Investigation of In-Flight Melting Behavior of Granulated Glass Raw Materials by Hybrid Heating of Twelve-Phase AC Arc with Oxygen Burner

Manabu Tanaka[1], Yaping Liu[1], Yosuke Tsuruoka[2], and Takayuki Watanabe[1]

[1] *Dept. Environmental Chemistry and Engineering, Tokyo Institute of Technology, Japan*
[2] *Dept. Energy Sciences, Tokyo Institute of Technology, Japan*

Abstract: An innovative in-flight glass melting technology was developed for a purpose of energy saving and environmental protection. Granulated glass raw materials with small diameter were treated by hybrid heating of multi-phase AC arc combined with an oxygen burner. A multi-phase AC arc and hybrid plasma were compared to study the in-flight melting behavior of the granulated raw materials. Furthermore, the effect of primary size of SiO_2 in the raw material was investigated. The well-vitrified glass powders with inhibited volatilization were prepared by the hybrid plasma heat-treatment.

Keywords: in-flight melting, multi-phase AC arc, hybrid plasma

1. Introduction

Typical melting system for glass production is refractory-lined melting furnace, fired by air and natural gas or oil as fuel, which has been used over 140 years in glass industry [1]. Glass industry is one of the most energy intensive industries, only second to the aluminum industry. During glass production, lots of emissions like CO_2, NO_x, etc. are produced due to the usage of fossil fuel, which brings more environmental pollution. Current glass melting technology, based on the Siemens furnace developed in 1860s, has evolved in response to manufacturing requirements. However, few revolutionary changes to the basic technology have occurred.

Under the support of New Energy and Industrial Technology Development Organization (NEDO) project in Japan, our previous works of innovative in-flight melting technology with an induction thermal plasma and a multi-phase AC arc have solved the above-mentioned shortcomings [2-7]. Furthermore, our most recent work [8] has reported about the technology called "hybrid plasma", which combines the multi-phase AC arc and the oxygen burner for the purpose of the enhancement of energy efficiency and the in-flight glass productivity. This so-called "hybrid" concept aims at exploiting the benefits of both heating sources.

The objective of this work is to investigate the in-flight glass melting behavior by hybrid plasma. The melting behavior of the granulated glass raw materials was compared between the in-flight treatment by the multi-phase AC arc and the hybrid plasma. Furthermore, the effect of primary size of SiO_2 in the granulated glass raw materials was investigated.

2. Experimental

2.1 Raw Material Preparation

The raw materials of alkali-free glass were prepared into granulated powders by using the spray-drying method as shown in Figure 1. The components of raw material for alkali-free glass with the composition of $49SiO_2$-$15B_2O_3$-$10Al_2O_3$-$25BaO$ in wt% were prepared by SiO_2, H_3BO_3, Al_2O_3, and $BaCO_3$. Two kinds of the granulated raw materials were prepared from SiO_2 particles with different sizes (7-8 μm and 1-2 μm). The average diameters of these granulated raw materials were 114 μm and

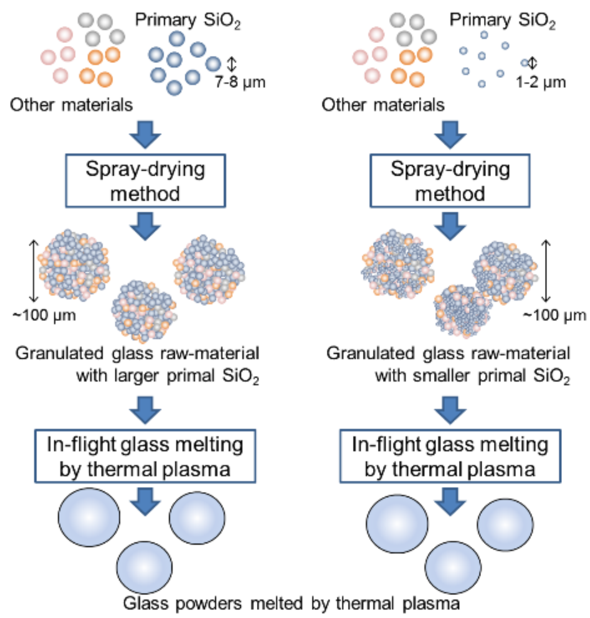

Figure 1. Schematic diagram of sample preparation.

105 µm, respectively. Then the prepared glass raw-materials were fed into the multi-phase AC arc and the hybrid plasma.

2.2 In-flight melting system

The schematic illustration of the hybrid plasma apparatus is shown in Figure 2. It combines a multi-phase AC arc plasma reactor with an oxygen burner. The multi-phase AC arc consists of 12 electrodes, a reaction chamber, a powder feeder and an AC power supply. The 12 electrodes were divided into two layers to increase the plasma volume, upper six inclined electrodes and lower six horizontal electrodes, and the angle between upper and lower electrodes was 30º. The electrodes were made of thoriated tungsten with diameter of 3.2 mm. The host of electrodes and powder feeder nozzle were cooled by water. Argon gas with purity of 99.99% was injected around the electrodes to prevent them from oxidation at a flow rate of 5 L min^{-1} for each electrode. Total power was about 50 kW which the total current was 300 A and total voltage was 200 V. The distance from powder feed nozzle to arc was 150mm and the diameter of arc was 100 mm and the powders were quenched on the bottom of an ion container at a distance of 1050 mm below the nozzle.

The oxygen burner, which consisted of a nozzle with cooling water, fuel and oxygen feeding system, is installed above the furnace chamber. Propane (C_3H_8) was used as the fuel. The flow rates of C_3H_8 gas, the primary oxygen and the secondary oxygen were 6, 6, and 24 L min^{-1}, respectively. The combustion power

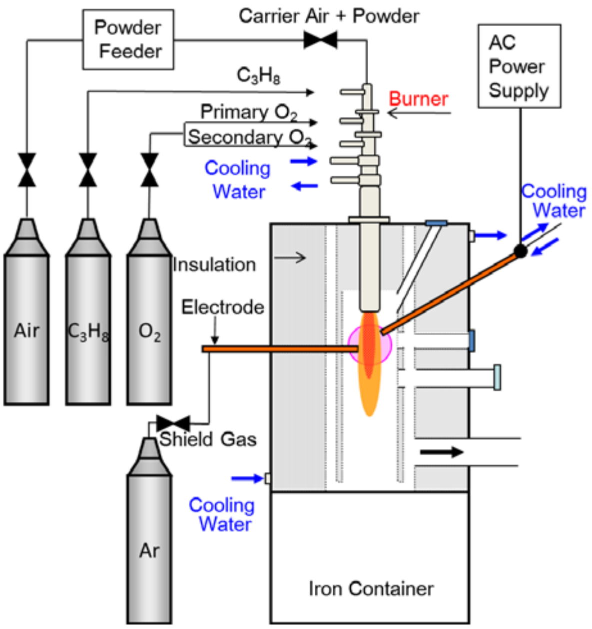

Figure 2. Schematic illustration of hybrid plasma melter.

of oxygen burner was 9 kW. More detail about the hybrid plasma apparatus was explained in Ref [8].

2.3 Analysis

The complicated process including many reactions which convert raw material into glass-like amorphous solid is called vitrification. In this paper, the vitrification degree is simply defined as the reaction ratios of SiO_2 in the melted powders to the total crystal SiO_2 in the raw material by using X-ray diffractometry (XRD, Miniflex, Rigaku, Japan). The collected particles after in-flight heat treatments were mixed with ZnO powder as an internal standard, and the diffraction peaks due to quartz and ZnO were compared. The morphology of the particles was performed with optical microscope on VHK-1000 (Keyence, Japan). Chemical analysis for the average compositions of melted particles was carried out by X-ray fluorescence analyzer.

3. Comparison of different heat sources

Figure 3 shows the optical micrograph images of the glass raw material (a) and the particles after in-flight heat treatment by hybrid plasma (b). While the glass raw materials have bumpy shape and no optical transparency, the melted particles have spherical shape and optical transparency, which indicates the raw materials were well-vitrified during the in-flight heat treatment.

The vitrification degrees of the particles melted by the multi-phase AC arc and the hybrid plasma are presented in Figure 4. The vitrification degree of the particles melted by the hybrid plasma is obviously higher than that of the multi-phase AC arc. This is because the residence time in the hybrid plasma heat treatment would be longer than that in the multi-phase arc treatment due to the oxygen flame, which has longer high temperature region than that of the multi-phase arc.

Figure 5 shows the relationship between the powder feed rate and the volatilization degree of B_2O_3, which was calculated from the comparison of the

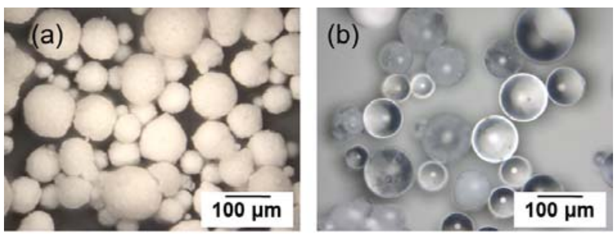

Figure 3. Optical microscope images of (a) raw material and (b) melted particles after hybrid plasma heating.

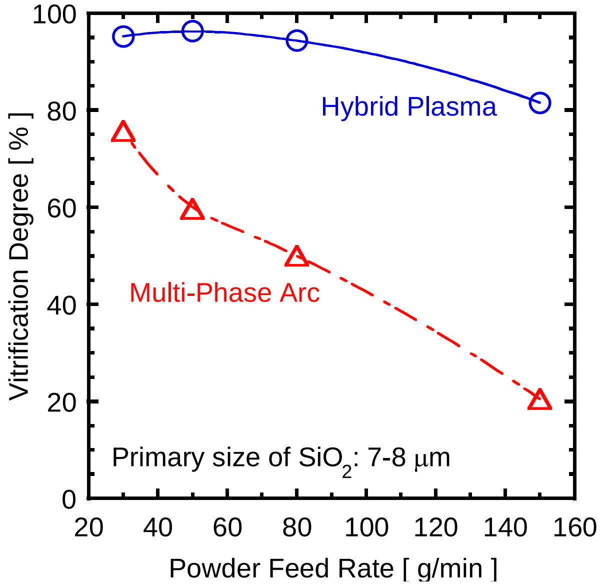

Figure 4. Effect of powder feed rate on vitrification degree of collected particles for larger primary size of SiO_2.

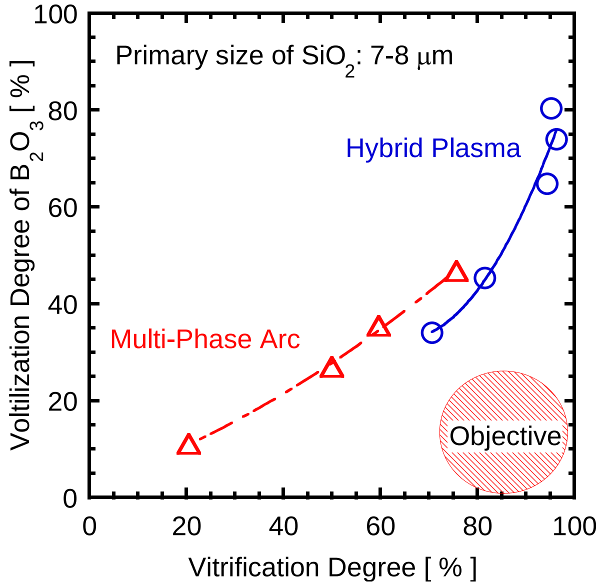

Figure 6. Relationship between vitrification degree and volatilization degree for larger primary size of SiO_2.

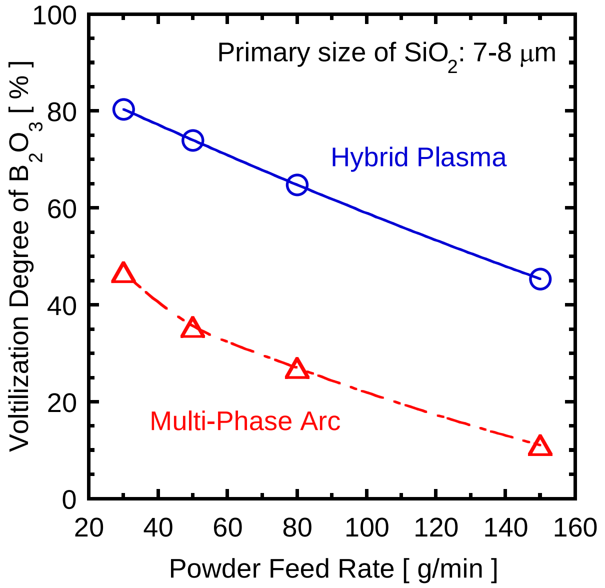

Figure 5. Effect of powder feed rate on volatilization degree of B_2O_3 for larger primary size of SiO_2.

chemical composition between the raw materials and the melted particles. As well as the vitrification degree, the volatilization degree of the particles melted by the hybrid plasma is higher than the particles treated by the multi-phase arc.

Figure 6 shows the relationship between the vitrification degree and the volatilization degree of the particles after heat treatment by different heat sources. Increasing the vitrification degree leads to the higher volatilization degree, resulting from the increase of the energy transfer from the heat sources to the particles. The hybrid plasma heating leads to the lower volatilization degree than the multi-phase arc in the case with the same vitrification degree. This is because the different temperature history and the residence time between the hybrid plasma and the multi-phase arc heating. For the in-flight melting method, it must be important to control the temperature history and the residence time. Compared to the case of the only multi-phase arc system, it is possible to control the balance between the multi-phase arc and the oxygen burner in the case of the hybrid plasma system.

4. Comparison of different raw materials

Figure 7 shows the relationship between the powder feed rate and the vitrification degree of the collected particles by the hybrid plasma for different primary size of SiO_2. The vitrification degree of the particles with smaller primary size of SiO_2 was higher than that with larger SiO_2, resulting from the enhanced reaction rate due to the decrease of the SiO_2 size in the raw material.

The volatilization degrees of B_2O_3 of the melted particles for different raw materials were almost same because the volatilization would be mainly affected by the particle size of the granulated raw material. Figure 8 shows the relationship between the vitrification degree and the volatilization degree of the melted particles by hybrid plasma heating for different sizes of Primary SiO_2. Smaller size of primary SiO_2 leads to higher vitrification degree than larger one in the case with the same

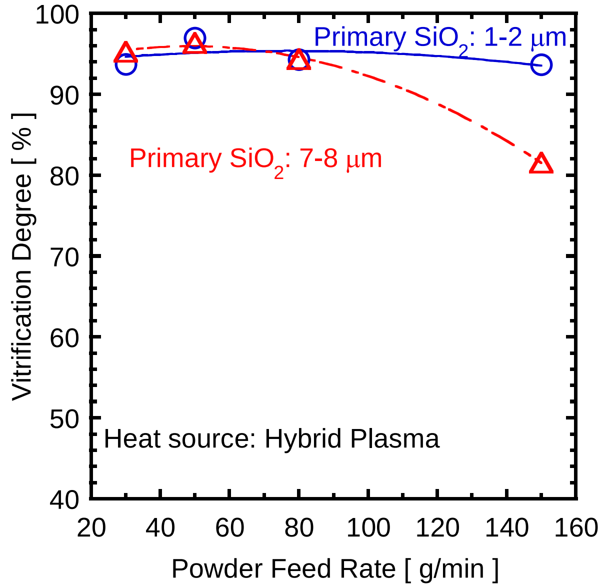

Figure 7. Effect of powder feed rate on vitrification degree of melted particles by hybrid plasma.

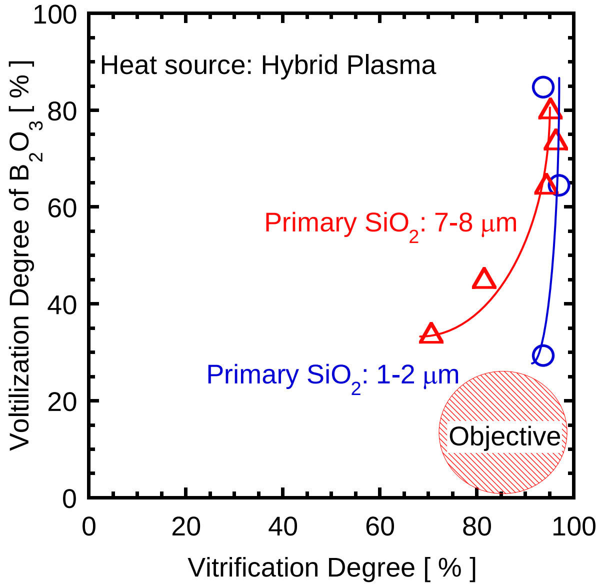

Figure 8. Relationship between vitrification degree and volatilization degree of melted particles by hybrid plasma.

volatilization degree. This is because the primary size of SiO_2 is important on the reaction rate inside the individual particles during the in-flight melting process.

The obtained results indicate that the optimization of the particles size and the primary size of the species (SiO_2) must be important to produce well-vitrified glass with inhibited volatilization of the species during in-flight melting heat-treatment, as well as the optimization of the temperature history and the residence time by the improvement of the hybrid plasma-heating system. These will be our future works.

5. Conclusion

The melting behavior of the alkali-free glass raw material during in-flight melting processes was investigated. The comparison between the different heat sources suggested the hybrid plasma heating leads to higher vitrification degree than the multi-phase arc in the case with the same volatilization degree. Furthermore, the comparison between the different primary sizes of SiO_2 in the granulated raw materials indicated that the primary SiO_2 greatly affect to the vitrification during in-flight melting process. The optimization of the particle size and the primary SiO_2 size would be important to melt the glass in-flight processes, as well as the optimization of the balance between the multi-phase AC arc and the oxygen burner.

References

[1] C. P. Ross, Am. Ceram. Soc. Bull. **83,** 18-20 (2004).

[2] Y. Yao, M. M. Hossain, T. Watanabe, T. Matsuura, F. Funabiki, and T. Yano, Chem. Eng. J. **139,** 390-397 (2008).

[3] Y. Yao, M. M. Hossain, T. Watanabe, T. Tsujimura, F. Funabiki, and T. Yano, Thin Solid Films **516,** 6622-6627 (2008).

[4] Y. Yao, K. Yatsuda, T. Watanabe, F. Funabiki, and T. Yano, Cheml. Eng. J. **144,** 317-323 (2008).

[5] Y. C. Yao, T. Watanabe, T. Yano, T. Iseda, O. Sakamoto, M. Iwamoto, and S. Inoue, Sci. Technol. Adv. Mat. **9,** 8 (2008).

[6] M. M. Hossain, Y. Yao, T. Watanabe, F. Funabiki, and T. Yano, Chem. Eng. J. **150,** 561-568 (2009).

[7] M. M. Hossain, Y. C. Yao, and T. Watanabe, IEEJ Trans. Electr. Electron. Eng. **4,** 504-509 (2009).

[8] Y. Liu, M. Tanaka, Y. Tsuruoka, and T. Watanabe, Thin Solid Films, in press