Liquidus Surface and Isothermal Section Diagram at 973 K in BaO-Fe₂O₃-(0-50 mol%)B₂O₃ Pseudo-ternary System[†]

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The isothermal section at 973 K and the liquidus surfaces of the phase diagram of the pseudo-ternary BaO-Fe₂O₃-B₂O₃ system were determined in a composition range of less than 50 mol% B₂O₃ by thermal analyses and X-ray diffraction. The liquidus surface which is in equilibrium with Fe₃O₄ spreads over a wide composition range and that in equilibrium with BaO·6Fe₂O₃ extends long and narrow to BaO·B₂O₃. The pseudo-ternary BaO-Fe₂O₃-B₂O₃ compound does not exist in the composition range investigated. The ternary eutectic reaction is $L \rightarrow \text{BaO} \cdot 6\text{Fe}_2\text{O}_3 + \text{BaO} \cdot \text{B}_2\text{O}_3 + 3\text{BaO} \cdot \text{B}_2\text{O}_3$, occurring at 1033 K at the composition 48 mol%BaO-19 mol%Fe₂O₃-33 mol%B₂O₃.

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I. Introduction

The amorphous(glass)-crystallization method is very useful to producing ultrafine powders of ferrite magnet materials such as SrO·6Fe₂O₃ and BaO·6Fe₂O₃ (M-type ferrite) from the pseudo-ternary SrO-Fe₂O₃-B₂O₃ and BaO-Fe₂O₃-B₂O₃ systems⁽¹⁾⁻⁽¹²⁾. In the previous paper, investigations on the heat-treatment of samples prepared by a melt spinning or a water-quenching method were conducted on the relation between the kinds of precipitated crystalline phases and relative quantities of them and on the effects of starting composition and cooling rate on the magnetic properties of the SrO·6Fe₂O₃ or BaO·6Fe₂O₃ particles⁽¹³⁾⁻⁽¹⁵⁾. The process of particle growth of the SrO·6Fe₂O₃ was observed using a transmission electron microscope (TEM), and the relationship between the average particle size and the magnetic properties of SrO·6Fe₂O₃ particles was examined⁽¹⁶⁾⁽¹⁷⁾. However, the ranges of composition and temperature at which the individual phases including the magnetic material of BaO·6Fe₂O₃ are in stable equilibria have not been clarified yet. Phase diagrams are important in the study of relationships between properties and microstructures of materials and also in the designing of BaO·6Fe₂O₃ magnet. In the previous paper, an isothermal section and the liquidus surfaces of the pseudo-ternary phase diagram of the SrO-Fe₂O₃-B₂O₃ system have been determined⁽¹⁸⁾⁽¹⁹⁾. Although the pseudo-binary phase diagrams of the BaO-Fe₂O₃ and BaO-B₂O₃ systems⁽²⁰⁾⁽²¹⁾ are available, however, the pseudo-ternary phase diagram of the BaO-Fe₂O₃-B₂O₃ system has not been reported. In the present study, an isothermal section and the liquidus surfaces of the pseudo-ternary phase diagram of the BaO-Fe₂O₃-B₂O₃ system have been determined in a

composition range of the B₂O₃ content less than 50 mol% using the thermal analyses and the X-ray diffraction.

II. Experimental Procedure

The starting materials used were reagent-grade BaCO₃, α-Fe₂O₃ and H₃BO₃ powders. These powders were well mixed to obtain the prescribed compositions. A mixed powder was melted in air in a Pt crucible placed in an electric furnace. Specimens were prepared by pouring the molten samples directly on a steel plate after heating for 2.7 ks at 1723 K. Specimens were then heat treated at 1073 K for 3.6 to 108 ks. Figure 1 shows compositions of the specimens together with the pseudo-binary phase diagrams of BaO-Fe₂O₃ and BaO-B₂O₃ systems⁽²⁰⁾⁽²¹⁾. Several vertical pseudo-binary sections of pseudo-ternary BaO-Fe₂O₃-B₂O₃ phase diagram are defined as follows. The section through the line connecting the compositions of BaO·6Fe₂O₃ and BaO·B₂O₃ is named Section A. Similarly, the section connecting (BaO)_{0.862}·(B₂O₃)_{0.138} and (Fe₂O₃)_{0.862}·(B₂O₃)_{0.138}, is called Section B, and those of BaO·6Fe₂O₃-3BaO·B₂O₃, 2BaO·Fe₂O₃-B₂O₃, 3BaO·Fe₂O₃-B₂O₃, BaO·Fe₂O₃-3BaO·B₂O₃, BaO·Fe₂O₃-BaO·B₂O₃, BaO·Fe₂O₃-B₂O₃, (BaO)_{0.80}·(B₂O₃)_{0.20}-(Fe₂O₃)_{0.80}·(B₂O₃)_{0.20}, and (BaO)_{0.90}·(Fe₂O₃)_{0.10}-(B₂O₃)_{0.90}·(Fe₂O₃)_{0.10} are named Section C, D, E, F, G, H, I and J, respectively.

The isothermal section at 973 K and the liquidus surfaces of the phase diagram of the pseudo-ternary BaO-Fe₂O₃-B₂O₃ system were determined by thermal analysis, differential thermal analysis (DTA), and X-ray diffraction. In DTA, the specimens of about 10 × 10⁻³ kg were heated in air to the specified temperatures at the fixed heating rate 0.17 K/s. Al₂O₃ was employed as a standard sample for DTA. The temperatures detected by the DTA were compared with the known melting points of standard substances, and the difference were within 10 K in

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Fig. 1 Compositions in the pseudo-ternary BaO-Fe₂O₃-B₂O₃ system.

both heating and cooling operations.

In order to identify existing phases in the specimens, X-ray diffraction analysis was conducted for specimens prepared under various conditions, i.e. quenched at various annealing temperatures. When the primary phase was hardly identified in the composition range of the high BaO or Fe₂O₃ content, a rapid quenching method was conducted by pouring the molten sample on a rotating twin rollers, and the primary phases were confirmed. The primary phases of 3BaO·Fe₂O₃, 2BaO·Fe₂O₃, BaO·Fe₂O₃, BaO·6Fe₂O₃, 3BaO·B₂O₃, BaO·B₂O₃, Fe₂O₃ and Fe₃O₄ were identified from the results of X-ray diffraction analysis using the JCPDS Powder diffraction file. In X-ray diffraction, the samples were pulverized to about 50 μm in diameter, and the X-ray of CuKα was used at a voltage of 40 kV and at an electric current of 30 mA.

III. Experimental Results and Discussion

1. Isothermal section at 973 K

According to the experimental results, the isothermal section at 973 K was determined in a composition range of less than 50 mol%B₂O₃ as shown in Fig. 2. In the region enclosed by the lines connecting the compositions of Fe₂O₃, BaO·B₂O₃, 3BaO·B₂O₃, 2BaO·Fe₂O₃, and Fe₂O₃, the following four regions of the three phase equilibria are present;

Fig. 2 Isothermal section at 973 K of the phase equilibrium diagram of the pseudo-ternary BaO-Fe₂O₃-B₂O₃ system.

2. Vertical sections of phase diagram

The vertical pseudo-binary sections were determined by thermal analyses and X-ray diffraction measurements. The quantities of the existing phases were estimated from the intensities of X-ray diffraction analysis and were referred to as the determination of eutectic points. Moreover, the liquidus lines and the eutectic points in the pseudo-binary sections for which a small number of experimental points were obtained were determined by extrapolating the liquidus lines connecting the measured points in the other pseudo-binary sections so that no inconsistency may occur in the ternary phase diagram.

Figure 3 shows the BaO·6Fe₂O₃-BaO·B₂O₃ section (Section A). The circles (○) in this figure represent the measured points using thermal analysis. Regions of two-phase equilibria of Fe₃O₄+L, BaO·6Fe₂O₃+L and BaO·B₂O₃+L exist where L denotes a liquid phase. A eutectic reaction occurs between BaO·B₂O₃ and BaO·6Fe₂O₃ at

the composition of 41.9 mol%BaO-19.4 mol%Fe₂O₃-38.7 mol%B₂O₃ at 1185±10 K.

Figure 4 shows the (BaO)_{0.862}·(B₂O₃)_{0.138}-(Fe₂O₃)_{0.862}·(B₂O₃)_{0.138} section (Section B). Regions of two phase equilibria of Fe₃O₄+L, BaO·6Fe₂O₃+L, BaO·Fe₂O₃+L, 2BaO·Fe₂O₃+L, and BaO+L exist. Three eutectic reactions are observed between BaO·Fe₂O₃ and BaO·6Fe₂O₃ with the composition of 40.7 mol%BaO-45.5 mol%Fe₂O₃-13.8 mol%B₂O₃ at 1424±10 K, between 2BaO·Fe₂O₃ and BaO·Fe₂O₃ with the composition of 57.2 mol%BaO-29.0 mol%Fe₂O₃-13.8 mol%B₂O₃ at 1373±10 K, and between BaO and 2BaO·Fe₂O₃ with the composition of 68.2 mol%BaO-18.0 mol%Fe₂O₃-13.8 mol%B₂O₃ at 1406±10 K.

The result of Section C is shown in Fig. 5. Regions of two-phase equilibria of BaO·6Fe₂O₃+L, BaO·Fe₂O₃+L and 3BaO·B₂O₃+L exist. Two eutectic reactions are found between BaO·6Fe₂O₃ and BaO·Fe₂O₃ with the composition of 36.8 mol%BaO-54.0 mol%Fe₂O₃-9.2

Fig. 3 BaO·6Fe₂O₃-BaO·B₂O₃ section of the phase diagram. (A Section)

Fig. 4 (BaO)_{0.862}·(B₂O₃)_{0.138}-B₂O₃ of the phase diagram. (B Section)

Fig. 5 $\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$ – $\text{BaO} \cdot \text{B}_2\text{O}_3$ section of the phase diagram. (C Section) Fig. 7 $3\text{BaO} \cdot \text{Fe}_2\text{O}_3$ – B_2O_3 – B_2O_3 section of the phase diagram. (E Section) Fig. 6 $2\text{BaO} \cdot \text{Fe}_2\text{O}_3$ – B_2O_3 section of the phase diagram. (D Section) Fig. 8 $\text{BaO} \cdot \text{Fe}_2\text{O}_3$ – $3\text{BaO} \cdot \text{B}_2\text{O}_3$ section of the phase diagram. (F Section)

$\text{mol}\% \text{B}_2\text{O}_3$ at 1413 ± 10 K and between $\text{BaO} \cdot \text{Fe}_2\text{O}_3$ and $3\text{BaO} \cdot \text{B}_2\text{O}_3$ with the composition of $55.7 \text{ mol}\% \text{BaO}$ – $33.0 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $15.3 \text{ mol}\% \text{B}_2\text{O}_3$ at 1331 ± 10 K.

The result of Section D is shown in Fig. 6. Regions of two phase equilibria of $\text{BaO} \cdot 6\text{Fe}_2\text{O}_3 + \text{L}$, $3\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{L}$ and $2\text{BaO} \cdot \text{Fe}_2\text{O}_3 + \text{L}$ exist. Two eutectic reactions occur between $3\text{BaO} \cdot \text{B}_2\text{O}_3$ and $\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$ with the composition of $46.9 \text{ mol}\% \text{BaO}$ – $23.1 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $30.0 \text{ mol}\% \text{B}_2\text{O}_3$ at 1101 ± 10 K and between $2\text{BaO} \cdot \text{Fe}_2\text{O}_3$ and $3\text{BaO} \cdot \text{B}_2\text{O}_3$ with the composition of $57.6 \text{ mol}\% \text{BaO}$ – $28.6 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $13.8 \text{ mol}\% \text{B}_2\text{O}_3$ at 1373 ± 10 K.

The result of Section E is shown in Fig. 7. Regions of two-phase equilibria of $\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{L}$, $3\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{L}$ and $2\text{BaO} \cdot \text{Fe}_2\text{O}_3 + \text{L}$ exist. Two eutectic reactions are observed between $\text{BaO} \cdot \text{B}_2\text{O}_3$ and $3\text{BaO} \cdot \text{B}_2\text{O}_3$ with the composition of $50.0 \text{ mol}\% \text{BaO}$ – $16.7 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $33.3 \text{ mol}\% \text{B}_2\text{O}_3$ at 1087 ± 10 K and between $2\text{BaO} \cdot \text{Fe}_2\text{O}_3$ and $3\text{BaO} \cdot \text{B}_2\text{O}_3$ with the composition of $21.5 \text{ mol}\% \text{BaO}$ – $64.5 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $14.0 \text{ mol}\% \text{B}_2\text{O}_3$ at 1420 ± 10 K. Though the region marked with the dotted line has not been confirmed yet, it is expected that a eutectic reaction exists in the region.

The result of Section F is shown in Fig. 8. Regions of two-phase equilibria of $\text{BaO} \cdot \text{Fe}_2\text{O}_3 + \text{L}$, $2\text{BaO} \cdot \text{Fe}_2\text{O}_3 + \text{L}$

and $3\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{L}$ exist. Two eutectic reactions are found between $\text{BaO} \cdot \text{Fe}_2\text{O}_3$ and $2\text{BaO} \cdot \text{Fe}_2\text{O}_3$ with the composition of $56.5 \text{ mol}\% \text{BaO}$ – $37.0 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $6.5 \text{ mol}\% \text{B}_2\text{O}_3$ at 1528 ± 10 K and between $2\text{BaO} \cdot \text{Fe}_2\text{O}_3$ and $3\text{BaO} \cdot \text{B}_2\text{O}_3$ with the composition of $64.0 \text{ mol}\% \text{BaO}$ – $22.0 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $14.0 \text{ mol}\% \text{B}_2\text{O}_3$ at 1425 ± 10 K.

The result of Section G is shown in Fig. 9. Regions of two-phase equilibria of $\text{BaO} \cdot \text{Fe}_2\text{O}_3 + \text{L}$, $3\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{L}$ and $\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{L}$ exist. Two eutectic reactions are found between $3\text{BaO} \cdot \text{B}_2\text{O}_3$ and $\text{BaO} \cdot \text{Fe}_2\text{O}_3$ with the composition of $50.0 \text{ mol}\% \text{BaO}$ – $34.0 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $16.0 \text{ mol}\% \text{B}_2\text{O}_3$ at 1343 ± 10 K and between $\text{BaO} \cdot \text{B}_2\text{O}_3$ and $3\text{BaO} \cdot \text{B}_2\text{O}_3$ with the composition of $50.0 \text{ mol}\% \text{BaO}$ – $16.7 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $33.3 \text{ mol}\% \text{B}_2\text{O}_3$ at 1087 ± 10 K.

The result of Section H is shown in Fig. 10. Regions of two-phase equilibria of $\text{BaO} \cdot 6\text{Fe}_2\text{O}_3 + \text{L}$ and $\text{BaO} \cdot \text{Fe}_2\text{O}_3 + \text{L}$ exist. A eutectic reaction is found between $\text{BaO} \cdot \text{Fe}_2\text{O}_3$ and $\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$ with the composition of $43.0 \text{ mol}\% \text{BaO}$ – $43.0 \text{ mol}\% \text{Fe}_2\text{O}_3$ – $14.0 \text{ mol}\% \text{B}_2\text{O}_3$ at 1443 ± 10 K.

The result of Section I is shown in Fig. 11. Regions of two-phase equilibria of $\text{BaO} \cdot 6\text{Fe}_2\text{O}_3 + \text{L}$ and $3\text{BaO} \cdot$

Fig. 9 $\text{BaO} \cdot \text{Fe}_2\text{O}_3$ – $\text{BaO} \cdot \text{B}_2\text{O}_3$ section of the phase diagram. (G Section)

Fig. 12 $(\text{BaO})_{0.90} \cdot (\text{Fe}_2\text{O}_3)_{0.10}$ – $(\text{B}_2\text{O}_3)_{0.90} \cdot (\text{Fe}_2\text{O}_3)_{0.10}$ section of the phase diagram. (J Section)

Fig. 10 $\text{BaO} \cdot \text{Fe}_2\text{O}_3$ – B_2O_3 section of the phase diagram. (H Section)

Fig. 11 $(\text{BaO})_{0.80} \cdot (\text{B}_2\text{O}_3)_{0.20}$ – $(\text{Fe}_2\text{O}_3)_{0.80} \cdot (\text{B}_2\text{O}_3)_{0.20}$ section of the phase diagram. (I Section)

$\text{B}_2\text{O}_3 + \text{L}$ exist. A eutectic reactions is found between $3\text{BaO} \cdot \text{B}_2\text{O}_3$ and $\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$ with the composition of 48.0 mol% BaO –32.0 mol% Fe_2O_3 –20.0 mol% B_2O_3 at 1350 ± 10 K.

The result of Section J is shown in Fig. 12. Regions of

two-phase equilibria of $\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{L}$ and $3\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{L}$ exist. A eutectic reactions is found between $3\text{BaO} \cdot \text{B}_2\text{O}_3$ and $\text{BaO} \cdot \text{B}_2\text{O}_3$ with the composition of 55.0 mol% BaO –10.0 mol% Fe_2O_3 –35.0 mol% B_2O_3 at 1123 ± 10 K.

3. Liquidus surfaces of pseudo-ternary phase diagram

The liquidus surfaces of the pseudo-ternary phase diagram of the BaO – Fe_2O_3 – B_2O_3 system were determined according to the results shown in Fig. 3 to Fig. 12. The resultant liquidus surfaces of the pseudo-ternary phase diagram are shown in Fig. 13 in the composition range of less than 50 mol% B_2O_3 . No ternary BaO – Fe_2O_3 – B_2O_3 compound is found in this region. The following phase transformations occur in cooling on the lines of L_1 to L_9 shown in Fig. 13;

For the pseudo-binary BaO – Fe_2O_3 or BaO – B_2O_3 system, phase transformations occur at the points of e_1 , e_2 , e_3 , e_4 and p_1 in Fig. 13 which are expressed as follows;

The transformation points of e_1 , e_2 and p_1 were reported

Fig. 13 Phase diagram of the pseudo-ternary BaO-Fe₂O₃-B₂O₃ system, showing the liquidus surface.

by Zioulouski⁽²⁰⁾ and that of e_3 and e_4 were reported by Levin and Ugrinic⁽²¹⁾. In the region examined in the present study, a ternary eutectic reaction (E_1) is $L \rightarrow \text{BaO} \cdot 6\text{Fe}_2\text{O}_3 + \text{BaO} \cdot \text{B}_2\text{O}_3 + 3\text{BaO} \cdot \text{B}_2\text{O}_3$, occurring at 1033 K with the composition of 48 mol% BaO-19 mol% Fe₂O₃-33 mol% B₂O₃. The points of R_1 , R_2 and R_3 are those of phase transformations obtained from the results of this study. The points of R_1 , R_2 and R_3 of forth phase equilibria of $\text{BaO} + 3\text{BaO} \cdot \text{B}_2\text{O}_3 + 2\text{BaO} \cdot \text{Fe}_2\text{O}_3 + L$, $3\text{BaO} \cdot \text{B}_2\text{O}_3 + 2\text{BaO} \cdot \text{Fe}_2\text{O}_3 + \text{BaO} \cdot \text{Fe}_2\text{O}_3 + L$ and $3\text{BaO} \cdot \text{B}_2\text{O}_3 + \text{BaO} \cdot \text{Fe}_2\text{O}_3 + \text{BaO} \cdot 6\text{Fe}_2\text{O}_3 + L$ exist.

R_1 : $1501 \pm 10 \text{ K}$

R_2 : $1373 \pm 10 \text{ K}$

R_3 : $1313 \pm 10 \text{ K}$.

The region where the liquidus surfaces are in equilibrium with Fe₃O₄ and BaO·6Fe₂O₃ spreads toward BaO·B₂O₃. The region where the liquidus surface is in equilibrium with Fe₃O₄ widely spreads toward BaO·B₂O₃. The liquidus surface equilibrated with BaO·6Fe₂O₃ also extends long and narrow in the same direction.

Figure 14 shows the isothermal lines on the liquidus surface of the pseudo-ternary phase diagram of the BaO-Fe₂O₃-B₂O₃ system. The temperature at which liquidus phase and the BaO·6Fe₂O₃ or Fe₃O₄ primary phase are in equilibrium is relatively high in the region of less than 20 mol% B₂O₃ and abruptly decreases when the B₂O₃ content exceeds 20 mol%. The liquidus surface in equilibrium with BaO·6Fe₂O₃ forms a steep slope, while the slopes of the liquidus surface equilibrated with BaO·B₂O₃ or 3BaO·B₂O₃ are more gentle than those in equilibria with Fe₃O₄ and BaO·6Fe₂O₃. The outline, which was reported by Haberey⁽²²⁾, of the three-dimen-

Fig. 14 Phase diagram for the pseudo-ternary BaO-Fe₂O₃-B₂O₃ system, showing isotherms of the liquidus surface.

sional region of primary crystallization of $\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$ is similar to this result.

IV. Conclusions

The isothermal section at 973 K and the liquidus surface of the pseudo-ternary phase diagram of the BaO – Fe_2O_3 – B_2O_3 system have been determined in a composition range of the B_2O_3 content less than 50 mol%. No pseudo-ternary compound exists in this range. The ternary eutectic reaction is $\text{L} \rightarrow \text{BaO} \cdot 6\text{Fe}_2\text{O}_3 + \text{BaO} \cdot \text{B}_2\text{O}_3 + 3\text{BaO} \cdot \text{B}_2\text{O}_3$, occurring at 1033 K at the composition 48 mol% BaO –19 mol% Fe_2O_3 –33 mol% B_2O_3 .

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