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Trace elements study of high purity nanocrystalline silicon carbide (3C-SiC) using k_0-INAA method

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ABSTRACT

Silicon carbide (3C-SiC) nanoparticles have been irradiated by neutron flux (2×10^{13} $n\cdot cm^{-2}\cdot s^{-1}$) at TRIGA Mark II type research reactor. After neutron irradiation, the radioisotopes of trace elements in the nanocrystalline 3C-SiC were studied as time functions. The identification of isotopes which significantly increased the activity of the samples as a result of neutron radiation was carried out. Nanocrystalline 3C-SiC are synthesized by standard laser technique and the purity of samples was determined by the k_0-based Instrumental Neutron Activation Analysis (k_0-INAA) method. Trace elements concentration in the 3C-SiC nanoparticles were determined by the radionuclides of appropriate elements. The trace element isotopes concentration have been calculated in percentage according to k_0-INAA method.

1. Introduction

Silicon carbide is one of the materials which has great importance in nuclear and cosmic technologies. Therefore, nowadays nano-sized SiC and its different components started to be learned theoretically and practically by the researchers [1–11]. Cubic silicon carbide (3C-SiC) is widely used in different fields of science and technics for its unique physical, physical-chemical properties and radiation resistance [12–16]. High temperature resistance, excellent structure, mechanical resistance, low oxidation potential of SiC expand its application area as a nuclear and cosmic material [17–22]. The combination of excellent mechanical and functional properties of SiC is the base of its wide application in modern electronics as a semiconductor. There are more than 200 polytypes of SiC. The most widely used of these in electronic systems are cubic (3C - SiC) and hexaganal (4H-SiC and 6H-SiC) polytypes. Cubic nano SiC has wide usage in microelectronics as a nanoparticle for its wide band gap (2.26 eV), thermal and electrical properties [9–14]. Therefore, cubic 3C-SiC nanoparticles (also, it is known as β-SiC) were used in all experiments in this work.

The application opportunity of synthesized materials in different technologies depends directly on its purity. Until today the purity of synthesized materials is being increased with several methods and means. Intensive researches are conducted in the directions of improving purity of materials with the purpose of enlarging application opportunity of nanomaterials in the technologies [23–28]. At the present work, nanocrystalline 3C-SiC synthesized by standard laser method and irradiated by neutron flux (2×10^{13} $n\cdot cm^{-2}\cdot s^{-1}$) up to 20 h. Trace element isotopes activities of 3C-SiC nanomaterial and "cooling time" (up to 500 h) dependence, the time required for fission after neutron irradiation have been studied. The quality and quantity identification of mixtures in silicon carbide nanomaterial was conducted according to the results. Today nano silicon carbide with the purity of 99–99,9% is considered as high purity SiC nanomaterial and the samples with this purity are widely applied. According to the experiments it was found out that the nanomaterial had approximately 99.35% of purity and it could be considered as high purity SiC nanomaterial. However, trace elements in the nanomaterial could be greater than 0.65% and it seems insignificant at first sight but it is a big value in nanoscale and atomic composition. So, if we consider that there are about $1,7\times10^{22}$ particles in 1 g of SiC nanopowder on atomic level, then it can be clear how big value is 0.65 per cent (about 1.1×10^{20} mixed particles). Indeed, 0.65% of mixture influences the physical parametres of the sample very little, but this mixture expresses itself clearly during radiation in the reactor and the study of their structure is very important. So, the activity of the samples used in the experiment reached approximately 3GBq as a result of the effect of this mixture radioisotopes. In this case, until the activity of samples decreases (approximately after 500 h), it is impossible to carry out other experiments. Simultaneously, it is important to note "cooling time" on the other scientific researches [29–33]. At the present work, active

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isotopes and they standard decreasing curves has been given.

2. Experimental

Nanomaterial used in the experiment is cubic silicon carbide nanoparticles which is has 120 $m^2 \cdot g^{-1}$ specific surface area (SSA), 18-nm-sized particles and 0.03 $g \cdot cm^{-3}$ density (true density is 3.216 $g \cdot cm^{-3}$) (US Research Nanomaterials, Inc., TX, USA). The samples used in the experiments were irradiated at Jozef Stefan Institute TRIGA Mark II research reactor with neutron flux (central chanel, 2×10^{13} $n \cdot cm^{-2} \cdot s^{-1}$) at full power (250 kW thermal power). Parameters for neutron flux existed in central chanel at full power are 5.107×10^{12} $n \cdot cm^{-2} \cdot s^{-1}$ (1 ± 0.0008, $E_n < 625$ eV) for thermal neutrons, 6.502×10^{12} $n \cdot cm^{-2} \cdot s^{-1}$ (1 ± 0.0008, E_n ~ 625 eV ÷ 0.1 MeV) for epithermal neutrons, 7.585×10^{12} $n \cdot cm^{-2} \cdot s^{-1}$ (1 ± 0.0007, $E_n > 0.1$ MeV) for fast neutrons and lastly 1.920×10^{13} $n \cdot cm^{-2} \cdot s^{-1}$ (1 ± 0.0005) for all neutrons. Moreover, flux uncertainty (1 ± 0.0008, 1 ± 0.0007 and 1 ± 0.0005) is statistical part of Monte Carlo N-Particle Transport Code (MCNP) calculation [34–41].

For k_0-INAA an aliquot about 0.051 g of SiC in nano powder form was sealed into a pure polyethylene ampoule (SPRONK system, Lexmond, The Netherlands). For determination of short-lived radionuclides an aliquot and standard Al-0.1%Au (IRMM-530R) were stacked together, fixed in the polyethylene vial in sandwich form and irradiated for 5 min in the carousel facility (CF) of the TRIGA reactor with a thermal neutron flux of 1.1×10^{12} $n\ cm^{-2}\ s^{-1}$. For determination of long-lived radionuclides an aliquot and standard Al-0.1%Au were prepared on the same way as above and irradiated for 12 h in the CF of the TRIGA reactor.

After short irradiation (5 min) the aliquot was measured after 6, 25 and 180 min cooling time on an absolutely calibrated HPGe detector with 45% relative efficiency. After long irradiation (12 h) the aliquot was measured after 2, 7 and 28 days cooling time on the same HPGe detector. For peak area evaluation, the HyperLab 2002 program was used. The values f = 27.11 (thermal to epithermal flux ratio) and $\alpha = -0.0042$ (epithermal flux deviation from the ideal 1/E distribution) were used to calculate element concentrations. For elemental concentrations and effective solid angle calculations the software package Kayzero for Windows was applied. For QA/QC purposes for k_0-INAA the BCR-320R channel sediment was used (the results are presented together with certified values).

For the other physical experiments nanocrystalline 3C-SiC was irradiated in special aluminum cylinder. SiC nano powder which had the density of $\rho_{powder} = 0{,}03$ g/cm^3 (density was about ~0,1 g/cm^3 in container) and the mass of about ~ 1,28 g shaped in a special form and its parametres were like ρ_{tablet} = ~ 3.2 g/cm^3, V_{tablet} ~ 0,4 cm^3, S_{tablet}~ 4.5 cm^2. The samples were irradiated with the neutron flux intensity of 2×10^{13} $n \cdot cm^{-2} \cdot s^{-1}$. Absorption dose value of studied samples which were powder and tablet was determined according to the geometric measures, radiation intensity, radiation periods, the density of neutron flux effect and energetic spectrums of neutrons. The neutron flux value for the samples in the form of tablet changes between $1{,}3338\times10^{17}$ ÷ $2{,}6676\times10^{18}$ neutron/tablet intervals.

The radionuclides formed in nano SiC after mutual influence of neutron were analyzed in "Ortec HPGe detectors (Coaxial, Low and Well-Type)" and "Canberra coaxial HPGe detector" spectrometers. Radioactivity, isotope composition and mixed elements concentration of irradiated samples were determined according to [42–45] methodics.

3. Results and discussion

Owing to the neutron irradiation new excited radioactive nucleus generated in the nanomaterial. If we accept the initial number of these radioactive nucleus as N, the number of nucleus decreases according to the following conformity as a result of radioactive decay:

$$\frac{dN}{dt} = -\lambda N \tag{1}$$

here, λ is decay constant. We can get the following equality by simplifying the Eq. (1):

$$\ln N = -\lambda t + C \tag{2}$$

If we accept the number of radioactive isotopes as N_0 at the start ($t=t_0$), we can write Eq. (2) like the following:

$$\ln N = -\lambda t + \ln N_0$$

$$\ln\left(\frac{N}{N_0}\right) = -\lambda t$$

$$N = N_0 \exp(-\lambda t) \tag{3}$$

The last equation is exponential radioactive decay equation. Half-life period (half-life $t_{1/2}$) can be calculated following equations:

$$\frac{N}{N_0} = 0.5 = \exp(-\lambda t_{1/2})$$

$$t_{1/2} = \frac{\ln 2}{\lambda} = \frac{0.693}{\lambda} \tag{4}$$

Identification of radioactive isotopes was studied according to gamma spectroscopy method. Ray intensity γ appropriate to nuclear transmutation in gamma spectrum is different depending on radiation period and decay constants.

After neutron irradiation, activity of the trace elements in the samples was studied up to 500 h (for k_0-INAA up to 672 h). It was found out that the initial radioactivity of the irradiated samples changed between 0.02 kBq and 3 GBq. The activity of newly formed radioactive isotopes observed in irradiated samples change appropriately to decay constants. The initial activities of different radionuclides defined as a result of 20 days activity analysis change in a wide range. On the other hand, half-life of radionuclides existed in the mixture changes in a wide range like 0.037 ~ $8.5\cdot10^8$ h. So, the experiments were conducted in two stages: the long-lived radioisotopes were obtained from the samples irradiated 12 h and the short-lived isotopes were obtained from the samples irradiated 5 min. Therefore, here, these elements are divided into two groups. Observed radioactive isotopes are divided into long and short-lived radioisotopes according to their activity and half-life period. The dependence of initial activities of observed radioactive isotopes on observation time was studied in (Figs. 1–3) by groups. Firstly, let's look through radionuclides formed in nanomaterial after neutron flux effect which are long-lived and have the activity up to 163 MBq (Fig. 1).

Long-lived radionuclides were analyzed during 20 days (about 500 h). There were totally 8 types of radionuclides here and their half-life changes between 12.7 and 8.6E8 hours. Here the initial activities of existed radionuclides change between 0.3 kBq and 163MBq. It was found out from the analysis that long-lived radionuclides could be divided into two different groups according to their initial activities. The first group includes radionuclides which have the activity up to 0.35 kBq (Fig. 1a). Here half-live of the observed radionuclides is like 1023 h (^{181}Hf), 1077 h (^{59}Fe), 7508 h (^{54}Mn), $5.8\cdot10^8$ hour (^{93}Zr), $6.7\cdot10^8$ hour (^{59}Ni) and $8.6\cdot10^8$ hour (^{41}Ca). The activity of all radionuclides was less than 0.35 kBq after about 20 days. There are two long-lived radionuclides in the other group (Fig. 1b). The initial activities of these radionuclides are about 163 MBq. The radionuclides observed in this group have half-life like ^{64}Cu 12.7 h and ^{24}Na 15 h. The activity of both radionuclides decreased to zero and it is the result of low half life of radionuclides. The long-lived radionuclides in the other group which had different activity were described in Fig. 2a. Here existed radionuclides can be sorted like 27 h (^{121}Sn), 676 h (^{51}Cr) and 1238 h (^{89}Sr) for their half-life. The activity of observed radionuclides reduces to 8.8 kBq after about 500 h. The dependence of the activity of short-lived radionuclides which had medium activity (~kBq) on measurement period was described in Fig. 2b. We can sort these