Structural Analysis of $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ Melts by Pulsed Neutron Total Scattering Method and Molecular Dynamics Simulation

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In order to obtain direct information on the structure of sodium borate melts which are of use in metallurgical processes, pulsed neutron total scattering measurements have been carried out on 10mol% Na_2O - and 30mol% $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melts at 1 073 K, in addition to the previous measurement on pure B_2O_3 melt. Molecular dynamics (MD) calculations have also been made on pure B_2O_3 and 30mol% $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melts at 1 073 K.

With increase in Na_2O content, the first peak of $RDF(r)$, representing B–O correlation, spread asymmetrically to larger r side, and the coordination number of O atoms around a reference B atom $n_{\text{B-O}}$ increased from 3.0 to 3.5, showing variation of B–O correlation. This increase in $n_{\text{B-O}}$ could be explained by the simple assumption that two BO_3 triangle structure units are converted to two BO_4 tetrahedron structure units with the addition of one Na_2O molecule in the same way as in $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ glasses. These $n_{\text{B-O}}$ values also agreed well with those obtained by MD calculations, which showed the clear evidence of the appearance of BO_4 units that the distribution of O–B–O bond angle spread from 120° to smaller angle side for 30mol% $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$.

From these results, it is safely concluded that a part of BO_3 triangle structure units is converted to BO_4 tetrahedron structure units with the addition of Na_2O up to 30 mol% to B_2O_3 melt in the same way as in $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ glasses.

KEY WORDS: $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melt; sodium borate; structure; structure unit; pulsed neutron scattering; molecular dynamics, MD calculation.

1. Introduction

A precise knowledge of the structure and properties of B_2O_3 -based melts and glasses is required increasingly from the fundamental and practical viewpoints. Among binary borate glasses, alkali borate glasses have long been absorbing the attention of many researchers because of “borate anomaly” shown in the composition dependence of some physical properties. $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ system has also been attractive for its practical use in metallurgical processes. Therefore, many investigations have been performed on the structure of $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ glasses by means of X-ray diffraction,¹⁾ Raman spectroscopy,^{2,3)} NMR^{4,5)} and thermal expansion.⁶⁾

Previously, one of the authors carried out a pulsed neutron scattering study on the temperature dependence of the structure of pure B_2O_3 glasses and melts.⁷⁾ Several other diffraction studies have been reported on the structure of pure B_2O_3 melt.^{8–11)} However, high temperature diffraction studies on the structure of

$\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melts have not yet been reported to the authors' knowledge, although physical properties such as the density,^{12–14)} viscosity,^{15,16)} surface tension¹⁷⁾ and the velocity of sound¹⁸⁾ have been measured widely on $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melts. An attempt has been made recently by Derno *et al.*¹⁹⁾ to reveal the structure of $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melts by comparing the X-ray diffraction data on rapidly quenched $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ glasses and normally cooled glasses.

In the present study, pulsed neutron total scattering experiments have been carried out on $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melts, in order to investigate the structural change of these melts with the addition of Na_2O . Molecular dynamics (MD) simulation has also been made on these melts for comparison with the diffraction experiments.

2. Experimental

Pulsed neutron scattering experiments have been carried out on 10mol% Na_2O - and 30mol% $\text{Na}_2\text{O}-$

Fig. 1. The phase diagram of $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ system.²⁰⁾ Arrows show the temperatures and compositions of the specimens measured.

B_2O_3 melts at 1073 K with time-of-flight (TOF) method by using the high intensity total scattering spectrometer (HIT) installed at a pulsed spallation neutron source at National Laboratory for High Energy Physics (KEK) in Tsukuba. The temperatures and compositions of measured specimens are indicated in Fig. 1 showing the phase diagram of $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ system.²⁰⁾

The samples were prepared by the following procedure. B_2O_3 glass was prepared from boric acid (H_3BO_3) enriched with 99.61 at% ^{11}B , in order to avoid extremely high neutron absorption of ^{10}B . H_3BO_3 was melted and dehydrated in a platinum crucible in a vacuum furnace at 1073 K under 10^{-3} Torr for more than 8 h to produce B_2O_3 glass. The mixture of Na_2CO_3 (99.5%) and B_2O_3 with a given composition was heated and melted for dehydration and decarboxylation in a Pt crucible with a lid at 1373 K for more than 4 h, and then poured into cylindrical copper mold to form a cylindrical sample of 7 mm diameter.

In the scattering experiment, the sample was contained in a thin-walled cylindrical Pt cell (8 mm in inner diameter and 0.1 mm thick), and held at 1073 K by heating with infrared image furnace under reduced pressure of about 0.1 Torr. The scattering intensity spectra were obtained for the sample contained in the cell, and for the blank cell. The incident neutrons were scattered by the sample, and the scattered neutrons were detected by the ^3He counters located at several scattering angles, i.e. $2\theta = 8^\circ, 13^\circ, 24^\circ, 30^\circ, 42^\circ, 90^\circ$ and 150° . The time-of-flight scattering intensity spectra were recorded on a hard disk using a computer-controlled data-acquisition system, and then processed by use of a computer at KEK. As the time-of-flight corresponds to the wave length of a pulsed neutron, λ , the scattering intensity can be obtained as a function of the magnitude of the scattering vector or the momentum transfer, $Q = 4\pi \sin\theta/\lambda$, where 2θ is the scattering angle.

The scattering intensity for the sample in the Pt cell was corrected for the cell, background, absorption, and multiple and incoherent scatterings. The coherent scattering intensities $I(Q)$ were normalized using the scattering intensities from the vanadium rod, since coherent scattering length of V is negligibly small.

The interference function $S(Q)$ in the Faber-Ziman form²¹⁾ is obtained as follows:

$$S(Q) = \frac{[I(Q) - \{\sum (c_i b_i^2) - (\sum c_i b_i)^2\}]}{(\sum c_i b_i)^2} \quad \text{.....(1)}$$

where b_i is the coherent scattering length of the i -th nuclei, ($b_{\text{B}} = 6.65$, $b_{\text{O}} = 5.805$ and $b_{\text{Na}} = 3.63 \times 10^{-15} \text{ m}^{22})$ and c_i the atomic fraction. The pair distribution function $g(r)$ is obtained by Fourier transform of the structure function $Q(S(Q) - 1)$:

$$g(r) = 1 + \frac{1}{2\pi^2 r \rho_0} \int_0^{Q_{\text{max}}} Q(S(Q) - 1) \sin(Qr) dQ \quad \text{....(2)}$$

where the number density ρ_0 was evaluated from the density values¹²⁾ of $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melts. The radial distribution function $RDF(r)$ is determined by the definition.

$$RDF(r) = 4\pi r^2 \rho_0 g(r) \quad \text{.....(3)}$$

3. Molecular Dynamics Simulation

Molecular dynamics (MD) simulation has been carried out on pure B_2O_3 melt and 30mol% $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melt at 1073 K, in order to investigate the detailed 3-dimensional configuration of atoms in the melts.

The MD calculations were performed in a similar way to that used previously by one of the authors.²³⁾ The pair potential functions were assumed to consist of a simplified Coulombic and a repulsion terms:

Fig. 2. Experimental structure functions $Q(S(Q)-1)$ for Na_2O - B_2O_3 melts at 1073 K.

Table 1. Peak positions of $S(Q)$ curves.

Melt/mol%	1st/nm ⁻¹	2nd/nm ⁻¹	3rd/nm ⁻¹
B_2O_3	13.1	29.4	57.0
10 Na_2O -90 B_2O_3	13.1	29.4	56.9
30 Na_2O -70 B_2O_3	13.2	29.6	56.9

$$\phi_{ij} = \frac{z_i z_j e^2}{r_{ij}} + f_0 (\beta_i + \beta_j) \exp \left[\frac{(a_i + a_j - r_{ij})}{(\beta_i + \beta_j)} \right] \quad \text{.....(4)}$$

where, z_i : the formal charge number of ion i (e.g., +3 for B^{3+} ion)
 e : the unit charge
 r_{ij} : the distance between ion i and j
 f_0 : a force constant arbitrarily taken here to be 1 kcal·mol⁻¹·Å⁻¹ (= 6.948 × 10⁻⁶ dyn)
 a_i, β_i : the crystal radius and compressibility of ion i , respectively.

The potential parameters proposed by Xu *et al.*²⁴⁾ were used for the MD calculations. The edge length of the basic cell (a cube) was calculated from the density¹²⁾ of the melt. The number of particles (atoms) within the basic cell was about 1000 (B: 400; O: 600 for B_2O_3 , and B: 280; O: 480; Na: 120 for 30mol% Na_2O - B_2O_3). In evaluating the potential energy and the force, the Coulombic term was calculated by the Ewald method. The MD calculations were carried out at the computer center of Government Industrial Research Institute, Osaka. The results of calculation were compared with those of neutron diffraction experiments.

4. Results and Discussion

$Q(S(Q)-1)$ curves obtained from the experiments for B_2O_3 ,⁷⁾ 10mol% Na_2O - and 30mol% Na_2O - B_2O_3 melts at 1073 K are compared in Fig. 2 in the wide range of Q up to 300 nm⁻¹. The peak positions of $S(Q)$ curves are given in Table 1. With the addition of Na_2O to B_2O_3 melt, the peak positions and the overall shapes of $Q(S(Q)-1)$ curves remained almost unchanged, and the oscillatory behavior of $Q(S(Q)-1)$ in the high Q region is clearly observed, with appreciable damping in

Fig. 3. Radial distribution functions $RDF(r)$ for Na_2O - B_2O_3 melts at 1073 K.

Table 2. Peak positions of $RDF(r)$ and FWHM of the first peak.

Melt/mol%	1st/nm	2nd/nm	3rd/nm	FWHM/nm
B_2O_3	0.137	0.238	0.360	0.018
10 Na_2O -90 B_2O_3	0.138	0.241	0.370	0.021
30 Na_2O -70 B_2O_3	0.139	0.244	0.364	0.026

amplitude with increase in Na_2O content.

The radial distribution functions $RDF(r)$ obtained by Fourier transform of the experimental $Q(S(Q)-1)$ truncated at about 300 nm⁻¹ are compared in Fig. 3. The three peaks are clearly observed for each of $RDF(r)$. The peak positions and the full width at half maximum (FWHM) of the first peak are given in Table 2. With increase in Na_2O content from zero to 30 mol%, the first peak position of $RDF(r)$, representing the atomic spacing of B-O, shifted gradually from 0.137 to 0.139 nm, and the first peak broadened asymmetrically, spreading to larger r side as shown in Fig. 3.

Fig. 4. Experimental total pair distribution function $g(r)_{\text{expt}}$ compared with partial pair distribution function $g_{ij}(r)_{\text{MDcalc}}$ and distribution of coordination numbers $n_{ij}(r)_{\text{MDcalc}}$ by MD calculations. (B_2O_3 melt at 1073 K)

The total pair distribution functions $g(r)$ obtained from the experimental $Q(S(Q)-1)$ for B_2O_3 ⁷⁾ and 30mol% $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ are compared in Figs. 4 and 5 with the partial pair distribution function $g_{ij}(r)$ and the distribution of the coordination number $n_{ij}(r)$ obtained from MD calculations. The experimental $g(r)$ corresponds well to the calculated $g_{ij}(r)$, which serve as a reference in attributing the peaks of $\text{RDF}(r)$ to specific atomic bonds. In Fig. 6, $Q(S(Q)-1)$ for B_2O_3 ⁷⁾ and 30mol% $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ obtained from the experiments are compared with those obtained from inverse Fourier transform of $g(r)$ of MD calculations. Both of $Q(S(Q)-1)$ are coincide with each other fairly well.

As described in the previous paper on the structure of B_2O_3 melt,⁷⁾ and also as can be seen in Figs. 4 and 5, the first peak of $\text{RDF}(r)$ is attributed to the B-O bond. The coordination number of O atoms around a reference B atom, $n_{\text{B-O}}$, is determined from the area, A_1 , under the first peak of experimental $\text{RDF}(r)$ using a relationship as follows:

$$A_1 = \frac{2b_{\text{B}}b_{\text{O}}}{\langle b \rangle^2} c_{\text{B}} n_{\text{B-O}} \quad \dots\dots\dots (5)$$

The coordination number of B atoms around a reference O atom was evaluated from the relationship $c_{\text{O}} n_{\text{O-B}} = c_{\text{B}} n_{\text{B-O}}$. The experimental values of the coordination numbers $n_{\text{B-O}}(\text{expt})$ and $n_{\text{O-B}}(\text{expt})$ determined by Eq. (5) are shown in Table 3. With the addition of Na_2O to

Fig. 5. Experimental total pair distribution function $g(r)_{\text{expt}}$ compared with partial pair distribution function $g_{ij}(r)_{\text{MDcalc}}$ and distribution of coordination numbers $n_{ij}(r)_{\text{MDcalc}}$ by MD calculations. (30mol% $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melt at 1073 K)

Fig. 6. Comparison of structure functions $S(S(Q)-1)$ obtained by experiments and by MD calculations, for B_2O_3 and 30mol% $\text{Na}_2\text{O}-\text{B}_2\text{O}_3$ melts at 1073 K.

B_2O_3 , the coordination number $n_{\text{B-O}}$ increased from 3.0 to 3.5, showing variation of the configuration of O atoms around a reference B atom. These values correspond well to those of MD calculations, as seen from

Figs. 4 and 5 as a plateau of $n_{B-O}(r)$ curves (3.0 for B_2O_3 and 3.3 for 30 mol% Na_2O).

As regards the structure of alkali borate glasses, $R_2O-B_2O_3$, it has been established that the structure unit, BO_3 triangle, constructing boroxol ring and partly random network structure in B_2O_3 glasses, is converted to tetrahedron BO_4 by the addition of R_2O to B_2O_3 up to 33%. According to the Raman^{2,3)} and NMR^{4,5)} spectroscopic studies, by the addition of one molecule of Na_2O to the B_2O_3 glass, two BO_3 units are converted into two BO_4 units as shown below schematically:³⁾

If the structure units in $Na_2O-B_2O_3$ melts are converted from BO_3 to BO_4 by the addition of Na_2O in the same way as in $Na_2O-B_2O_3$ glasses, the fraction of three-coordinated and four-coordinated B atoms, N_3 and N_4 , respectively, can be expressed as follows:

$$N_3 = \frac{1-2x}{1-x}, \quad N_4 = \frac{x}{1-x} \quad \dots\dots\dots(6)$$

where x is the mole fraction of Na_2O . Accordingly, the coordination number of O atoms around a reference B atom is:

$$n_{B-O} = 3N_3 + 4N_4 = 3 + \frac{x}{1-x} \quad \dots\dots\dots(7)$$

The coordination number of B atoms around a reference O atom is:

$$n_{O-B} = \frac{c_B}{c_O} n_{B-O} = 2 \quad \dots\dots\dots(8)$$

Table 3. Area under the first peak of $RDF(r)$, A_1 , and the experimental and calculated coordination number n_{B-O} and n_{O-B} .

Melt/mol%	A_1	$n_{B-O}(\text{expt})$	$n_{B-O}(\text{calc})$	$n_{O-B}(\text{expt})$	$n_{O-B}(\text{calc})$
B_2O_3	2.43	3.0	3.0	2.0	2.0
10 Na_2O -90 B_2O_3	2.58	3.2	3.1	2.0	2.0
30 Na_2O -70 B_2O_3	2.59	3.5	3.4	2.0	2.0

The values of the coordination numbers $n_{B-O}(\text{expt})$ and $n_{O-B}(\text{expt})$ determined from the experimental $RDF(r)$ are compared in Table 3 with $n_{B-O}(\text{calc})$ and $n_{O-B}(\text{calc})$ calculated from Eqs. (7) and (8).

The experimental values of the coordination number n_{B-O} and n_{O-B} agree well with those calculated from Eqs. (7) and (8), respectively. These results suggest that the structural change in $Na_2O-B_2O_3$ melts with the addition of Na_2O up to 30 mol% is similar to that in $Na_2O-B_2O_3$ glasses. It should be noted that the coordination number of B atoms around a reference O atom n_{O-B} remains a constant value, *i.e.* two, in other words, B-O bonds in $Na_2O-B_2O_3$ melts are not broken with the addition of Na_2O up to 30 mol%. This means that the network structure of $Na_2O-B_2O_3$ is stable, being not broken but only modified partially by the coordination of oxygen atoms provided from Na_2O around a boron atom. Seemingly, this is not compatible with the behavior of physical properties such as the viscosity,^{15,16)} which decreases rapidly with the addition of a small amount of Na_2O to B_2O_3 melt. Further investigation will be needed for a clear understanding of the detailed structure of $Na_2O-B_2O_3$ melts.

In relation to the above description on the conversion of the structure unit from BO_3 to BO_4 , it should be noted that the first peak of $RDF(r)$ becomes broader and spreads asymmetrically to larger r side with the addition of Na_2O as shown in Fig. 3. It is supposed that the asymmetry in $RDF(r)$ may reflect the appearance of the BO_4 unit. The first peak of $RDF(r)/r$ was decomposed into two Gaussian peaks with keeping all the parameters, *i.e.* the height, FWHM, and the position, variable. The resultant Gaussian curves are converted to $RDF(r)$ and depicted in Fig. 7. For 10 and 30 mol% Na_2O melts, the left peak is located at 0.137 and 0.138 nm respectively, while the right peak is found at 0.150 and 0.152 nm respectively. It is interesting that these values correspond fairly well to the values of B-O spacings of BO_3 and BO_4 unit in $Na_2O-B_2O_3$ crystals. (0.137 and 0.148 nm respectively²⁵⁾)

In Fig. 8, the histogram of O-B-O and B-O-B bond angles obtained by MD calculations are shown for B_2O_3 and 30 mol% Na_2O melts. The histogram of O-B-O bond angles for B_2O_3 is symmetric and has a sharp maximum at 119.4°, while that for 30 mol%

Fig. 7. Decomposition of the first peak of $RDF(r)$ into Gaussian profile components of $RDF(r)/r$. The peak positions of the resultant curves are 0.138 nm for B_2O_3 , 0.137 and 0.150 nm for 10 mol% Na_2O , and 0.138 and 0.152 nm for 30 mol% Na_2O melts.

Fig. 8. The histograms of probability distribution $P(\theta)$ of O-B-O and B-O-B bond angles in B_2O_3 and 30mol% Na_2O - B_2O_3 melts.

Table 4. The coordination number of B atoms around a reference B atom, n_{B-B} , and that of O atoms around an O atom, n_{O-O} .

Melt/mol%	n_{B-B}	n_{O-O}
B_2O_3	3.0	4.0
10 Na_2O -90 B_2O_3	3.1	4.2
30 Na_2O -70 B_2O_3	3.4	4.9

Na_2O is asymmetric and spreads to a lower angle side, corresponding to 109° of a tetrahedron, showing the conversion of a part of the BO_3 triangles to BO_4 tetrahedra.

It can be safely deduced from the results mentioned above, that a part of the BO_3 triangle structure units is converted to BO_4 tetrahedron units with the addition of Na_2O to B_2O_3 melt in the same way as in Na_2O - B_2O_3 glasses.

The second peak in $RDF(r)$ also provides information on the structure of Na_2O - B_2O_3 melts. The second peak is considered to reflect mainly O-O, B-B, and Na-O atomic correlations, as judged from the partial pair distribution functions by MD calculations shown in Fig. 5. At first, as to B-B correlation, it is reasonable to suppose that each of four-coordinated B atoms has four neighboring B atoms, while each of three-coordinated B atoms has three neighboring B atoms. Accordingly,

$$n_{B-B} = 4N_4 + 3N_3 = 3 + \frac{x}{1-x} \quad \text{.....(9)}$$

Secondly, as to O-O correlation, the coordination number of O atoms around a reference O atom was evaluated under the following assumptions: Each of

Table 5. The area under the second peak of $RDF(r)$, A_2 , that of B-B and O-O contribution, A'_2 and the difference, ΔA_2 .

Melt/mol%	A_2	A'_2	ΔA_2
B_2O_3	4.0 ₀	3.5 ₅	0.4 ₅
10 Na_2O -90 B_2O_3	4.0 ₃	3.7 ₁	0.3 ₂
30 Na_2O -70 B_2O_3	5.3 ₄	4.1 ₂	1.2 ₂

four-coordinated B atoms has four nearest B atoms, while each of three-coordinated B atoms has three nearest B atoms. Furthermore, both of these nearest B atoms consist of four-coordinated and three-coordinated B atoms, the ratio of which is proportional to their concentrations. Under these assumptions, the following equation can be derived:

$$\begin{aligned} n_{O-O} &= N_4(6N_4 + 5N_3) + N_3(5N_4 + 4N_3) \\ &= \left(\frac{x}{1-x}\right) \cdot \left(\frac{5-4x}{1-x}\right) + \left(\frac{1-2x}{1-x}\right) \cdot \left(\frac{4-3x}{1-x}\right) \end{aligned} \quad \text{.....(10)}$$

n_{B-B} , n_{O-O} calculated by Eqs. (9) and (10) are shown in Table 4. With the addition of Na_2O up to 30 mol% into B_2O_3 , the coordination numbers, n_{B-B} and n_{O-O} , increase from 3.0 to 3.4 and from 4.0 to 4.9, respectively. These values correspond well to those of MD calculations, as seen from Figs. 4 and 5 as plateaus of $n_{B-B}(r)$ and n_{O-O} curves, respectively. The area, A'_2 , under the second peak contributed by n_{B-B} and n_{O-O} is calculated from the following relation:

$$A'_2 = c_B \frac{b_B^2}{\langle b \rangle^2} n_{B-B} + c_O \frac{b_O^2}{\langle b \rangle^2} n_{O-O} \quad \text{.....(11)}$$

The area A_2 under the second peak of the experimental $RDF(r)$ is defined here as the integration from 0.20 nm to the second minimum of the RDF curve, i.e., 0.276, 0.288 and 0.282 nm respectively for B_2O_3 , 10 mol% Na_2O and 30 mol% Na_2O melts. The values of A_2 and A'_2 are listed in Table 5, with the difference between them: $\Delta A_2 = A_2 - A'_2$. If ΔA_2 of 10 and 30 mol% Na_2O is regarded as the area contributed by Na-O bond, the coordination number of O atoms around a reference Na atom, n_{Na-O} , can be evaluated from the following relation:

$$\Delta A_2 = 2 \cdot c_{Na} \frac{b_{Na} \cdot b_O}{\langle b \rangle^2} n_{Na-O} \quad \text{.....(12)}$$

The values of n_{Na-O} calculated from Eq. (12) are 6.7 for 10 mol% Na_2O and 7.1 for 30 mol% Na_2O melts. Thus, roughly speaking, each Na atom is coordinated by about 6-7 oxygen atoms.

5. Conclusions

Pulsed neutron total scattering measurements have been carried out on 10mol% Na_2O - and 30mol% Na_2O - B_2O_3 at 1073 K. Data on pure B_2O_3 melt were used from the previous experiment at the same conditions. Molecular dynamics (MD) calculations were also made

on pure B_2O_3 and 30mol% $Na_2O-B_2O_3$ melts for reference.

The first peak of $RDF(r)$, attributed to B-O bond, spread asymmetrically to larger r side, and the coordination number of O atoms around a reference B atom n_{B-O} increased from 3.0 to 3.5 with increase in Na_2O content, showing the change of B-O correlation.

The experimental values of n_{B-O} agreed well with those calculated on the assumption that two triangle BO_3 units are converted to two BO_4 tetrahedron units with the addition of one Na_2O molecule as in $Na_2O-B_2O_3$ glasses.

Sufficient agreement was also found between experiment and MD calculation, on the results of n_{B-O} , experimental $g(r)$ and calculated $g_{ij}(r)$, and $Q(S(Q)-1)$. MD calculations also showed that the distribution of bond angle of O-B-O for B_2O_3 melt was concentrated about 120° , while that for 30mol% $Na_2O-B_2O_3$ melt shifted to lower angle side, showing appearance of BO_4 tetrahedra.

From these results, it is deduced that a part of BO_3 triangle units in B_2O_3 melt is converted to BO_4 tetrahedron units with increase in Na_2O content up to 30mol%, as in $Na_2O-B_2O_3$ glasses.

The coordination numbers n_{B-B} and n_{O-O} were found to be similar to those from MD calculations. The coordination number of O atoms around a reference Na atom, n_{Na-O} , was estimated to be about 6-7.

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