

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 SiO₂ powder being pressed at 7kN/cm² 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 $f(\epsilon) = f(\nu)$ 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.

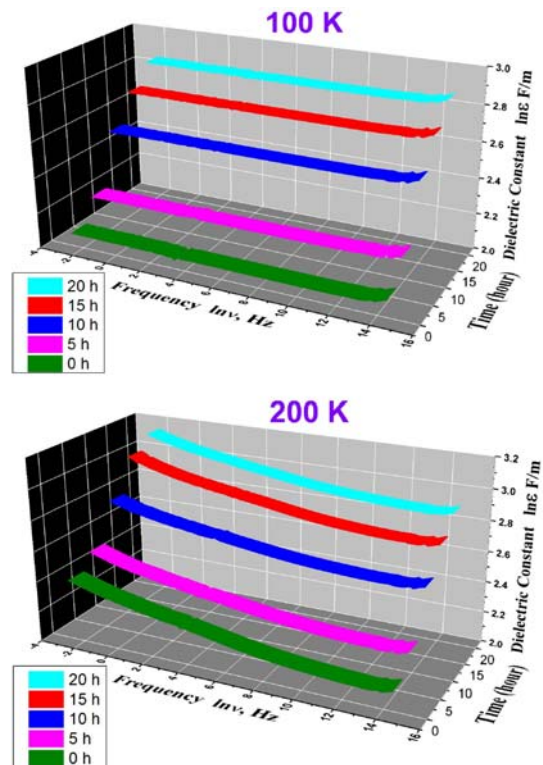


Figure 1: Dielectric constant and frequency dependencies of silica nanoparticles at 100K and 200K temperatures (0h – before irradiation, 5h, 10h, 15h and 20h – after irradiation).

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 chaoticization 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 chaoticization is observed in a certain degree at 200K value of temperature as well [19].

On the other side the chaoticization 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].

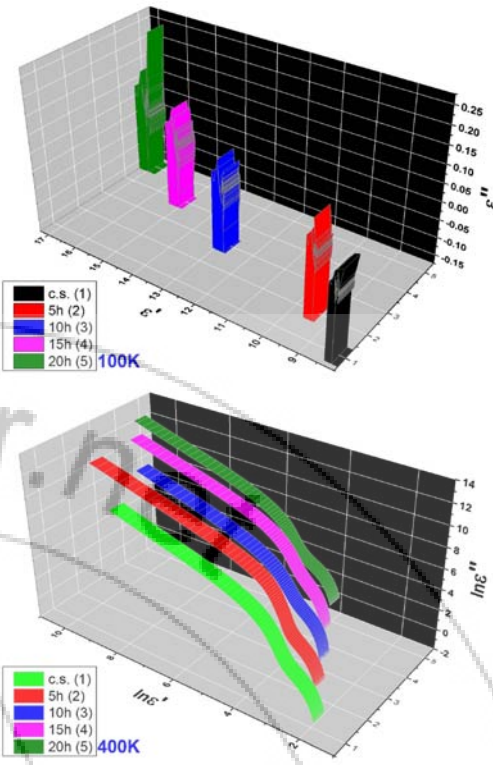
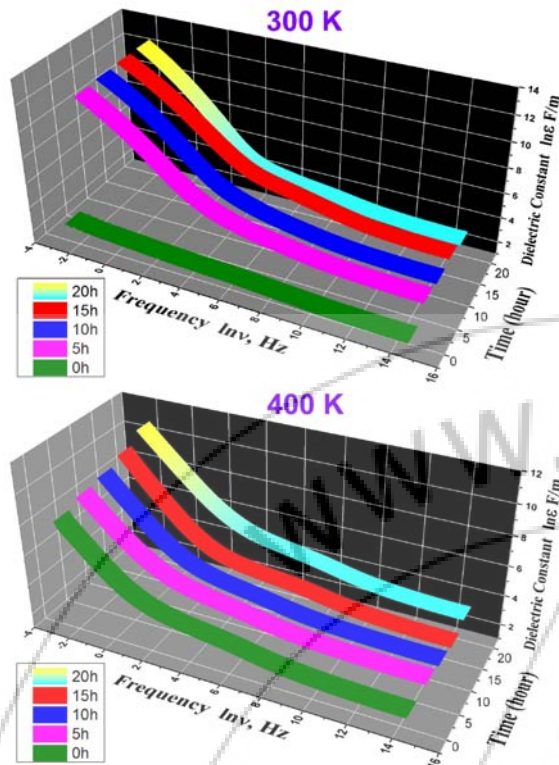


Figure 2: Dielectric constant and frequency dependencies of silica nanoparticles at 100K and 200K temperatures (0h – before irradiation, 5h, 10h, 15h and 20h – after irradiation).

Figure 3: Mutual dependence of dielectric constant of silica nanoparticles

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 nano particles.

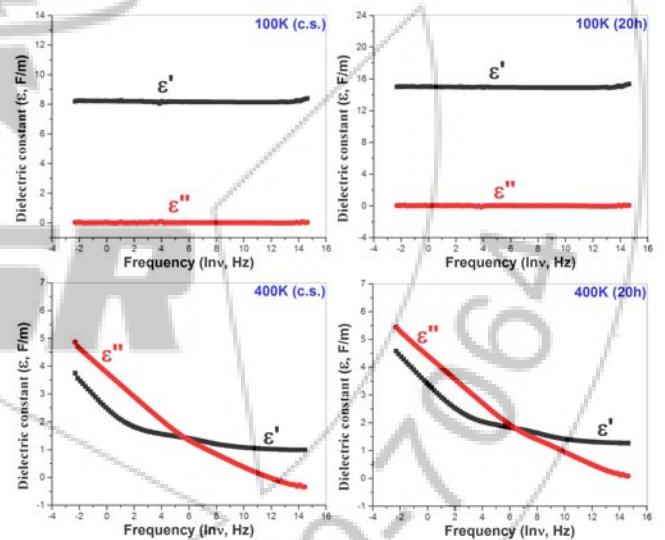


Figure 4: Real and imaginary parts of dielectric constant and frequency dependencies of silica nanoparticles.

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 stationary 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]. It has been revealed from the conducted analyses that the numerical values of the real and imaginary parts of dielectric constant increase with temperature increase.

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 of nanoparticles 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 Jozef 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

- [1] Jason M. Larkin and Alan J. H. McGaughey "Thermal conductivity accumulation in amorphous silica and amorphous silicon" *Phys. Rev. B* 89, 144303 (2014)
- [2] D. Farnesi, A. Barucci, G. C. Righini, S. Berneschi, S. Soria, and G. Nunzi Conti "Optical Frequency Conversion in Silica-Whispering-Gallery-Mode Microspherical Resonators" *Phys. Rev. Lett.* 112, 093901 (2014)
- [3] Dagmara Sokolowska, Daniel Dziob, Urszula Gorska, Bartosz Kieltyka and Jozef K. Moscicki "Electric conductivity percolation in naturally dehydrating, lightly wetted, hydrophilic fumed silica powder" *Phys. Rev. E* 87, 062404 (2013)
- [4] O. Morandi, P.-A. Hervieux and G. Manfredi "Bose-Einstein condensation of positronium in silica pores" *Phys. Rev. A* 89, 033609 (2014)
- [5] Yong-Ho Kim, Byunghwan Lee, Kwang-Ho Choo, Sang-June Choi "Adsorption characteristics of phenolic and amino organic compounds on nano-structured silicas functionalized with phenyl groups" *Microporous and Mesoporous Materials* 185, 121–129 (2014)
- [6] John Breuer, Roswitha Graf, Alexander Apolonski and Peter Hommelhoff "Dielectric laser acceleration of nonrelativistic electrons at a single fused silica grating structure: Experimental part" *Phys. Rev. ST Accel. Beams* 17, 021301 (2014)
- [7] Adarsh Shekhar, Ken-ichi Nomura, Rajiv K. Kalia, Aiichiro Nakano and Priya Vashishta "Nanobubble Collapse on a Silica Surface in Water: Billion-Atom Reactive Molecular Dynamics Simulations" *Phys. Rev. Lett.* 111, 184503 (2013)
- [8] Mark Wilson, Avishek Kumar, David Sherrington and M. F. Thorpe "Modeling vitreous silica bilayers" *Phys. Rev. B* 87, 214108 (2013)
- [9] Hugo Bergeron, Jean-Raphaël Carrier, Vincent Michaud-Belleau, Julien Roy, Jérôme Genest, and Claudine Ni. Allen "Optical impulse response of silica microspheres: Complementary approach to whispering-gallery-mode analysis" *Phys. Rev. A* 87, 063835 (2013)
- [10] T. Glomann, G. J. Schneider, J. Allgaier, A. Radulescu, W. Lohstroh, B. Farago and D. Richter "Microscopic Dynamics of Polyethylene Glycol Chains Interacting with Silica Nanoparticles" *Phys. Rev. Lett.* 110, 178001 (2013)
- [11] Indresh Yadav, Sugam Kumar, V. K. Aswal and J. Kohlbrecher "Small-angle neutron scattering study of differences in phase behavior of silica nanoparticles in the presence of lysozyme and bovine serum albumin proteins" *Phys. Rev. E* 89, 032304 (2014)
- [12] Joseph Chamieh, Yvan Zimmermann, Anne Boos, Agnès Hagège "A simple cladding process to apply monolithic silica rods in high performance liquid chromatography" *Journal of Chromatography A* 1217, Issue 45, 7172–7176 (2010)
- [13] A.T.Abdul Rahman, R.P.Hugtenburg, Siti Fairus Abdul Sani, A.I.M.Alalawi, Fatma Issa, R.Thomas, M.A.Barry, A.Nisbet, D.A.Bradley "An investigation of the thermoluminescence of Ge-doped SiO₂ optical fibres for application in interface radiation dosimetry" *Applied Radiation and Isotopes* 70, 1436–1441 (2012)
- [14] S.M.Jafari, D.A.Bradley, C.A.Gouldstone, P.H.G.Sharpe, A.Alalawi, T.J.Jordan, C.H.Clark, A.Nisbet, N.M.Spyrou "Low-cost commercial glass beads as dosimeters in radiotherapy" *Radiation Physics and Chemistry* 97, 95–101 (2014)
- [15] B.Brighard, A.L.Tomashukb, H.Oomsa, V.A.Bogatyrvov, S.N.Klyamkin, A.F.Fernandez, F.Berghmansa, M.Decreton "Radiation assessment of hydrogen-loaded aluminium-coated pure silica core fibres for ITER plasma diagnostic applications" *Fusion Engineering and Design* 82, 2451–2455 (2007)
- [16] D.A.Bradley, R.P.Hugtenburg, A.Nisbet, Ahmad Taufek Abdul Rahman, FatmaIssa, Noramaliza Mohd Noor, Amani Alalawi "Review of doped silica glass optical fibre: Their TL properties and potential applications in radiation therapy dosimetry" *Applied Radiation and Isotopes* 71, 2–11 (2012)
- [17] P.Miiller, M.Schvoerer, R.Berger and N.Jacquet-Francillon "Coupled thermostimulated luminescence/electron spin resonance study in pure vitreous silica: application to alpha

- radiation damage study" *Journal of Non-Crystalline Solids* 159, 154-161 (1993)
- [18] Yadan Ding, Xueying Chu, Xia Hong, Peng Zou and Yichun Liu "The infrared fingerprint signals of silica nanoparticles and its application in immunoassay" *Applied Physics Letters* 100, 013701 (2012)
- [19] Elchin Huseynov, Adil Garibov, Ravan Mehdiyeva "Temperature and frequency dependence of electric conductivity in nano-grained SiO₂ exposed to neutron irradiation" *Physica B: Condensed Matter*, 450, 77–83 (2014)
- [20] Elchin Huseynov, Adil Garibov and Ravan Mehdiyeva "Study of blend composition of nano silica under the influence of neutron flux" *Nano Convergence* 2014 1:21
- [21] E.M.Huseynov, A.A.Garibov, R.N.Mehdiyeva "Calculation of the specific surface area of SiO₂ nano powder and getting nano-SiO₂-H₂O systems" *Azerbaijan Journal of Physics* ISSN 1028-8546, Volume XIX, Number 1, p. 10-14, Azerbaijan 2013
- [22] E.M. Huseynov, A.A.Garibov, R.N.Mehdiyeva, "Synthesis methods of nano SiO₂ powder" *Transactions of National Academy of Sciences of Azerbaijan, Series of Physics – Mathematical and Technical Sciences, Physics and Astronomy*, ISSN 0002-3108 Vol. XXXII N5, p 83-88/152, Azerbaijan 2012
- [23] E.M.Huseynov, A.A.Garibov, R.N.Mechdiyeva "Temperature and frequency dependences of dielectric properties of nano SiO₂ compound" *Journal of Qafqaz University – Physics*, Volume 1, Number 2, Pages 191-199, 2013
- [24] Luka Snoj, Gasper Zerovnik, Andrej Trkov "Computational analysis of irradiation facilities at the JSI TRIGA reactor", *Applied Radiation and Isotopes*, 70 (2012) 483–488
- [25] L. Snoj, A. Trkov, R. Jačimović, P. Rogan, G. Žerovnik, M. Ravnik, "Analysis of neutron flux distribution for the validation of computational methods for the optimization of research reactor utilization", *Appl. Radiat. Isotopes*, Vol. 69, pp. 136-141, 2011
- [26] L. Snoj et al. "Calculation of kinetic parameters for mixed TRIGA cores with Monte Carlo", *Ann. Nucl. Energy*, 37 (2) (2010), pp. 223–229
- [27] L. Snoj et al., "Testing of cross section libraries on zirconium benchmarks", *Ann. Nucl. Energy*, 42 (2012) 71–79
- [28] Luka Snoj, Matjaž Ravnik, "Power peakings in mixed TRIGA cores" *Nuclear Engineering and Design*, Volume 238, Issue 9 (2008) 2473-2479
- [29] Jazbec Anže, Žerovnik Gašper, Snoj Luka, Trkov Andrej, "Analysis of tritium production in TRIGA Mark II reactor at JSI for the needs of fusion research reactors" *Atw. Internationale Zeitschrift für Kernenergie*, iss. 12, vol. 58 (2013) 701-705
- [30] Vladimir Radulović, Žiga Štancar, Luka Snoj, Andrej Trkov, "Validation of absolute axial neutron flux distribution calculations with MCNP with ¹⁹⁷Au(n,γ)¹⁹⁸Au reaction rate distribution measurements at the JSI TRIGA Mark II reactor" *Applied Radiation and Isotopes*, Volume 84 (2014) 57-65
- [31] G. Žerovnik et al. On normalization of fluxes and reaction rates in MCNP criticality calculations, *Ann. Nucl. Energy*, 63 (2014) 126–128
- [32] Shiyun Xiong, Kaike Yang, Yuriy A. Kosevich, Yann Chalopin, Roberto D'Agosta, Pietro Cortona and Sebastian Volz "Classical to Quantum Transition of Heat Transfer between Two Silica Clusters" *Phys. Rev. Lett.* 112, 114301 (2014)
- [33] Y.Satoh, Y.Matsuda, T.Yoshiie, M.Kawai, H.Matsumura, H.Iwase, H.Abe, S.W.Kim, T.Matsunaga "Defect clusters formed from large collision cascades in fcc metals irradiated with spallation neutrons" *Journal of Nuclear Materials* 442, S768–S772 (2013)
- [34] M.L.Gamez, M.Velarde, F.Mota, J.Manuel Perlado, M.Leon, A.Ibarra "PKA energy spectra and primary damage identification in amorphous silica under different neutron energy spectra" *Journal of Nuclear Materials* 367–370, pp. 282–285 (2007)
- [35] R.Chakarova, I.Pazsit "Fluctuations and correlations in sputtering and defect generation in collision cascades in Si" *Nucl. Instrum. and Meth. B* 164&165, 460 – 470 (2000)
- [36] Moussab Harb, Pierre Labéguerie, Isabelle Baraille, and Michel Rérat "Response of low quartz SiO₂ to the presence of an external static electric field: A density functional theory study" *Physical Review B* 80, 235131 (2009)
- [37] Harry J.Whitlow, Sachiko T.Nakagawa "Low-energy primary knock on atom damage distributions near MeV proton beams focused to nanometre dimensions" *Nuclear Instruments and Methods in Physics Research B* 260 (2007) 468–473 (2007)
- [38] Q.Xu, T.Yoshiie, H.Watanabe, N.Yoshida "Effects of oversized element Sn on diffusion of interstitial clusters in Ni irradiated by ions and neutrons" *Journal of Nuclear Materials* 367–370 (2007) 361–367 (2007)
- [39] H.J.Stein "Energy Dependence of Neutron Damage in Silicon" *Journal of Applied Physics*, Volume 38, N1, p.204-210 (1967)
- [40] Sugam Kumar, M.-J. Lee, V. K. Aswal, and S.-M. Choi "Block-copolymer-induced long-range depletion interaction and clustering of silica nanoparticles in aqueous solution" *Phys. Rev. E* 87, 042315 (2013)
- [41] P.R. Rejikumar, P.V.Jyothy, Siby Mathew, Vinoy Thomas, N.V.Unnikrishnan "Effect of silver nanoparticles on the dielectric properties of holmium doped silica glass" *Physica B: Condensed Matter* 405, Issue 6, 1513–1517 (2010)
- [42] E.Axelrod, A.Givant, J.Shappir, Y.Feldman and A.Sa'ar "Dielectric relaxation and transport in porous silicon" *Phys. Rev. B* 65, 165429 (2002)
- [43] Dagmara Sokolowska, Daniel Dziob, Urszula Gorska, Bartosz Kieltyka, and Jozef K. Moseicki "Electric conductivity percolation in naturally dehydrating, lightly wetted, hydrophilic fumed silica powder" *Phys. Rev. E* 87, 062404 (2013)

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