Influence of Neutron Flux on Frequency Dependence of Dielectric Properties of Nano SiO$_2$

Elchin Huseynov, Adil Garibov, Ravan Mehdiyeva

Institute of Radiation Problems of Azerbaijan National Academy of Sciences
AZ 1143, B.Vahabzadeh 9, Baku, Azerbaijan

Abstract: At the present work nano SiO$_2$ has been irradiated by $2 \times 10^{13}$ cm$^{-2}$s$^{-1}$ neutron flux at different times up to 20 hours. It has been comparatively analyzed the frequency dependencies of dielectric constant of the nanomaterial exposed to neutron flux influence and initial state. From analysis results it has been revealed that the dielectric constant of nano SiO$_2$ increases in general tendency with influence of neutron flux. The complex dependence of the real and imaginary parts of dielectric constant of nano silica particles has been reviewed at 100 K and 400K. Similar Cole-Cole diagrams existing in the dependencies it has been revealed that the value of the relaxation period is compatible with polarization of the nano silica.

Keywords: nanomaterial, nano SiO$_2$, neutron effect, dielectric property, permittivity
PACS: 61.80.Hg, 61.80.-x, 77.22.Ch

1. Introduction

Nano-size SiO$_2$ and its different-type mixtures have been in the focus point of world researchers in recent years [1-11]. Also, silica and its oxide compounds are widely applied as a sorbent and radiation-resistant material in electronics and detection of ionizing rays [12-18]. An oxide layer usually with nano size is formed on silica materials in application fields. Surface oxide layer protects silica from external influences, as well as affects its physical and surface physicochemical properties. On the other side SiO$_2$ in nano sizes has a wide application field in medicine and technology for its simple composition, easy obtain and also as an oxide dielectric resistant to external influences and as a material with different purpose. For these properties, silicon oxide is of great importance in space techniques and nuclear technology as well. Recently it has been defined the dependence of physical and surface physicochemical properties of oxide dielectrics on particle size, particularly, strong influence of volume electron excitation in nano size, of defects and other factors on surface. Thus, nano-size samples of classic oxide dielectric SiO$_2$ are again in the focus of attention of researchers in modern times. For this purpose, ionizing ray effect on physical and surface physicochemical properties of nano SiO$_2$ is studied and suggestions are prepared on application of the samples in different fields [1-18].

The defects being generated during the irradiation of nano SiO$_2$ by neutron flux create important changes in dielectric properties. The submitted article is devoted to the study of neutron effect on real and imaginary parts of dielectric constant (permittivity) of nano SiO$_2$. It has been determined that the dielectric constant of neutron-irradiated nano SiO$_2$ on irradiation period and frequency at different temperature changing field. The defects generated in nano-compound exposed to continuous neutron irradiation at various periods cause changes in dielectric properties of the sample. Thus, with increase of effect period of neutron flux, the numerical values of real and imaginary parts of dielectric constant change and in its turn this change is more in real part of dielectric constant than in imaginary one. As a main reason for the change in the work it has been mentioned the generation of extra charges in the sample under neutron flux influence. The generated extra charges change the system polarization and thus real and imaginary parts of permittivity. In the work it has also been observed a decrease in real and imaginary parts of permittivity in the samples with increase of external field frequency. This has been explained as a destruction of charges with different barrier energy in the system with frequency effect and thus, as a decrease of polarization.

2. Experiment

From previous studies it is known that the specific surface area of nano material used in the experiment is 160m$^2$/g, dimensions are 20nm and purity is 99.5% and some parameters of the used sample has been studied [19-23, parameters of the used sample has been studied [19-23, manufacturing company: SkySpring Nanomaterials, Inc. Houston, USA]. Nano SiO$_2$ has been irradiated at full power mode (250 kW) by neutron flux with $2 \times 10^{13}$ cm$^{-2}$s$^{-1}$ flux density in central channel (channel A1) at TRIGA Mark II light water pool type research reactor in “Reactor Centre’’ of Jozef Stefan Institute (JSI) in the city of Ljubljana of Slovenia. It is important to note that the JSI TRIGA reactor has been thoroughly characterized [24-29] and the computational model used for computational characterization has been thoroughly verified and validated [30,31] against several experiments. While working at full power mode, the neutron flux has the following composition parts [24,26]: $5.107 \times 10^{12}$ cm$^{-2}$s$^{-1}$ (1±0.0008, $E_n < 625$eV) for thermal neutrons, for epithermal neutrons – as $6.502 \times 10^{12}$ cm$^{-2}$s$^{-1}$ (1±0.0008, $E_n \approx 625$eV), for fast neutrons - 7.585x$10^{12}$ cm$^{-2}$s$^{-1}$ (1±0.0007, $E_n > 0.1$ MeV) and finally for all the neutrons in central channel flux density is as 1.920x$10^{13}$ cm$^{-2}$s$^{-1}$ (1±0.0005).

Dielectric parameters of neutron-irradiated nano SiO$_2$ have been measured in “Novocontrol Alpha High Resolution Dielectric Analyzer” device at 0.09Hz – 2.3MHz range of
frequency and 100K – 400K temperature range in the laboratory of “Condensed Matter Physics F5” at JSI. Determination of characteristic parameters of neutron-irradiation and neutron flux has been conducted with the methodology known from literature. Nano SiO$_2$ powder being pressed at 7kN/cm$^2$ pressure in special conditions in the laboratory of “Thin Films and Surfaces F3” at JSI, was made in the form of tablet with 550 μm height and 5.5mm diameter and then placed in aluminium container appropriate to the channels of the reactor. The prepared samples have been continuously irradiated at central channel for 5, 10, 15 and 20 hours. Activity of the samples has increased up to 1.5GBq under neutron flux influence. Therefore all the measurements have been carried out approximately 200 hours after neutron flux influence. After irradiation silver contacts have been fixed on the surface of samples in special condition and examined its quality. It has been used Cr/Au electrodes obtained on the top layer by spray method. Then the obtained samples have been placed in between two platinum plates and measurements taken. Permittivity of the samples has been measured in “Novocontrol Alpha High Resolution Dielectric Analyzer” device at alternative current (AC ~0,5V) at 100-400 K temperature range. During the measurements storage accuracy of temperature in any degree was up to 0,01K and the accuracy was obtained with the method of bridge. From the experiments it has been measured the capacity and resistance of the samples and taking into account the known parameters of the samples, the permittivity has been calculated. All of the results obtained compatible with calculated values have been graphically depicted in “OriginPro 9.0” program.

3. Result and Discussions

At present work, within the measurements the frequency dependencies of dielectric properties of the samples have been reviewed at 4 different constant values of temperature. The experiments have been carried out within the range of 0,09 – 2260000 Hz at 95 different constant values of frequency and during the measurements it has been revealed, that at various values of temperature the frequency dependency of dielectric constant are different. It has been reviewed the frequency dependence of the dielectric constant at 100K, 200K, 300K and 400K constant values of temperature. Firstly it has been reviewed frequency dependencies of dielectric constant at 100K and 200K temperature ranges (Fig. 1). At 100K value of temperature from $f(ε) = f(ν)$ dependency it is obviously observed an increase of dielectric constant with increase of influence period of neutron flux. The dielectric constant of the initial sample to be minimum gives ground to say that the polarization in this case also is minimum. Under neutron flux influence extra charge carriers appear in the samples, so they cause extra general polarization in the samples.

Figure 1: Dielectric constant and frequency dependencies of silica nanoparticles at 100K and 200K temperatures (0h – before irradiation, 5h, 10h, 15h and 20h – after irradiation).

In the result of these charge carriers the dielectric constant increases. The similar process occurs at 200K value of temperature. In this temperature decrease of dielectric constant with frequency increase is the result of combined effects of temperature and frequency [19]. At 100K temperature the choation existing in frequency dependence of the electric conductivity is maybe due to the clusters formed in the system at this temperature [19, 32-40]. At the same time that choationization is observed in a certain degree at 200K value of temperature as well [19].

On the other side the choationization to be more at low frequency range is due to the nanomaterial to be more inclined to polarization at this range [19]. At relatively high temperatures it is observed more obviously especially in the samples exposed to neutron flux influence (Fig. 2). As it seem from figure, at 300K and 400K values of temperature the dielectric constant gets its maximum values at low frequencies. This maximum is observed more sharply in the samples exposed to neutron flux influence at 300K temperature. The obtained results can be accepted as formation of nanoparticles in dipole state at low frequencies [19, 41].
At 100K value of temperature the fact of increase of the dielectric constant with increase of neutron flux is confirmed once again from complex dependence of the real and imaginary parts of permittivity at 100 K and 400 K (Fig. 3, c.s. – before irradiation, 5h, 10h, 15h and 20h – after irradiation). On the other hand it is observed oscillation of dielectric constant around zero value of imaginary part. At low temperatures the clusters being formed inside the sample decompose under frequency influence and cause the device to detect negative resistance [19]. This state has been observed also during analysing the electric conductivity of the samples and explained by similar cluster model [19, 32-40]. At 400K temperature the existing clusters are destructed under the influence of frequency and heat and there are no negative cases in imaginary part of permittivity. At this temperature at the range of about 0.15 – 0.25 values of imaginary part of permittivity it is observed the cases similar to “Cole-Cole” diagram at average frequency range, and it indicates to the existence of polarization which can be relaxed at that range. In this case the calculated relaxation period decreases directly proportional to irradiation period and the values are compatible with the polarization of nanoparticles.

It can be argued that extra charge carriers are formed in the system with irradiation influence and it reduces the relaxation period. At 400 K temperature in each curve it is observed two states similar to “Cole-Cole” diagram. It allows to say that at this temperature there may be two types of relaxors different for their stationar states [19]. From the dependencies of real and imaginary parts of dielectric constant on frequency on a same coordinate system, it is obviously seen that at 100 temperature the real part is several times more than the imaginary part (Fig. 4). On the other hand though there is no considerable change in the imaginary part of dielectric constant in the samples exposed to neutron flux influence for 20 hours, the real part of dielectric constant has increased approximately 2 times.
Also, at 100K value of temperature the real and imaginary parts of dielectric constant almost are not dependent on the frequency. At 400K temperature at low frequencies the imaginary part of dielectric constant is larger than the real one, but in contrast at high frequencies it is less. It is usually taken as dancing presence of charge carriers, accumulation of charges and charging-discharging current inside the sample [42,43]. In this case the existence of dancing-proton conductivity inside nano SiO2 is expected [42, 43].

Also, at 100K value of temperature the dielectric constant increases with increase of neutron flux influence period regardless of frequency. It has been revealed that the clusters formed at 100K temperature induce the device to show dielectric loss as negative value. At other temperatures (400K) from the cases similar to Cole-Cole diagrams it has been revealed that the value of relaxation period is compatible with the polarization of nano particles.

4. Conclusion

In the result of the conducted researches it has been revealed that under neutron flux influence the dielectric constant of nano silica increases in general tendency. At 100K value of temperature the dielectric constant increases with increase of neutron flux influence period regardless of frequency. It has been revealed that the dielectric constant depend on frequency inversely proportionally at relatively high temperature (400K). After neutron flux influence the increase in dielectric constant is more severe at low frequencies. From complex dependences of real and imaginary parts of dielectric constant it has been revealed that the clusters formed at 100K temperature induce the device to show dielectric loss as negative value. At other temperatures (400K) from the cases similar to Cole-Cole diagrams it has been revealed that the value of relaxation period is compatible with the polarization of nano particles.

5. Acknowledgements

The work has been carried out on the base of agreement signed between the Institute of Radiation problems Azerbaijan National Academy of Sciences (ANAS) and Jožef Stefan Institute, Slovenia. We express our gratitude to colleagues from the Institute of Radiation Problems of ANAS and "Reactor Infrastructure Centre (RIC)" and "Condensed Matter Physics Department" at Jožef Stefan Institute for assisting us with implementation of the work. We wish to thank Dr. Luka Snoj and Anže Jazbec for irradiated samples in TRIGA Mark II type research reactor and Asst. Prof. Vid Bobnar and Andreja Eršte for all the fruitful discussions us.

Reference


**Author Profile**

Elchin Huseynov received the B.S. and M.S. degrees in Physics and Quantum electronics from Baku State University in 2007 and 2010, respectively. During 2010–2011, he was junior researcher at Republic Nuclear Security and Radiation Safety Examination Center of Institute of Radiation Problems of Azerbaijan National Academy of Sciences. At present he's PhD student and young scientist at Physics of Nuclear Reactors Group of Azerbaijan National Academy of Sciences.