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RADIOACTIVITY STUDY OF 3C-SiC NANOPARTICLES UNDER THE NEUTRON FLUX AT THE TRIGA MARK II TYPE RESEARCH REACTOR

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Abstract. Nano silicon carbide (3C-SiC) particles are continuously irradiated up to 20 h by neutron flux with $2 \cdot 10^{13}$ n/cm^2·s density at the TRIGA Mark II type research reactor. The change of activities of SiC nanoparticles have been studied after neutron irradiation within 500 h. It was determined that, the initial activity of irradiated nanomaterial are increased up to 3GBq andit's due to the radioisotopes of trace elements inside nanomaterial. As a result of activity analysis of silicon carbide nanoparticles trace elements has been determined appropriate to radioisotopes.

Keywords: *nano 3C-SiC, nanomaterial, nanopowder, radioactivity, neutron irradiation.*

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1. Introduction

Silicon carbide is very important material in nuclear and space technology. For this reason, recently nanoSiC and it's various components are detailed investigated in theoretically and experimentally by world scientists (Koyanagi *et al*., 2014; Meenakshi *et al*., 2016; Van Laar *et al.*, 2015; Bae *et al*., 2015; Pradhan *et al*., 2015; Xie *et al*., 2002; Dinh *et al.*, 2015; Cui *et al*., 2013). Cubic modification silicon carbide (3C-SiC) is widely applying in different fields of science and technology due to unique physical, physical-chemical properties and radiation resistance (Bruzzi *et al*., 2001; Aguado & Baonza, 2006; McMullin *et al*., 1993; Singh *et al*., 2009; Singh *et al*., 2016; Ivanov *et al*., 2006; Katoh *et al*., 2012). High temperature stability, perfect structure, mechanical stability, low oxidation properties are increase application potential of SiCas a nuclear and space material. The combination of perfect mechanical and functional properties of SiC is the base of wide application in modern electronics. SiC has known more than 200 polytypes. Among them the cubic (3C-SiC) and hexagonal (4H and 6H) phase polytypes are more widely applied in the electronics systems. The cubic modification nanoSiC has more application potential in microelectronics due to the band gap (2.26 eV) as well the thermal and electrical properties (Yan-Kun *et al.*, 2012; Mallick *et al.*, 2007; Zhonghua *et al.*, 2015; Cui *et al.*, 2013). Therefore, we have been used cubic

modification nano 3C-SiC (also known as β-SiC) for all experiments in the presented work. Nano 3C-SiC particles have been continuously irradiated by neutron flux with $2x10^{13}$ n/sm^{2}s flux density up to20 hours in various period at the TRİGA Mark II type research reactor.

In the presented article, we have been investigated the dependences on integral doses and decay times after irradiation of radioactive nucleus activities which formed in the 18 nm size SiC nanomaterials as result of the influence of neutron flux with the $2x10^{13}$ n/sm^{2}s intensity. The type of the trace elements in the silicon carbide nanomaterial has been determined in the base of obtained results. These elements are clearly appearing during the irradiation in the nuclear reactors and the study of their compositions is very important. The activity of experimental samples is increase to 3GBq as a result of the influence of these elements. In this case, it's impossible to carrying out of other experiments until the decreasing an activity of nanomaterial (approximately during the 500 hours) irradiated with neutron flux. So that, the "cooling time" is marked in the similar experiments at TRIGA Mark II research reactor (Huseynov *et al*., 2019; Huseynov, 2018a; Huseynov, 2018b; Huseynov, 2018c; Huseynov, 2018d; Huseynov & Jazbec, 2017; Huseynov, 2017; Huseynov & Garibov, 2017; Huseynov, 2016). In the presented work, are given isotopes that generated activity and the standard reduction regularity of the activity of corresponding isotopes.

2. Experimental methods

Cubic modification silicon carbide nano particles used at the present experiment, which has 120 m^2/g specific surface areas, 18nm particle size and 0.03g/cm^3 bulk density (true density is 3.216 g/cm^3). The samples used during experiment were irradiated by neutron flux ($2x10^{13}$ n/cm^{2}s) at full power (250kW) in the channel A1 of TRIGA Mark II light water pool type research reactor in the "Reactor Center" of Institute Jozef Stefan (IJS) in Ljubljana, Slovenia. When the reactor is running at full power, the parameters of neutron flux in channel A1 is $5.107x10^{12}$ n/cm^{2}s (1±0.0008, E_n< 625eV) for thermal neutrons, $6.502x10^{12}$ n/cm^{2}s (1±0.0008, E_n ~ 625eV ÷ 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 it is $1.920x10^{13}$ n/cm^{2}s (1±0.0005) for all neutrons (Snoj *et al*., 2012; Žerovnik *et al*., 2014a; Žerovnik *et al*., 2015; Henry *et al*., 2015; Snoj *et al*., 2010; Filliatre *et al*., 2015; Žerovnik *et al*., 2014b; Kolšek *et al*., 2015).

For the other physical experiments nanocrystalline 3C-SiC was irradiated in special aluminum cylinder. SiCnano powder which had the density of ρ_{powder} = 0,03g/cm^3 (density was about ~0,1q/sm^3 in container) and the mass of about ~ 1,28g shaped in a special form and its parameters were like ρ_{tablet} = ~ 3.2g/cm^3, V_{tablet} ~ 0,4cm^3, S_{tablet}~ 4.5cm^2. The samples were irradiated with the neutron flux intensity of$2x10^{13}$ n/sm^2. Absorption dose value of studied samples which were powder and tablet was determined according to the geometric measures, radiation intensity, radiation periods, and 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 radionuclide's formed in nanoSiC after mutual influence of neutron were analyzed in "Ortec HPGe detectors" and "Canberra coaxial HPGe detector" spectrometers. Radioactivity, isotope composition and mixed elements concentration of irradiated samples were determined according to (Frontasyeva, 2011; Ishak-Boushaki *et al*., 2012) methodics.

3. Results and discussions

The most probable process of interaction neutrons with substance is radiation capture (Huseynov & Jazbec, 2017; Frontasyeva, 2011; Ishak-Boushaki *et al.*, 2012).

$$n + {}^{A}Z \rightarrow {}^{A+1}Z^{*} \rightarrow {}^{A+1}Z + \gamma \qquad (1)$$

here, ${}^{A}Z$ is an irradiated isotope, ${}^{A+1}Z^{*}$ is an excited nucleus as a result of neutron capture and γ – is second gamma rays. Radioactive excited samples are forming on irradiated samples as a result of mentioned processes. An identification of these samples was carried out by gamma spectroscopy method. The intensity of γ-rays in gamma spectrometer which corresponds to nuclear transitions is different depending on irradiation time and decay constants.

The change of activities was investigated after 500 hours from neutron irradiation of samples. It was determined that, the initial activity of irradiates samples are changed in the range of 0.02kBq – 3GBq. The determination of density of the elements is carried out in the basis of activities which formed in corresponding energetic range. In the absolute variant, the activity is determined on the basis of the following formula based on nuclear constants (Huseynov & Jazbec, 2017; Frontasyeva, 2011; Ishak-Boushaki *et al.*, 2012).

$$A = \sigma\Phi\left(\frac{m}{M}\right)N_A\Theta P_\gamma\xi\left(1-\exp(-\lambda\tau_{irrad.})\right)\left(1-\exp(-\lambda\tau_{meas.})\right)\exp(-\lambda\tau_{cool.}) \qquad (2)$$

here, A is the measured activity (Bq), σ is the cross-section of activation of determined isotopes (sm^2), Φ is a neutron flux (n/sm^2 san), m is the mass of determined elements (g), M is the atomic mass of determined element (g/mol), N_A is the Avogadro number, Θ is a distribution of activated isotopes, P_γ is a distribution probability of E energy gamma quanta, ξ is the efficiency of defects based on E energy, λ is the decay constants of formed isotopes, $\tau_{irrad.}$, $\tau_{meas.}$, $\tau_{cool.}$ is the irradiation, measurements and cooling time ranges of appropriate samples.

The activity of the new formed radioactive isotopes which observed in irradiated samples are change corresponding to decay constants. As a result of activation analyzes for 20 days was determined that, the initial activities of various type of determined radionuclides change very wide range. Moreover, the half-life of radionuclide's in the mixture are change in range of $0.037 \sim 8.5 \cdot 10^8$ hours. So that, observations were conducted in two stages: long life-time radioisotopes were found from up to 20 hours irradiated samples, short life-time radioisotopes were found from 5 minutes irradiated samples. For this reason, these elements conditionally are divided into two groups (long and relatively short life-time radioisotopes). The observed radioactive isotopes are grouped according to their activity and life-time. The dependence of initial activation observation time on conventional groups of radioactive isotopesis shown in Figures 1-5. In the first, relatively short life-time and up to 7kBq activities of the radionuclides in the irradiated nanomaterial have been considered (Figure 1).

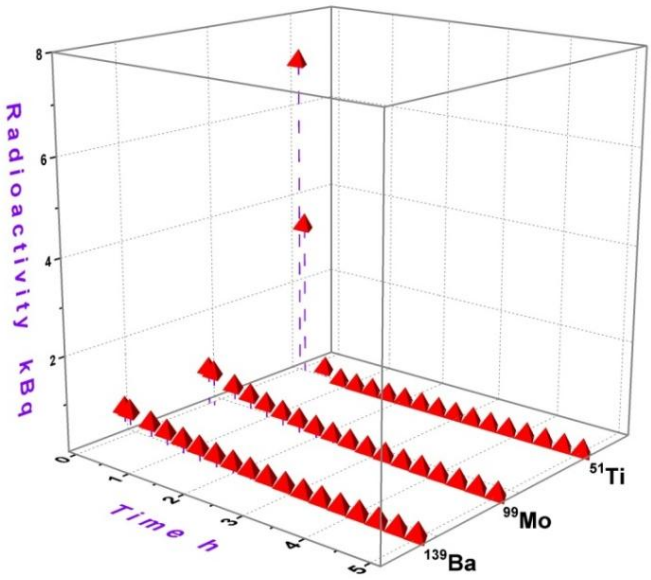

Figure 1. The dependences of activity on measurement times of relatively low activity (~kBq) and short life-time radionuclide's formed in nanoSiC under the neutron flux

In general, here 3 types of radionuclides were observed. Their half-life changes from 0.095 to 1.38 hours. The initial activities of existing radionuclides are changes from 0.6 to 7 kBq. The life-time of observed radionuclide's are ^{51}Ti (0.095 h), ^{99}Mo (1.1 h) and^{139}Ba (1.38 h), respectively.

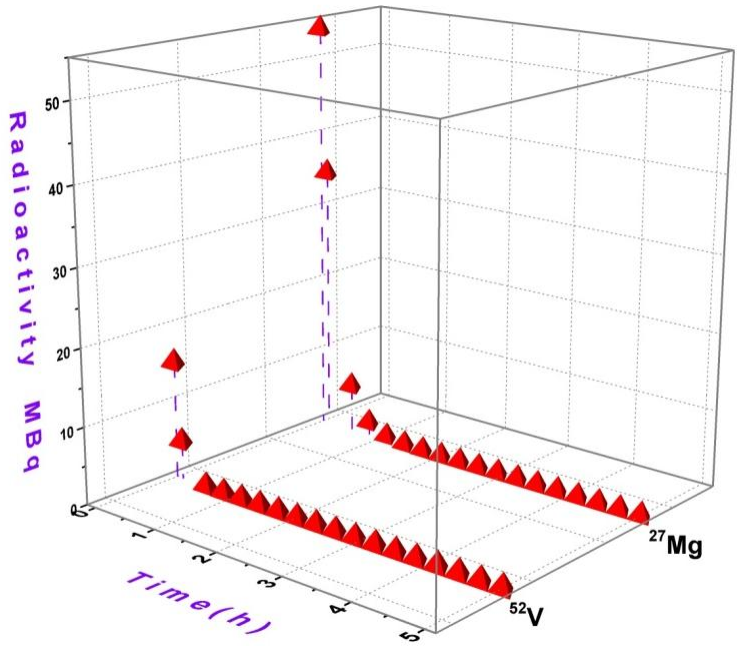

Figure 2. The dependences of activity on measurement times of relatively medium activity (~MBq) and short life-time radionuclide's formed in nanoSiC under the neutron flux

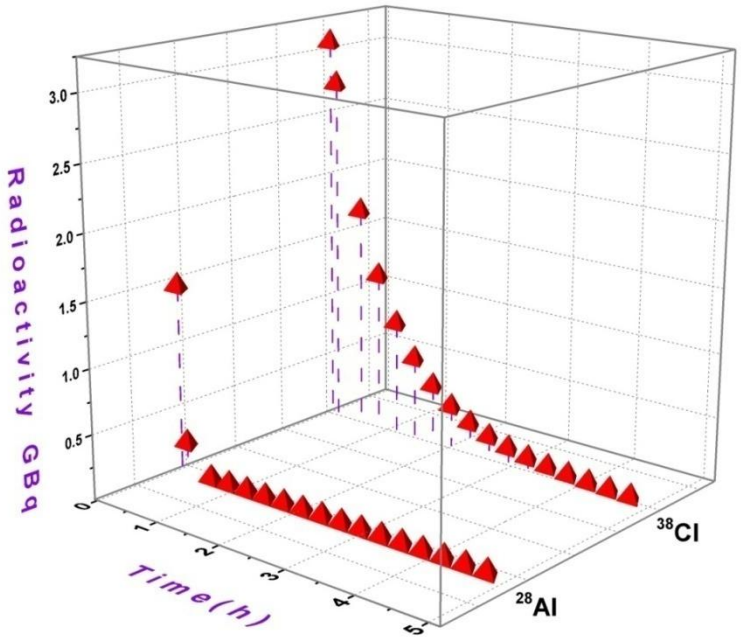

Figure 3. The dependences of activity on measurement times of relatively high activity (~GBq) and short life-time radionuclide's formed in nanoSiC under the neutron flux.

The initial activity of the short life-time other radionuclide's approximately up to 54 MBq (Figure 2). Here two types of radionuclide's (^{52}V and^{27}Mg) were observed and their half lifeis 0.062 h and 0.16 h respectively to ^{52}V and ^{27}Mg. As seen from figure 2,the activity of both radionuclides has decreased approximately to zero after 5 hours. The initial activities of other groups of short life-time radionuclides approximately have up to 3GBq. The half-life of observed ^{38}Cl high activity isotope is 0.62 hour and the activity of this isotope has decreased to 11.5 kBq at the end of 5 hours of measurements (Figure 3).The initial activity of ^{28}Al isotopes included in this group is 1.4GBq and half-life is 0.037 hours (the activity of ^{28}Al isotope estimate has decreased to zero at the end of 5 hours of measurements). The ^{38}Cl isotope, which has several times more activity, is a major part of the mixture.

Long life-time radionuclides were analyzed for 20 days (approximately 500 hours). As a result of these analyzes, it has been established that the long life-time radionuclides can be divided into two groups based on their initial activities. The first group of radionuclide's are includes about up to 15kBq activities (Figure 4). In here, the half-life of observed radionuclide's are 27 hours (^{121}Sn), 676 hours (^{51}Cr), 1023 hours (^{181}Hf), 1077 hours (^{59}Fe), 1238 hours (^{88}Sr), 7508 hours (^{54}Mn), $5.8\cdot10^{8}$ hours (^{93}Zr), $6.7\cdot10^{8}$ hours (^{59}Ni) and $8.6\cdot10^{8}$ hours (^{41}Ca), respectively. The activity of all radionuclide's after 20 days estimate were less than 8.8 kBq.

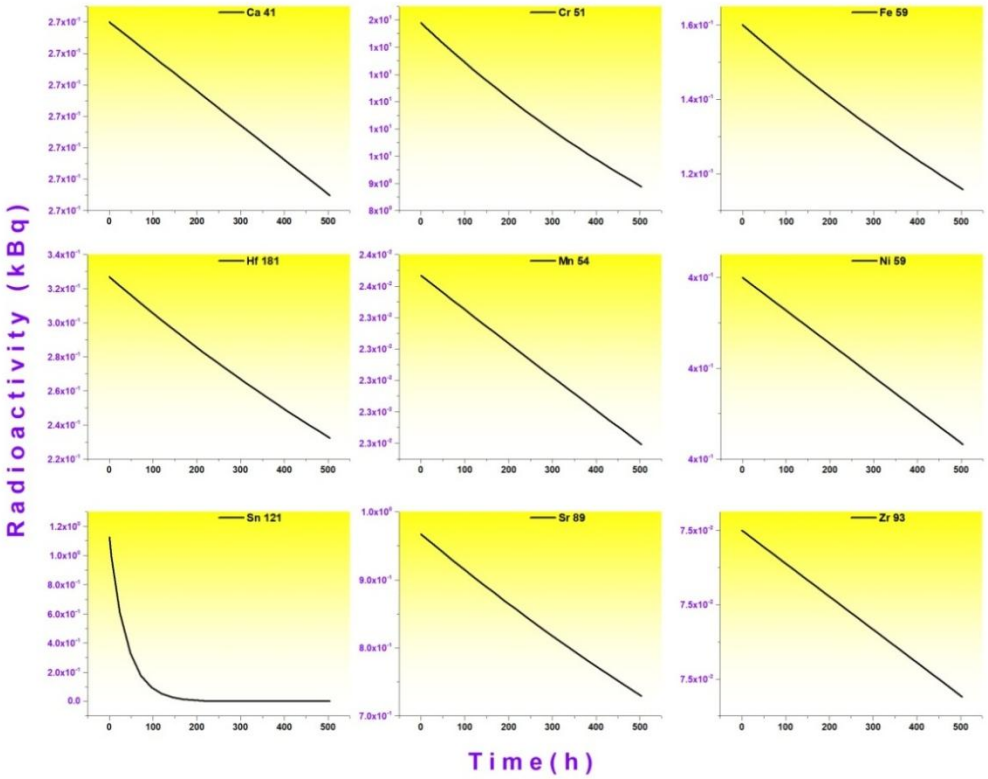

Figure 4. The dependences of activity on measurement times of relatively low activity and long life-time radionuclide's formed in nanoSiC under the neutron flux

There were two long life-time radionuclide's included in other group (Figure 5). Initial activities of these radionuclides approximately were 163 MBq. An observed radionuclide's in this groups have ^{64}Cu 12.7 hours and ^{24}Na 15 hours half-life. The activity both radionuclideshas decreased to zero due to the relatively short half-life of radionuclides.

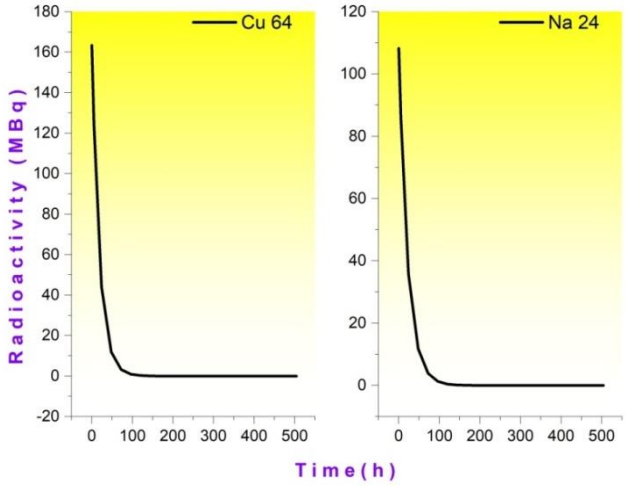

Figure 5. The dependences of activity on measurement times of relatively high activity and long life-time radionuclide's formed in nanoSiC under the neutron flux

It seems that given dependences in figures 4,5, the activity of radioactive low decay constant and long half-time radionuclide's are slightly change during the experimental observation and the effects of their radioactive decay are expected during the next study of the properties of nanoSiC. Overall, the calculated radioactivity in various times of radionuclide's of trace elements during the effect of the neutron flux on nanoSiC particles are presented in tables (Table 1 and Table 2)

Table 1.Relatively long life-time radioisotopes formed in SiC nanomaterial under the neutron flux

Time	Activity (kBq)									Activity (MBq)	
(hour)	***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 2.Relatively short life-time radioisotopes formed in SiC nanomaterial under the neutron flux

Time	Activity (kBq)			Activity (MBq)		Activity (GBq)	
(hour)	***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

Relatively long life-time radioisotopes and their activities with kBq and MBq are given in table 1. Relatively short life-time radioisotopes and their activities are listed in table 2 with kBq, MBq and GBq.

4. Conclusions

An identification of radioactive isotopes formed in SiC under the neutron flux was carried out. Irradiated samples investigated as a function of time. It was established that, the initial activities and life-times of trace elements in the sample are sharply differs each others. So that, the initial activities of trace isotopes in the nanoSiC irradiated with neutron flux are change in the range of 0.02kBq – 3GBq. The dependence of radioactivity of the detected isotopes on observation time of and the type of trace elements are determined. Relatively long life-time isotopes were detected in investigated nanoSiC samples, which are suggested to be considered to description of the physical properties of nanoSiC after neutron irradiation.

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