

# Effect of neutron flux on the frequency dependencies of electrical conductivity of silicon nanoparticles

Elchin Huseynov and Aydan Garibli

*Department of Nanotechnology and Radiation Material Science, National Nuclear Research Center,*

*AZ 1073, Inshaatchilar pr. 4, Baku, Azerbaijan*

*e-mail: elchin.huse@yahoo.com; e.huseynov@mntm.az*

Received 27 January 2016; accepted 17 February 2016

It has been reviewed the frequency dependencies of electrical conductivity of nanoparticles affected by neutron flux at different times and initial state, at various constant temperatures such as 100 K, 200 K, 300 K and 400 K. Measurements have been carried out at each temperature at the different 97 values of frequency in the 1 Hz - 1 MHz range. From interdependence between real and imaginary parts of electrical conductivity it has been determined the type of conductivity. Moreover, in the work it is given the mechanism of electrical conductivity according to the obtained results.

*Keywords:* Nanostructures; electrical properties; radiation damage; defects; semiconductivity.

PACS: 61.80.Hg; 61.46.Hk; 72.15.Eb; 74.25.fc; 61.80.-x

## 1. Introduction

Over the past few years, nano-sized Si has been at the focus of researchers in the world and its properties have been theoretically and practically studied [1-10]. Micro sizes silicon has wide application field in nuclear technology and cosmic electronics. In the future, these application fields' to bein nano-scale is inevitable and therefore, study of the influence of neutron irradiation on electrical conductivity of these compounds in the point of interest. Thereby, micro and nano silicon has wide applications in electronic devices employed for neutron and cosmic radiation measurements. The defects appeared due to neutron irradiation make important changes on the electrophysical properties of the samples as it will be reported in the section of results. Firstly, let's consider the defects formed in nanoparticles exposed to neutron irradiation continuously at different periods. So, Si nanoparticles are composed of Si atoms with cubic modification, that's why within the neutron flux influence on lattice atoms there can be observed high energy recoils (high energy recoils, primary knock-on) in some atoms. Within energy exchange between Primary Knock-on Atom (PKA) and other atoms displaced atoms from its lattice site by neutron irradiation, point defects or clusters are formed and they are the basis of fundamental defects. The defects formed under neutron flux influence, can be migrated and are real place for storage of any charge at different periods [11-14]. These formed defects are as if a "trap" for storing other charges (ex. electrons) occurred under neutrons' influence. However, there is very little probability that random elements or gases adsorbed from atmosphere may be caught in these traps [14,15]. In the submitted work the mechanism of electrical conductivity of the samples has been explained by known methods [16-22].

Silicon is the basis of almost all electronics systems which are used today. So, this material is widely applied in oscillators, sensors, electronics, computers, networks,

phones, detectors, etc. and their application in nano sizes in above-mentioned fields in future is real. Considering that the majority of the above-mentioned fields are widely used in used in nuclear and outer space technologies, then it can be clearly seen the importance of studying the electrical properties of Si nanoparticles.

## 2. Experimental

The nanomaterial used in the experiment consists of cubic modification silicon nanoparticles which is has, 80 m<sup>2</sup>/g specific surface area (SSA), 100 nm particles' size and 0.08 g/cm<sup>2</sup> density. Some parameters of oxide form of used samples (silica) have been studied similar experiments [19-26]. It has been reviewed the frequency dependencies of electrical conductivity within initial state and after neutron irradiation. The samples have been irradiated at full power mode (250 kW) by neutron flux ( $2 \times 10^{13} \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$ ) in central channel (channel A1) of TRIGA Mark II light water pool type research reactor in "Reactor Centre" of "Institute Jozef Stefan" (IJS) in the city of Ljubljana, Slovenia. The parameters of neutron flux at full power mode in central channel are as follows:  $5.107 \times 10^{12} \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-2}$  ( $1 \pm 0.0008$ ,  $E_n \sim 625 \text{ eV}$ ) for thermal neutrons,  $6.502 \times 10^{12} \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$  ( $1 \pm 0.0008$ ,  $E_n \sim 625 \text{ eV} \div 0.1 \text{ MeV}$ ) for epithermal neutrons,  $7.585 \times 10^{12} \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$  ( $1 \pm 0.0007$ ,  $E_n > 0.1 \text{ MeV}$ ) for fast neutrons and finally for all the neutrons in central channel flux density is as  $1.920 \times 10^{13} \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$  ( $1 \pm 0.0005$ ) [27-34]. Thus, the average energy of neutrons in central channel is compatible with nearly the energy of epithermal neutrons ( $E_n \sim 625 \text{ eV} \div 0.1 \text{ MeV}$ ). Nano Si particles being pressed at 3 kN/cm<sup>2</sup> press machine in special conditions in the "Thin Films and Surfaces F3" department of IJS, was made in the form of tablet with 2.5 mm height and 5.5 mm diameter and then placed in aluminium container appropriate

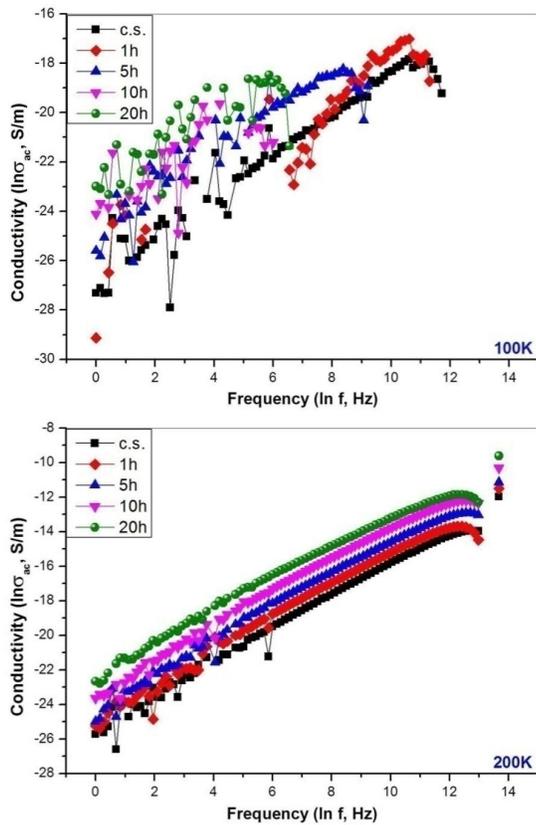


FIGURE 1. Frequency dependencies of electrical conductivity of nano silicon particles at the 100 K and 200 K temperature (control sample (c.s.) and after 1, 5, 10 and 20 hours of neutron irradiation).

to the channels of the reactor. The prepared samples have been continuously irradiated in central channel for 1, 5, 10 and 20 hours. Activity of the samples has increased up to 3.1 GBq under neutron flux influence. Therefore all the measurements have been carried out approximately 400 hours after neutron flux influence. After irradiation, silver contacts have been fixed on the surface of samples in special condition and examined its quality (Ögussa, Leitsilber 200). It has been used Cr/Au electrodes obtained on the top layer by spray method. Then using platinum top and bottom of tablet form samples and electrical conductivity of the samples has been measured in “Novocontrol Alpha High Resolution Dielectric Analyzer” device for alternating current (AC  $\sim$  0.5 V) at 100-400 K temperature and 1 Hz - 1 MHz frequency range. During the measurements storage accuracy of temperature in any degree was up to  $10^{-2}$  K and the accuracy was obtained with the bridge method (Pt100 resistor) [35]. All of the results compatible with the values obtained from experiments have been graphically depicted employing “OriginPro 9.0” program.

### 3. Result and discussions

Within the measurements it has been reviewed the frequency dependencies of electrical conductivity of samples at various constant values of temperature. The experiments have been

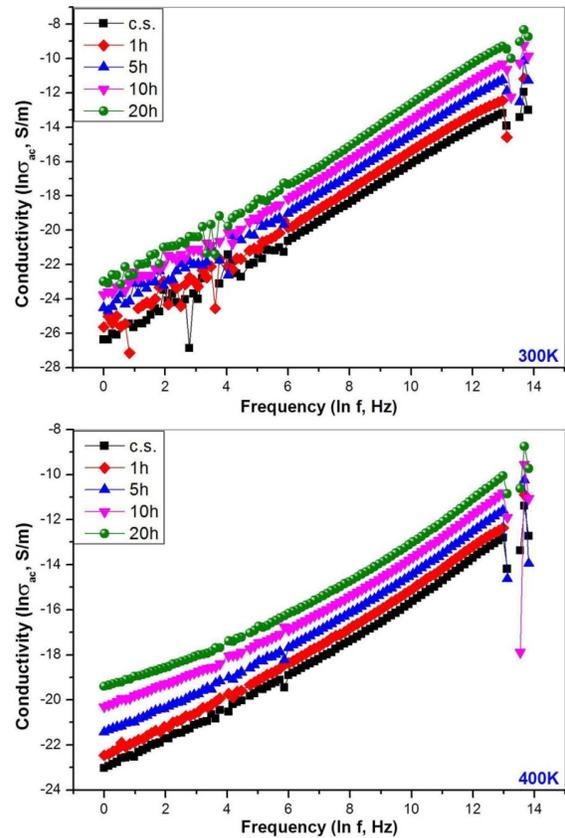


FIGURE 2. Frequency dependencies of electrical conductivity of nano silicon particles at the 300 K and 400 K temperature (control sample (c.s.) and after 1, 5, 10 and 20 hours of neutron irradiation).

conducted within the  $1 - 10^6$  Hz frequency range and during the measurements it has been revealed that the frequency dependencies of electrical conductivity are different at various values of temperature. Here firstly, we have reviewed the frequency dependencies of electrical conductivity irradiated at different periods and four values of temperature within the frequency range 1 Hz - 1 MHz. First of all, let's see the frequency dependencies of electrical conductivity at 100 K and 200 K values of temperature (Fig. 1). As it is seen from figure, at 100 K temperature there is a chaotization in frequency dependence of electrical conductivity.

However, in general tendency there is an increase in electrical conductivity with increase of effect term of neutron flux. The chaotization can be explained by the theory of cluster formed in the sample under neutron flux influence [15-21]. At 200 K temperature the existing chaotization is relatively eliminated due to the temperature. Under the influence of temperature and frequency the chaotization is almost fully recombined at the frequency range  $\ln f \geq 5$  (Fig. 1). The similar cases are observed at 300K temperature, as well (Fig. 2).

However, at low frequency range at 400 K temperature this chaotization is fully recombined with temperature and frequency influence (Fig. 2). In the result of the measurements carried out at all temperatures it has been established that, the frequency dependencies of electrical conductivity

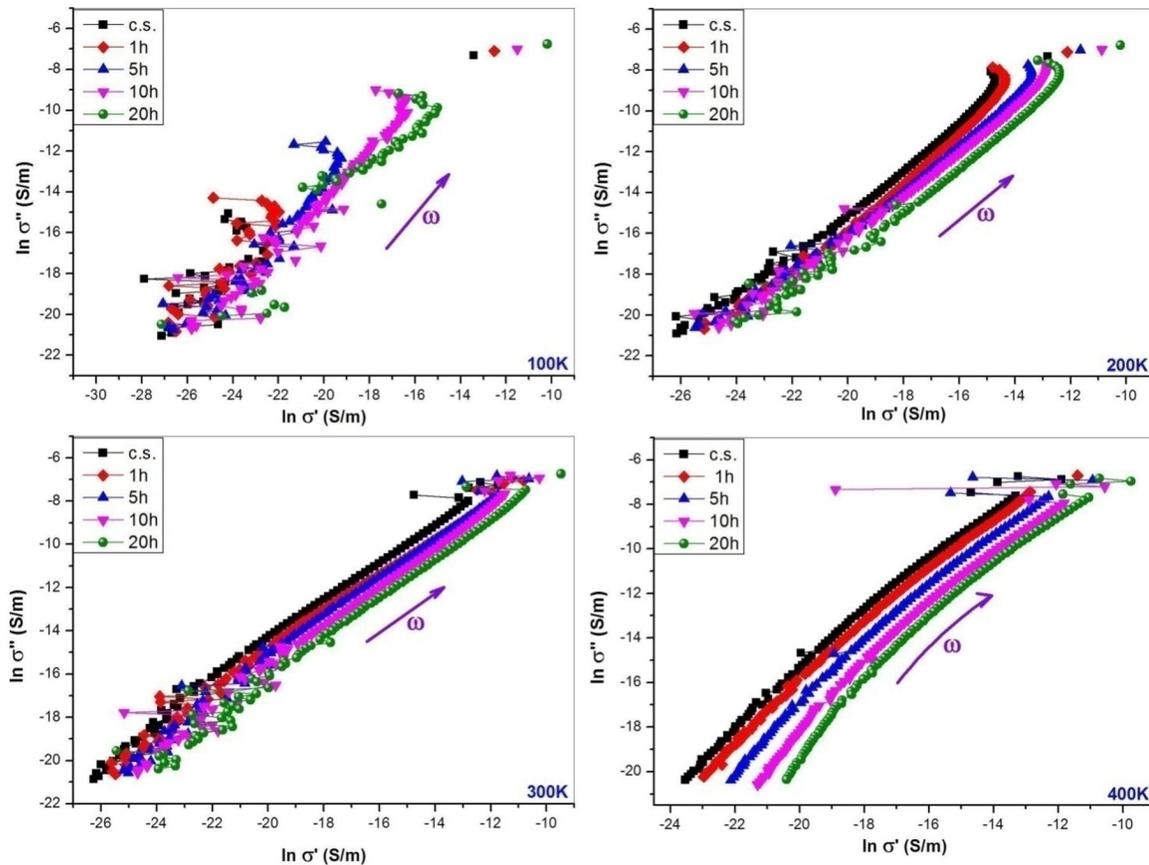


FIGURE 3. Correlations between real and imaginary parts of electrical conductivity of nano Si particles at initial state (control sample-c.s.) and after 1, 5, 10 and 20 hours of neutron irradiation at 100 K, 200 K, 300 K and 400 K temperatures.

are of linear character. As it is seen from Figs. 1 and 2 that, at 200 K, 300 K and 400 K temperatures the electrical conductivity of the samples increase about 20 times under the influence of neutron flux. Its reason is the active participation of the charge carriers or electrons existing in local states in conductivity due to irradiation.

At all temperatures sharp deviations and peaks are observed at high frequency range (Fig. 1-2). It can be explained by resonance phenomenon which appeared in the result of coincidence of intrinsic frequency of measuring device with the frequency of the sample. Thus, in measuring system the resistance or current deviations in inductance can be cause a breakthrough in experimentally recording dielectric data [36,37]. In this case general impedance can be determined as follows:

$$\frac{1}{Z} = \left[ \frac{1}{G + i\omega C} + R + i\omega L \right]^{-1} \quad (1)$$

where:

$$G_{\text{meas}} = \frac{G + R(G^2 + \omega^2 C^2)}{(1 - \omega^2 LC + GR)^2 + (\omega LG + \omega CR)^2}$$

$$C_{\text{meas}} = \frac{C - L(G^2 + \omega^2 C^2)}{(1 - \omega^2 LC + GR)^2 + (\omega LG + \omega CR)^2} \quad (2)$$

in measuring device resonance state in inductance is expressed by the equation  $\omega^2 LC = 1$ . This resonance state is easily observed at high frequency range in Fig. 1, 2. It should be mentioned that,  $\omega = 1/\sqrt{LC}$  resonance frequency depends on the material type which is used in measurement [37]. If the measurements are carried out at the frequency range where resonance can be obtained, then the inductance of the system becomes practically zero ( $L = 0$ ). In this case the parameters of the Eq. (1) can be given as follows [37]:

$$G_{\text{meas}} = \frac{G + R(G^2 + \omega^2 C^2)}{(1 + GR)^2 + (\omega CR)^2}$$

$$C_{\text{meas}} = \frac{C}{(1 + GR)^2 + (\omega CR)^2} \quad (3)$$

The results obtained in quite small resistances of measuring device are explained by dielectric effect in the frequency  $\omega = \sqrt{2/CR}$  by the relaxation of artificial Debye. Although this relaxation frequency is usually very high, in this approach it is not right (in available experiments the Eq. (3) can be used for some Ohm resistance). However, in measuring chamber the current frequency of high-capacity samples can be reduced. Using from the literature, [38,39].

$$\begin{aligned}
 \sigma &= \varepsilon_0 \omega \varepsilon'' \\
 \sigma' &= \sigma \cos \varphi \\
 \sigma'' &= \sigma \sin \varphi
 \end{aligned}
 \tag{4}$$

known expressions, the permittivity obtained from experiment and  $f(\sigma') = f(\sigma'')$  dependencies relevant to the values obtained from the frequency which is used within the measurements, have been depicted in Fig. 3. When considering interdependence of the real and imaginary parts of electrical conductivity at various temperature values before and after the irradiation, we can see the chaotization more obviously at 100 K temperature (Fig. 3).

These dependencies also confirm the facts depicted in Figs. 1 and 2: decrease of the current chaotization with temperature increase and its fully recombination at 400 K. From interdependence of real and imaginary parts of electrical conductivity it is observed that the real part of the electrical conductivity changes more than the imaginary part due to neutron flux influence. In Fig. 3 it is observed a semicircle-like case at 400 K temperature, and it can be accepted as a part of Cole-Cole diagram [21]. In this case, approximately the values  $10^8 - 10^{10}$  Hz of frequency correspond to the peak of imaginary extension of the semicircle. Interdependence of real and imaginary parts of electrical conductivity, it has been found out the conductivity is atomic or dipolar ion type.

#### 4. Conclusions

In the result of comparative analysis of frequency dependencies of electrical conductivity of the samples affected by neu-

tron flux with different periods and initial state, it has been established that, clusters are formed inside the samples under neutron flux influence at 100 K temperature. As a result of the generated clusters it is observed a sharp chaotization at this temperature. At the 200 - 300 K temperatures the clusters are reduced under the influence of temperature and frequency and fully recombined at 400 K. From comparative analysis of frequency dependencies at various temperatures it has been revealed that the value of electrical conductivity increases up to 20 times due to neutron flux influence. From interdependence of real and imaginary parts of electrical conductivity it has been found out those real parts of electrical conductivity has changed more than the imaginary parts after neutron irradiation. The conductivity to be atomic or dipolar ion type, has been found out from interdependence of real and imaginary parts of electrical conductivity.

#### Acknowledgments

The work has been conducted following the agreement signed between the Institute of Radiation Problems (IRP) of Azerbaijan National Academy of Sciences (ANAS), and Institute Jožef Stefan (IJS), Slovenia. We gratefully acknowledge the assistance of our colleagues from National Nuclear Research Center (NNRC), IRP of ANAS, "Reactor Infrastructure Centre" and "Condensed Matter Physics Department" at IJS. We would like to thank Asst. Prof. Dr. Luka Snoj and Anže Jazbec for providing irradiated samples in TRIGA Mark II type research reactor and Assoc. Prof. Dr. Vid Bobnar and Dr. Andreja Eršte for encouraging discussions.

1. Fang Cao *et al.*, *Solar Energy Materials and Solar Cells* **141** (2015) 132-138.
2. Ashok Kumar Kherodia, Ashish K. Panchal, *Energy Procedia* **75** (2015) 2193-2198.
3. Gumjae Park *et al.* *Journal of Alloys and Compounds* **639** (2015) 296-300.
4. N.S. Bennett *et al.*, *Nano Energy* **16** (2015) 350-356.
5. L. Boodhoo *et al.* *Microelectronic Engineering* **145** (2015) 66-70.
6. ShameelFarhan, Rumin Wang, Hao Jiang, *Materials Letters* **159** (2015) 439-442.
7. K.E. SarathRaghavendraBabu, MuthukannanDuraiselvam, *Applied Surface Science* **353** (2015) 1112-1116.
8. JinyoungJeong *et al.*, *Carbon* **94** (2015) 120-123.
9. S. Azimi, Z.Y. Dang, M.B.H. Breese, *Microelectronic Engineering* **121** (2014) 156-161.
10. Jin Nyoung Jang, Dong Hyeok Lee, MunPyo Hong, *Current Applied Physics* **14** (2014) 901-904.
11. Y. Satoh *et al.*, *Journal of Nuclear Materials* **442** (2013) S768-S772.
12. M.L. Gamez *et al.*, *Journal of Nuclear Materials* **367-370** (2007) 282-285.
13. R. Chakarova, I. Pazsit, *Nucl. Instrum. and Meth. B* **164-165** (2000) 460-470.
14. J.G. Mihaychuk, N. Shamir, and H.M. van Driel, *Physical Review B* **59** (1999) 2164-2173.
15. C. Richard and Dieter M. Kolb, *Advances in Electrochemical Science and Engineering*, Vol. **8**, Book Printed in the Federal Republic of Germany (2003)
16. H.J. Stein, *Journal of Applied Physics* **38** (1967) 204-210.
17. M.Alurralde, *Journal of Applied Physics* **95** (2004) 3391-3396.
18. Moussab Harb, Pierre Labéguerie, Isabelle Baraille, and Michel Rérat, *Physical Review B* **80** (2009) 1-7.
19. Elchin Huseynov, Adil Garibov, Ravan Mehdiyeva, *Physica B: Condensed Matter* **450** (2014) 77-83.
20. Elchin Huseynov, Adil Garibov, Ravan Mehdiyeva, *Journal of Electrostatics* **74** (2015) 73-78.
21. Elchin Huseynov, Adil Garibov, Ravan Mehdiyeva, Eršte Andreja, Anar Rustamov, *AIP Advances* **4** (2014) 117122.

22. Elchin Huseynov, Adil Garibov, and Ravan Mehdiyeva, *International Journal of Modern Physics B* **28** (2014) 1450213.
23. Elchin Huseynov, Adil Garibov and Ravan Mehdiyeva, *Nano Convergence* **1** (2014) 21.
24. Elchin Huseynov, Adil Garibov, Ravan Mehdiyeva, Efsane Huseynova, *Modern Physics Letters B* **30** (2016) 1650115.
25. Elchin Huseynov and Aydan Garibli, *Effect of Neutron Flux on the Frequency Dependence of Permittivity of Nano Silicon Particles* (Silicon, 2016).
26. Elchin Huseynova, Adil Garibova, Ravan Mehdiyeva, *Journal of Materials Research and Technology* (2016).
27. Luka Snoj, Gasper Zerovnik, Andrej Trkov, *Applied Radiation and Isotopes* **70** (2012) 483-488.
28. Gasper Zerovnik, Manca Podvratnik, Luka Snoj, *Ann. Nucl. Energy* **63** (2014) 126-128.
29. G. Žerovnik, *et al.*, *Applied Radiation and Isotopes* **96** (2015) 27-35.
30. R. Henry, I. Tiselj, L. Snoj, *Applied Radiation and Isotopes* **97** (2015) 140-148.
31. Luka Snoj, Andrej Kavcic, Gasper Zerovnik, Matjaz Ravnik, *Ann. Nucl. Energy*, **37** (2010) 223-229.
32. P. Filliatre *et al.*, *Annals of Nuclear Energy* **83** (2015) 236-245.
33. G. Žerovnik, *et al.*, *IEEE Transactions on Nuclear Science*, **61** (2014) 2527-2531
34. A. Kolšek, V. Radulović, A. Trkov, L. Snoj, *Nuclear Engineering and Design*, **283** (2015) 155-161.
35. Andreja Eršte, *Investigations and separation of various contributions to dielectric response of advanced ceramic and polymeric materials* (Jozef Stefan Institute, Ljubljana, Slovenia, 2012)
36. Kremer Friedrich, Schönhals Andreas, *Broadband Dielectric Spectroscopy* Springer, Berlin, (2003) p. 729.
37. Andreja Eršte, Barbara Malič, Brigita Kužnik, Marija Kosec and Vid Bobnar, *Advances and applications in electroceramics II: Ceramic transactions* **235** (2012) 23-29.
38. A.K. Himanshu, D.C. Gupta, and T.P. Sinha, *Indian Journal of Pure & Applied Physics (IJPAP)*, **44** (2006) 391-397.
39. LeeSlater, *Surveys in Geophysics* **28** (2007) 169-197.