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Temperature Dependence of AC Conductivity and Complex Dielectric Constant of $Cd_2Si_{1-x}Ge_xO_4$ Compound

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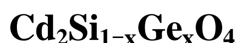
Abstract
In this work, samples of $Cd_2Si_{1-x}Ge_xO_4$ prepared by powder technology for (x = 0, 0.3, 0.6) were studied. The effect of (Ge) additives at different ratio of Ge (x=0, 0.3, 0.6) on the behavior of dielectric constant, dielectric loss and a,c conductivity were measured as a function of temperature at a selected frequencies (0.01 – 10) MHz in the temperature range 298 K to 473 K. The dielectric constant and dielectric loss obtained different behavior with the additives of (Ge). The activation energy for the electrical conduction process was studied.

Keywords: Cd alloys, Cd compounds, Si compounds, Powder Technology, a.c conductivity, Dielectric Properties

اعتماد التوصيلية المتناوبة وثابت العزل المركب على درجة الحرارة لمركب



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الخلاصة
في هذا العمل , درست نماذج $Cd_2Si_{1-x}Ge_xO_4$ المحضرة بتكنولوجيا المساحيق للنسب (x = 0, 0.3, 0.6) . درس تاثير اضافة Ge لنسب مختلفة من Ge (x = 0, 0.3, 0.6). على ثابت العزل , فقد العازل والتوصيلية المتناوبة كدالة لدرجة الحرارة ولترددات مختارة MHz (10 – 0.01) في مدى درجات الحرارة 298 كلفن الى 473 كلفن. اكتسب ثابت العزل وفقد العازل سلوك مختلف باضافة Ge . تم دراسة طاقة التنشيط لعملية التوصيل الكهربائي.

Introduction
Germanium is a chemical element with symbol Ge and atomic number 32. It is a lustrous, hard, grayish-white metallloid in the carbon group, chemically similar to its group neighbors like tin and silicon. Purified germanium is a semiconductor, with an appearance most similar to elemental silicon. Like silicon, germanium naturally reacts and forms complexes with oxygen in nature. Unlike silicon, it is too reactive to be found naturally on Earth in the free (native) state. Two oxides of germanium are known: germanium dioxide, (GeO_2, germania) and germanium monoxide, (GeO). The dioxide, GeO_2 can be obtained by roasting germanium disulfide (GeS_2), and is a white powder that is only slightly soluble in water but reacts with alkalis to form germanates.[1]. The monoxide, germanous oxide, can be obtained by the high temperature reaction of GeO_2 with Ge metal. The dioxide (and the related oxides and germanates) exhibits the unusual property of having a high

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refractive index for visible light, but transparency to infrared light. Silicon-germanium compounds are rapidly becoming an important semiconductor material, for use in high-speed integrated circuits. Circuits utilizing the properties of Si-SiGe junctions can be much faster than those using silicon alone.[2] Silicon-germanium is beginning to replace gallium arsenide (GaAs) in wireless communications devices.[3] The SiGe chips, with high-speed properties, can be made with low-cost, well-established production techniques of the silicon chip industry [4]. The aim of work is to study the effect of temperature on the behavior of a.c conductivity, activation energy, type of mechanism, and complex dielectric constant of this type of compounds. The measurements of the temperature-dependence of dielectric constant of $Cd_2Si_{1-x}Ge_xO_4$ samples were carried out through the temperature range (298 – 473) K and frequency range from 0.1 kHz to 10 MHz. In general, the relation [5] gives the complex relative dielectric permittivity:

$$\varepsilon_r(\omega) = \varepsilon_1(\omega) - i\varepsilon_2(\omega) \quad (1)$$

($\varepsilon_r = \varepsilon / \varepsilon_o$; where $\varepsilon_o = 8.85 x 10^{12}$ F/m is the permittivity of free space and ω is the angular frequency) .
The dielectric constant ε_1 was calculated from the relation:

$$\varepsilon_1 = C/C_o \quad (2)$$

where C_o is vacuum capacitance ($C = \varepsilon_o A/d$, where d is the thickness of the sample and A is the cross sectional area).Also the dielectric loss ε_2 was calculated from the relation:

$$\varepsilon_2 = G / \omega C_o \quad (3)$$

where G is the conductance ($G = \frac{1}{R}$).

Experimental Work

Samples of $Cd_2Si_{1-x}Ge_xO_4$ were prepared by powder technology by mixtures of SiO_2 (purity 99.9% from Ted Pella), GeO_2 (purity 99.9% Koch Light Ltd., England) and CdO (purity 99.9% Loba Chemicals Ltd., England), the three oxides pre – calcined at 500 ^oC for 1 hour to remove hydroxide and carbonate impurities. After being weighted out in the appropriate molar ratios, (x = 0, 0.3, 0.6) , the powder were intimately mixed by hand grinding and reacted at 900 ^oC for 12 hour . Then pressed into a 13 mm diameter pellet at 5 ton for 5 min and lightly sintered in air at 850 ^oC for 48 h. To study the behavior of the dielectric constant and a.c conductivity of $Cd_2Si_{1-x}Ge_xO_4$ samples, we put it between two Cu electrodes into electrical oven which is connected to LCR system of a programmable automatic LCR bridge (PM 60304 Philips) in order to measure the capacitance C and the conductance G as a function of temperatures and frequency .

Result and discussion

Atomic Force Microscopy (AFM)

Figures. 1a, b, c show the 3D atomic force microscopy (AFM) images for x= 0, 0.3, 0.6) of Ge doped in $Cd_2Si_{1-x}Ge_xO_4$. From the figure, it was observed that the topography of x = 0 figure. 1a possesses average grain size of 99.22 nm with grain number 130. The incorporation of Ge into the compound figure. 1b at x = 0.3 showed an increase in grain size to become 110 nm with grain number 58. Figure. 1c at x = 0.6 showed more increasing in grain size to become 139 nm with grain number 65. It was noticed that $Cd_2Si_{1-x}Ge_xO_4$ samples were denser with more uniform granular grains with introducing of Ge atoms. To explain it, there is four electrons in the valence orbital of Si and Ge atoms. So, a new bonds form with the additives of Ge and this increases the grain size and its numbers or merging the small grain to become larger, then enhanced electronic transport [6]. Table 1 indicates such results.

a b

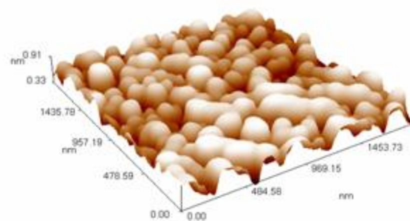

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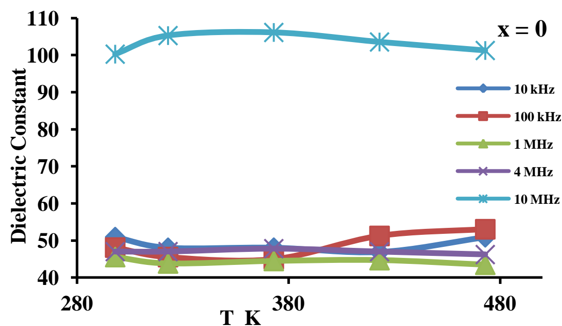
Figure 1 (a, b, c)-3D-AFM images for $Cd_2Si_{1-x}Ge_xO_4$

Table 1-Values of average grain size and no. of grain size of different ratio of Ge

x-values	Average grain size (nm)	No. of grain
0	99.22	130
0.3	110	58
0.6	139	65

Temperature Dependence of Complex Dielectric constant

Figures. 2 a, b and c show the temperature - dependence of dielectric constant ε_1 of $Cd_2Si_{1-x}Ge_xO_4$ at various frequencies with different ratio of x. The behavior of the dielectric constant slightly varies in the temperature range (298 K – 473 K) at selected frequencies of all ratio of Ge. At 10 MHz, the dielectric constant has higher values of all x values (i.e x = 0, 0.3, 0.6). At lower frequencies, this can be explained by the static dielectric constant (dc component that means frequency independent). At higher frequencies (10 MHz), the dielectric constant is frequency dependent which means the reorientation of carriers with the direction of alternating field and this increasing the polarization , then that increase of dielectric constant [7, 8]. In Fig.2c, there is a peak in the plot of dielectric constant and this is to response of the carriers with the alternating field due to the enhancement of grains arrangement [9]. This behavior can be certified by the variation of dielectric loss with temperature and frequency as shown in Figures. 3 a, b and c.



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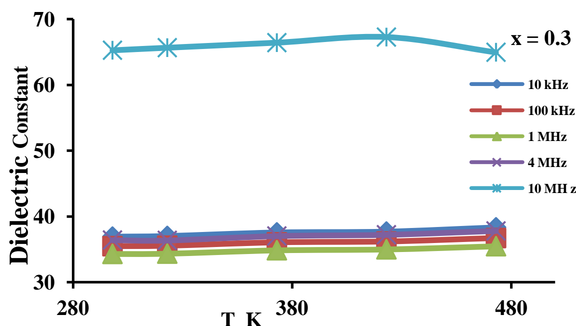

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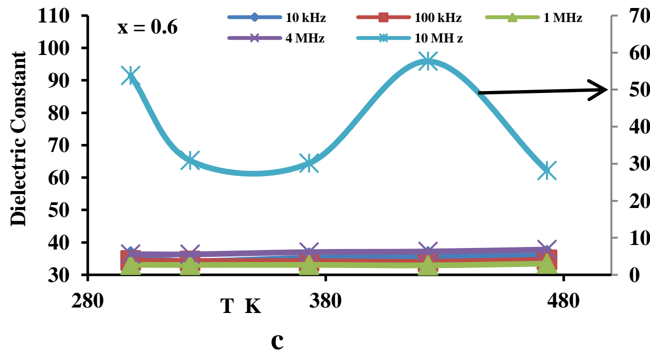

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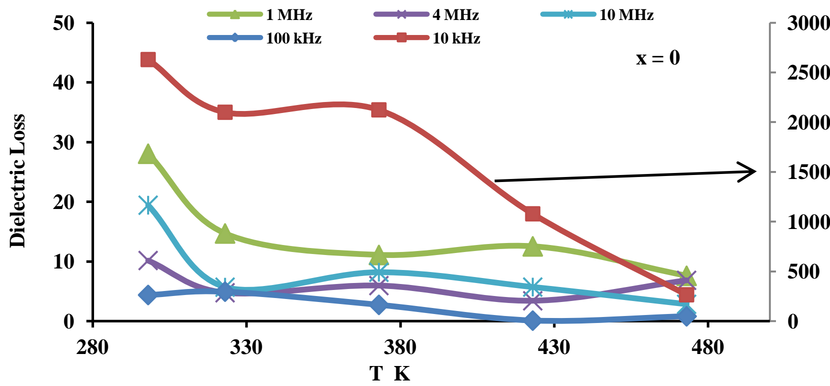
Figure 2 a, b and c-Temperature - dependence of dielectric constant of $Cd_2Si_{1-x}Ge_xO_4$ compound

In figure. 3a the dielectric loss decreases with the increase of temperature at selected frequencies for Cd_2SiO_4 at x = 0. The different behavior of dielectric loss for $Cd_2Si_{0.7}Ge_{0.3}O_4$ and $Cd_2Si_{0.4}Ge_{0.6}O_4$ (i.e x = 0.3 and 0.6) are shown in figures. 3 b and c which refers to a remarkable effect of Ge additive. The dielectric loss is temperature independent and frequency independent, except at 10 MHz, the dielectric loss increases with the increase of temperature and this is attributed as mentioned before the increase of grain size as shown in the images of atomic force microscopy [10].



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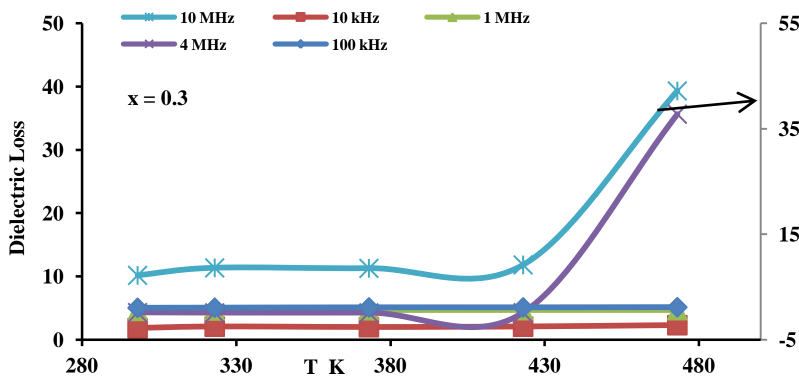

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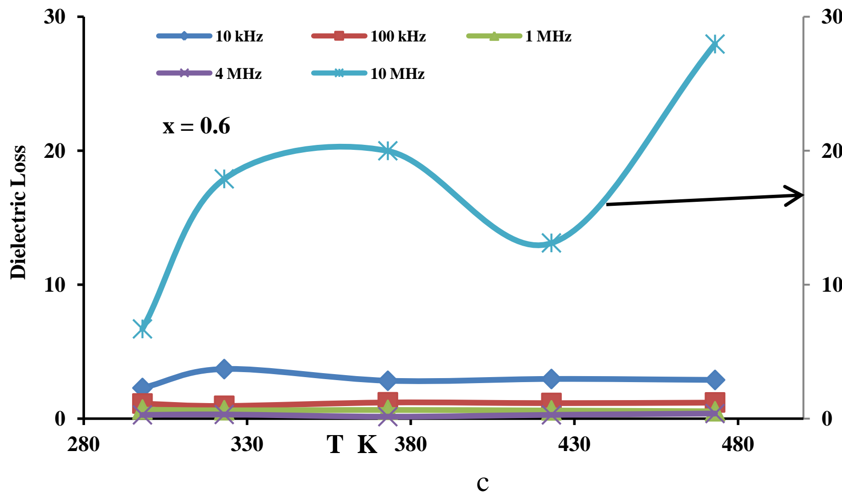

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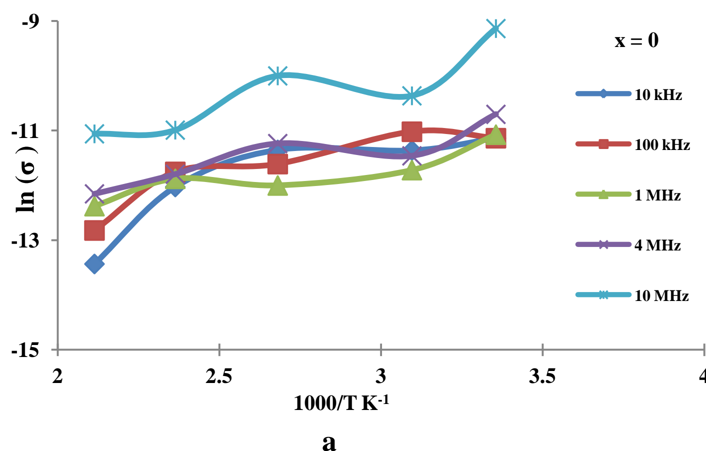
Figure 3 a, b and c-Temperature dependence of dielectric loss of $Cd_2Si_{1-x}Ge_xO_4$ compound

A.C. Conductivity

Figures. 4 a, b and c show the frequency- dependence of a.c conductivity σ (ω) versus reciprocal temperature at various frequencies for $Cd_2Si_{1-x}Ge_xO_4$ compound at different ratio of Ge . The pattern of temperature dependence conductivity plot can be said to follow the Arrhenius relation [11]:

$\sigma = \sigma_o \exp(-E_A/kT)$ (4)

Where σ_o is the pre-exponential factor, E_A is the activation energy, k is Boltzmann constant and T is absolute temperature . From relation 4, the activation energy E_A for the conduction process can be calculated from the slope of the linear part at low and high temperatures for all selected frequencies. As shown in Fig. 4a, the ac conductivity depends significantly on the temperature and frequency. It decreases with the increase of temperature and has high values at higher frequencies (10 MHz) for Cd_2SiO_4 at x = 0, and this can be interpreted to the deficiency of oxygen ions that make traps for carriers to jump in it [11]. With the increase of x ratio (i.e., 0.3, and 0.6), the ac conductivity is approximately constant at low temperature and it increases at higher temperature (10 MHz) with the increase of frequency as shown in figure. 4 b and c. The difference behavior of a.c conductivity at 10 MHz can be attributed to localize hopping, which is due to larger grain size with incorporation of Ge elements, and re-orientation motion, which is expected to be the dominated type of a.c conduction. So the mechanism of a.c conductivity is the co-related barrier height [12].



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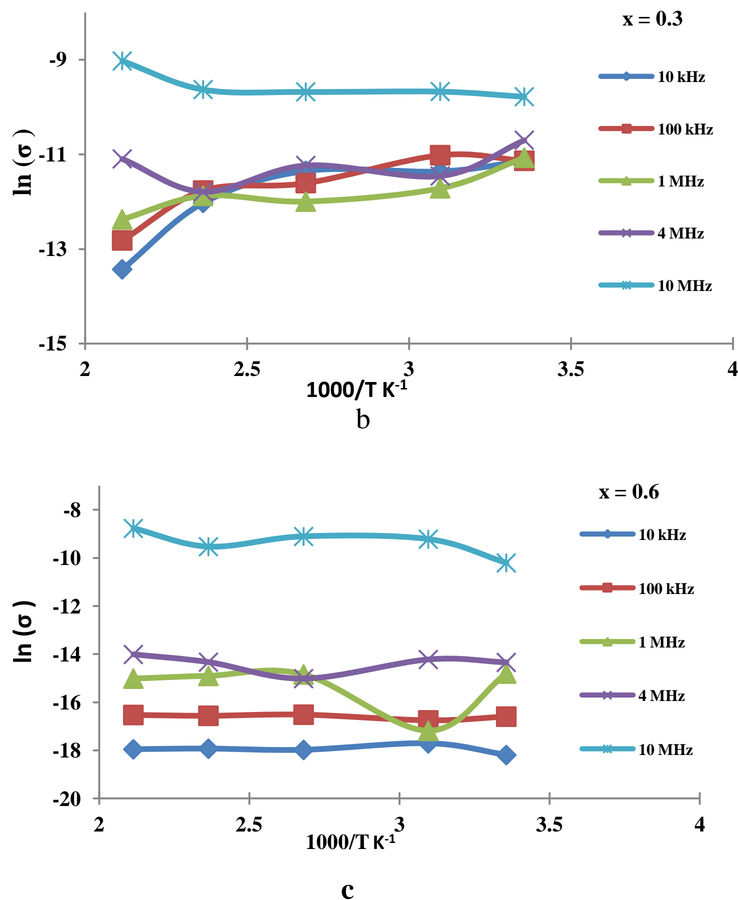

Figure 4 a, b and c-Temperature dependence of conductivity for $Cd_2Si_{1-x}Ge_xO_4$ compound

Figure. 5 represents Frequency dependence of the activation energy $E_A(\omega)$ which was calculated from the slope of the linear part of a.c conductivity plot. From the figure, the activation energy decreases with the increase of Ge ratio due to the responding the carriers and re-orientation itself with a.c field. The conduction process at (x = 0) can be attributed to the hopping of carriers between the deficiencies of oxygen ions that make traps for carriers. With the additives of Ge, the conduction process belongs to correlated barrier hoping between two sites [13].

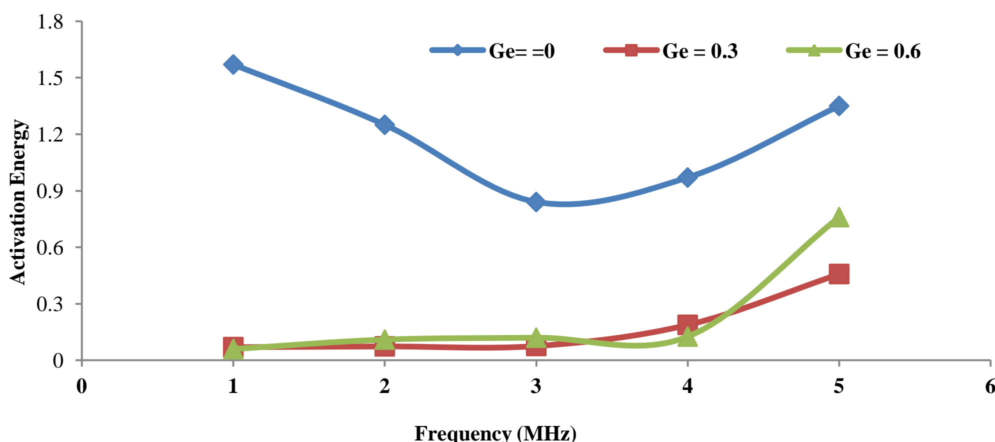

Figure 5-Frequency dependence of the activation energy $E_A(\omega)$

Conclusions

Samples of $Cd_2Si_{1-x}Ge_xO_4$ at (x = 0, 0.3, 0.6) were prepared by powder technology. The additive of Ge in the compound increased the grain size and its numbers and then enhanced the electronic transport. The parameters like dielectric constant, dielectric loss, a.c conductivity is slightly varies at

low temperature and frequency range up to 4 MHz but such parameters have larger values at frequency 10 MHz at (x = 0, 0.3, 0.6). From the results, the compound can be used at high frequencies in different applications. The mechanism of a.c conductivity is matched with correlated barrier height.

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