



# Composition dependence of crack formation probability in aluminoborosilicate glass



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## ABSTRACT

We investigate the effect of glass composition on crack formation probability in aluminoborosilicate (ALBS) glass. The ALBS glass base composition is  $13\text{Na}_2\text{O}-4\text{K}_2\text{O}-6\text{MgO}-16\text{Al}_2\text{O}_3-12\text{B}_2\text{O}_3-49\text{SiO}_2$  (mol%). Various glass compositions are prepared by replacing  $\text{SiO}_2$  in the base composition with other oxides. Mechanical properties, including crack formation probability, are evaluated and their compositional variation is discussed in terms of glass structure. Lower crack formation probability results from larger concentrations of bridging oxygen atoms around Si in glass.

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## 1. Introduction

Glass with reliable strength is a fascinating industrial material, with regard to safety. The theoretical value of glass strength is very high ( $>10$  GPa); however, usable glass strength is generally much lower, due to flaws on the glass surface [1]. There are two main approaches in designing stronger glasses: suppressing crack initiation and preventing crack propagation. Chemical strengthening is a common and promising method of preventing crack propagation [2,3]. To suppress crack initiation – in other words, to make glass less brittle – several methods have been implemented, such as controlling plastic flow, densification under stress [4], and fictive temperature dependence of elastic modulus [5].

Although the chemical strengthening process is promising in preventing crack propagation, all industrial glasses face potential damages during processing and handling before strengthening. Thus, glasses with high crack resistance (low crack formation probability) are highly desirable. In a previous study, we developed a glass with high crack resistance and demonstrated that chemical strengthening is an effective method for attaining stable strength [6].

Indeed, chemical strengthening is a mature technique; however, there is still plenty of room for the improvement of crack resistance since knowledge on the relationship between crack resistance and

glass structure is limited, even though many studies have been conducted [7–13] to obtain higher-crack-resistant glass.

Glass is considered a brittle material and shows no plastic flow during fracture. However, microindentation, such as Vickers indentation, introduces permanent deformation of the glass surface. In some cases, indented glass samples show cracks around the indentation created under a higher load. Wada et al. reported, in their pioneering study, that crack formation probability depends on glass composition [9]. They proposed a new index, the “crack resistance (CR)”, which is defined as the load at which crack formation probability is 50% at a given load. Sehgal et al. [4] developed less brittle (LB) glass with high crack resistance and reported these results based on easier stress relaxation by both densification and plastic flow under an indenter.

Continuous studies toward the mechanism of crack initiation have gradually revealed the relationship between crack initiation tendency and glass structure. Through Vickers indentation tests of glass, Arora et al. reported that crack morphology depends on the deformation mechanism, plastic flow, or densification [10]. A normal glass mainly shows plastic flow, which is shear-driven, volume-conservative, and results in irreversible deformation. On the contrary, an anomalous glass predominantly shows densification mainly by hydrostatic pressure. Since densification volume can be recovered by annealing, Yoshida et al. estimated the ratio of densification volume to total deformation volume by measuring the recovered volume using atomic force microscopy [11]. It has also been reported that densification affects the residual stress after indentation [12]. Densification reduces the crack initiation tendency of glass. Thus, higher densification tendency contributes to

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lower crack formation probability. Rouxel et al. reported that Poisson's ratio ( $\nu$ ) is a good measure for representing crack formation tendency [13,14]. A lower Poisson's ratio, or a lower atomic packing density, results in higher resistance to radial/median cracking.

In B<sub>2</sub>O<sub>3</sub>-containing glass, indentation induces a distinct change, from three-fold to four-fold boron, which can be confirmed through Raman spectroscopy. Winterstein-Beckmann et al. conducted a Raman spectroscopic study in alkaliborosilicate glass and reported that indentation-induced structural changes depend on not only borate concentration but also silicate sub-network connectivity [15]. They also reported that the largest value of CR is not obtained for the composition that shows the highest values of densification to total indentation volume ratio. They concluded that structural evolution of boron, formation of non-bridging oxygen ions, and connectivity between the silicate and borate sub-networks are also important factors controlling crack initiation behavior. These reports indicate that crack initiation tendency depends on not only packing density but also network connectivity.

Although numerous studies have been conducted to provide insight into crack initiation mechanisms, there are few reports on the relationships among glass structure, physical property, and crack resistance, especially for multicomponent aluminoborosilicate glass, which is now widely used for electronic devices such as flat panel display base and cover glass. Thus, the present study attempts to shed some light on the relationships among crack formation tendency, glass composition, and glass structure. In this study, a wide range of glass compositions are prepared and various physical properties, including crack formation probability, are investigated. In addition, glass structures are evaluated using Raman spectroscopy and the relationship between crack resistance and glass structure is discussed.

## 2. Experimental procedure

The base glass composition employed was 13Na<sub>2</sub>O-3K<sub>2</sub>O-5MgO-12B<sub>2</sub>O<sub>3</sub>-15Al<sub>2</sub>O<sub>3</sub>-52SiO<sub>2</sub> in mol%, which is the ALBS glass composition considered in our previous study [6]. Thirty-five different glass compositions were prepared, as shown in Table 1, by replacing SiO<sub>2</sub> in the base glass composition with other oxides (Al<sub>2</sub>O<sub>3</sub>, B<sub>2</sub>O<sub>3</sub>, Na<sub>2</sub>O, K<sub>2</sub>O, and MgO). The glasses were named "Series A", "Series B", "Series N", "Series K", and "Series M", respectively. The number in the sample name denotes the molar content of each component; for instance, A12 denotes the glass containing 12 mol% of Al<sub>2</sub>O<sub>3</sub> at the batch composition. The replacing components appear in bold characters. The batches were prepared from reagent-grade Na<sub>2</sub>CO<sub>3</sub>, NaNO<sub>3</sub>, K<sub>2</sub>CO<sub>3</sub>, KNO<sub>3</sub>, MgO, Na<sub>2</sub>B<sub>4</sub>O<sub>7</sub>, Al(OH)<sub>3</sub>, and SiO<sub>2</sub> powders to obtain 500 g of glasses. The powder was weighed appropriately, mixed thoroughly, and roughly melted in clay crucibles at 1350–1500 °C for 1.5 h in an electric furnace. Then, the melted glasses were quenched into water and dried at room temperature. The roughly melted glass was remelted in Pt–Au crucibles at 1350 °C for 4 h in an electric furnace. The glasses were heated up to temperatures around its glass transition temperature (T<sub>g</sub> + 30 K; T<sub>g</sub>: glass transition temperature), held for 30 min, and then cooled by 3 K/min to obtain annealed glass. The T<sub>g</sub> of each glass was measured using a thermomechanical analyzer (TMA, TA Instruments Q400). The obtained samples were grounded, lapped with SiC slurry, and then polished with cerium oxide to obtain optically smooth surfaces. The samples were ca. 37 mm in diameter and 3 mm in thickness. The glass compositions were analyzed using a wavelength dispersive X-ray fluorescence spectrometer (Rigaku ZSX100e). The analysis was conducted as semi-quantitative using the fundamental parameter method. The B<sub>2</sub>O<sub>3</sub> content was determined using the calibration curve method in order to increase the results' accuracy. Table 1 summarizes the analyzed compositions of each glass. Each value in the table represents an average of at least two.

The glass density was measured using Archimedes' method with distilled water as an immersion liquid. The molar volume of oxygen (V<sub>(O)</sub>) was also determined from the density and chemical composition of the glass. The Vickers hardness, Hv, was measured at 0.29 N in air

**Table 1**

Analyzed chemical compositions of the glasses (in mol%). The replacing components appear in bold characters.

	Name	Na <sub>2</sub> O	K <sub>2</sub> O	MgO	B <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Total
Series A	A8	13.6	3.7	6.4	11.1	<b>7.8</b>	57.1	100
	A10	13.8	4.0	6.2	11.5	<b>9.9</b>	54.5	100
	A12	13.8	4.1	6.3	11.2	<b>11.9</b>	52.6	100
	A14	13.9	4.3	6.3	11.1	<b>14.0</b>	50.3	100
	A16	13.2	4.5	6.2	11.2	<b>16.1</b>	48.7	100
	A18	14.1	4.4	6.1	11.6	<b>17.8</b>	45.9	100
	A20	14.0	4.3	6.2	11.5	<b>19.7</b>	44.1	100
Series B	B4	13.2	4.6	6.1	<b>4.2</b>	15.9	55.8	100
	B6	13.2	4.6	6.0	<b>6.1</b>	15.9	54.0	100
	B8	13.3	4.7	6.1	<b>8.0</b>	16.0	51.9	100
	B10	13.3	4.6	6.1	<b>9.9</b>	15.9	50.0	100
	B12	13.1	4.5	6.2	<b>11.4</b>	16.0	48.4	100
	B14	13.3	4.4	6.1	<b>13.3</b>	16.1	46.6	100
	B16	13.1	4.3	6.1	<b>15.0</b>	16.1	45.1	100
Series N	N9	<b>9.1</b>	4.5	6.2	11.3	16.0	52.8	100
	N11	<b>10.9</b>	4.5	6.2	11.2	16.1	51.0	100
	N13	<b>13.3</b>	4.6	6.1	11.5	16.0	48.4	100
	N15	<b>15.5</b>	4.6	6.6	11.7	16.2	45.3	100
	N17	<b>17.2</b>	4.2	6.2	11.8	15.9	44.6	100
	N19	<b>18.9</b>	4.1	6.2	11.1	16.2	43.2	100
	N21	<b>20.8</b>	3.7	6.4	10.2	16.5	42.2	100
Series K	K1	13.1	<b>0.9</b>	6.0	10.8	15.7	53.2	100
	K3	12.9	<b>2.7</b>	6.1	11.2	15.9	51.0	100
	K5	13.2	<b>4.5</b>	6.2	11.4	16.1	48.6	100
	K7	14.6	<b>6.5</b>	7.2	12.3	17.4	41.9	100
	K9	13.3	<b>7.7</b>	6.4	11.8	16.3	44.3	100
	K11	13.3	<b>9.4</b>	6.4	11.6	16.3	42.8	100
	K13	13.3	<b>10.9</b>	6.3	11.1	16.6	41.7	100
Series M	M3	13.1	4.6	<b>2.6</b>	11.4	16.1	52.2	100
	M5	13.5	4.9	<b>4.6</b>	11.7	15.8	49.3	100
	M7	13.1	4.5	<b>6.2</b>	11.1	16.2	48.8	100
	M9	13.0	4.4	<b>7.8</b>	11.1	16.2	47.4	100
	M11	13.2	4.3	<b>9.7</b>	10.9	16.3	45.5	100
	M13	13.0	4.2	<b>11.7</b>	10.8	16.3	43.8	100
	M15	13.0	4.0	<b>13.6</b>	10.6	16.3	42.2	100
Experimental error		±1	±1	±1	±1	±1	±2	

using a Vickers hardness tester (Akashi MVK-H1) for 10 s as dwell time. Young's modulus was measured through the pulse echo method [16] with a DPR300 Pulser/Receiver (JSR Ultrasonics). The longitudinal and shear wave sound velocities (V<sub>l</sub> and V<sub>s</sub>, respectively) of the test piece were determined from the sample thickness, *d*, and the roundtrip transit time, *s*, using the following equation.

$$V_l \text{ or } V_s = \frac{2d}{s} \quad (1)$$

Young's modulus, *E*, was calculated from the density,  $\rho$ , longitudinal wave sound velocity, and shear wave sound velocity using the following equation.

$$E = V_s^2 \rho \frac{3V_l^2 - 4V_s^2}{V_l^2 - V_s^2} \quad (2)$$

The shear modulus (*G*), Poisson's ratio ( $\nu$ ), and bulk modulus (*K*) were calculated using the following equations.

$$G = \rho V_s^2 \quad (3)$$

$$\nu = \frac{1}{2} \frac{V_l^2 - 2V_s^2}{V_l^2 - V_s^2} \quad (4)$$

$$K = \frac{E}{3(1-2\nu)} \quad (5)$$

Crack formation probability was determined using the hardness tester shown above. The indentation loads were varied from 0.98 to 9.8 N.

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