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Effect of phase separation on the fracture toughness of SiO_2–B_2O_3–Na_2O glass

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Abstract. Fracture toughness of glass is usually poor, due to the absence of grain boundaries and discontinuities. The compositions of the glass studied are in the phase separated region of SiO_2–B_2O_3–Na_2O system. The interface between the glass in glass separation enhances the fracture toughness. The increase in the connectivity of phase separated regions causes increase of fracture toughness from 0·98 through 1·43 to 1·54 MPam$^{1/2}$.

Keywords. Glass; phase separation; fracture toughness; knoop hardness.

1. Introduction

The ternary Na_2O–B_2O_3–SiO_2 is a very important system, which forms the basis of many important commercial glasses, particularly around the phase-separated region. The phase separated characteristics of this ternary system have been thoroughly investigated (Haller *et al* 1970). Glass is a brittle material and the fracture toughness is usually very poor. Recently, Ray and Dutta (1999) studied in detail the fracture toughness of soda lime silica glasses. The reported value of fracture toughness of this glass is 0·89 ± 0·02 MPam$^{1/2}$ and about 0·8–0·9 MPam$^{1/2}$ for sodium–boro–silicate glasses. The glass is an isotropic homogeneous material with no grain boundary. Thus once a crack is formed, there is no barrier to arrest the propagation of crack and this accounts for the poor fracture toughness. As a whole for a ceramic material, the basic problem is the low fracture toughness and the goal of materials science is to develop ever-increasing stronger, tougher, and lighter materials (Suryanarayana 1994). Many experimental methods have been developed to measure the macro fracture toughness (Davidge 1979; Jenkins *et al* 1987a,b; Sakai 1988), e.g. single edge notch beam (SENB), chevron notched specimen (CNS), all of them requiring specific sample geometries. Compared to these techniques, micro vickers indentation technique to compute the local micro fracture toughness, is gaining steadily increasing importance due to simplicity of test and requiring no specific sample geometry. Depending on the type of the cracks palmqvist or median, a number of equations have been proposed to compute the micro fracture toughness (Sakai *et al* 1992). The system under investigation is sodium–boro–silicate glass which shows glass in glass phase separation. It would be interesting to find out whether the discontinuities may improve the fracture toughness of glass. We have used the micro vicker indentation technique to evaluate the micro fracture toughness of sodium boro silicate glasses as a function of composition as well as phase separation. In this paper we report the results.

2. Experimental

Table 1 gives the batch compositions of glass which have been used in this investigation and each composition falls in the phase separated region of Na_2O–B_2O_3–SiO_2 ternary system.

The batches for melting to form glasses are prepared from silica, boric acid and sodium carbonate. For each composition the corresponding batch is made up by weighing the components in physical balance and components are mixed thoroughly in acetone in an agate mortar. The thoroughly mixed batch is taken in a alumina crucible and melted at about 1250°C for 1 h in an electrically heated furnace. The homogeneous liquid is cast in an aluminium mold of 1 × 0·5 × 10 cm and the bar is immediately transferred to an annealing furnace kept at 250°C for 30 min followed by furnace cooling. Small samples are cut from the cast and annealed glass bars. Samples made from glass composition 4 are subjected to the following heat treatment as summarized in table 2.

The opposite faces of each sample, as cast or heat treated are made flat and parallel by grinding successively

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with finer SiC abrasive powder and finally made mirror finished by polishing on a rotating wheel with diamond paste of one micron size. The Wolpert Instron micro hardness tester has been used to determine the Knoop hardness and micro fracture toughness based on micro Vickers indentation.

2.1 *Determination of Knoop hardness*

For each load, fifteen indentations are taken at different places of the sample. From these the average characteristic length (d) of the Knoop indentation in micron is measured. The following linear relationship is observed between the characteristic length, d, in micron and square root of load, p, expressed in Newton

$$d = (14229/H_0)^{1/2}\, p^{1/2} + d_e, \quad (1)$$

where H_0 is the true hardness in GPa and d_e a constant. Thus the slope of d vs $p^{1/2}$ plot will yield H_0, the true hardness of glass.

2.2 *Determination of fracture toughness*

When load applied to the Vickers indenter is greater than the critical value, the crack initiates from the corners of the Vickers indentation as shown schematically below in figure 1.

For each load fifteen such impressions are taken and average values of l, a and c are evaluated. Depending on the type of cracks, the fracture toughness of the glass sample has been evaluated from the experimental data l, a and c by using the equation available in the literature (Sakai *et al* 1992).

3. Results and discussion

Figure 2 represents the plots of d vs $p^{1/2}$ for composition numbers 1 to 4. The best straight line is fitted to each plot. The slope of each straight line yields the true hardness (1) and the true hardness data are summarized in table 3 along with the correlation coefficient and standard deviation for each linear fit.

It is observed from the above table that as the silica replaces more and more boric oxide from compositions 1–4 (table 1), hardness increases. This is expected because the silica glass is harder than the borate glass. Table 4 contains the c and a of Vickers indentations (figure 1) as a function of indentation load for different glass compositions. The difference between c and a gives the crack length, l.

Figure 3 shows a typical plot of crack length, l ($l = c–a$) in micron vs load, p in Newton. It shows a knee in the curve at approximately 3 N load for glass composition 1. For all other compositions the knee appears in the range of 3 to 4 Newtons. Thus the type of cracks with the indentation load greater than 3 to 4 Newton is median (Sakai *et al* 1992). So the equations developed for median type cracks will be appropriate for computation of micro fracture toughness. In this paper we have used the equation for median crack developed by Lawn and Fuller (1980) to compute the microfracture toughness for all the glass compositions.

The equation is given by

$$K_{IC} = (1/p^{3/2}\, \tan\psi)p/c^{3/2}, \quad (2)$$

here ψ is half of the angle of Vickers indenter.

Making use of data in table 4 and (2), the micro fracture toughness data have been evaluated for all the glass com-

Table 1. Batch composition of glass.

Composition no.	Components (wt%)		
	Na_2O	B_2O_3	SiO_2
1.	10	70	20
2.	10	60	30
3.	10	40	50
4.	10	30	60

Table 2. Heat treatment given to the glass samples having composition 4.

Sample no.	Heat treatment schedule
4A	As cast annealed
4B	600°C for 64 h
4C	650°C for 16 h
4D	700°C for 24 h

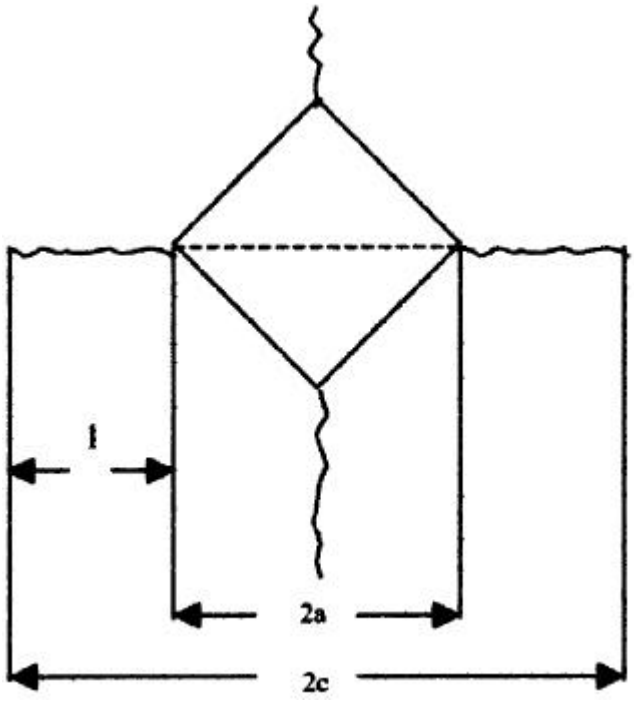

Figure 1. Characteristics length of Vickers indentation.

positions and as a function of indentation load. The results have been summarized in table 5 below.

For a particular glass composition, the K_{IC} is almost unaffected with increasing indentation load. Thus the K_{IC} value for glass is independent of indentation load. However, as the silica content of glasses increases the K_{IC} value increases and becomes of the order of 1 MPam$^{1/2}$. This is in agreement with the reported K_{IC} value of soda–lime–silicate glasses (Ray and Dutta 1999). This may be attributed to the high bond strength as well as high coordination number of silica compared to boric oxide.

It is observed from table 6 that as the heat treatment temperature increases from 600–700°C the fracture toughness decreases. The plausible explanation for this is as follows. The TEM photographs (Haller *et al* 1970) reveal that for the glasses having the same composition as glass 4, and heat treated at the same temperature and time as are used for the present investigation, it is observed that as the heat treatment temperature increases from 600°C through 650°C to 715°C the interconnectivity of phase separated interface decreases and the separated phase finally turns into spherical morphology. Thus it is expected that the glass, heat treated at higher temperature, provides less barrier for arresting the cracks propagating through glass matrix and thereby accounts for the decrease in the fracture toughness for the glasses 4B to 4D. The glass corresponding to composition 4 is in the phase separation region of SiO_2–B_2O_3–Na_2O system. Thus during casting and annealing itself the glass 4A appears to undergo a phase separation of the spinodal

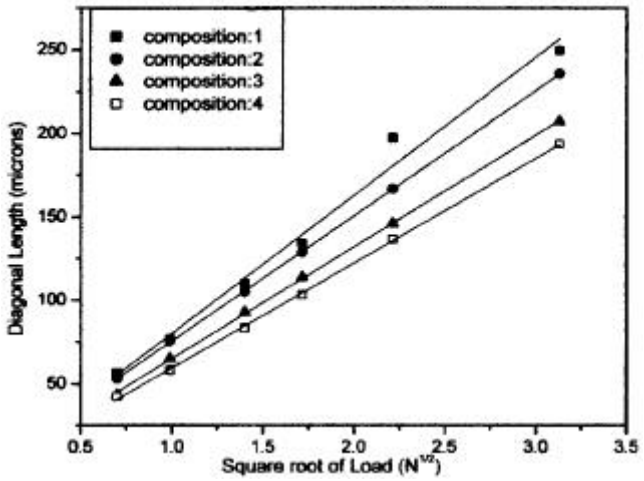

Figure 2. Relationship between characteristics knoop indentation length and square root of test load for compositions 1–4.

Table 3. True hardness of the sodium boro silicate glass as a function of composition.

Composition	True hardness (GPa)	Correlation coefficient	Standard deviation
1.	2·07	0·99306	9·68825
2.	2·53	0·99997	0·58324
3.	3·17	0·99984	1·17576
4.	3·60	0·99975	1·38258

Table 4. *c* and *a* values of Vickers indentations as a function of load for different glass compositions.

Load (N) composition		1·96	2·94	3·92	4·90	5·88
1.	*a* (μm)	19·00	24·00	26·00	30·50	32·00
	c (μm)	34·93	52·75	62·65	72·39	75·28
2.	*a* (μm)	17·50	21·50	25·50	28·00	30·50
	c (μm)	35·86	44·40	50·00	60·83	63·00
3.	*a* (μm)	16·50	20·50	21·50	26·00	30·00
	c (μm)	39·30	36·44	45·50	51·57	60·40
4.	*a* (μm)	15·50	17·50	19·50	22·50	25·00
	c (μm)	24·50	30·85	40·50	48·42	51·28

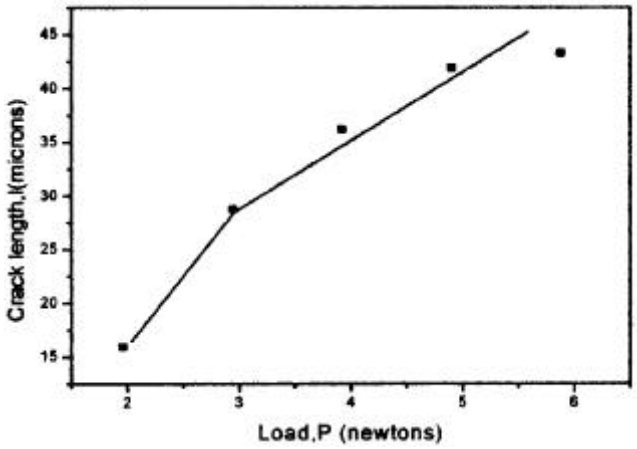

Figure 3. Relationship between crack length, *l* and test load, *P* for composition, 1.

Table 5. Micro fracture toughness (K_{IC}) data of different glasses.

Glass no.	Load (N)	K_{IC} (MPam$^{1/2}$)
1.	3·92	0·574
	4·90	0·580
	5·88	0·650
2.	3·92	0·800
	4·90	0·750
	5·88	0·850
3.	3·92	0·892
	4·90	0·960
	5·88	0·900
4.	3·90	1·100
	4·90	1·180
	5·88	1·160

Table 6. Comparison of K_{IC} values of same glass composition heat-treated at different temperatures at a load of 4·90 N.

Sample no.	Heat treatment schedule	Fracture toughness (K_{IC}) ($MPam^{1/2}$)
4A	As cast annealed	1·18
4B	600°C for 64 h	1·54
4C	650°C for 16 h	1·43
4D	700°C for 24 h	0·98

type to some extent. The increase of fracture toughness of samples 4B and 4C compared to 4A (as cast) is probably due to more of interconnected phase separation which provides more of discontinuities to arrest cracks and thus explains the increases in fracture toughness.

4. Conclusions

Based on the above discussion it can be concluded that:

(I) With the increase in silica content in sodium boro silicate glasses both the fracture toughness and hardness increase.
(II) Phase separation having higher interconnectivity enhances the fracture toughness of glasses.

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