Effect of magnetic field on thermal convection of phosphate glass melts

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Abstract. Flow behaviour has been observed in binary alkali phosphate glass melts (xR₂O-2P₂O₅; R=Li, K; x=1, 3) under vertical magnetic field up to 3 T. The flow velocity of thermal convection decreased gradually to about one-third of the velocity at 0 T with an increase in magnetic field up to 1 T in the melt of low alkali content glass, and reached the constant value, under the condition of \( \frac{\partial B}{\partial z} < 0 \). In contrast, the flow velocity was almost unchanged in the melt of high alkali content glass up to 3 T. On the other hand, the flow velocity decreased gradually to half of the velocity at 0 T with an increase in magnetic field in the melts of high alkali metal content glasses up to 3T, under the condition of \( \frac{\partial B}{\partial z} > 0 \). The velocity decreased more gradually in the lithium containing melt than in the potassium containing melt.

It was deduced from these observations that the flow behaviour under magnetic field was the resultant of balance between the thermomagnetic convection under the gradient of field and the electro-magnetic force by induced current.

1. Introduction

Recently, studies of magnetic effects on material transfer in fluid are accelerated with a recent progress of the generation of high magnetic fields above 10T using superconducting magnet [1-2]. Especially, behaviour of convection flow of metal and semiconductor melts under magnetic fields is important for their production process. The flow behaviour has been explained by magnetohydrodynamics (MHD) [3-5]. On the other hand, the flow and convection of non-magnetic melts with low electronic conductivity are thought to be less affected by magnetic field according to MHD, because the melt itself consists of both positively and negatively charged ions without conduction electrons, and is neutrally charged as a whole. Nevertheless, Miyazawa et al.[6] have reported that the flow behaviour of non-magnetic insulator melts is changed by the application of a magnetic field due to an imbalance in electromagnetic forces acting on the positively and negatively charged ions. It is interesting to study the effect of magnetic field on the flow behaviour of glass melts which is a kind of non-magnetic insulator melts, because the control of the flow and convection of glass melt is an important subject in the production of homogeneous and clear glass materials. There is, however, no systematic studies regarding the effect of magnetic fields on the glass melts. In a previous study, we developed an in-situ observation system of the high temperature melt under high magnetic fields (up to 800°C and 10T) and observed the suppression of convection flow of a potassium phosphate glass melt, which has negative magnetic susceptibility, for the first time [7]. In general, glass melt consists of positively charged modifier ions and negatively charged structural units.
constructing networks which correspond to potassium ions and PO₄ chains in the potassium phosphate glass melt, respectively. It is expected that the kinds of alkali ions and the size of PO₄ chain would be affected the flow behaviour under magnetic field. In the present study, therefore, we have studied the flow behaviour of melts of lithium and potassium phosphate glasses under magnetic fields. The effect of magnetic field on the convection flow is discussed in terms of concentration and kinds of alkali ions in the melts.

2. Experimental procedure

Binary alkali phosphate xR₂O·2P₂O₅ (R=Li, K; x=1, 3) glasses were prepared from Li₂CO₃, K₂CO₃ and (NH₄)₂HPO₄ as starting materials by usual liquid-quenching method. Glass pieces were re-melted in a quartz crucible at 600-650°C in an *in-situ* observation furnace as shown in figure 1. Some pieces of thin gold film were added to the glass melt as markers for the visualization of convection flow. Bubbles in the melt were also used as markers. Convection flow was observed by the CCD camera from upper side through quartz windows. The observation system under magnetic field has been described in detail elsewhere [7]. The magnetic field was applied along the vertical direction by using a superconducting magnet 15T-CSM at the Institute for Materials Research, Tohoku University, Japan. Magnetic field was increased stepwise and kept constant for several minutes to eliminate the influence of the sweeping magnetic field. Then moving picture was recorded for about 5 minutes.

In the first experiment, the crucible was set at a position where the bottom of the crucible is located at the centre of magnet as shown by (A) in figure 2. This brought a negative field gradient (\( \frac{dB}{dz} < 0 \)) which generates an upward magnetic force of an order of \( 10^{-2} \) N in the melt. Flow velocity was in the range of 100-200 \( \mu m/s \) at 0T in any run. The velocity was

3. Results and Discussion

A simple flow pattern of thermal convection was observed in the glass melt in any runs due to the temperature distribution of the melts in the crucible as shown in figure 2. The distribution might be due to slight off-center arrangement of the crucible and be within a few tens degree. The melt flowed upwards at one (high-temperature) side of the crucible wall and downwards at the opposite (low-temperature) side. The flow velocity of the melt was calculated from the motion of markers at the surface of the melt from the recorded movie image.

At first, the crucible was set at a position where the bottom of the crucible is located at the centre of magnet (A). Susceptibility of the present glasses is estimated to be in the range from -5.3 to -6.5x10⁻⁹ m³/kg by their ionic ones. This setting brought a negative field gradient (\( \frac{dB}{dz} < 0 \)) which generates an upward magnetic force of an order of \( 10^2 \) N in the melt. Flow velocity was in the range of 100-200 \( \mu m/s \) at 0T in any run. The velocity was
normalized by the velocity at 0T and is plotted against magnetic field up to 3T in figure 3. The flow velocity of the convection decreased gradually to about one-third of the velocity at 0 T with an increase in magnetic field up to 1T in the melt of low lithium content (Li,O-2P,O) glass, and reached the constant value. In contrast, the flow velocity was almost unchanged in the melt of high lithium content (3Li,O-2P,O) glass up to 3T. These behaviours were independent of the kinds of alkali ions. Kitamura et al. [8] have suggested the decrease in negative magnetic susceptibility of the optical glass with increasing temperature from the magnetic levitation experiment. This implies that upward magnetic force acting on the melt in the high temperature region in the crucible should be higher than the force in the lower temperature region. Therefore the convection should be accelerated by magnetic force. The velocity was, however, suppressed by applying magnetic fields in the melt of low alkali content and was unchanged in the melt of high alkali content. This discrepancy between the experimental results and the consideration based on the magnetic force suggests electromagnetic force stronger than or comparable to the magnetic force acts on the melt.

Structure of P,O, glass has random network of PO₄ tetrahedra which connect with each other at 3 corners of the tetrahedra. Addition of alkali oxides brake the linkage between PO₄ tetrahedra with producing negatively charged terminal (non-bridging) oxygen ions in the network and alkali ions neighboring the negatively charged oxygen ions balance the charge like as ≡P-O^- R^+ R^+ -O-P≡. In other words, alkali phosphate glasses consist of positively charged alkali ions and negatively charged structural units consisting of PO₄ tetrahedra (the latter is called PO₄ clusters or clusters hereafter for convenience) and the size of PO₄ clusters decreases with an increase in alkali oxide content. The network structure in the melt is thought to be similar to that in glass, while some parts of the network are broken by thermal disturbance. Therefore, the melt of low alkali content (R₂O-2P₂O₅) consists of large PO₄ clusters (long or infinite cross-linked chains) and alkali metal ions. Although, both of negatively charged clusters and positively charged alkali ions act as charge carriers under magnetic field, it is expected that the large PO₄ clusters is much less mobile than alkali ions. Thus, only the alkali ions should act as charge carrier like as electrons in conductive fluids in the melt of low alkali content glass. It is known that electric current is induced toward direction perpendicular to the flow at the position where the strength of magnetic field perpendicular to the flow changes as shown by "I" in figure 2, according to the MHD. The Lorentz force was generated by the electric current due to the movement of alkali ions, resulting in the deceleration of the flow velocity. The direction of the flow alternated between the vertical and transverse direction in the convection cycle in the present study. In other word, the field strength perpendicular to the flow alternated in the convection cycle. Therefore, it was deduced that the Lorentz force acted on the glass melt toward the counter direction of the flow at the position where the strength of magnetic field perpendicular to the flow direction increased in the convection cycle. However, the ionic attractive force between alkali ions and the non-bridging oxygen

**Figure 3.** Normalized flow velocity of the melts of low and high alkali content phosphate glasses under magnetic field with negative gradient field (BdB/dz<0).

**Figure 4.** Normalized flow velocity of the melts of high alkali content phosphate (3R₂O-2P₂O₅) glasses under magnetic field with positive gradient field (BdB/dz>0).
ions should restrict the movement of alkali ions, leading to the suppression of electric current and the suppression of the resultant Lorentz force under lower magnetic field. Thus, it was deduced that the movement of alkali ions got rid of the restriction by the clusters above 1T.

On the other hand, small PO₄ clusters such as P₄O₁₃ is dominant in the melt of high alkali content (x=3) glass. The small PO₄ clusters are mobile and act as carriers as same as alkali ions. Nevertheless, the mobility of small PO₄ clusters would be lower than that of alkali ions. Since the direction of the movement of the clusters is opposite to that of alkali ions, the electromagnetic force to suppress the flow should be reduced. Consequently, the velocity was insensitive to the magnetic field.

Secondary, the crucible was set at a position where the surface of the melt is located at the centre of magnet (B). This brought a positive field gradient ($\frac{dB}{dz} > 0$) which generates a downward magnetic force of an order of $10^{-2}$ N in the melt. The flow velocity of the melt of high alkali content (3R₂O-2P₂O₅) glass is plotted against magnetic field up to 3T in figure 4. The flow velocity decreased gradually to half of the velocity at 0T with an increase in magnetic field in the melts up to 3T, as shown in the figure. Since the negative magnetic susceptibility should decrease with increasing temperature. In a positive field gradient, it is reasonable that downward magnetic force acts on the melt in the high temperature region is higher than in the lower temperature region, which suppresses the convection flow. In addition, the electromagnetic force to suppress the convection flow should be the same as the former case. Thus both the forces decelerate the thermal convection. The flow velocity decreased more gently in the 3Li₂O-2P₂O₅ glass melt than in the 3K₂O-2P₂O₅ glass melt. The detail mechanism is, however, yet unclear.

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References
[1] Kitazawa K, Hirota N, Ikezoe Y and Uetake H 2000 Proc. of the 3rd International Symposium on Electromagnetic Processing of Materials p 9
[2] Wakayama N I, 1999 SPIE Proc. 3972 p 102
[3] Hunt J C R, 1965 Fluid Mech. 21 577
[4] Witt A F, Herman C J and Gatos H G, 1970 J. Mater. Sci. 5 822
[5] Hoshi K, Suzuki T, Okano Y and Iwasa N, 1980 Extended Abstract of ECS Meeting p 811
[6] Miyazawa Y, Morita S and Sekiwa H, 1996 J. Cryst. Growth 166 286
[7] Kitamura N, Fukumi K, Nishii J, Takahashi K, Mogi I, Awaji S and Watanabe K, 2004 J. Ceram. Soc. Jpn. 112 S1210
[8] Kitamura N, Makihara M, Hamai M, Sato T, Mogi I, Awaji S, Watanabe K and Motokawa M, 2000 Jpn. J. Appl. Phys. 39 L324