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STUDY OF DIELECTRIC PROPERTIES OF SOME BULK Cu-As-Se-I CHALCOGENIDES

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The impedance spectroscopy technique was used to study the frequency dependence of some dielectric parameters in the frequency range from 100 Hz to 1 MHz for the bulk amorphous semiconducting glasses from the system $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$ (x=0, 1, 5, 10 and 15 at. % Cu). The measurements were carried out at different temperatures (298–398 K). Values of the real and imaginary part of dielectric permittivity (ε' and ε'') were found to decrease with frequency and increase with temperature. The obtained results are interpreted in terms of the Debye theory of molecular dipoles. They indicated the existence of different types of dipoles that determine the mechanism of dielectric behavior for the investigated compounds. The parameters of dielectric permittivity are explained according to a model of hopping over the potential barrier between the localized sites. The maximum barrier height W_M was calculated from the dielectric measurements in accordance with Giuntini equation.

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1. Introduction

Chalcogenide glasses have received a lot of attention because of their potential and current use in various solid-state optical and electrical devices [1,2]. Amorphous chalcogenide materials are generally semiconductors, usually involving covalent bonding in the structural units. Their dielectric properties depend on the combination of composition and purity of starting components, synthesis conditions, rate of melt annealing, thermal treatment, and some other factors. It has been found that the majority of physical effects observed in the glassy chalcogenides are related to the existence of localized energy states in the mobility gap, i.e. at the mid-point of the band gap and in the tails of the energy bands, due to the absence of long-range structural order, as well as to various inherent defects [3]. These inherent defects originate from the rearrangements taking place at the chain ends (charged dangling bonds or valence-alternation pairs). In view of the fact that these characteristics are a consequence of the different defect types in the glass structure, it can be expected that more complex compositions will have a richer spectrum compared to that of the binary and ternary glasses, which may be the cause of activation of different charge transfer mechanisms. The doping of chalcogenide glassy semiconductors with d-elements such as Ag and Cu is an effective way of changing their electrical and switching properties in a definite direction. By means of such doping it is possible to overcome the difficulty of high electrical resistivity, which is characteristic of most chalcogenide glasses. Copper has been used as a chemical modifier in arsenic selenides, in which it shows a marked increase in the electrical conductivity [4]. It has been found that the presence of Cu in amorphous arsenic selenides leads to the increase in density, microhardness [5], softening temperature and thermal stability [6], refractive index, and to a

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decrease of the optical band gap [7], activation energy of electrical conductivity [8], as well as to the changes in the electrical switching properties of the glass to a definite direction [9].

The aim of this contribution was to study of dielectric properties of the semiconducting glassy system $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$ (x=0, 1, 5, 10 and 15 at. % Cu) in the temperature range from 298 to 398 K.

2. Experimental

The studied bulk alloys were synthesized by the melt-quenching method, using high-purity elementary components (99.999 %), and amorphous character of the samples was checked by X-ray diffraction as described in detail in [10]. To ensure correct electrical contacts, the investigated samples were polished with alumina powder of 0.3 and 0.05 μm grain size until a high gloss surface was obtained. The rectangular-shaped samples had a cross-section of ~ 20 mm^2 and the thickness between 0.5 and 1 mm. Silver paste was applied on the opposite parallel surfaces of the samples.

The measurements were carried out in a specially constructed measuring cell with two electrodes. The samples were spiral springs loaded between two brass electrodes to ensure good electrical connection and constant pressure contact during the measurements. The dielectric properties were measured in the frequency range of 10^2–10^6 Hz at the temperatures from 298 to 398 K on a computer-controlled AC-impedance system (HP 4194A Impedance/Gain-Phase Analyzer).

The measurements above room temperature were performed by placing the measuring cell in a thermally controlled furnace. The ambient temperature was regulated with a temperature controller BECKMAN CTC 250 (PID type). The temperature was measured using a copper-constantan thermocouple with an accuracy of ±0.5 K. All measurements were done in air and in dark.

3. Results and discussion

It is well known that the complex dielectric function ε of a material is represented by two parts, $\varepsilon = \varepsilon' + i\varepsilon''$, where ε′ is the real part and ε″ is the imaginary part (dielectric loss). These dielectric parameters were calculated from the measured values of the capacitance C and resistance R of investigated glasses according to the following equations:

$$\varepsilon' = \frac{d}{S} \cdot \frac{C_p(\omega)}{\varepsilon_0} \qquad \varepsilon'' = \frac{1}{R_p(\omega)\,\omega C_0} \tag{1}$$

where d is the sample thickness, S is the cross-sectional area, ω is the angular frequency and C_0 is the free space permittivity.

Figs. 1-3 show the frequency dependence of the real part of dielectric permittivity ε′ of the some samples of studied system $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$ measured at different temperatures. It can be noticed that quantity ε′ for all the samples shows dispersion behaviour in the given frequency range. Frequency dependence is especially pronounced at higher temperatures and lower frequencies.

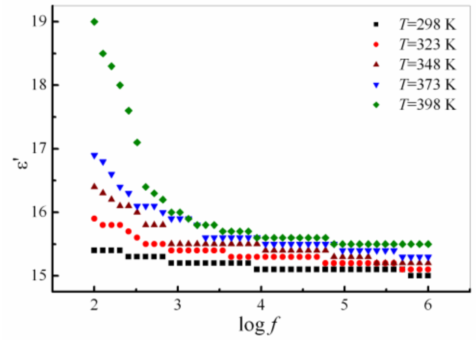

Fig. 1. Frequency dependence of ε′ for $(As_2Se_3)_{0.9}(AsI_3)_{0.1}$ *at different temperatures*

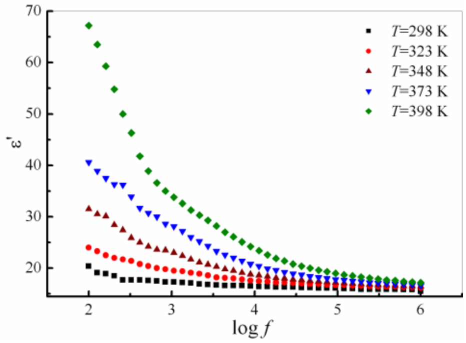

Fig. 2. Frequency dependence of ε′ for $Cu_5[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{95}$ *at different temperatures*

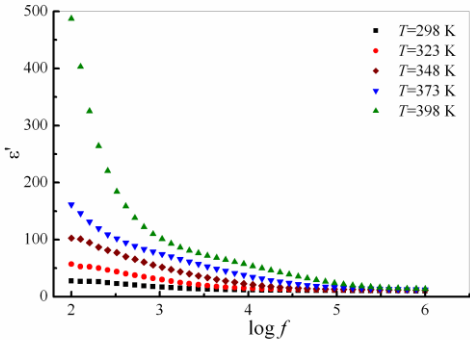

Fig. 3. Frequency dependence of ε′ for $Cu_{15}[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{85}$ *at different temperatures*

The origin of polarization in investigated samples for the given frequency range is a rather complex issue. It is known that there are generally four types of polarization: electronic, ionic (or atomic), dipole and space charge polarization. However, it is not likely that the intermediate polarization could be involved in the studied materials since there are no interfacial surfaces or grain boundaries in the structure where space charge would appear, at least not in the amount which would contribute significantly to the overall polarization. The investigated glasses are the compounds with predominantly covalent bonds and an extremely low percentage of ionic bonds. Therefore, the ionic type of polarization could also be neglected rather than taken as a significant factor. Namely, if one calculates the percentage of covalency according to Pauling relation [11] for all possible chemical bond in the studied chalcogenides, it comes out that a minimal percentage of covalency (86.6 %) is characteristic of the bond Cu—I.

However, with the increase of copper content in amorphous matrix As_2Se_3–AsI_3, the bonds of this element with selenium (whose covalency amounts to 90.0 %) are more probable than those with iodine. Namely, copper prevailingly forms structural unit of the type CuSe, Cu_2Se, $CuSe_2$, $CuAsSe_2$, Cu_3AsSe_4, in which covalent bonds are dominant [12]. Also, the percentage of the ionic character of chemical bonds for arsenic selenide is lower that 9 % which suggests that the contribution of ionic polarization to dielectric properties of the investigated materials is not significant.

Therefore, it can be concluded that decrease in the parameter ε' at higher frequencies is a consequence of the hindered rotation of dipoles (structural units) due to fast changes in the field direction. Electronic polarization is practically always present in the overall polarization of material in a wide range of field frequencies (up to 10^{17} Hz). In the area 10^{14}–10^{17} Hz this mechanism is the only factor that contributes to the polarization and according to Maxwel electromagnetic theory the real part of dielectric permittivity is equal to the square of the refractive index, $\varepsilon' = n^2$ [13]. In that manner, based on the refractive index measurements in the long-wave part of the spectrum it is possible to estimate the percentage share of the electronic mechanism in the overall polarization. Table 1 lists the values of the refractive index for some investigated compounds [7], as well as corresponding values of the real part of dielectric permittivity to which funcional dependence $\varepsilon'(f)$ tends to reach in the range of high field frequencies at room temperature. The calculated values of electronic polarization share for the compositions with $x \leq 10$ at. % Cu are close to 50 %. On the other hand, for the composition with 15 at. % Cu, apart from a lower value of ε', the representation of electronic polarization share is significantly higher. This could be explained with the changes of the mutual ratio of structural units that gradually change from the dominant two-component As_2Se_3 and AsSe for the glasses with $x \leq 10$ at. % to the three-component ones with copper in its composition [14]. At higher frequencies (Table lists the data for f=1 MHz) these constituent elements are less able to follow the changes in the field direction and their contribution to the overall polarization is smaller than in the case where the prevailing structural units are As–Se.

Table 1. Refractive index n [7] and real part of dielectric permittivity ε' (f=1 MHz) for $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$ glassy system.

x [at. % Cu]	n	ε'	n^2/ε' [%]
0	2.68	15.0	47.9
1	2.71	15.1	48.6
5	2.77	15.5	49.5
10	2.83	17.1	46.8
15	2.88	10.2	81.3

Besides, it can be noted from the Table 1 that the real part of dielectric permittivity at low field frequencies shows almost constant increase with the increase in the copper concentration. It is known that the addition of copper (and of d-transition metals in general) to chalcogenide glasses increases concentration of bond defects that is charged defect centers (D^+ and D^-) in the structure. This also causes an increase in the density of localized energy states in the mobility gap around the Fermi level [15], but also in the tails of the bands. The mentioned defects are the main factor responsible for the electrical conduction of this class of amorphous semiconductors, which occurs due to charge carriers hopping between the localized states in the gap. Hence, the increase of copper content enhances the conductivity, as evidenced in the previous investigations [8], causing also an increase of the quantity ε'.

Deviation from this behaviour is noticed for composition $Cu_{15}[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{85}$. The content of the new (three-component but also two-component) structural units in this compound that may enhance dipole polarization is obviously insufficient to overcome their weak mobility, even in low-frequency fields. The decreased mobility of these structural elements is a consequence of stronger chemical bonds formed by copper atoms primarily with selenium, compared with the bonds between arsenic and selenium (AsSe and As_2Se_3) which more easily follow the changes in the field direction. If one considers the energy of chemical bonds between the constituent elements of the glass network, it comes out that the bonds of copper with selenium and iodine are characterised with the higher energy. Thus, this fact is one of the main reasons for the decreased permittivity of the investigated materials.

Figs. 4–6 show the temperature dependence of the real part of dielectric permittivity for certain glass samples (x=0, 5 and 15 at. % Cu). The increase in the permittivity at higher temperatures, especially pronounced in low-frequency range, is a consequence of the fact that the orientational polarization is associated with the thermal mobility of molecules. Hence, at lower temperatures dipoles can not be fully oriented. The increase in temperature allows dipole orientation and the orientation polarization share in the overall polarization increases, also reflecting on the increase in the real part of dielectric permittivity [16,17].

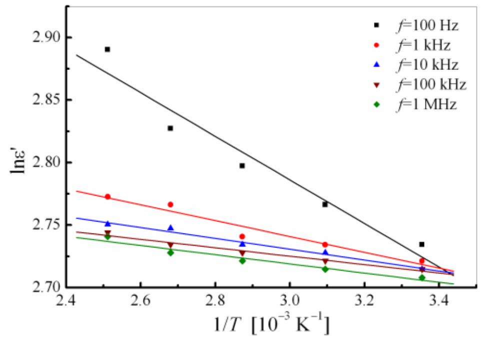

Fig. 4. Temperature dependence of ε' *for* $(As_2Se_3)_{0.9}(AsI_3)_{0.1}$ *at different frequencies*

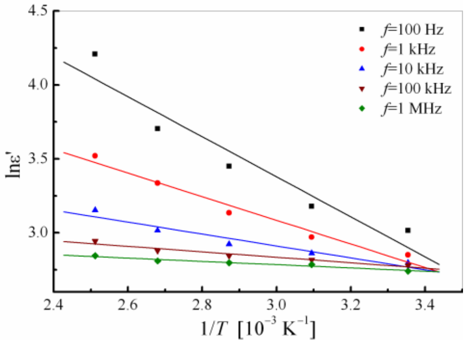

Fig. 5. Temperature dependence of ε' *for* $Cu_5[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{95}$ *at different frequencies*

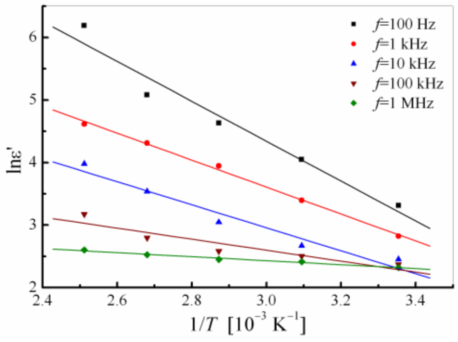

Fig. 6. Temperature dependence of ε' *for* $Cu_{15}[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{85}$ *at different frequencies*

Linear dependence of the function $\ln \varepsilon' = f(1/T)$ is characteristic of the systems with molecular dipoles described with Debye theory [17] on the dependence of the viscosity on the relaxation time. According to this theory, ε′ rises exponentially with increase in temperature. Such dependence was obtained for all investigated samples. Therefore, it can be concluded that the dielectric properties of the system $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$ can be successfully explained with Debye theory.

The frequency dependence of the imaginary part of the dielectric permittivity (dielectric loss), measured at the different temperatures is presented in Figures 7–9. The expected increase of the dielectric losses at higher temperature is commonly observed for this class of materials [18]. This behaviour can be interpreted with a higher resistance of the viscous medium to dipole rotation in the alternating electric field.

In a more comprihensive approach to the analysis of the observed characteristics it is necessary to mention the Stevels interpretation of the increase in the dielectric losses with heating of the material [19]. According to this author, there are three groups of relaxation, depending on its origin. Thus, there are the losses due to the conductivity, dipole and vibrational losses. At lower temperatures the losses due to conductivity have small values; more exactly, they reach a minimum. With the increase in temperature the conductivity also increases, so as the corresponding losses, having influence on the increase of ε''.

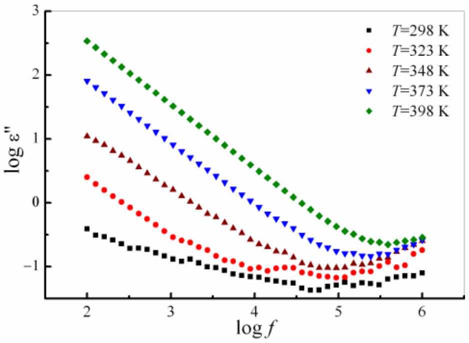

Fig. 7. Frequency dependence of dielectric loss ε″ for $(As_2Se_3)_{0.9}(AsI_3)_{0.1}$ at different temperatures

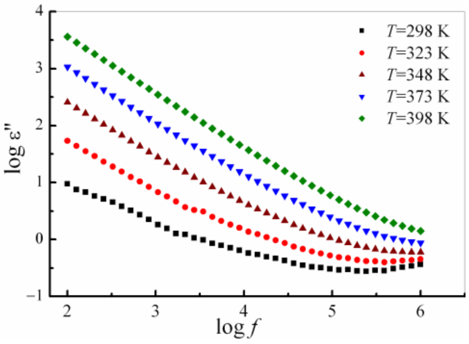

Fig. 8. Frequency dependence of dielectric loss ε″ for $Cu_5[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{95}$ at different temperatures

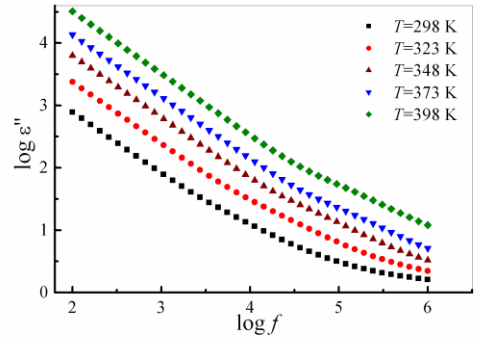

Fig. 9. Frequency dependence of dielectric loss ε'' *for* $Cu_{15}[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{85}$ *at different temperatures*

Also, it should be noticed that ε'' increases with the copper content in the investigated samples. Since the dielectric losses in chalcogenide glasses depend on the number of localized states [20] it can be concluded that the concentration of defect states increases with the amount of copper in the system $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$.

As far as the frequency dependence of $\varepsilon''(f)$ is concerned, it should be noted that this value decreases in the whole range of field frequency. Therefore, it is assumed that the maximum of dielectric losses in dispersion area, if there is one, should be outside the range of measurements using the instrument at disposal. The absence of peaks becomes evident when there exists a wide distribution of relaxation times, which also could be expected in the case of the investigated glasses. Such behavior has been also observed for some other classes of amorphous materials [21]. Therefore, the determination of the characteristic field frequency ω_m (or f_m) for which the loss factor exhibits a maximum was not possible in this case.

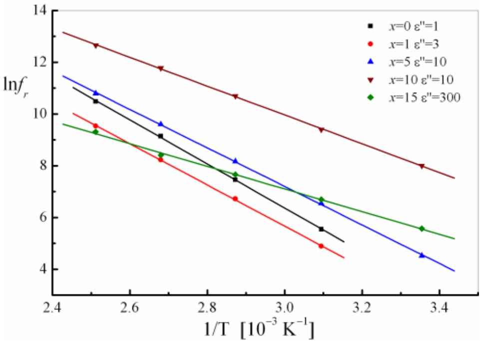

Fig. 10. Temperature dependence of charactestic frequencies f_r *of* ε' *for glasses* $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$.

However, based on the analysis of temperature dependence of characteristic frequency f_m it was noticed that da Arrhenius equation is also applicable for the frequencies f_r, lower or higher than f_m [22,23]:

$$f_r = f_{r0} \exp\left(-\frac{W_f}{kT}\right) \tag{2}$$

Therefore, the reading was done for those values of the electric field frequency f_r at which the dielectric loss factor at a given temperature reaches the selected value instead of maximum. Figure 10 shows the linear dependence of the values $\log f_r = f(1/T)$ for the chosen values of dielectric loss. Fitting these curves it was possible to determinate the activation energy of relaxation process W_f. The obtained average values of this parameter are shown in Table 2, together with activation energy values of dc conductivity, determined in previous investigations [8]. It can be noted that presented values are mutually very comparable. This means that the mechanisms of conductivity and relaxation in the investigated frequency range are the same.

Table 2. The activation energies associated with dielectric loss process W_f and for dc conductivity E_σ [8].

x [at. % Cu]	W_f [eV]	E_σ [eV]
0	0.73(1)	0.72(1)
1	0.66(1)	0.67(1)
5	0.64(1)	0.62(1)
10	0.45(1)	0.54(2)
15	0.38(1)	—

The analyses of the frequency dependence of ε'' for many amorphous materials indicate that it often satisfies the equation [24]:

$$\varepsilon'' = A\omega^m \tag{3}$$

where A is a temperature-dependent constant and m is the negative exponent of the power function of the angular frequency ω.

The experimental measurements presented in this study allowed determination of the parameter m and potential barrier height W_M (energy required to move electron from a site to infinity) based on the slope of the linear function $\log\varepsilon'' = f(\log f)$. The obtained values are listed in Table 3. The calculations were carried out using the Giuntini model for description of dielectric dispersion in chalcogenide glasses [25]. This model is based on Elliott idea of charge carrier hopping over the potential barrier between the charged defect states (D^+ and D^-) [26]. It is assumed that each pair of states forms a dipole whose relaxation time τ depends on energy needed to overcome the potential barrier. In accordance with this model, at a given temperature T and field frequency ω, ε'' is described with the following expression:

$$\varepsilon'' = (\varepsilon'_0 - \varepsilon'_\infty)\,2\pi^2 N\left(\frac{ne^2}{\varepsilon'_0}\right)^3 kT\,\tau_0^m W_\mathrm{M}^{-4}\,\omega^m \tag{4}$$

where ε'_0 and ε'_∞ are the respective static and optical dielectric permittivity (at 'infinitely high' frequencies), n is the number of hopping electrons, N is the concentration of localized states, and τ_0 is the pre-exponential factor in the Arrhenius equation for the temperature dependence of relaxation time. The exponent m is a negative quantity since $\varepsilon''(\omega)$ is a decreasing function, and it is related to temperature according to the following expression [25]:

$$m = -\frac{4kT}{W_\mathrm{M}} \tag{5}$$

Table 3. Values of parameter W_M *for the samples of the system* $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$.

	x=0	x=1	x=5	x=10	x=15
T [K]	W_M [eV]	W_M [eV]	W_M [eV]	W_M [eV]	W_M [eV]
298	0.321	0.313	0.205	0.141	0.125
323	0.161	0.215	0.162	0.135	0.128
348	0.154	0.165	0.146	0.138	0.130
373	0.141	0.153	0.144	0.138	0.135
398	0.140	0.150	0.145	0.144	0.143

It can be noticed that the values of potential barrier height are significantly smaller than the activation energies of hopping conductivity between the localized states in the band tails, whose values for investigated glasses are in the interval of 0.54–0.72 eV [12]. Rather, it might be said that they correspond to the mechanism of charge carrier hopping between the localized states in the vicinity of the Fermi level. These states are due to the existence of charged defect centers (bipolarons, D^+ and D^-) in the structure, proposed for the class of chalcogenide glasses [27]. The obtained values of W_M correspond to the values also observed for some other chalcogenide glasses [24,28].

Besides, it should be noted that there is an increase in the slope of the function $\log \varepsilon'' = f(\log f)$ with increase in temperature (that is the increase in the parameter m), and a decrease in the activation energies W_M for the samples with x=0–5 at. % of Cu. The changes in potential barrier height can be explained with the decrease of density of localized states in the vicinity of the Fermi level at higher temperature [29]. Namely, the heating of the material causes that some of charged defect states, to which one can ascribe the higher values of the W_M, can transform into single-polaron states (D^0) by charge carrier hopping between them. These states are characterised with the lower values of parameter W_M. For the glass samples with x=10 and 15 at. % Cu the changes in the energy W_M are insignificant, or even go the opposite direction. This indicates either that the structure of charged defect states remains unchanged at higher temperatures or that there exists a weak opposite trend associated with the increase in the content of three-component structural units with copper. Concerning the effect of copper content on the parameter W_M it can be noted that at lower temperatures (298 and 323 K) its value decreases due to the increase in the concentration of localized states in the conductance gap, which is a consequence of increased disorder of the structure.

4. Conclusions

Bulk samples of $Cu_x[(As_2Se_3)_{0.9}(AsI_3)_{0.1}]_{100-x}$ chalcogenide glassy system, obtained by melt quenching method, were subjected to the measurement of their dielectric properties in the temperature interval of 298–398 K and in the frequency range from 100 Hz to 1 MHz. The introduction of copper into the system increased the values of dielectric parameters. The enhancement of dielectric quantities was also observed with the increase in temperature. The interpretation of the results indicated that the primary reason for such trend is the increase in the density of localized energy states as a consequence of dopping with copper. Certain deviation from this behavior for the compound with x=15 at. % Cu was explained with structural changes manifesting as an increased presence of the domains with copper whose mobility is lower compared to that of As–Se structural units. The decrease in the dielectric function parameters with increase in frequency of the applied field is a consequence of the hindered dipole orientation, i.e. lower orientation polarization of the material. Based on linear dependence of the function $\ln \varepsilon' = f(1/T)$ it was concluded that the dielectric properties of the investigated system can be successfully explained with Debye theory. The frequency dependence of the dielectric losses was described with the hopping mechanism of charge carriers over the potential barrier between the charged defect centers (D^+ and D^-). The barrier height was determined using the Giuntini equation based on the Elliott model for chalcogenide glasses. For all investigated glasses, this barrier corresponds to the difference between the energy states around the Fermi level. It was found that the activation energy of relaxation process W_f is very comparable with the activation energy of dc conductivity. Therefore, the mechanisms of these two processes in investigated materials for given frequency range are the same.

Acknowledgements

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