# An Effect of Transfer Velocity on Formation of Porous Alumina during Unidirectional Solidification

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## ABSTRACT

An effect of transfer velocity on the formation of porous alumina during the unidirectional solidification under pressurized 50%$H_2$-50%Ar mixed gas was examined. The porosity of the samples increases with the transfer velocity up to 400 mm/h (111 μm/s) and then, it gradually decreases. The pore morphology such as positional distribution in the samples and pore direction against solidification direction varies with the transfer velocity. When the transfer velocity is 400 mm/h, the sample with a maximum porosity was obtained where the angle between solidification direction and pore direction is zero. The angle between solidification direction and pore direction increases with the transfer velocity.

## 1. INTRODUCTION

*Porous alumina with cylindrical pores can be* candidate for a gas filter and catalyst carriers for high-temperature use because of its high thermal and chemical stabilities at high temperatures /1/. The authors proposed a fabrication method of porous alumina with cylindrical pores (lotus-type porous alumina) using optical floating zone method under pressurized hydrogen atmosphere /2/. The pores are formed at solid-liquid interface due to hydrogen solubility gap between solid and liquid phase at the melting point during the unidirectional solidification /2-4/. The pore morphologies such as porosity, pore size and positional distribution of pores can be controlled by experimental conditions such as hydrogen partial pressure, total pressure and addition of impurity phase in feed rod /2,5/.

Though the pores in lotus-type porous alumina show cylindrical shape, the pore direction for each pore is not always correspond with solidification direction /2/. Since the pores are formed at solid-liquid interface and grow along the heat gradient between solid and liquid phases during the solidification, it is expected that the pore directions can be controlled by controlling the shape of solid-liquid interface. Besides it is easy to expect that the shape of solid-liquid interface change *with transference velocity in the unidirectional* solidification process.

In this paper, the relation between pore direction and transference velocity was examined.

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## 2. EXPERIMENTAL PROCEDURE

For the preparation of alumina feed rod, high purity alumina (99.99% purity, Sumitomo Chemical Co. Ltd.) powder was used as starting materials. The slurry of the powder with a binder was poured into the mold. After drying in air, calcination was performed at 1473K for 7.2 ks in air and then, the feed rods with 8 mm diameter and 150 mm length were prepared.

Porous alumina was fabricated using optical floating zone apparatus (Euro-System Co. Ltd., SFZ-20065XV) in 50%$H_2$-50%Ar mixed gas. The total pressure, rod rotation rate and hydrogen partial pressure were fixed to 0.6 MPa, 20 rpm and 50%$H_2$ during the experiment. Xenon lamp was used as an optical source as shown in Figure 1. The infrared light emitted from the Xenon lamp is refracted by the elliptical mirror and focused into the central part in order to melt the feed rod of alumina. The feed rod was hooked on upper shaft and another feed rod was fixed with lower shaft. This melting system was set up in a quartz tube which was filled with mixture gas of hydrogen and argon. The transfer velocity was controlled from 5 mm/h to 3,060 mm/h.

The samples were cut in perpendicular and parallel to the solidification direction. The transversal and longitudinal cross-sections of the solidified samples were observed by SEM (JEOL, JSM-6360T). The porosity was calculated by computer program WinRoof (ver.5, Mitani Co. Ltd.) using the transversal cross section images.

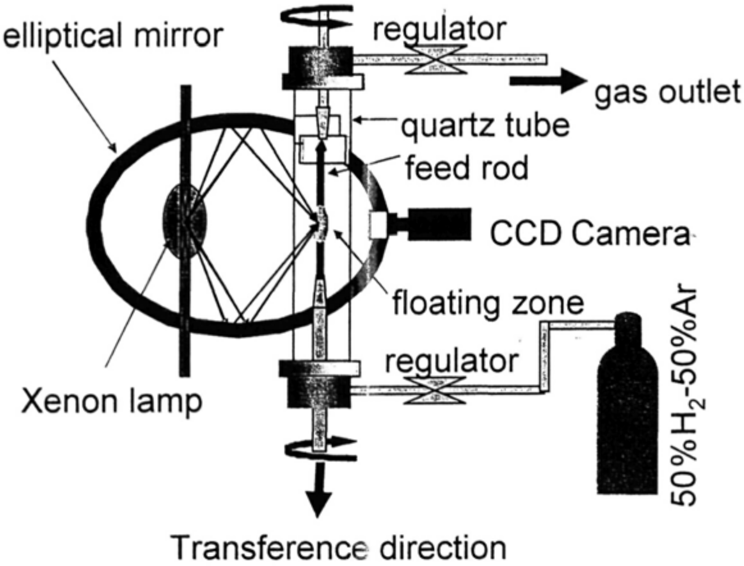

**Fig. 1:** Schematic drawing of optical floating zone furnace

## 3. RESULTS AND DISCUSSION

Figure 2 shows the transversal cross sectional views of the samples as a function of the transfer velocity. In the mechanism of pore growth during the solidification due to hydrogen gas solubility gap between solid and liquid phase at the melting point, an excess hydrogen gas in the solid phase near solid-liquid interface diffuses back to the liquid phase or diffuses into the pores. When the transfer velocity is slow, almost all of the excess hydrogen diffuses back to the liquid phase without forming the pores. In this experiment, a non-porous alumina was obtained for transfer velocity of 5 mm/h. Thus, the positional pore distribution, porosity and pore shape of the samples varied with the transfer velocity. For the transfer velocity less than 200 mm/h, the pores tend to be located in the center of the sample. On the other hand, for the transfer velocity more than 400 mm/h, pores are distributed in the brink of the sample.

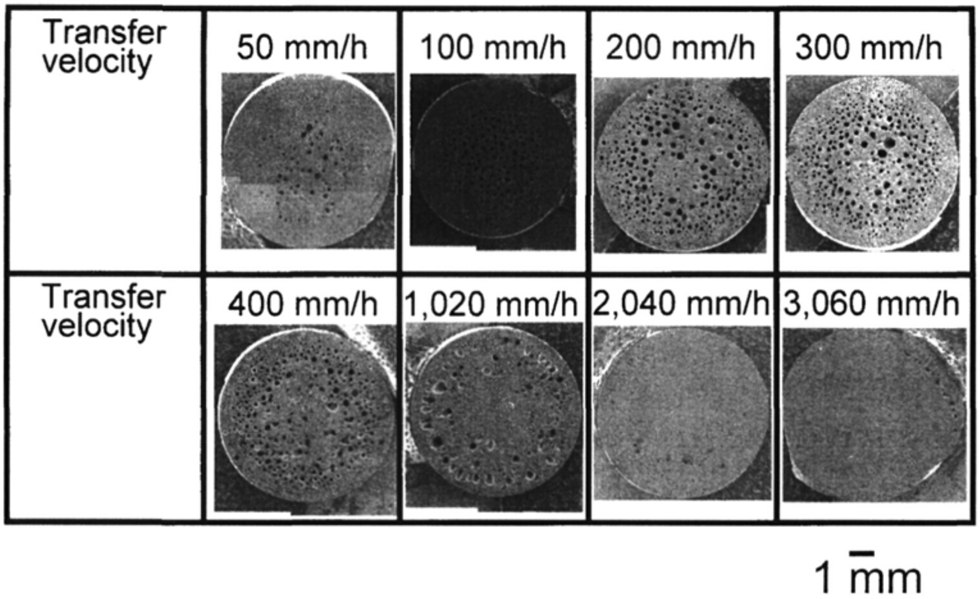

**Fig. 2:** Transversal cross sectional views of the samples for the transfer velocity of 50, 100, 200, 300, 400, 1,020, 2,040 and 3,060 mm/h.

Figure 3 shows the porosity change with transfer velocity where the porosity was measured from six different kinds of transversal cross sections for each sample. The porosity increases with increasing transfer velocity up to transfer velocity of 400 mm/h and then, it decreases gradually.

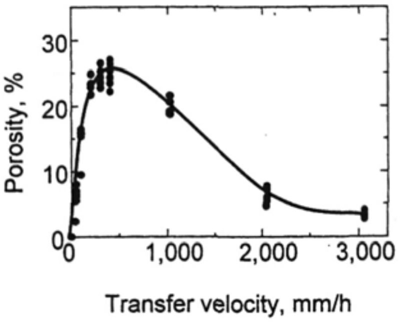

**Fig. 3:** Transfer velocity dependence of the porosity of the samples.

Figures 4 (a), (b) and (c) show the longitudinal cross sectional views of the samples transfer velocity of 100, 400 and 1,020 mm/h, respectively. For transfer velocity of 400 mm/h sample, all cylindrical pores are elongated in the solidification direction. However, for transfer velocity of 100 and 1,020 mm/h samples, the pores near the sample surface are titled from the solidification direction as indicated by arrows. The pores for the former and later cases are elongated toward outer and inner of the sample, respectively.

Figure 5 shows the angle between pore direction and solidification direction where the angle is calculated using ten pores near the sample surface. Since the number of pores is less than ten for transfer velocity of 5 and 3,060 mm/h samples, in the data of the angles for these samples are not plotted in the figure. The angle increases with decreasing transfer velocity blow transfer velocity of 400 mm/h. On the other hand, the angle increases with increasing transfer velocity above transfer velocity of 400 mm/h in the opposite direction.

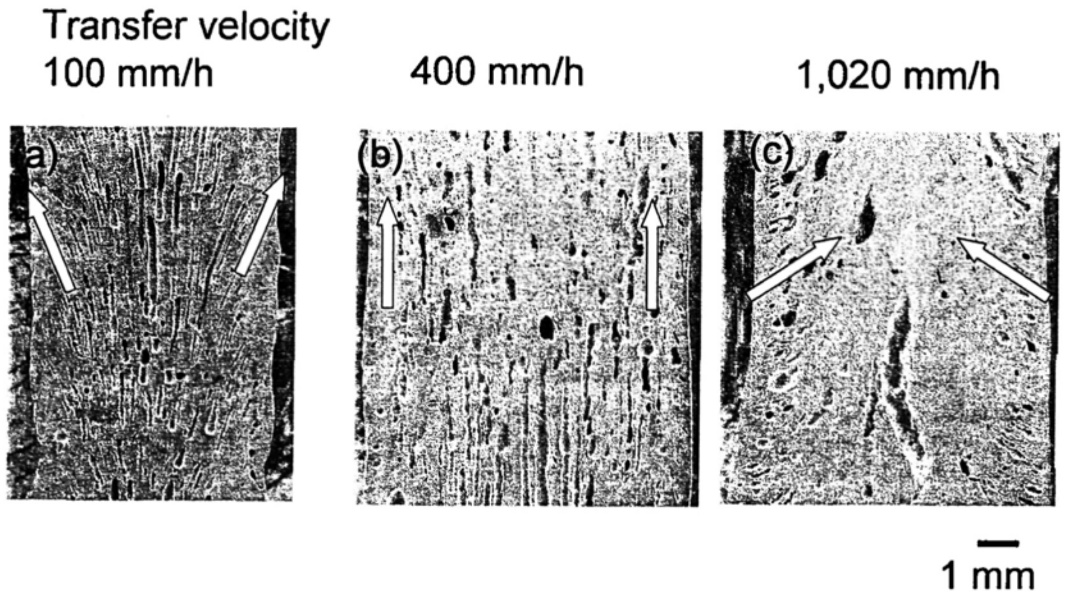

**Fig. 4:** Longitudinal cross section views of samples for the transfer velocity of (a) 100, (b) 400 and (c) 1,020 mm/h.

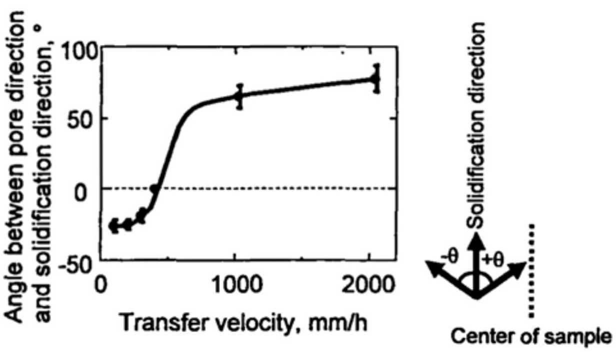

**Fig. 5:** Transfer velocity dependence of the angle between pore direction and solidification direction.

On the unidirectional solidification via optical floating zone method, the heat balance on the surface of liquid phase is achieved by heating from optical irradiation and heat loss onto environmental gas. Since the heat quantities of optical heating and cooling by environmental gas at the liquid surface are not always same, it is easy to understand that the heat balance on the liquid surface varies with the transfer velocity. According to the variety of heat balance on the liquid surface due to transfer velocity, the morphology of solid-liquid interface, namely, the direction of heat gradient against solidification direction also varies with the transfer velocity during the unidirectional solidification and then, the formed pores are titled from the solidification direction as shown in Figures 4 and 5. In the present study, the morphology of solid-liquid interface varies from convex shape to concave shape at the transfer velocity of 400 mm/h.

Since the pores grow toward sample surface on solid-liquid interface during the unidirectional solidification blow transfer velocity of 400 mm/h, a large amount of hydrogen gas is released from the sample surface on the interface and then, the porosity decreases with decreasing transfer velocity as shown in Figure 3 and pores are located at the center of the sample as shown in Figure 1. On the other hand, the excess hydrogen gas released into the liquid phase above transfer velocity of 400 mm/h, the porosity decreases with increasing transfer velocity and the pores are distributed on the brink of the sample as shown in Figure 1.

## 5. CONCLUSION

Since the morphology of solid-liquid interface varies from convex shape to concave shape at the transfer velocity of 400 mm/h for unidirectional solidification of alumina with 8 mm diameter, the pore growth direction tilt from the solidification direction. The angle between pore growth direction and solidification direction increases with decreasing transfer velocity blow transfer velocity of 400 mm/h. On the other hand, the angle increases with increasing transfer velocity above transfer velocity of 400 mm/h in the opposite direction.

## ACKNOWLEDGMENTS

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