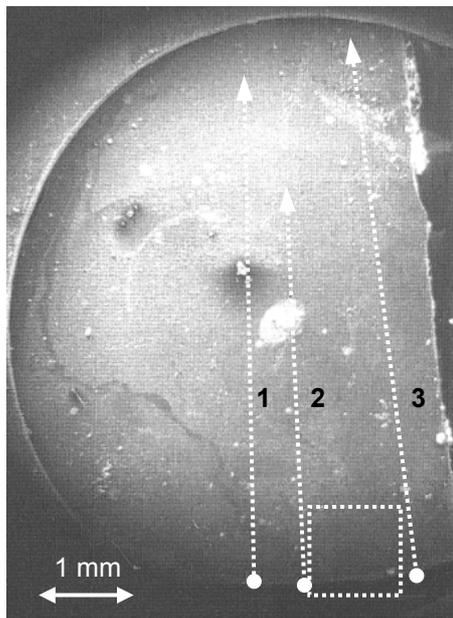


Fig.1 Photomicrography (SEM) of the absorber fragment (dotted lines are used for demonstration of some routes in examination and one of the ion image registration area)

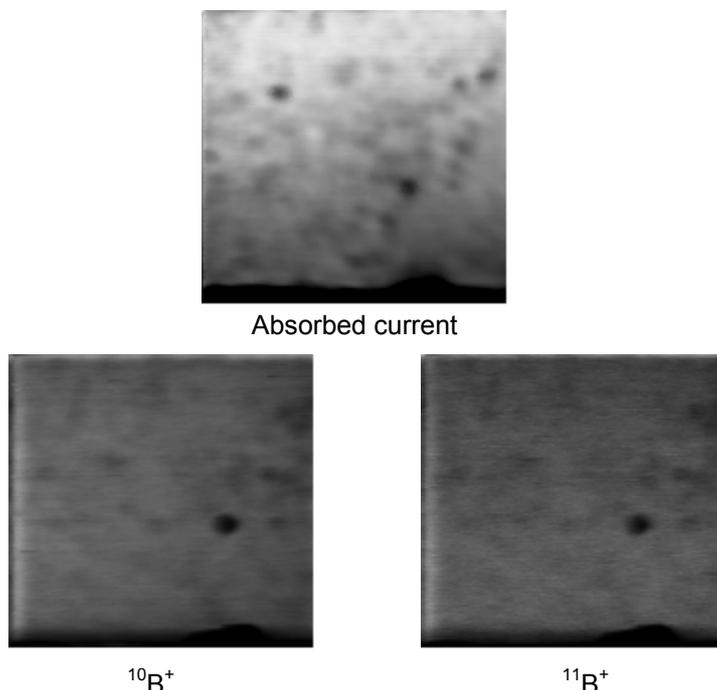


Fig. 2 A set of ion images registered on the sample surface (Fig. 1). Image resolution is 150x150 points

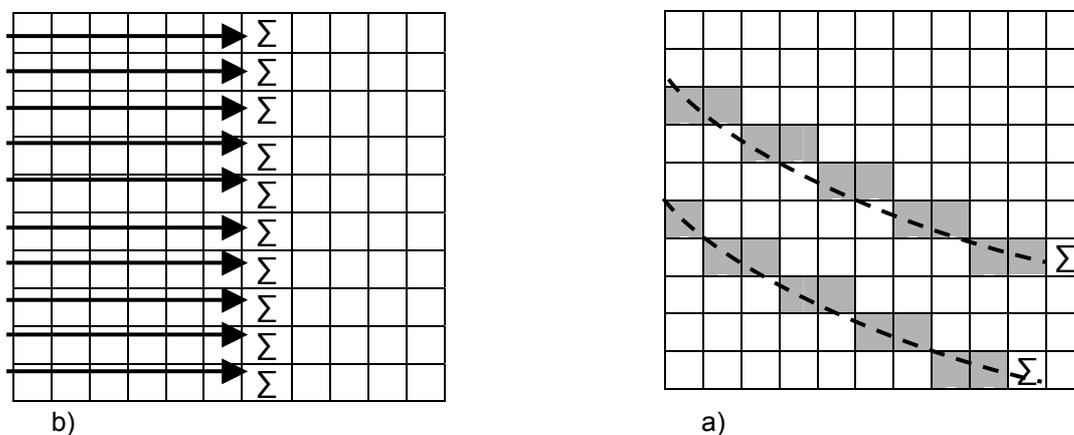


Fig. 3 Possible options of the ion image processing in examination of irradiated absorber:

a) averaging of the ratio matrix (each square corresponds to the image point intensity in ¹⁰B compared with the intensity of the corresponding image point in ¹¹B) b) averaging of the ratio matrix over imaginary circumferences whose centre coincides with the pellet centre

The ¹⁰B and ¹¹B ratio was measured using a tradition approach also that was points found in segments connecting the pellet edges and supposed centre (Fig. 1). The distance was varied within 30 and 1000 μm among the points. A number of measurements were in the range of 30 to 60 at each point of the measurement route. They comprised a consecutive registration of the mass-spectrum area incorporating the ¹⁰B and ¹¹B mass peaks. The relative measurement errors (calculated with the use of the Student t-criterion with a confidence probability of 0.95) were no higher than 0.3 relative units at each measurement point. Approximately this value corresponds to 0.3 atomic percent.

Results and discussion

Figs. 4 and 5 demonstrate radial distributions of the ^{10}B relative content resulted from a set of specific ion images and for two specific routes, correspondingly.

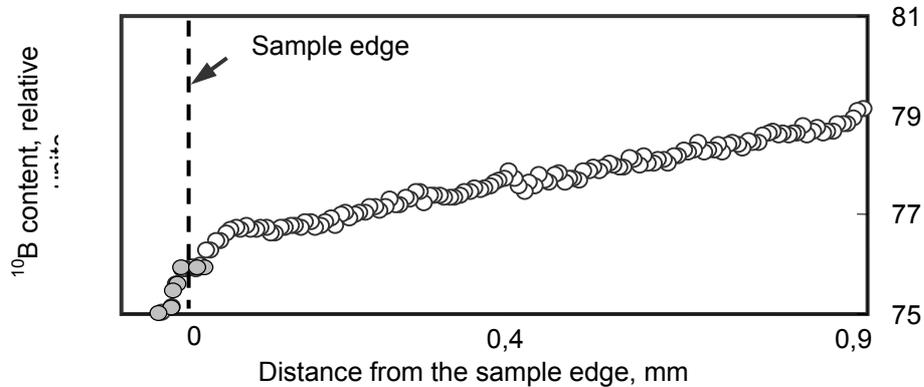


Fig. 4 Radial distribution of the ^{10}B relative content resulted in numerical processing of a set of ion images (Fig. 3)

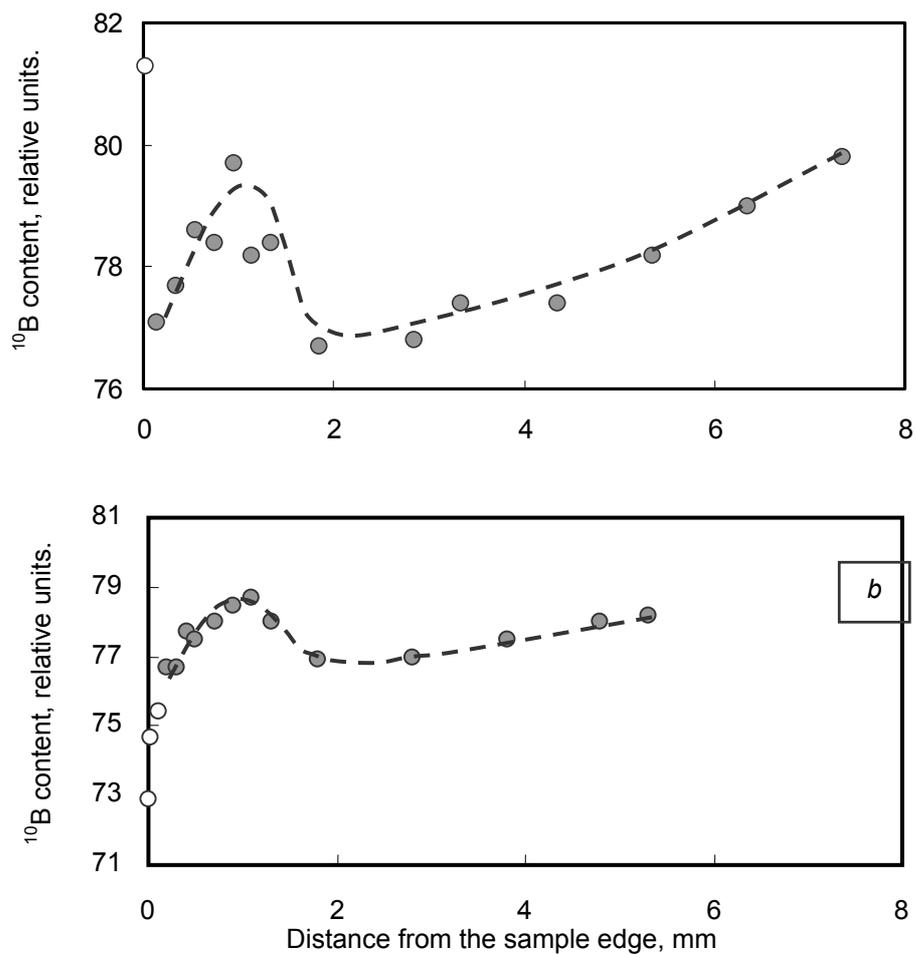


Fig. 5 Radial distributions of the ^{10}B relative content for two specific routes (a, b – route 1 and 2, respectively on Fig. 1)

The SEM data demonstrate that only a part of the ion probe (non-localized sliding beam) was present at the shaded points (Fig. 4) and unshaded points (Fig. 5) on the sample surface, correspondingly. Such points were excluded from the obtained routes.

Each point of Fig. 4 represents the average of 150 values. So a potential effect of the topographic contrast was corrected statistically that was averaging of a great number of ¹⁰B and ¹¹B isotope ratio values.

The qualitative form of the relationship obtained after the ion image processing (Fig. 4) corresponds to the curve trends, which are shown on Fig. 5. It might be well to point out that time required for getting one set of ion images (Figs. 2 and 4) corresponds with the time required for getting one point of the route (Figs. 1 and 5) when the measurement error is congruent.

Comparison between the relationships presented on Fig. 4 and similar data obtained in the course of examinations [3] (Fig. 6) and [1] (Fig. 7) reveals that the isotope ratio distributions in boron carbide pellets is significantly different after irradiation in fast and thermal reactors.

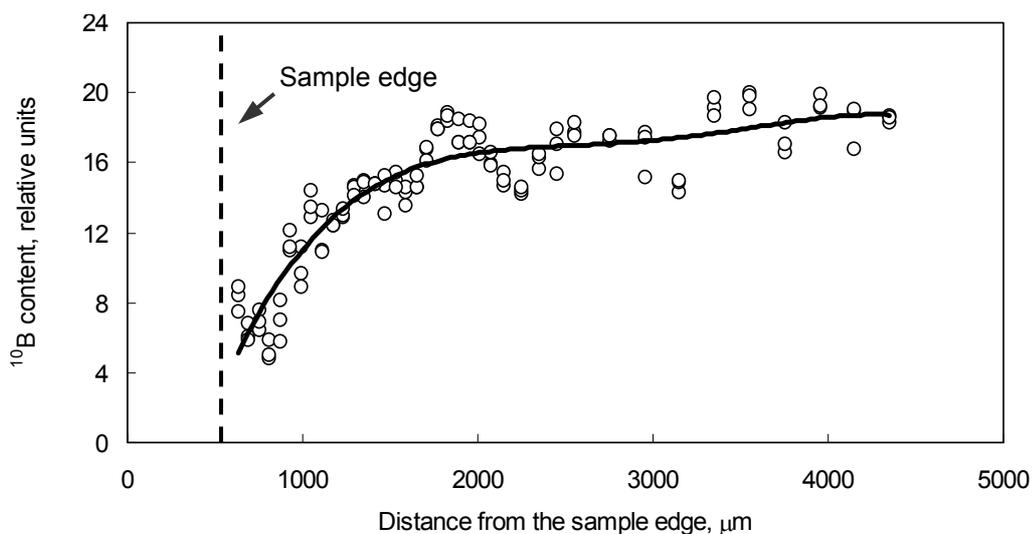


Fig. 6 Radial distribution of the ¹⁰B relative content in the power reactor absorber (B₄C powder of natural isotope composition enclosed in the stainless steel cladding) [3]

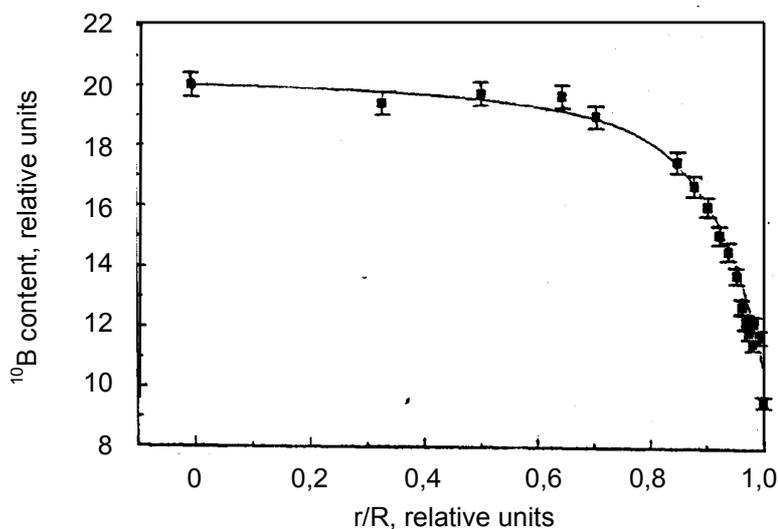


Fig. 7 Radial distribution of the ¹⁰B relative content in the absorber (B₄C sintered powder) irradiated in a thermal neutron flux of $3,0 \times 10^{12}$ neutron/(cm²·s) and fast neutron flux of ($E_n > 1$ MeV) $1,0 \times 10^{14}$ neutron/(cm²·s)) [1]

The phenomenon value is much higher than measurement errors. It makes up about two relative units (approximately it corresponds to 2 atomic %) both close to the edge of the object under examination and its centre.

One of the possible explanations for this phenomenon arises from the fact that ^{11}B and carbon isotopes as well as transmutation elements, which decrease fast neutron energy and cause their efficient absorption with the ^{10}B isotopes, are present in the material except for ^{10}B . Quantitative characteristics of the isotope ratio on the absorber radius can vary with a specific neutron spectrum and B_4C composition.

Conclusion

The secondary ion mass spectrometry method allows for getting a radial distribution of the boron isotope ratio in boron carbide pellets with a micron resolution. It was experimentally revealed that a radial distribution of isotope ratio in the boron carbide pellets is significantly different after irradiation in fast and thermal reactors. The discovery of such phenomenon points to the SIMS appropriateness for obtaining data on the isotope composition change in irradiated absorbers.

It is also reasonable to use ion images for such examinations. The resulted isotope maps allow for qualitative analysis of the isotope ratio with a resolution equal to the ion probe diameter. It is necessary to consider the topographic contrast effect accurately in order to make a quantitative analysis of the isotope ratio. So it is necessary to improve the available technique that resides in correction of the isotope ratio matrix with the use of images registered in the absorbed ion current. It will allow for getting a matrix of true isotope ratio values. The quantitative analysis of the isotope ratio will require less time significantly (if the accuracy is comparable with the results of a linear analysis) but the analysis itself will be performed with a maximum allowable lateral resolution for this particular type of the SIMS equipment.

References

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