

Thermal Conductivity of Lithium-Boro Silicate Glasses

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ABSTRACT: Three systems of lithium borosilicate (LBS) glasses namely SI $42.5\text{Li}_2\text{O} : (57.5-x) \text{B}_2\text{O}_3 : x\text{SiO}_2$, SII $42.5\text{Li}_2\text{O} : x\text{B}_2\text{O}_3 : (57.5-x) \text{SiO}_2$ where $x=0, 5, 10, 20$, and 30 , and SIII $(100-2x) \text{Li}_2\text{O} : x\text{B}_2\text{O}_3 : x\text{SiO}_2$ where $x=30, 28.75, 27.5, 25$, and 22.5 , are prepared using conventional melt quenching technique. Glass has been studied under the heading of solid electrolytes. Glass is a kind of matter (solid electrolyte), which also gives the conductance at high temperature, due to phonon-phonon interaction. Here, the $\text{Li}_2\text{O}-\text{B}_2\text{O}_3-\text{SiO}_2$ glass systems have been studied, in case of thermal conductivities. The variations in rate of heat flow (mW) and heat capacity ($\text{mJ}/^\circ\text{C}$) per min with respect to rate of temperature ($^\circ\text{C}/\text{sec}$) are studied because theoretically these two change simultaneously in DSC scans. Thus values of K are determined from the DSC data.

KEYWORDS: Thermal conductivity, Phonon-Phonon interaction, Impurities, Solid electrolyte.

I. INTRODUCTION

Thermal Conductivity is a measure of the ability of a material to transfer heat. Given two surfaces on either side of a material with a temperature difference between them, the thermal conductivity is the heat energy transferred per unit time and per unit surface area, divided by the temperature difference. It is a well known fact that the glass is a bad conductor of heat. In materials, the conduction of heat has been due to phonon-phonon interaction. When heat energy is supplied, crystal expands and due to this expansion, amplitude of atomic vibrations also increases, which have been responsible for increasing the coefficient of thermal expansion.

In pure metals the total conduction has been due to electrons and phonons, however, in poor metals, containing large amount of impurities, the lattice conductivity has been important. A Debye's¹ theory of thermal conductivity of insulators shows that the lattice vibrations may be described by a model, in which elastic waves are propagated through a continuum. Since, solids expand on heating, the elastic waves is a combination of harmonic and an-harmonic types of waves. The an-harmonic waves acts as source of coupling between the lattice and electronic waves, so that the mutual scattering becomes possible. Peierls and Umklapp⁽²⁾ extended these ideas in terms of phonon-phonon interaction. The phonon-phonon collisions restore the thermal equilibrium, due to temperature gradient of solid, resulting into thermal resistance. Peierls and Umklapp² propagated that the thermal conductivity decreases exponentially with temperature, T, in the low-temperature region and decreases as reciprocal of temperature, in the high-temperature region. This evoked enough interest to under take the study on thermal conductivity of especially amorphous type which is Meta stable and very sensitive to temperature. This study provides understanding of thermal process in terms of phonon contribution, in the disordered materials like glasses. Therefore, a schematic study of thermal conductivity is undertaken.

II. EXPERIMENTAL

The starting materials B_2O_3 , Li_2O and SiO_2 ; taken in appropriate proportions in mole percent of these materials; by weighing to the accuracy of 0.01mg using a, Metter AE 163 balance, mixed thoroughly under acetone, after grinding materials, are taken in porcelain crucible and introduced into an electrical furnace maintained at 1100°C . The mixture is melted for about 30-40 minutes and melt was shaken frequently to achieve homogeneity. The melt was quickly poured onto a hot stainless steel plate (maintained at 373°K) and pressed with a steel rod. The fresh samples are annealed at the temperature 373°K to relieve internal stresses. The glasses formed are clear, transparent with light

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yellowish tint. The flat porcelain of the specimen *i.e.* glass (obtained after polishing) were sputtered (platinum) to ensure good adhesion. The compositions of the glass samples are given in table.1. The bubble-free glass samples of thickness 0.25 to 1mm are employed in measurements.

Table 1. Series I, II and III with compositions:

Serie s	Composion	Serie s	Composion	Serie s	Composion
I	Li ₂ O:B ₂ O ₃ :Si O ₂	II	Li ₂ O:B ₂ O ₃ :SiO ₂	III	Li ₂ O:B ₂ O ₃ :SiO ₂
SI.1	42.5 : 57.5 : 0	SII.1	42.5 : 0 : 57.5	SIII.1	40 : 30 : 30
SI.2	42.5 : 52.5 : 5	SII.2	42.5 : 5 : 52.5	SIII.2	42.5 : 28.75: 28.75
SI.3	42.5 : 47.5 : 10	SII.3	42.5 : 10 : 47.5	SIII.3	45 : 27.5 : 27.5
SI.4	42.5 : 37.5 : 20	SII.4	42.5 : 20 : 37.5	SIII.4	50 : 25 : 25
SI.5	42.5 : 27.5 : 30	SII.5	42.5 : 30 : 27.5	SIII.5	55 : 22.5 : 22.5

Table 1 show the sample prepared in mol% in three series namely, SI, SII and SIII; with different compositions denoting by SI.1, SII.1 and SIII.1 and so on.

III. ESTIMATION OF THERMAL CONDUCTIVITY FROM DSC.

The time, temperature, heat flow, reversible heat flow, non-reversible heat flow and heat capacity, of all the samples were estimated from the DSC data. These were used to evaluate the rate of heat flow and rate of change of temperature with distance in the direction of heat flow (*i.e.* temperature gradient). Thermal conductivity for each sample was determined by using the Fourier's law,

$$\frac{\Delta Q}{\Delta t A} = -K \frac{\Delta T}{\Delta x}$$

Where, $\frac{\Delta Q}{\Delta t A}$ = power per unit area transported,

$\frac{\Delta T}{\Delta x}$ = temperature gradient, and $-K$ = Thermal conductivity

Calculated values of thermal conductivities with different temperatures are noted in Table 1, 2 & 3.

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Table 1: Temperature in °c and Thermal conductivity for system I (SI).

T °C	SI.1	SI.2	SI.3	SI.4	SI.5
300	2.9E-05	4.5E-05	1.7E-04	1.6E-04	7.3E-05
325	2.7E-05	4.2E-05	1.5E-04	1.3E-04	6.9E-05
350	2.5E-05	3.9E-05	1.4E-04	9.5E-05	6.6E-05
375	2.3E-05	3.7E-05	1.2E-04	6.4E-05	6.2E-05
400	2.2E-05	3.4E-05	1.0E-04	3.6E-05	5.8E-05
425	2.0E-05	3.3E-05	9.0E-05	1.1E-05	5.4E-05

Table1 indicates thermal conductivity K ($\text{Wm}^{-1}\text{k}^{-1}$), of system one denoting by SI and for different compositions as SI.1 - SI.5; along with temperatures in °c having maximum value, of thermal conductivity for system SI.3.

Table 2: Temperature in °c and Thermal conductivity for system II (SII).

T °C	SII.1	SII.2	SII.3	SII.4	SII.5
300	3.0E-04	2.8E-04	8.5E-04	1.5E-04	1.8E-04
325	2.4E-04	2.2E-04	6.9E-04	1.3E-04	1.7E-04
350	1.8E-04	1.5E-04	5.5E-04	1.1E-04	1.6E-04
375	1.3E-04	9.8E-05	4.0E-04	9.5E-05	1.5E-04
400	8.9E-05	6.4E-05	2.7E-04	8.4E-05	1.5E-04
425	5.3E-05	2.0E-05	1.3E-04	7.2E-05	1.4E-04

Table 2 indicates thermal conductivity K ($\text{Wm}^{-1}\text{k}^{-1}$), of second system denoting by SII, for different compositions as SII.1 - SII.5; along with temperatures in °c having maximum value, of thermal conductivity for system SII.3.

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Table 3: Temperature in °C and Thermal conductivity for system III (SIII).

T °C	SIII.1	SIII.2	SIII.3	SIII.4	SIII.5
300	1.4E-04	1.4E-05	1.3E-04	9.5E-05	8.0E-05
325	1.1E-04	1.3E-05	1.1E-04	8.7E-05	7.3E-05
350	8.4E-05	1.2E-05	8.2E-05	8.0E-05	7.0E-05
375	6.1E-05	1.2E-05	6.5E-05	7.3E-05	6.2E-05
400	3.8E-05	1.1E-05	4.8E-05	6.8E-05	5.9E-05
425	2.1E-05	1.0E-05	3.5E-05	6.0E-05	5.5E-05

Table 3 indicates thermal conductivity K ($\text{Wm}^{-1}\text{k}^{-1}$), of system third, denoting by SII, for different compositions as SIII.1 - SIII.5; along with temperatures in °C having smaller values, of thermal conductivity then system SI.3 and SII.3 due to the same values of two glass forms.

IV. EXPERIMENTAL RESULTS

Figure 1, 2 & 3 shows the reciprocal values of temperature and logarithmic thermal conductivity K ($\text{Wm}^{-1}\text{k}^{-1}$) for system SI, SII and SIII respectively with compositional dependence of activation energy is depicted in the inset of the same figure denoting by (a) and (b).

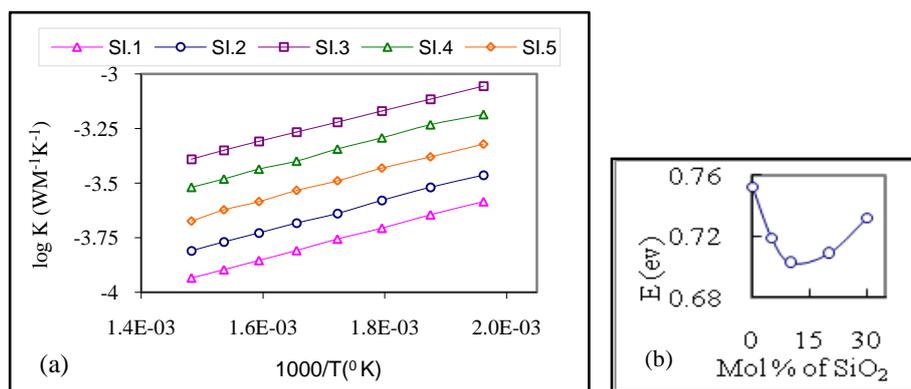


Figure 1: [SI (a,b)] Functional dependence of thermal conductivity on temperature and variation of activation energy with mol % of SiO_2 .

Figure 1(a) shows the reciprocal values of temperature and logarithmic thermal conductivity K ($\text{Wm}^{-1}\text{k}^{-1}$) for system SI, it indicates that SI.3 having higher values of thermal conductivity and (b) shows the variation of activation energy with mol % of SiO_2 minimum activation energy at 10 mol% of SiO_2 , because the addition of SiO_2 increases the tetrahedral network of SiO_4 at the cost of BO_4 . In the glass matrix, this seems to be optimum ratio of the solute (the content of formers) and the solvent the modifier, which in turn gives maximum hopping sites for the Li.

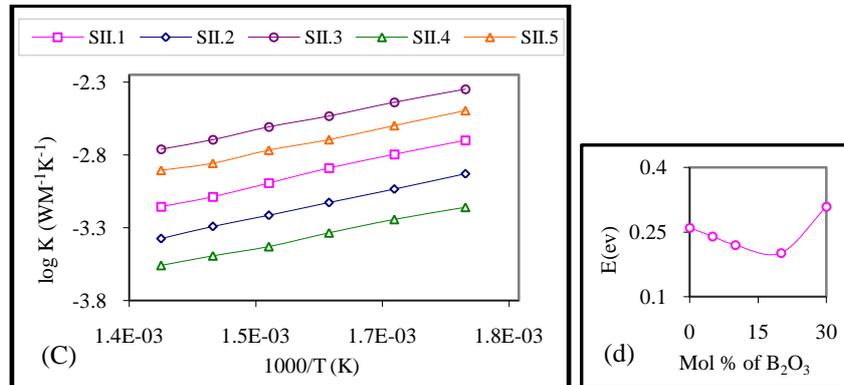


Figure 2: [SII (c, d)] Functional dependence of thermal conductivity on temperature and variation of activation energy with mol % of B_2O_3 .

Figure 2(c) shows the reciprocal values of temperature and logarithmic thermal conductivity $K(\text{Wm}^{-1}\text{k}^{-1})$ for system SII, it indicates that SII.3 having higher values of thermal conductivity and (d) shows the variation of activation energy with mol % of B_2O_3 minimum activation energy at 20 mol% of B_2O_3 .

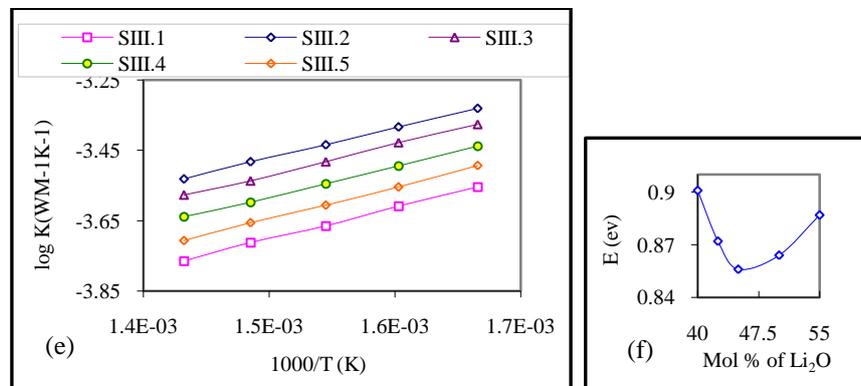


Figure 3: [SIII (e, f)] Functional dependence of thermal conductivity on temperature and variation of activation energy with mol % of Li_2O .

Figure 3(e) shows the reciprocal values of temperature and logarithmic thermal conductivity $K(\text{Wm}^{-1}\text{k}^{-1})$ for system SIII, it indicates that SIII.2 having higher values of thermal conductivity and (f) shows the variation of activation energy with mol % of Li_2O minimum activation energy at 55 mol% of Li_2O .

V. RESULT AND DISCUSSION

V.1. EFFECT OF COMPOSITION

Effect of compositions of lithium-boro silicate glasses are also studied in thermal conductivity with different temperatures and it gives very good satisfactory results. It is discussed for three systems as;

Glasses are not only the super cooled liquid but also thermal disorder predominates with increasing temperature. This indicates temperature sensitiveness of the glass structure. It is understood from Figure 1 that $\text{Li}_2\text{O}-\text{SiO}_2$ glass (SL.1) is having high thermal conductivity than that of SII.1 and SIII.3. This is due to the difference in their micro structure. In this case of $\text{Li}_2\text{O}-\text{SiO}_2$, the tetrahedral of SiO_2 are short linked and viscosity of this glass is higher than that of other two and so in this T_g is higher than that of SII.1 and SIII.3. The $\text{Li}_2\text{O}-\text{B}_2\text{O}_3$ system is a soft glass wherein, BO_3 and BO_4

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linkage persists. Due to BO_3 to BO_4 conversion, Li-ions has high mobility. Therefore in this glass thermal conductivity is higher than that of SII.1 and SIII.3. However, in SIII.3 because of $\text{Li}_2\text{O}-\text{B}_2\text{O}_3$ and $\text{Li}_2\text{O} - \text{SiO}_2$ phases present together to cause some sort of liquid-liquid immiscibility. Due to presence of two types of micro structure the thermal conductivity is lower in magnitude than in SII.1 and SIII.3.

It is well known that the glass is a bad conductor of heat. In such materials (insulators) conduction of heat is due to phonon-phonon interaction. Atomic vibration represents phonons when heat energy in a given solid expands and due to expansion amplitude of atomic vibration also increases, which is responsible for increasing coefficient of thermal conductivity.

In amorphous solids factors like randomness in structure and presence of short range ordering lowers down thermal conduction.

V.2. TEMPERATURE DEPENDENCE OF THERMAL CONDUCTIVITY

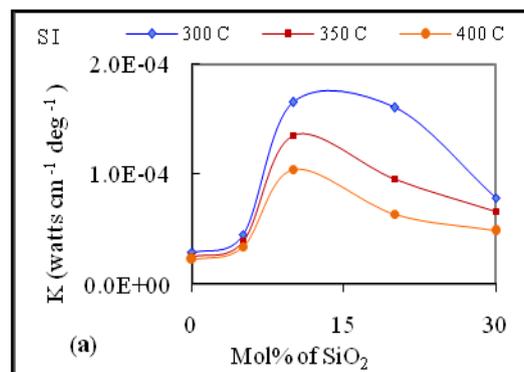


Figure 4 (a): Variation of thermal conductivity with mol % of SiO_2 at different temperature.

From Figure 4 (a) it is observed that thermal conductivity gradually increasing riches maximum at 10 mol % of SiO_2 and then goes on decreasing on increasing mol % of SiO_2 ; which verifies the result obtained from Figure 1.

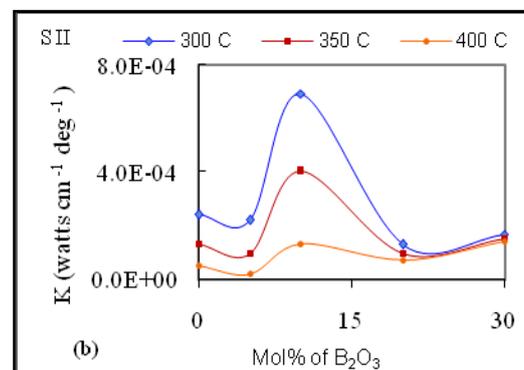


Figure 4 (b): Variation of thermal conductivity with mol % of B_2O_3 at different temperature.

From Figure 4 (b) it is observed that thermal conductivity initially decreasing and then gradually increasing riches maximum at 10 mol % of B_2O_3 and then goes on decreasing on increasing mol % of B_2O_3 ; which verifies the result obtained from Figure 2.

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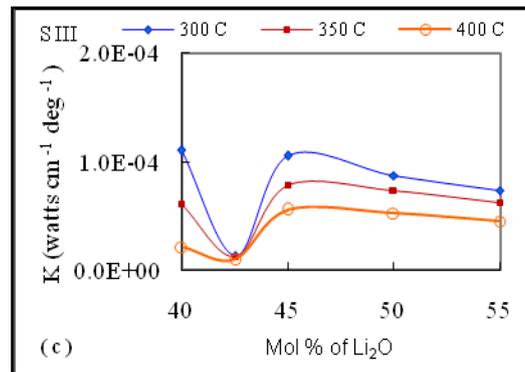


Figure 4 (c): Variation of thermal conductivity with mol % of B₂O₃ at different temperature.

From Figure 4 (c) it is observed that thermal conductivity initially decreasing and then gradually increasing reaches maximum at 45 mol % of Li₂O and then slowly goes on decreasing on increasing mol % of Li₂O; which verifies the result obtained from Figure 3.

Hence from Figure 4 (a, b, c), It is observed that K of different compositions of glasses decreased with increasing temperature, which is in agreement with the findings of Peierls and Umklapp¹. It is well known that the glasses with their completely non crystalline structure are found to have a phonon mean free path, which is limited to the order of inter atomic distance by the random structure. The conductivity increases at low temperature and then reaches a nearly constant value for temperature above a few degrees centigrade. Conductivity of insulator ranges from 10⁻⁸ and 10⁻¹⁸, of metals ranges from 10⁻² to 10⁻⁴ and that of semiconductor lies between these two. The fixing of the mean path by the structure leads to a much more limited range of thermal conductivity values for glasses than for crystals. From the above discussion it is concluded that thermal conductivity depends on an-harmonic coupling. The magnitude of the an-harmonic coupling increases with increase in the vibration amplitude, decreases thermal conductivity.

VI. CONCLUSION

From the above discussion it concludes that thermal conductivity is a dependence of an-harmonic coupling. The magnitude of the an-harmonic increases with increase in the vibration amplitude, which would, decreases the thermal conductivity. Vibration amplitude increases with rise in temperature, thermal conductivity decreases with rise in temperature. Heat transfer across materials of high thermal conductivity occurs at a higher rate than across materials of low thermal conductivity. Correspondingly materials of high thermal conductivity are widely used in heat sink applications and materials of low thermal conductivity are used as thermal insulation. Thermal conductivity of materials is temperature dependent. When heat is applied to a portion of a material, that heat will move through the material. This movement of heat through a material is called the thermal current. Depending on the composition of the atoms of that material, the heat may move very slowly, or it may move very quickly.

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