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NEUTRON ACTIVATION ANALYSIS OF 3C-SiC NANOPARTICLES UNDER THE NEUTRON FLUX

E.M. Huseynov
Institute of Radiation Problems of ANAS
elchin.h@yahoo.com

Abstract: Silicon carbide (3C-SiC) nanoparticles have been irradiated by neutron flux (2x10^{13} n/cm^{2}s) at the nuclear research reactor. After neutron irradiation, the radioisotopes of active elements in the 3C-SiC nanoparticles were studied. The identification of isotopes which significantly increased the activity of the samples as a result of neutron radiation was carried out. The methodology of neutron activation analysis of 3C-SiC nanoparticle has been studied.

Keywords: nano 3C-SiC, nanomaterial, radioactivity, neutron activation analysis, neutron irradiation

1. Introduction

Over the past decade, silicon carbide and its various composites have been widely studied by the researchers in the world [1-14]. SiC with attractive physical and chemical resistance has a wide range of applications in extrinsic environments [15-19]. The change of the bandwidth in the 2.4eV – 3.2eV ranges has led to a wide use of SiC as a semiconductor in electronic systems [20-27]. In the general approach, SiC is a covalent bonded semiconductor. In terms of structure, each Si atom is covalently bonded to four carbon atoms and vice versa. Si and C atoms in SiC have combined in different modifications and have led to the formation of more than 200 polytypes. The most common of these are cubic (3C-SiC) and hexagonal (4H-SiC and 6H-SiC) polytypes. In this study, nanocrystalline 3C-SiC were used, in which the bandgaps was 2.2eV at room temperature.

The activity of the samples used in the experiment reached approximately 3GBq as a result of the neutron irradiation effect on the mixture radioisotopes. In this case, until the activity of samples decreases (approximately after 500 hours), it is impossible to carry out other experiments. Simultaneously, it is important to note "cooling time" on the other scientific researches [28-35]. At the present work, active isotopes and they standard decreasing table has been given.

2. Experimental

At the present work, research object is silicon carbide nanoparticles, which is has special surface area (SSA) of 120 m^2/g, the particle size of 18nm and the density of 0.03g/cm^3 (true density 3.216 g/cm^3) (US Research Nanomaterials, Inc., TX, USA). Samples irradiated by neutron flux (2x10^{13} n/cm^{2}s) in the central canal (canal A1) of the TRIGA Mark II light water pool type research reactor at full power (250kVt) in the Reactor Center of Institute Jozef Stefan (IJS) in Ljubljana, Slovenia. It is important to note that, if the reactor is working at full power then neutron flux parameter as followed: 5.107x10^{12} n/cm^{2}s (1±0.0008, $E_n < 625$eV) for thermal neutrons, 6.502x10^{12} n/cm^{2}s (1±0.0008, $E_n \sim 625$eV ÷ 0.1MeV) for epithermal neutrons, 7.585x10^{12} n/cm^{2}s (1±0.0007, $E_n > 0.1$ MeV) for fast neutrons and finally, the flux is 1.920x10^{13} n/cm^{2}s (1±0.0005) for all neutrons in the central canal [36-43].

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 1,3338x10^{17} ÷ 2,6676x10^{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 [44-47] methodics.

3. Results and Discussion

At the present work, the activity of mixture elements in the 3C-SiC nanoparticle has been studied under the neutron irradiation. 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 equation (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 equation (2) like the following:

$$\begin{gathered} \ln N = -\lambda t + \ln N_0 \\ \ln\left(\frac{N}{N_0}\right) = -\lambda t \\ N = N_0 \exp(-\lambda t) \end{gathered} \tag{3}$$

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

$$\begin{gathered} \frac{N}{N_0} = 0.5 = \exp\left(-\lambda t_{1/2}\right) \\ t_{1/2} = \frac{\ln 2}{\lambda} = \frac{0.693}{\lambda} \end{gathered} \tag{4}$$

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.

At the present work, clearly, described the analysis procedure of gamma-spectrum to the determination of the amount of activity. The acquisition of sample gamma spectrum has been described in Fig 1. Via the spectroscopy, it is possible to identify the radionuclides produced and their amounts of radioactivity in order to derive the target elements from which they have been produced and their masses in the activated sample. The spectrum analysis starts with the determination of the location of the (centroids of the) peaks. Secondly, the peaks are fitted to obtain their precise positions and net peak areas.

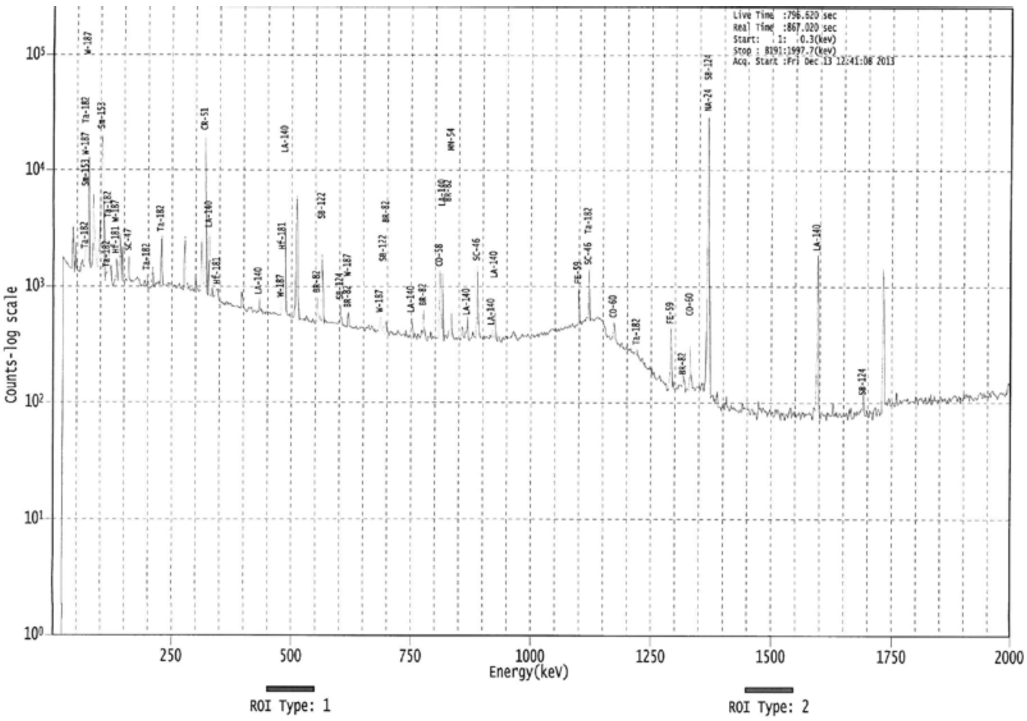

Fig 1. Gamma spectrum of nanoparticles after the neutron irradiation.

The determination of concentrations of elements is conducted according to the activities in appropriate energetic interval. The reaction rate (R) of the neutrons in each nucleus is defined as below [44-47]:

$$R = \int_0^\infty \sigma(v)\phi'(v)dv = \int_0^\infty \sigma(E)\phi'(E)dE = \int_0^\infty n'(v)v\sigma(v)dv \tag{5}$$

σ(v) is effective cross-section (cm^2) if the neutron velocity is v, σ(E) is effective cross-section (cm^2) if the neutron energy is E (eV), $\phi'(v)$is neutron flux (cm^{-3}) whose velocity is v, $n'(v)$is neutron density (cm^{-4}) whose velicity is v, $\phi'(E)$is neutron flux (cm^{-2} s^{-1} eV^{-1}) whose energy is E. Radioactivity of 3C-SiC nanoparticle has been giving at the table for low and high half-life elements (Table 1).

***Table 1a**. Radioactivity of various elements in the 3C-SiC nanoparticles (for high half-life elements)*

Time (hour)	Radioactivity (kBq)									Radioactivity (MBq)	
	Ca 41	***Cr 51***	***Fe 59***	***Hf 181***	***Mn 54***	***Ni 59***	***Sn 121***	***Sr 89***	***Zr 93***	***Cu 64***	***Na 24***
0	0.273	14.8943	0.160034	0.326954	0.023668	0.351	1.126551	0.967156	0.075	163.419	108.2264
5	0.273	14.81813	0.15952	0.325848	0.023657	0.351	0.990842	0.964452	0.075	124.3904	85.89935
24	0.273	14.53224	0.157581	0.32168	0.023616	0.351	0.608371	0.954247	0.075	44.09882	35.7014
48	0.273	14.17899	0.155166	0.316492	0.023563	0.351	0.328539	0.94151	0.075	11.90012	11.77707
72	0.273	13.83432	0.152787	0.311387	0.023511	0.351	0.177421	0.928943	0.075	3.211262	3.884984
96	0.273	13.49803	0.150446	0.306364	0.023459	0.351	0.095812	0.916544	0.075	0.866563	1.281567
120	0.273	13.16991	0.14814	0.301422	0.023407	0.351	0.051742	0.90431	0.075	0.233843	0.422759
144	0.273	12.84977	0.145869	0.29656	0.023355	0.351	0.027942	0.89224	0.075	0.063103	0.139459
168	0.273	12.53741	0.143633	0.291777	0.023304	0.351	0.01509	0.880331	0.075	0.017028	0.046004
192	0.273	12.23265	0.141432	0.28707	0.023252	0.351	0.008149	0.868581	0.075	0.004595	0.015176
216	0.273	11.93529	0.139264	0.28244	0.023201	0.351	0.004401	0.856987	0.075	0.00124	0.005006
240	0.2729999	11.64516	0.137129	0.277884	0.023149	0.351	0.002376	0.845549	0.075	0.000335	0.001651
264	0.2729999	11.36209	0.135027	0.273402	0.023098	0.351	0.001283	0.834263	0.075	9.03E-05	0.000545
288	0.2729999	11.08589	0.132958	0.268992	0.023047	0.351	0.000693	0.823127	0.075	2.44E-05	0.00018
312	0.2729999	10.81641	0.13092	0.264653	0.022996	0.351	0.000374	0.81214	0.075	6.58E-06	5.93E-05
336	0.2729999	10.55348	0.128913	0.260384	0.022945	0.351	0.000202	0.8013	0.075	1.77E-06	1.96E-05
360	0.2729999	10.29694	0.126937	0.256184	0.022894	0.351	0.000109	0.790605	0.075	4.79E-07	6.45E-06
384	0.2729999	10.04664	0.124992	0.252052	0.022844	0.351	5.89E-05	0.780052	0.075	1.29E-07	2.13E-06
408	0.2729999	9.802421	0.123076	0.247986	0.022793	0.351	3.18E-05	0.769641	0.075	3.49E-08	7.02E-07
432	0.2729999	9.564139	0.121189	0.243986	0.022743	0.351	1.72E-05	0.759368	0.075	9.41E-09	2.32E-07
456	0.2729999	9.33165	0.119332	0.240051	0.022692	0.351	9.28E-06	0.749232	0.075	2.54E-09	7.64E-08
480	0.2729999	9.104812	0.117503	0.236179	0.022642	0.351	5.01E-06	0.739232	0.075	6.85E-10	2.52E-08
504	0.2729999	8.883488	0.115702	0.232369	0.022592	0.351	2.71E-06	0.729365	0.075	1.85E-10	8.31E-09

***Table 1b**. Radioactivity of various elements in the 3C-SiC nanoparticles (for low half-life elements)*

Time (hour)	Radioactivity (kBq)			Radioactivity (MBq)		Radioactivity (GBq)	
	Ba 139	***Mo 99***	***Ti 51***	***Mg 27***	***V 52***	***Al 28***	***Cl 38***
0	5.79E-01	7.71E-01	6.98E+00	5.36E+01	1.51E+01	1.43E+00	3.04E+00
0.1	5.51E-01	7.24E-01	3.36E+00	3.48E+01	4.95E+00	2.20E-01	2.72E+00
0.5	4.51E-01	5.62E-01	1.82E-01	6.15E+00	5.65E-02	1.23E-04	1.74E+00
0.8	3.88E-01	4.65E-01	2.04E-02	1.68E+00	1.97E-03	4.45E-07	1.24E+00
1.1	3.33E-01	3.85E-01	2.28E-03	4.57E-01	6.90E-05	1.61E-09	8.89E-01
1.4	2.87E-01	3.19E-01	2.56E-04	1.25E-01	2.41E-06	5.84E-12	6.36E-01
1.7	2.47E-01	2.64E-01	2.86E-05	3.40E-02	8.43E-08	2.12E-14	4.54E-01
2	2.12E-01	2.19E-01	3.21E-06	9.26E-03	2.94E-09	7.67E-17	3.25E-01
2.3	1.82E-01	1.81E-01	3.59E-07	2.52E-03	1.03E-10	2.78E-19	2.32E-01
2.6	1.57E-01	1.50E-01	4.03E-08	6.88E-04	3.60E-12	1.01E-21	1.66E-01
2.9	1.35E-01	1.24E-01	4.51E-09	1.88E-04	1.26E-13	3.65E-24	1.19E-01
3.2	1.16E-01	1.03E-01	5.06E-10	5.11E-05	4.39E-15	1.32E-26	8.50E-02
3.5	9.98E-02	8.49E-02	5.66E-11	1.39E-05	1.53E-16	4.80E-29	6.08E-02
3.8	8.59E-02	7.03E-02	6.35E-12	3.80E-06	5.36E-18	1.74E-31	4.34E-02
4.1	7.39E-02	5.82E-02	7.11E-13	1.04E-06	1.87E-19	6.30E-34	3.11E-02
4.4	6.35E-02	4.82E-02	7.97E-14	2.83E-07	6.55E-21	2.28E-36	2.22E-02
4.7	5.46E-02	3.99E-02	8.93E-15	7.70E-08	2.29E-22	8.28E-39	1.59E-02
5	4.70E-02	3.30E-02	1.00E-15	2.10E-08	8.00E-24	3.00E-41	1.14E-02

Nuclear transmutations are defined according to the number of nuclear fissions. Initially, the number of active nucleus ($N(t_i, t_d)$) can be calculated following equation [44-47]:

$$N(t_i, t_d, t_m) = \frac{RN_0}{\lambda}(1 - \exp[-\lambda t_i])\exp[-\lambda t_d] \tag{6}$$

The number of nuclear fissions during the measurement can be defined according to the following equation:

$$\Delta N(t_i, t_d, t_m) = \frac{RN_0}{\lambda}(1 - \exp[-\lambda t_i])\exp[-\lambda t_d](1 - \exp[-\lambda t_m]) \tag{7}$$

Here, t_d is decay or waiting time (the time between the end of irradiation and the beginning of measurement), t_i is end of the irradiation time, t_m is measurement time. According to the existed peaks in the spectrum we can write activation formula like the above:

$$N_p = \Delta N \gamma \varepsilon = \varphi_{th} \sigma_{eff} \frac{N_{av} m_x \theta}{M_a}(1 - \exp[-\lambda t_i])\exp[-\lambda t_d](1 - \exp[-\lambda t_m])I\varepsilon \tag{8}$$

Here, N_p is total number in gamma peak (E_γ), N_{av} is Avogadro's number, θ is isotopic abundance of target isotopes, m_x is mass of irradiated element in grams, M_a is atomic mass (g mol^{-1}), I is the gamma-ray abundance and ε is total energy detected in detector. Simply, element quantity can be calculated from the net peak area according to the following equation:

$$m_x = N_p \cdot \frac{M_a}{N_{av}\theta} \cdot \frac{\lambda}{\varphi_{th}\sigma_{eff} I\varepsilon(1 - \exp[-\lambda t_i])\exp[-\lambda t_d](1 - \exp[-\lambda t_m])} \tag{9}$$

Calculations are a bit complicated in k_0 comparator method [32].

4. Conclusions

Radioisotopes generated in the nanocrystalline 3C-SiC particles after neutron irradiation and it were studied. It was found out that initial activity and half life of mixed elements in the sample differed from each other significantly. The dependence of the radioactivity of isotopes on observation time were studied. Long half life isotopes were found in the nano SiC samples and it was suggested to consider them in the explanation of physical properties of nanocrystalline 3C-SiC particles after irradiation.

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НЕЙТРОННО-АКТИВАЦИОННЫЙ АНАЛИЗ НАНОЧАСТИЦ 3C-SiC ПОД НЕЙТРОННЫМ ПОТОКОМ

Э.М. Гусейнов

Резюме: Наночастицы карбида кремния (3C-SiC) были облучены потоком нейтронов (2x10^{13} н/см^{2}с) в ядерном исследовательском реакторе. После облучения нейтронами изучались радиоизотопы активных элементов в наночастицах 3C-SiC. Была проведена идентификация изотопов, которые значительно увеличили активность образцов в результате нейтронного излучения. Изучена методология нейтронно-активационного анализа наночастиц 3C-SiC.

Ключевые слова: нано-3C-SiC, наноматериал, радиоактивность, нейтронно-активационный анализ, нейтронное облучение

NEYTRON SELİNİN TƏSİRİ ALTINDA 3C-SiC NANOHİSSƏCİKLƏRİNİN NEYTRON AKTİVLƏŞMƏ ANALİZLƏRİ

E.M. Hüseynov

Xülasə: Silisium karbid (3C-SiC) nanohissəcikləri neytron seli ilə (2x10^{13} n/sm^{2}san) tədqiqat nüvə reaktorunda şüalandırılmışdır. Neytronlarla şüalanmadan sonra 3C-SiC nanohissəciklərində aktiv elementlərin radioizotopları öyrənilmişdir. Neytronlarla şüalanmanın təsiri nəticəsində nümunənin aktivliyini artıran izotoplar təyin edilmişdir. 3C-SiC nanohissəciklərində neytron aktivləşmə analizlərinin metologiyası analiz edilmişdir.

Açar sözlər: nano 3C-SiC, nanomaterial, radioaktivlik, neytron aktivasiya təhlili, neytron irradiasiyası