

Electrical Properties of Nano-Si/U₃O₈ Composites

Ramazanov MA^{1*}, Maharramov AM¹, Nuriyev MA² and Garibli AA¹

¹Department of Chemical Physics of Nanomaterials, Baku State University, Azerbaijan

²Department of Chemistry, ANAS Institute of Radiation Problems, Azerbaijan

Abstract

Have been studied the electrical properties of Nano-Si/U₃O₈ composites. Was found the change of electrical conductivity of Nano-Si/U₃O₈ composite properties mostly occurs via mechanism of spasmodic conductivity. The analysis of experiment results of dependency of electro physical properties of Nano-Si/U₃O₈ composite from frequency shows that the changes occur due to polarization of components on interphase border. In such kind materials the dielectric relaxation can be determined by exact impedance of ratio of real and imaginary content.

Keywords: Radio nucleotides; Uranium oxides; Nano composites; Polarization; Nanosized silica

Introduction

Radio nucleotides are widely applied in the field of nuclear energetics, and, generally uranium, plutonium oxides, thorium and other radioactive elements as well as high temperature resistant oxide mixtures are used as fuel. The energy of nuclear reaction and power capacity of reactor can be managed by variation of quantity of components in the composite mixture [1-4]. So, the study of the physical processes occurring in composite systems nano-Si/U₃O₈ is very important problem. The scientific researches in this direction are not new, the composites on the basis of oxides are widely used and this fact is proved with a lot of data in scientific literature [5-7]. The compounds on the basis of silica and uranium oxides can lead to preparation of new materials that would have practical and scientific importance in term of radiation material crafts. There are few literature data about composite materials on the basis of uranium and other metal oxides. We report about synthesis and properties of composite material with uranium oxide (U₃O₈) and nano sized silica (nano-Si) [8]. For the first time for synthesis and investigation of such kind composite materials was used nanosized silica. We supposed that interaction of uranium oxide and nano-Si would increase due to surface activity of nano-Si and this fact contributes to improving of its exploitation parameters. From the other side it is known nano-Si/U₃O₈ is a heterogeneous system and characterized by chaotic distribution of components and Maxwell-Wagner polarization [9]. It brings to forming of conductive channels, connecting opposite sites at definite content of filler, and as sequences it causes the percolation phase transfer. These factors can serve to development of new materials with controllable properties based on dependence of composite physical properties from concentration and applied electrical field's frequency.

There is no doubt that this approach is actual in terms of preparation and investigation of physical electrical properties of composite material on the basis of uranium oxide (U₃O₈) and nano sized silica (nano-Si). The investigation of electro physical properties of these materials as well is very important in term of forecast of its exploitation capacity and evaluation of its quality in nuclear reactors.

Materials and Methods

The object of investigation is nano-Si/U₃O₈ system composed of Silicon Nanoparticles Cubic ~100 nm sized and ~50 mkm U₃O₈ powders. Taken in various ratio the components were mixed for a while and pressed P=300 H/cm² to form a tablet. Then tablets were vacuumed in the quarts ampules and kept in the oven at 1273-1473

K during 100 hours. The surface of prepared this way tablets were covered by silver paste and equipped with electrodes. In prepared sandwich structures the dependence of dielectric parameters (C, tgδ, φ Z and R) from frequency was measured by means of E7-20 immitans. The samples' dependence of dielectric parameters from frequency was measured in screened measuring cell under vacuum and at constant temperature. The error of measuring results was not more than 5%. The other parameters (ε, ρ, Z' and Z'') were calculating from the results of carried out measuring. The frequency dependence was measured at 25-10⁶ Hz diapason.

Results of Experiments

On Figure 1a shown frequency dependence of U₃O₈ taken in various concentrations a)-10% U₃O₈ 20% U₃O₈ and 30% U₃O₈, b)-90%U₃O₈ and 80% U₃O₈) Z'=f(v) and Z''=f(v) for nano-Si/U₃O₈

As it seen the maximum value of frequency of Z''=f(v) and the frequency of descent area's centre of Z'=f(v) are almost coincide of each other, and this fact indicates on relaxation process occurring in composite. As it seen the half-width of Z'' maximum's broadened at

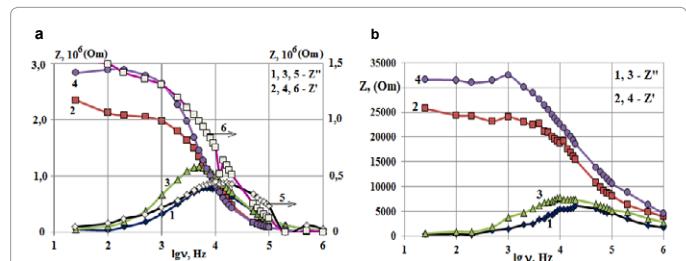


Figure 1: The dependence of Z=f(v), for nano-Si/U₃O₈ from frequency at T=300K a) 10%Si/10%U₃O₈ (1,2), 20%Si/20%U₃O₈ (3,4); 30%Si/30%U₃O₈ (5,6); b) 90%Si/10%U₃O₈ (1,2), 80%Si/20%U₃O₈ (3,4).

***Corresponding author:** Ramazanov MA, Dean, Department of Chemical Physics of Nanomaterials, Baku State University, AZ 1148, Baku, Azerbaijan, Tel: 994124390858; E-mail: mamed_r50@mail.ru

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relatively low frequency ($\nu \sim 4\text{-}5$ kHz) and concentrations of U₃O₈ (10% and 20% U₃O₈) and shift to high frequency ($\nu \sim 10\text{-}15$ kHz) with increasing of U₃O₈ concentration (30% U₃O₈). The comparison of parameters of samples with high concentrations of U₃O₈ (80% and 90% U₃O₈) (Figure 1b) reveals that relaxation process in composite starts at 25-30% concentration of U₃O₈. The reason of shifting of the frequency maximum most probably is increasing of conductivity of composite with increasing of U₃O₈ concentration as well as grows of the clusters size [10]. The shift of frequency dependence of maximums of impedance's imaginary part (Z'') measured at various temperatures proves the relaxation processes that takes place in composite (Figure 2). As it seen from dependences the maximums shift to higher frequency with increasing of temperature. The morphology of Nano-Si/U₃O₈ composite's samples was studied by scanning electron microscopy (SEM) (Figure 3). For comparison were taken two samples with various content of nano-Si/U₃O₈ system - 80%Si/20%U₃O₈ and 20%Si/80%U₃O₈. From these figures the nanoparticles in the composite present as agglomerate, not as separate particles. The comparison of agglomerate of 80%Si/20%U₃O₈ composite with agglomerate of 20%Si/80%U₃O₈ composite reveals that, composite with higher nano-silica content 80%Si/20%U₃O₈ led to preparation of small-sized structure. The reason of that is the main part of these agglomerates is a silica nanoparticles.

In other composite the main part of composite are U₃O₈ micron sized particles, and due to decreasing of special surface the numbers of pores also diminished. The Si nanoparticles in a few amounts are collected on the surface or in free cavities of U₃O₈ particles.

Consequently, the value of effective contact surface between components of composite 20%Si/80%U₃O₈ decreases. However, increasing the permeability and quantity of U₃O₈ particles leads to an increase a number of the charge carrier and cause dielectric relaxation of the process. On the other hand, increasing the particle U₃O₈ can bring to internal radiation and as a result lead to the formation of additional carriers. The participation of such carriers in polarization process influences the relaxation process.

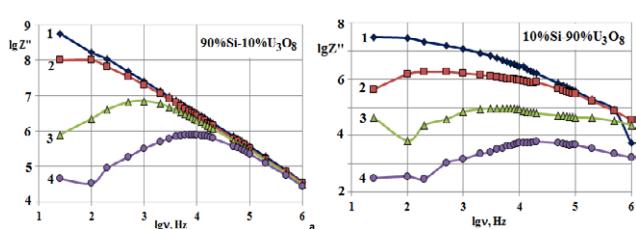


Figure 2: The frequency dependence of imaginary part of impedance (Z'') for nano-Si/U₃O₈ composite measured at various temperature: 1-100K; 2-160K; 3- 220K; 4- 285K

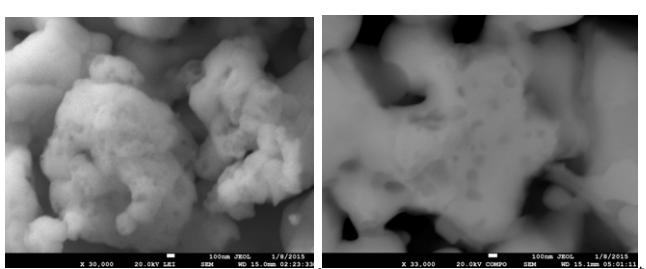


Figure 3: SEM images of samples of nano-Si/U₃O₈ system: a. 80%Si/20%U₃O₈; b. 20%Si/80%U₃O₈

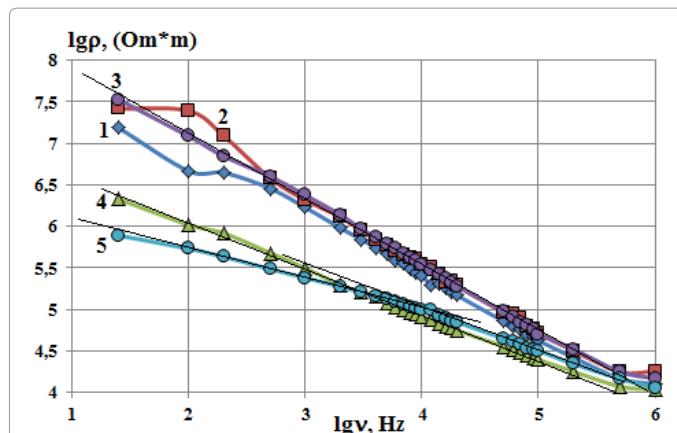


Figure 4: Dependence of $\lg p = f(\nu)$ for nano-Si/U₃O₈ system, (T=100K): 1-10% U₃O₈; 2-20% U₃O₈; 3-30% U₃O₈; 4-40% U₃O₈; 5-90% U₃O₈

S.No	nano-Si/U ₃ O ₈	the n parameter of $\rho \sim \omega^n$ equation	
		I orientation	II orientation
1	90% Si/10% U ₃ O ₈	0,84	-
2	80% Si/20% U ₃ O ₈	0,84	-
3	70% Si/30% U ₃ O ₈	0,84	-
4	20% Si/80% U ₃ O ₈	0,51	0,55
5	10% Si/90% U ₃ O ₈	0,48	0,37

Table 1: The value of n for nano-Si/U₃O₈ systems with various contents.

As we know, electrical field caused electron and ion polarization in the dielectric with ion solid structure. The increasing of temperature and decreasing of force of ion interactions led to growth of freedom degree and as a result the increasing of ion-relaxation polarization in nano-Si/U₃O₈ system. As we mentioned under an alternating electrical field the free charges are gathering and distributing on the interphase border (Maxwell-Wagner polarization) and created local electrical field. This field led to changes of electrical field. It is known the polarization in composite materials at low frequency is determined by its conductivity as well as at high frequency – dielectric permeability. So, the increasing of ϵ and Z' at high frequency can be explained by Maxwell-Wagner polarization caused by strong semiconductor U₃O₈ [11].

For clarifying the mechanism of electrical conductivity in such kind systems the dependence of specific resistance from frequency was applied. On Figure 4 there is the graph of dependence of specific resistance from frequency measured at low temperature (T=100 K) for the nano-Si/U₃O₈ system.

As it seen from Figure 4 for low content of U₃O₈ (10-30%) the dependence presents the straight line (1-3). For U₃O₈ with content 80-90% the dependence presents different orientation (4,5). In such kind unordered the frequency conductivity dependence is the function $\rho \sim \nu^n$ where n can be determined from conductive mechanism, depending of the parameters. In the Table 1 presented n for various content samples.

As it is seen from the obtained results the value of parameter n varies in the 0.4n<0.9 range. Variation in electrical conductivity by this regularity has allowed predict the existence of rapid conductivity mechanism in the nano-Si/U₃O₈ systems examples [12].

Conclusion

Thus the analysis of the experimental results of the dependence of electro-physical properties versus frequency for Nano-Si/U₃O₈

system composite show that changes in these properties are related to the polarization processes that occur in the phase boundary between components.

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