Effect of Magnetic Field on Thermal Convection of Glass Melt

Naoyuki KITAMURA, Kohei FUKUMI, Junji NISHII, Kohki TAKAHASHI*, Iwao MOGI*, Satoshi AWAIJ* and Kazuo WATANABE*

National Institute of Advanced Industrial Science and Technology, 1–8–31, Midorigaoka, Ikeda, Osaka  563–8577, Japan
*Institute for Materials Research, Tohoku University, 2–1–1, Katahira, Aoba, Sendai, Miyagi 980–8577, Japan

We have developed a system for in-situ observation of glass melts under high magnetic field up to 10 T. Thermal convection in binary potassium phosphate melts was observed under vertical and transverse magnetic fields. Above 1 T, the flow velocity of the convection decreased to one-third of the velocity at 0 T in the melt of low potassium content glass by applying vertical and transverse magnetic fields. Contrary to this, the flow velocity increased in the melt of high potassium content glass by applying transverse magnetic fields of around 1 T. It was deduced that different electro-magnetic forces act on positively charged potassium ions and negatively charged clusters consisting of PO₄ tetrahedra resulting in the attenuation or acceleration of the flow velocity of the melts.

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1. Introduction

After a recent progress in the generation of high magnetic fields above 10 T using superconducting and hybrid type magnets, studies on the magnetic and electronic properties of materials have been accelerated, and in addition, high magnetic fields have been applied to many of the fabrication processes for metals and semiconductors. For example, the effects of magnetic fields on the thermal convection of low viscosity melts of inorganic materials, gas absorption into liquid, motion of bubbles in fluid and so on, have been studied for the purpose of improvements to the fabrication processes. Generally speaking, the effects of magnetic field on flow and convection are based on magnetohydrodynamics (MHD) in metal and semiconductor liquids. In other words, it is expected from MHD, that the flow and convection are less affected by magnetic field in non-magnetic oxide melts with low electronic conductivity, because the melt itself consists of both positively and negatively charged ions without conduction electrons, and is neutralized as a whole. Nevertheless, Miyazawa et al. have reported that the flow behavior of non-magnetic liquids of insulators is changed by the application of a magnetic field. Their study suggests that magnetic fields can be utilized to control the flow and convection of glass melts, which is an important subject in the fabrication process of homogeneous and clear glass materials. There is, however, no systematic study regarding the effect of magnetic fields on the flow in glass melts. For the application of a magnetic field, it is important to clarify the effects of the field on flow and convection in terms of the kinds of structural units (glass network formers) and network modifier ions, coordination and so on. In the current paper, we have developed a system for in-situ observation of glass melts under high magnetic field up to 10 T. Thermal convection was studied in the melts of K₂O–P₂O₅ system under high magnetic field. The effect of high magnetic fields on the flow velocity and direction is discussed in relation to the structure of the melt.

2. Experimental procedures

We developed an electric furnace equipped with a video system, which works under magnetic field higher than 10 T. Figures 1(a) and (b) show a block diagram of in-situ observation system under a high magnetic field and a photograph of whole the system including a superconducting magnet, respectively. The components of the furnace are non-magnetic with the exception of a heater wire (Kanthal Gaderius). The ferromagnetism of the heater material, however, disappeared at temperatures higher than approximately 500°C. The majority of the heater wire was wired parallel to the vertical magnetic field in order to minimize the Lorentz force acting on it under a high magnetic field. An image of the glass melt in a silica glass crucible was recorded using a small CCD camera (ELMO, MN43H) through two quartz glass windows as shown in Fig. 1(a). The camera, which was settled adjacent to the top window, was cooled by airflow, so that it worked even when the temperature of the melt rose to over 900°C. Illumination was introduced into the furnace using a quartz glass rod.

Thermal convection was observed in K₂O–2P₂O₅ and 3K₂O–2P₂O₅ glass melts. Since the melts were transparent as shown in Fig. 2, some pieces of thin gold film were added to the melt as markers for the visualization of the convection flow. Bubbles in the melt were also used as markers. The glasses were melted in quartz crucibles at about 600°C. A vertical magnetic field below 10 T was applied using superconducting magnets 6 T–CSM and 15 T–CSM at the Institute for Materials Research, Tohoku University in Sendai, Japan. The vertical magnetic field was applied up to 4 T with increments of 1 T after holding the temperature for 30 minutes until the thermal convection stabilized. An image of the flow of the melt was recorded for 10 minutes at each step. The velocity of the flow was analyzed from the motion of the markers in the melt. The latter 5 minutes at each step was used for analysis of flow velocity and direction of the thermal convection to eliminate the flow behavior on the sweeping magnetic field. A transverse magnetic field up to 0.8 T was applied using a standard Weiss type water-cooled magnet (Bruker, B–E25).

3. Results

We have successfully performed an in-situ observation of glass melt under high magnetic field up to 10 T. One remaining problem is the cooling of the quartz glass window, which became apparent above around 8 T. Since evaporation from the glass melt accelerates under a high magnetic field, the cooling could be caused by the evaporation and condensation of the glass melt. However, even under a high magnetic field above 8 T, the behavior of the flow can be observed within a short period of time.

A simple pattern of thermal convection was observed in
both the glass melts in any of the runs due to the temperature distribution of the melts in the crucible. The melt flowed upwards at one side of the crucible wall and downwards at the opposite side. The velocity of the flow along the surface was in the range of 200 to 300 μm/sec at 0 T in K₂O-2P₂O₅ melt. Figure 3 shows two dimensional (2D) representations of the flow velocity and the direction of thermal convection at the surface of the K₂O-2P₂O₅ melt under a vertical magnetic field up to 4 T. The application of the vertical magnetic field suppressed the flow velocity to one-third of the flow velocity at 0 T, and turned the direction counterclockwise. The velocity is almost constant above 1 T. The velocity and direction of the flow changed back toward those at 0 T to some extent after the field reached 0 T, although the phenomenon was not perfectly reversed within the period of several tens of minutes. To confirm the effect of the direction of magnetic field on the flow, the observation of flow was carried out below 1 T using the Weiss type magnet. Figure 4 shows 2D representations of the flow vectors under transverse magnetic fields up to 0.8 T. The velocity decreased gradually at a critical field between 0.6 and 0.8 T to one-third of the velocity at 0 T. Rotation of the flow direction was not observed up to 0.8 T. The phenomenon on the flow velocity seemed to be independent to the direction of the magnetic field.

The observation was performed on 3K₂O-2P₂O₅ glass melt in the same manner as for the K₂O-2P₂O₅ glass melt. Almost all the gold markers sedimented and only a few bubbles remained in the melt due to low viscosity. Therefore the statistics are not high enough to analyze the flow behavior in detail, however, the tendency of the flow velocity under high magnetic field could be observed as shown in Fig. 5. The flow velocity in the 3K₂O-2P₂O₅ glass melt increased with increasing transverse magnetic field as seen in Fig. 5. The velocity increased approximately 6 times greater at the transverse field of 0.8 T than that at 0 T. On the other hand, the velocity was scarcely changed under the application of a vertical magnetic field up to 3 T. These findings in the 3K₂O-2P₂O₅ glass melt were much different from those in the K₂O-2P₂O₅ glass melt. It could not be confirmed whether the flow velocity returned...
back toward the initial value after decreasing the magnetic field to 0 T.

4. Discussion

4.1 K$_2$O–2P$_2$O$_5$ glass melt

The deceleration of the flow velocity by the application of a vertical magnetic field was observed in the K$_2$O–2P$_2$O$_5$ glass melt as shown in Fig. 3. It is known that electric current is induced toward the direction perpendicular to the flow of the melt at the position where the strength of magnetic field perpendicular to the flow changes, according to the magnetohydrodynamics (MHD). The electric current generates the Lorentz force toward the counter direction of the flow, resulting in the deceleration of the flow velocity. The direction of the flow alternated between the vertical and transverse directions in the convection cycle in the present study. In other words, 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 direction of flow was changed in the convection cycle. The K$_2$O–2P$_2$O$_5$ glass includes negatively charged clusters consisting of PO$_4$ tetrahedra with non-bridging oxygen ions and positively charged potassium ions. Since the average size of clusters should be larger than that of potassium ions in the K$_2$O–2P$_2$O$_5$ glass melt, the mobility of the clusters in the melt is considered to be smaller than that of potassium ions. Therefore, it was deduced that only potassium ions acted as charge carriers in the K$_2$O–P$_2$O$_5$ glass melt and that the Lorentz force was generated by the electric current caused by the movement of potassium ions. However, the ionic attractive force between potassium ions and the clusters should restrict the movement of potassium ions, leading to the suppression of electric current and the suppression of the resultant Lorentz force. Thus, it was deduced that the movement of potassium ions got rid of the restriction by the clusters below 1 T.

In addition to the Lorentz force mentioned above, the Lorentz force perpendicular to the flow direction should be generated at the surface of glass melt, because the positively charged ions and negatively charged clusters flow toward the direction perpendicular to the vertical magnetic field at the surface of glass melts. Therefore, it can be deduced that the force perpendicular to the flow direction acted on potassium ions and the clusters under vertical magnetic field, which caused the rotation of flow direction as seen in Fig. 3. This consideration is supported in the previous study by Mogi et al. They demonstrated that silver metal grown by the Diffusion-Limited-Aggregation (DLA) process formed a spiral pattern under a homogeneous vertical magnetic field due to the Lorentz force acting on moving silver ions. The rotation of the flow can be explained by the generation of the Lorentz force. However, the direction of rotation is a complicated problem. For instance, it has been shown that the change in the direction of flow patterns is opposite to each other in the LiNbO$_3$ and TiO$_2$ melts under vertical magnetic fields.

The deceleration of the flow velocity was found in the glass melt under transverse magnetic fields at about 0.8 T as shown in Fig. 4. This is also due to the generation of the Lorentz force at the position where the direction of flow was changed in the convection cycle, as in under a vertical magnetic field. The field at approximately 0.8 T is most likely the critical field at which potassium ions get rid of the restriction by clusters.

The deceleration was to some extent a reversible phenomenon. Full recovery may be difficult in a short period of time for the melt with high viscosity, because hysteresis was discovered even in the LiNbO$_3$ melt due to the viscosity. A change in direction of the flow was not found under a transverse magnetic field at the surface of the melt, because the field strength is low and the change in flow direction should be toward the vertical direction if it occurred due to the direction of the generated Lorentz force.

4.2 3K$_2$O–2P$_2$O$_5$ glass melt

In the observation of the 3K$_2$O–2P$_2$O$_5$ glass melt as shown in Fig. 5, the flow velocity accelerated by approximately 4 times with increasing transverse magnetic field up to 0.8 T, while it was scarcely changed under the application of a vertical magnetic field. These findings observed in the 3K$_2$O–2P$_2$O$_5$ glass melt were much different from those in the K$_2$O–2P$_2$O$_5$ glass. This difference in flow behavior between the K$_2$O–2P$_2$O$_5$ and 3K$_2$O–2P$_2$O$_5$ glass melts may be due to the difference in the mobility of clusters in the melts, since it has been proposed that the acceleration of flow velocity found in the viscous melt is due to the difference of the mobility between the positively charged carriers and the negatively charged carriers. Since the sedimentation of gold markers and the remainder of only few bubbles were observed only in the 3K$_2$O–2P$_2$O$_5$ glass melt, it can be deduced that the viscosity of the 3K$_2$O–2P$_2$O$_5$ glass melt is lower than that of the K$_2$O–2P$_2$O$_5$ glass melt, that is, the cluster size in the former is smaller than that in the latter. Reduction of cluster size leads to the increase of mobility of clusters. Therefore, it is expected that the negatively charged clusters act as charge carriers together with positively charged potassium ions in the 3K$_2$O–2P$_2$O$_5$ glass melt, although the mobility of the cluster might be small as compared with potassium ions. On the other hand, only potassium ions acted as a charge carrier in the K$_2$O–2P$_2$O$_5$ glass melt. The difference in the kinds of charge carriers between 3K$_2$O–2P$_2$O$_5$ and K$_2$O–2P$_2$O$_5$ glass melts may cause the difference in flow behavior. The detailed mechanism is, however, yet unclear.

5. Summary

We developed an in-situ observation system of glass melt under high magnetic field up to 10 T. The flow attenuation and acceleration of the thermal convection by applying a magnetic field are found in the melts of the K$_2$O–2P$_2$O$_5$ and 3K$_2$O–2P$_2$O$_5$, respectively. The effect of magnetic field on the flow mainly relates to the idea based on the MHD. However, since the acceleration of the flow velocity, which was found in the latter glass melt, is difficult to explain now, a model which provides a universal explanation for all behaviors in the present study and also in past studies of the high field effect of
the flow observed in the melts of inorganic materials is necessary in the future.

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References