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Pre-crystallization study of $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ (where $x = 6$ and 8 at. %) chalcogenide glasses using differential calorimetric technique

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ABSTRACT

Pre-crystallization kinetic study of the $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ system (where $x = 6$ and 8 at. %) has been carried out under non-isothermal conditions. Two endothermic peaks have been detected in the DTA traces of the system where each peak is characterized by an onset temperature (T_o) and peak temperature (T_p). Five models have been used to analyze our results. The apparent activation energy of glass transition at the onset temperature (E_o) and at the peak temperature (E_p) have been carried out by means of three methods (Kissinger, Mahadevan and Ozawa-Chen models), while Matusita and JMA models were used to deduce (E_o) and (E_p) by means of the area analysis method of the endothermic peaks. The results indicated that (T_p) was the most suitable temperature to deduce the apparent activation energy of glass transition when Kissinger, Mahadevan and Ozawa-Chen models were applied. Kissinger's model was found to be the most suitable one to analyze our results when the former three methods were applied, while Ozawa-Chen model was the most suitable model when all the five models were considered. Avrami index (n) and the reaction factor (K_o) were deduced, where two and one dimensional regimes of growth for the first and second endothermic peaks respectively were found.

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KEYWORDS

Chalcogenide glasses;
Glass transition;
Non-isothermal conditions;
Pre-crystallization kinetic.

1. INTRODUCTION

The study of phase transformation of chalcogenide glasses has gained great attention because of their important applications^[1-4]. Based on the fact that the chalcogenide glasses combine the characteristic features of the disordered systems and some properties of the crystalline semi-conducting materials, their wide promising technological applications in solid state devices particularly in the reversible phase change optical recording,

fabrication of the inexpensive solar cells, optical fibers, photo resists, photo conductors, have been given a considerable interest^[5,6]. The thermal analysis methods including differential thermal analysis (DTA) and differential scanning calorimetry (DSC) are particularly important as they: (a) are easy to carry out, (b) require little sample preparation, (c) are quite sensitive and, (d) are relatively independent of the sample geometry.

The crystallization kinetics of chalcogenide glasses plays an important role in determining both their practi-

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cal applications, thermal stability and their transport mechanisms. The calorimetric study techniques have been widely used to study the crystallization kinetics of glasses, they are isothermal and non-isothermal techniques^[7,8]. It has been found^[9-11] that the later one is the most suitable technique to gain insight into the thermal stability and the phase transformation behavior of chalcogenide glasses. In this technique, the sample is thermally annealed at a fixed rate of heating and the physical property is recorded as a function of temperature, where information of some aspects of the crystallization process can be obtained.

DTA was used to study the variation of both glass transition temperature (T_g) and crystallization temperature (T_c) with the heating rate (α)^[12-16]. Through the crystallization kinetics, the kinetic parameters such as, the activation energy of crystallization (E_c), the activation energy of glass transition (E_g), the Avrami exponent (n) and the frequency factor (K_0) can be deduced^[17-22].

Understanding the glass transition kinetics of chalcogenide glasses is very important to establish their stability, glass forming ability and ultimately to determine the useful range of operating temperature for specific technological application before the eventual crystallization takes place.

The aim of the present work is to analyze the endothermic (glass transition) peaks, in the same way as that made on the exothermic (crystallization) peaks, to obtain the pre-crystallization parameters for the system $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ (where $x = 6$ and 8 at. %). No reported studies on crystallization kinetics in these materials have been carried out. According to the above points of view, five different methods of analysis have been used to study the pre-crystallization kinetics of the investigated system.

2. EXPERIMENTAL

Bulk chalcogenide glasses of the $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ system (where $x = 6$ and 8 at. %) were prepared using the usual melt quenching technique. Elemental constituents of 5N purity weighed according to their atomic percentages and were sealed in an evacuated (10^{-5} Torr) silica tubes and heated gradually up to 1223 K for 15 hours. Continuous stirring of the melt was carried out to ensure homogeneity. The melt was then rapidly quenched in ice water. The glassy nature of the prepared samples was confirmed by X-ray diffraction (XRD) technique using Shimadzu XD-3 diffractometer

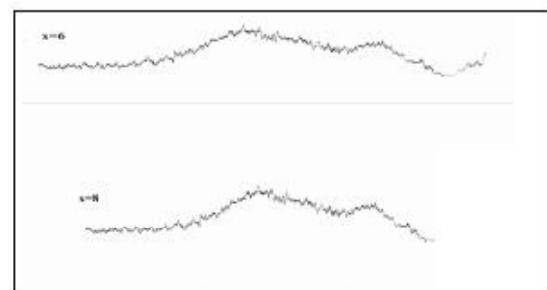


Figure 1: X-ray diffraction patterns for as-quenched $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ (where $x=6$ and 8 at. %) chalcogenide glasses

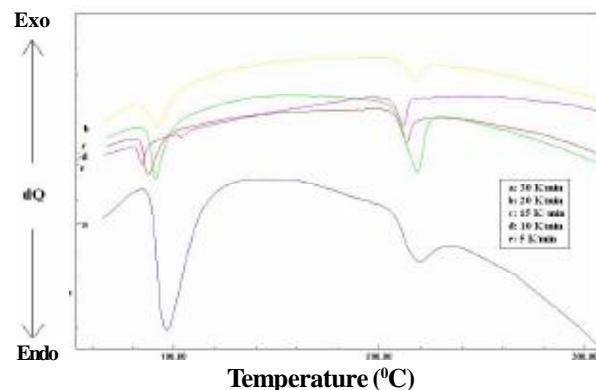


Figure 2 : DTA traces for $\text{Ge}_{15}\text{Te}_{79}\text{In}_x$ chalcogenide glasses

with scanning velocity of (20/min) and radiation source of Cu foil as shown in figure 1.

The prepared glasses were ground to obtain fine powder for DTA studies. The thermal behavior was investigated using a differential thermal analyzer (Shimadzu DTA-50). The measurements were carried out under nitrogen atmosphere. 5-10 mg of each sample was heated at constant heating rate and the changes in the heat flow with respect to an aluminum pan were recorded under a constant flow of nitrogen at a rate of 20 ml/min. The heating rates were varied from 5 to 30 K/min. The best fit for the obtained data points was made using the least square method.

3. RESULTS AND DISCUSSIONS

3.1. The dependence of the glass transition on the heating rate

DTA thermograms for $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ (where $x = 6$ and 8 at. %) chalcogenide glasses, at five different heating rates (5, 10, 15, 20 and 30 K/min), have been obtained as shown in figures 2 and 3, where two endothermic peaks are observed.

The differential thermal analysis of the sample $\text{Ge}_{15}\text{Te}_{77}\text{In}_6$ is shown in figure 4, as a representative curve, where the onset temperatures (T_{o1}, T_{o2}) and the peak temperatures (T_{p1}, T_{p2}), for the first and second endothermic peaks, can be observed.

The endothermic peak is the characteristic feature of the glass transition temperature. The appearance of two glass transition temperatures has been observed in many chalcogenide glasses^[23-28] which indicates an unusual phase separation process occurring in these glasses during the heat treatment. The appearance of the endothermic peak arises due to the abrupt increase in the specific heat of the samples. The first endothermic peak (FEP) and the second endothermic peak (SEP), which represent the strength or the rigidity of the glassy structure, are characterized by the onset temperature (T_o) and the peak temperature (T_p).

The dependence of (T_{o1}) and (T_{p1}) for FEP and SEP, on the heating rate (α) is shown in figures 5 and 6 for $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ (where $x = 6$ and 8 at. %) chalcogenide glasses. It can be noticed that the curves show linear relationships verifying the following empirical equation^[28]:

$$T_g = A + B \ln(\alpha) \quad (1)$$

Where A and B are constants for the heated samples and (α) is the heating rate in (K/min) employed in DTA runs.

The deduced values of A and B for the two endothermic peaks of $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ (where $x = 6$ and 8 at. %) are given in TABLE 1. The values of the constant B at (T_{o1}) and at (T_{p1}) are found to decrease with the increasing from (FEP) to (SEP) for either of $x = 6$ or $x = 8$ at. %. As the indium content increases in the sample, the deduced values of B show an increasing behavior at (T_{o1}) and (T_{p1}) for (FEP), while they show a decreasing behavior at the same temperatures for (SEP). It has been reported earlier^[27] that the value of B in equation (1) is related to the cooling rate of the melt: the lower the cooling rate of the melt, the lower the value of B, which indicates that B is related to the response of the configurationally changes with glass transition. For both of $x = 6$ and $x = 8$ at. % the intercept (A) is found to increase with the increasing in either the position of the endothermic peak or to the indium content in the sample which indicates that (A) is a composition and temperature dependent. From the values of (A), it can be noticed also that they represent the lowest values of transition temperatures at (T_{o1}) and (T_{p1}) which belong to either (FEP) or (SEP).

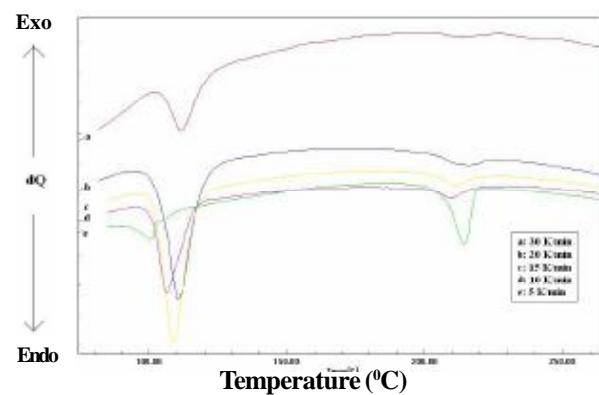


Figure 3: DTA traces for $\text{Ge}_{15}\text{Te}_{77}\text{In}_6$ chalcogenide glasses

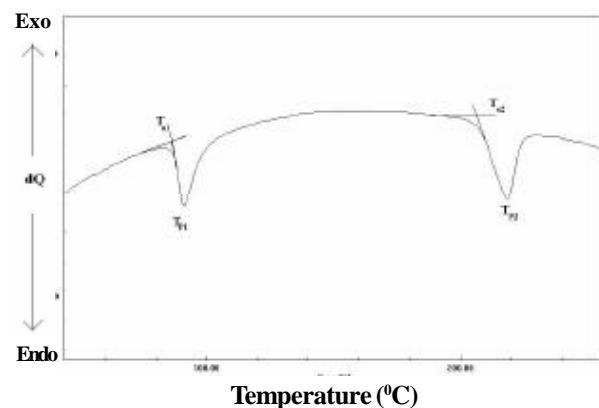


Figure 4: DTA trace for $\text{Ge}_{15}\text{Te}_{79}\text{In}_6$ at rate 15 K/min

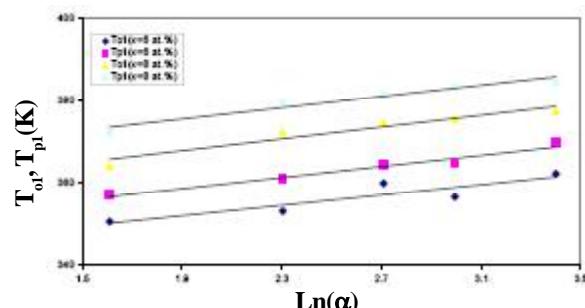


Figure 5 : T_{o1}, T_{p1} versus $\ln(\alpha)$ for $\text{Ge}_{15}\text{Te}_{79}\text{In}_6$ and $\text{Ge}_{15}\text{Te}_{77}\text{In}_8$ chalcogenide glass at FEP

TABLE 1

$\text{Ge}_{15}\text{Te}_{79}\text{In}_6$				$\text{Ge}_{15}\text{Te}_{77}\text{In}_8$			
FEP		SEP		FEP		SEP	
T_o	T_p	T_o	T_p	T_o	T_p	T_o	T_p
B	6.297	6.7408	1.651	4.628	7.261	6.848	1.256
A	340.01	345.72	475.85	476.79	353.96	362.42	473.99
							483.63

3.2. The activation energy of glass transition

In this study, the analysis and calculations will carry out for the glass transition temperatures as well as the crystallization temperatures. Five different models have

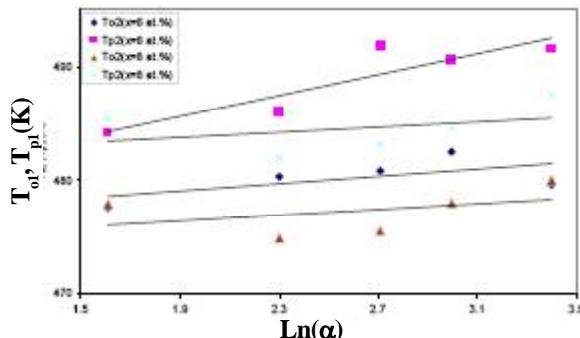
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Figure 6 : T_{o2} , T_{p2} versus $\ln(\alpha)$ (a) for $Ge_{15}Te_{79}In_6$ and $Ge_{15}Te_{77}In_8$ chalcogenide glass at SEP

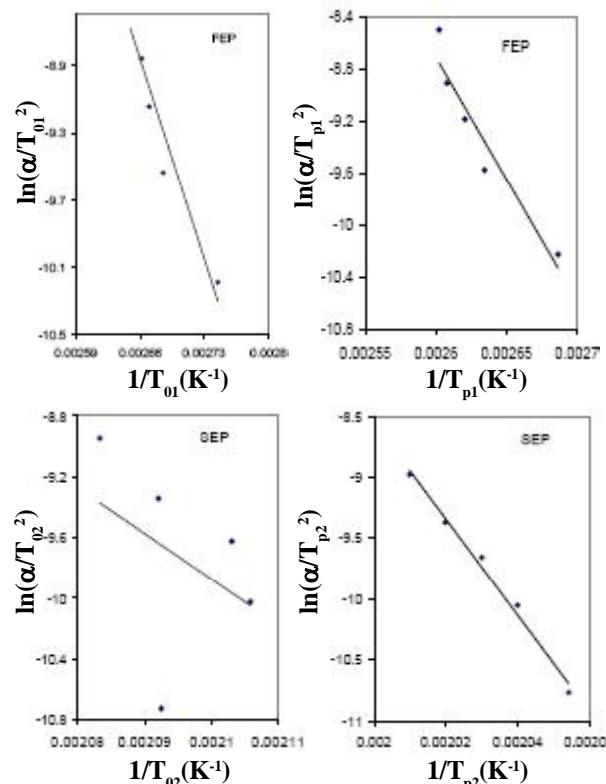


Figure 7: $\ln(\alpha/T^2)$ against $(1/T)$ for $Ge_{15}Te_{79}In_6$ chalcogenide glasses at FEP and SEP

been used to calculate the apparent activation energy of glass transition for the two endothermic peaks which appeared in the DTA traces of the system $Ge_{15}Te_{85-x}In_x$ (where $x = 6$ and 8 at. %). The transition process is generally understood in terms of the kinetic parameters such as the activation energy of glass transition (E_g), the Avrami index (n) and the frequency factor (K_o).

3.2.1. Kissinger model^[29]

This model has been widely used^[11,12,30,31,32,33] to determine the activation energy of glass transition (E_g). In the following, the activation energy of glass transition

Figure 8: $\ln(\alpha/T^2)$ against $(1/T)$ for $Ge_{15}Te_{77}In_8$ chalcogenide glasses at FEP and SEP

at (T_o) and (T_p) due to (FEP) and (SEP) of the $Ge_{15}Te_{85-x}In_x$ system (where $x = 6$ and 8 at. %) will be analyzed according to the following relation^[29].

$$\ln\left(\frac{\alpha}{T_g^2}\right) = \frac{-E_g}{RT_g} + \text{constant} \quad (2)$$

Where (E_g) is the apparent activation energy of glass transition and (R) is the gas constant. According to equation (2), plotting of $\ln(\alpha/T_g^2)$ against $(1/T_g)$ gives straight lines as shown in figures 7 and 8. From the slopes of these lines, the apparent activation energy of glass transition at the onset temperature (E_o) and at the peak temperature (E_p) for both FEP and SEP were deduced and given in TABLE 2.

3.2.2. Mahadevan model^[9]

Mahadevan et. al have been simplified Kissinger's equation to the following formula^[9]:

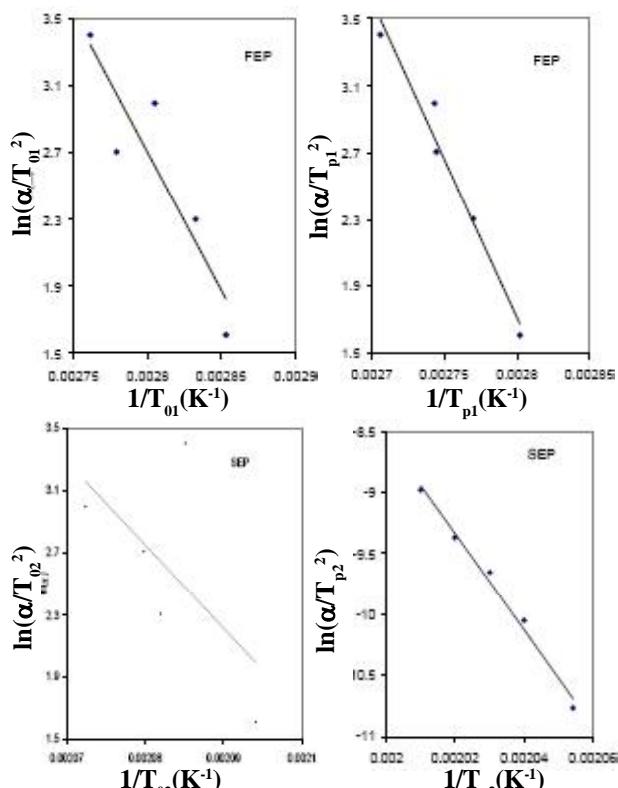
$$\ln(\alpha) = \frac{-E_g}{RT_g} + \text{constant} \quad (3)$$

The variations of $\ln(\alpha)$ against $(1/T_g)$ for the investigated samples at different heating rates are shown in figures 9 and 10 where straight lines verifying equation (3) are obtained.

The deduced values of the apparent activation en-

TABLE 2

Models	Ge ₁₅ Te ₇₉ In ₆				Ge ₁₅ Te ₇₇ In ₈			
	FEP		SEP		FEP		SEP	
	E _o (kJ/mol)	E _p (kJ/mol)	E _o (kJ/mol)	E _p (kJ/mol)	E _o (kJ/mol)	E _p (kJ/mol)	E _o (kJ/mol)	E _p (kJ/mol)
Kissinger	131.87	151.33	439.31	341.30	139.90	155.55	250.52	327.56
Mahadevan	137.79	157.36	447.29	349.41	142.89	161.83	258.45	328.51
Ozawa-Chen	134.83	154.34	443.30	354.36	142.98	158.69	254.49	328.03
Mean value using the above three models	134.83	154.34	443.30	348.36	141.92	158.69	254.49	328.03
Matusita		150.19		311.55		115.18		326.23
JMA		152.24		366.85		161.19		258.58
Mean value using the five models		153.89		344.69		150.49		313.78

Figure 9: ln(α) against (1/T) for Ge₁₅Te₇₉In₆ chalcogenide glasses at FEP and SEP

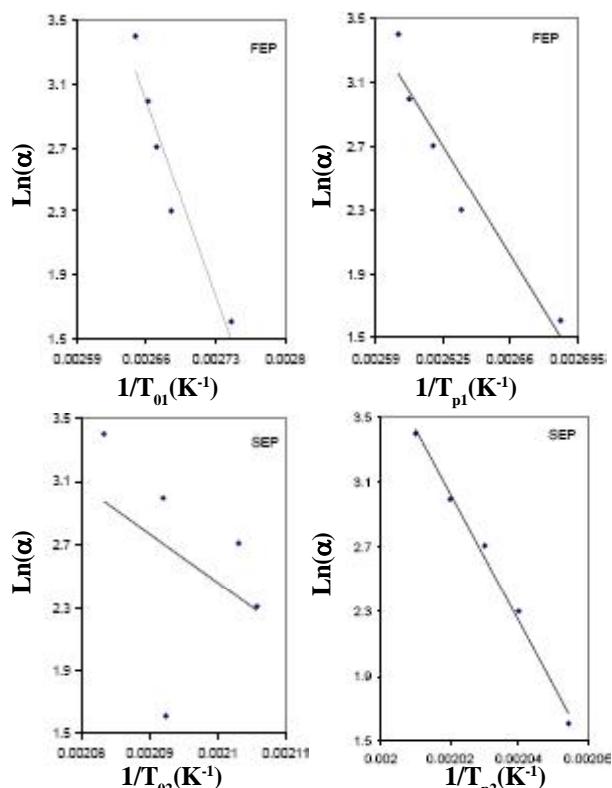
ergy of glass transition at the onset temperature (E_o) and at the peak temperature (E_p) for both FEP and SEP are given in TABLE 2.

3.2.3. Ozawa-Chen model^[34,35]

Ozawa^[34] and Chen^[35] have been proposed a model to determine the apparent activation energy of glass transition according to the equation:

$$\ln\left(\frac{\alpha}{T_g}\right) = \frac{-E_g}{RT_g} + \text{constant} \quad (4)$$

Plots of for the investigated samples are shown in figures 11 and 12. Straight lines could be fitted to give

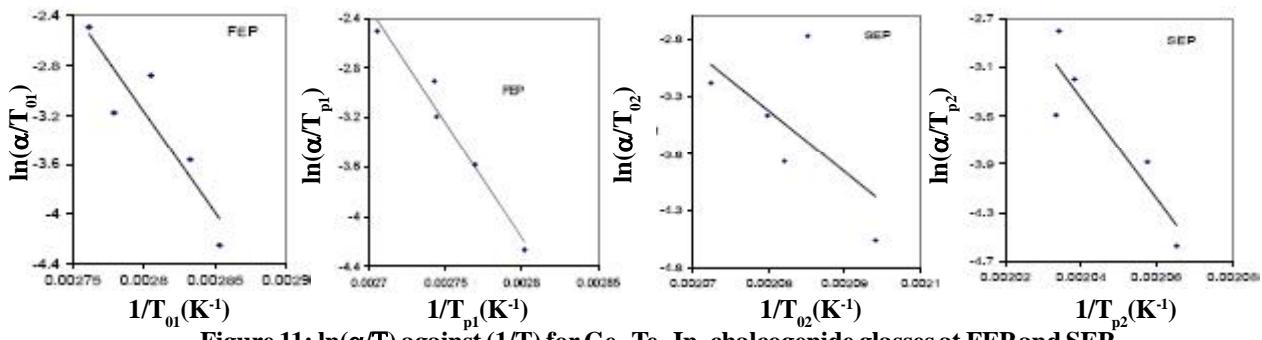
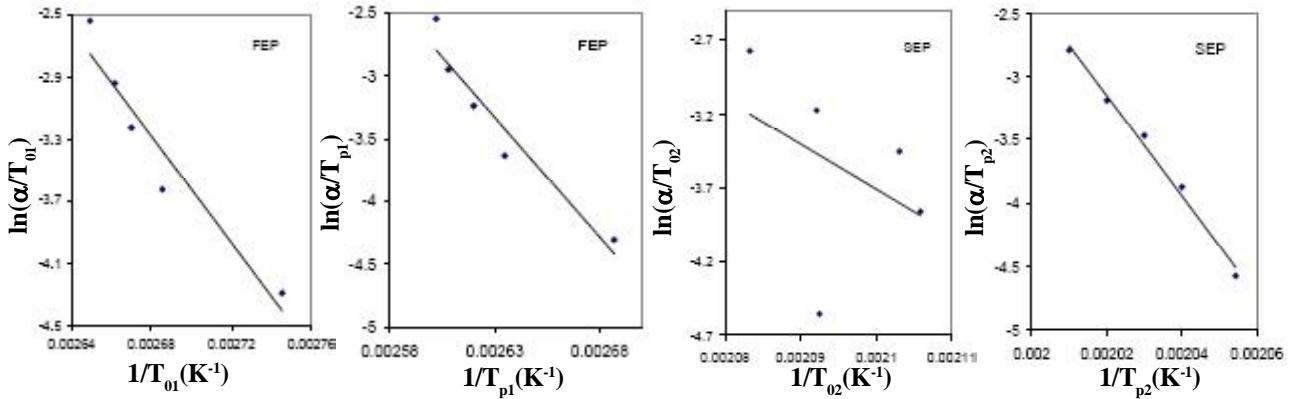
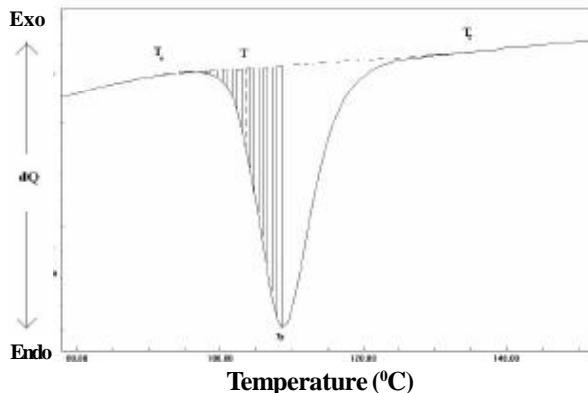
Figure 10: ln(α) against (1/T) for Ge₁₅Te₇₇In₈ chalcogenide glasses at FEP and SEP

the corresponding activation energies.

The deduced values of the activation energy of glass transition according to equation (4) at the onset and peak temperatures for the two endothermic peaks are given in TABLE 2.

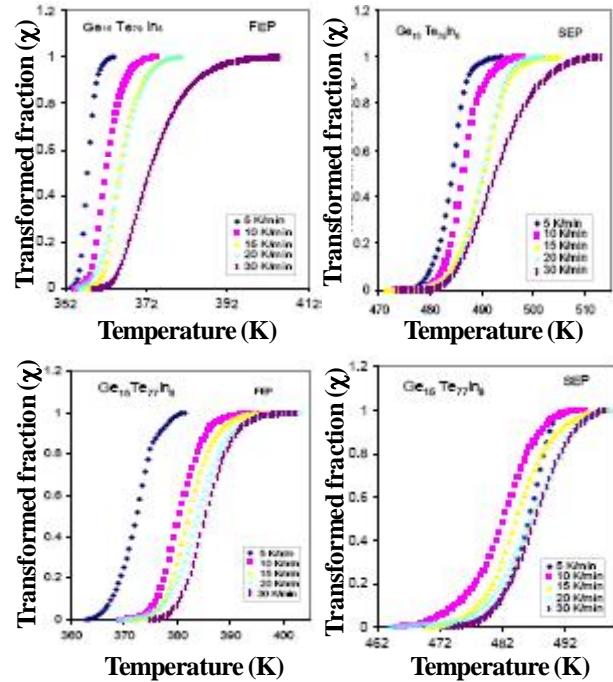
3.2.4. Matusita model^[36]

The crystallization kinetic parameters have been obtained using the model suggested specifically for non-isothermal crystallization by Matusita et al^[36]. According to DTA traces, we can say that the fraction (χ) that transformed into the glassy state at any temperature (T)

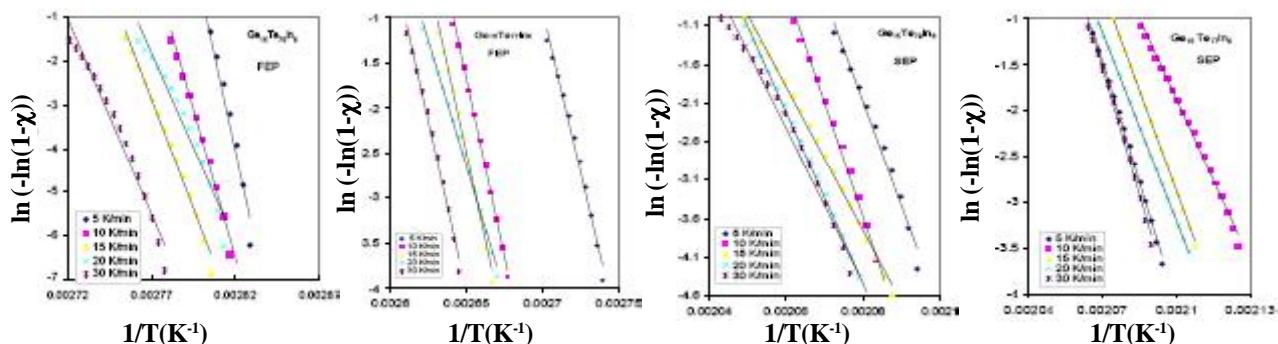
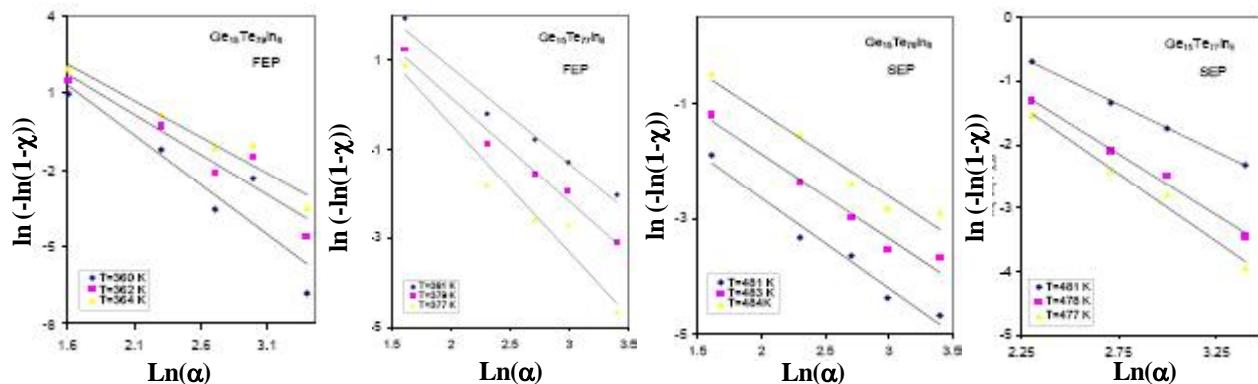
Full PaperFigure 11: $\ln(\alpha/T)$ against $(1/T)$ for $Ge_{15}Te_{79}In_6$ chalcogenide glasses at FEP and SEPFigure 12: $\ln(\alpha/T)$ against $(1/T)$ for $Ge_{15}Te_{77}In_8$ chalcogenide glasses at FEP and SEPFigure 13 : Representative curve for calculating the fraction (χ)

is given as where (A) is the total area of the endothermic peak between the onset temperature (T_o), at which the glass transition just begins and the temperature (T_f) at which the glassy state is completed and is the area between (T_o) and any temperature (T). A representative curve for calculating the fraction (χ) is shown in figure 13.

Figure 14 shows the relationship between the transformed volume fraction (χ) and the heating temperature for the investigated samples at different heating rates.

Figure 14: The transformed fraction (χ) versus temperature of the investigated samples at FEP and SEP

Matusita's model can be applied on the amorphous glass transition. The values of (χ) are related to the apparent

Figure 15: $\ln(-\ln(1-\chi))$ versus $1/T$ at different heating rates for the investigated samples at FEP and SEPFigure 16: $\ln(-\ln(1-\chi))$ versus $\ln(a)$ at different heating rates for the investigated samples at FEP and SEPTABLE 3 : The values of (n), the frequency factor (K_0) and the mean value of (nE_g) at (FEP) and (SEP) for the $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ system (where $x = 6$ and 8 at. %)

	$\text{Ge}_{15}\text{Te}_{79}\text{In}_6$		$\text{Ge}_{15}\text{Te}_{77}\text{In}_8$	
	FEP	SEP	FEP	SEP
n	3.11	1.486	2.461	1.84
nE_g (kJ/mol.)	468.31	462.966	283.476	600.26
K_0	3.132×10^{19}	1.71×10^{25}	1.675×10^{19}	2×10^{36}

TABLE 4 : The values of (n) and (m) for various crystallization mechanisms

Mechanism	n	m
Bulk nucleation		
Three – dimensional growth	4	3
Two – dimensional growth	3	2
One – dimensional growth	2	1
Surface – dimensional growth	1	1

activation energy of glass transition through the relation^[36]:

$$\ln[-\ln(1-\chi)] = -n \ln(\alpha) - 1.052 \left[\frac{mE_g}{RT} \right] + \text{constant} \quad (5)$$

Where (m) and (n) are related to the dimensionality of growth.

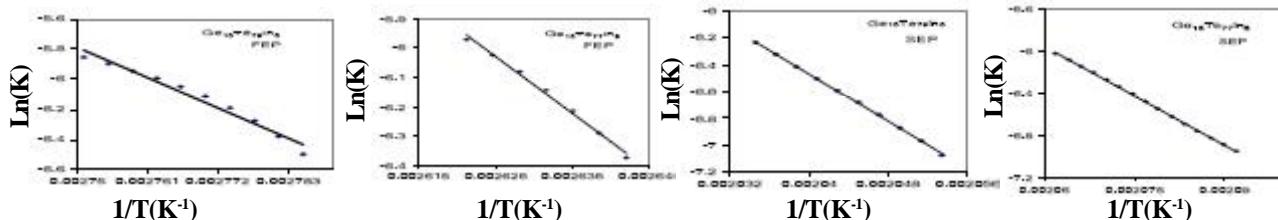
Figure 15 shows the relation between $\ln[-\ln(1-\chi)]$ and $(1/T)$ at different heating rates for the investigated samples. When the nuclei are formed during the thermal analysis, the factor (n) is equal to (m+1) and when nuclei are formed during any previous heat treatment

prior to thermal analysis (n) is equal to (m). The index (n) is considered to be equal to the parameter (m) because the amorphous-glass transition reaction occurs through one step^[33]. From the slope of each straight line, the value of (nE_g) is obtained and the average value for each endothermic peak of the investigated samples is deduced and given in TABLE 2.

In order to determine the Avrami index n, the relation between $\ln(-\ln(1-\chi))$ and $\ln(a)$ at fixed temperatures are plotted and given in figures 16 for $\text{Ge}_{15}\text{Te}_{79}\text{In}_6$ and $\text{Ge}_{15}\text{Te}_{77}\text{In}_8$ according to the formula^[37]:

$$\frac{d[\ln(-\ln(1-\chi))]}{d[\ln(a)]} = -n \quad (6)$$

The obtained values of the index (n) are given in TABLE 4. From TABLE 4 it can be noticed that, for the sample $\text{Ge}_{15}\text{Te}_{79}\text{In}_6$ the value of (n) is equal to 3.11 for (FEP) and equals to 1.48 for (SEP) which indicates that the mechanism of amorphous-glass transition is two and one dimensional regime for the former and the later one respectively. For $\text{Ge}_{15}\text{Te}_{77}\text{In}_8$ the deduced values of (n) are equal to 2.461 and 1.84 for (FEP) and (SEP) respectively, which indicates that the transformation mechanism is two and one dimensional regime for the former and the later one respectively. It is clear that, the

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transformation mechanisms of the investigated samples follow two types of regime: the first endothermic peak follows the two dimensional regime, while the second endothermic peak follows the one dimensional regime. From the values of (n) and the mean value of (nE_g) the associated activation energy of transition, for both of (FEP) and (SEP) due to the investigated samples, are deduced and given in TABLE 2. It can be noticed that the values of the activation energy of glass transition due to (FEP) are much smaller than those due to (SEP) which may be attributed to that the transformation mechanism at (FEP) is two dimensional regime while at (SEP) is one dimensional regime. Therefore, the process at (SEP) may leads to another further transition to attain the stable state with a lower activation energy as reported earlier^[38-40].

3.2.5. Johnson-Mehl-Avrami (JMA) model^[41-43]:

The relation between the volume fraction (χ) crystallized in time (t) and the reaction rate constant is given, on the basis of the theory of transformation kinetics, by the equation^[41-43]:

$$\chi(t) = 1 - \exp[-(K t)^n] \quad (7)$$

Where (n) is an integer or half-integer which depends on the mechanism of growth and dimensionality of the crystal growth^[44] and (K) is the reaction rate constant whose temperature dependence is expressed in Arhenian – type equation:

$$K = K_o \exp\left(\frac{-E_g}{R T}\right) \quad (8)$$

Where (K_o) is the frequency factor, (E_g) is the apparent activation energy of glass transition and (R) is the gas constant.

Equations (7) and (8) have been widely used to describe the non-isothermal crystallization kinetics. The application of JMA model, under the non-isothermal crystallization, assumes that the heat rate (α) is constant. Equation (7) can be rearranged and written in the form:

$$K = \frac{[-\ln(1-\chi)]^{1/n}}{t} \quad (9)$$

Equations (7), (8) and (9) can be applied to calculate the activation energy of glass transition at (FEP) and (SEP) for the $Ge_{15}Te_{85-x}In_x$ system (where $x = 6$ and 8 at. %). Substituting by the obtained values of (n) and the temperature (T) instead of the time (t), in equation (9), then the values of K at different temperatures can be obtained at constant heating rates. According to equation (8), plots of $\ln(K)$ versus $(1/T)$ for (FEP) and (SEP) are shown in figure 17 where straight lines can be observed and the corresponding activation energy of glass transition were deduced and given in TABLE 2. The intercepts of these straight lines with the vertical axis give the values of the frequency factor (K_o), which are given in TABLE 2. These values are found to be in good agreement with other published data^[33,45].

From TABLE 2 it can be noticed that five theoretical models were applied to analyze the non-isothermal amorphous-glass transition. According to the former three models (Kissinger, Mahadevan and Ozawa-Chen), the deduced activation energy of glass transition due to the onset temperature (E_{to}) is much less than that deduced due to the peak temperature (E_p). The mean values of (E_o) due to (FEP) and (SEP) for $Ge_{15}Te_{79}In_6$ are 134.83 kJ/mol and 443.30 kJ/mol while the mean values of (E_p) are found to be 154.34 kJ/mol and 348.36 kJ/mol for (FEP) and (SEP) respectively. The results of $Ge_{15}Te_{77}In_8$, which given in TABLE 2, indicate that the mean values of (E_o) due to (FEP) and (SEP) are 141.92 kJ/mol and 254.49 kJ/mol, while the mean values of (E_p) are 158.69 kJ/mol and 328.03 kJ/mol due to (FEP) and (SEP), respectively. From the above results and the mean values of the activation energy of transition it can be noticed that Ozawa-Chen model is the most suitable model that can be applied for our investigation of non- isothermal kinetic analysis if the former three models are only applied.

From TABLE 2 it can also be realized that the values of the activation energy of glass transition (E_o and E_p), according to Matusita and JMA models due to (FEP) and (SEP) for the investigated samples, are very

close to those obtained at (T_p) due to Kissinger, Mahadevan and Ozawa-Chen models. This means that the peak temperature of the endothermic peak (T_p) may be considered to be more suitable one to calculate the activation energy of glass transition than that at the onset temperature(T_o). Therefore, the mean value of the five activation energy due to the mentioned five models is taken as the mean value of (E_p) which are obtained using Kissinger, Mahadevan and Ozawa-Chen models and the values obtained using Matusita and JMA models. So, when the five models are applied on $\text{Ge}_{15}\text{Te}_{85-x}\text{In}_x$ (where $x = 6$ and 8 at.%) and comparing the mean value of the apparent activation energy due to these models with the value obtained by each model separately it can be noticed that Kissinger model is the most suitable one that characterizes our results efficiently.

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