# Innovative In-Flight Glass Melting Technology Using Thermal Plasmas

Takayuki Watanabe

*Department of Environmental Chemistry and Engineering, Tokyo Institute of Technology, Yokohama, Japan*

**Abstract:** An innovative in-flight melting technology was successfully developed to melt granulated glass raw materials. The granulated raw materials were heated by a 12-phase AC arc, an RF plasma, and an oxygen burner. The high degree of vitrification achieved within milliseconds reveals that the new in-flight melting technology can reduce energy consumption and shorten the glass production cycle.

**Keywords:** Thermal plasma, Multi-phase arc, RF plasma, In-flight melting, glass production

## 1. Introduction

Most glass has been produced by the typical Siemens-type melter fired in air with heavy oil or natural gas as the fuel. This type of melter has been used for more than 140 years because of its good large-scale performance and continuous melting system. The model of the conventional fuel-fired melter is shown in **Fig. 1**. The heat transfer from the burner flame to the raw material is low, therefore it usually takes several days to obtain an homogeneous glass product. A long time is required for the dissolution of $SiO_2$ in the melting process and to allow the bubbles in the glass to escape in the fining process.

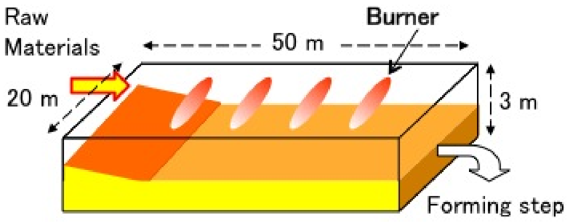

Fig. 1 Conventional glass production.

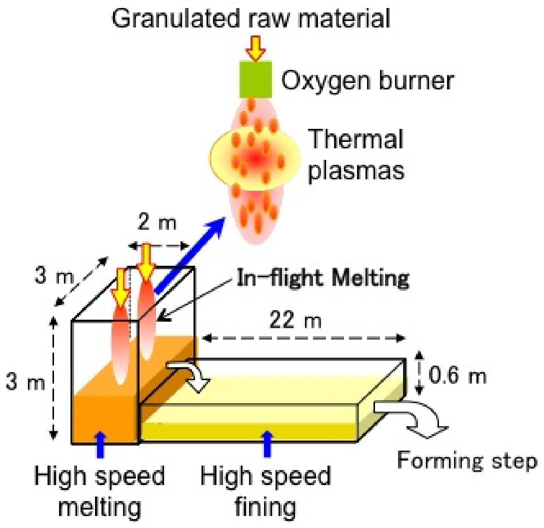

Fig. 2 Innovative in-flight melting for glass production.

Many improvements have been made in energy efficiency and furnace life, such as more efficient burner, better insulation, improved furnace design, and replacement of air using oxygen. However, these improvements have not changed the fundamental technology, therefore the improvement of energy conservation and reduction of emissions are limited.

Under the support of New Energy and Industrial Technology Development Organization (NEDO) project in Japan, an innovative in-flight glass melting technology was developed [1]. The concept of the in-flight melting for glass production is illustrated in **Fig. 2**. The granulated raw material with small diameter is dispersed in thermal plasmas and the powders contact fully with the plasma and burner flame. The high heat-transfer and temperatures of the plasma will melt the raw material quickly. In addition, the decomposed gas of carbonates is removed during the in-flight treatment to reduce the fining time considerably. Compared with the traditional glass production, the total vitrification time is evaluated only 2-3h in the same productivity as the fuel-fired melter.

In the present work, the granulated raw materials of glass were heated by a multi-phase AC arc, an RF plasmas, and an oxygen burner for the application of glass production. The vitrification degree, the morphology, the average diameter, and the composition of the melted powders were characterized. Also, the in-flight melting behavior of the glass powders was studied and the melting results with the three kinds of heating sources were compared for providing a guideline for the glass industry.

## 2. Experimental

The three kinds of heating sources, a multi-phase AC arc, an RF plasma, and an oxygen burner were used to melt the glass raw material to investigate the feasibility of in-flight melting technology for glass production. Two typical glass raw materials, soda-lime glass and alkali-free glass, were used in the experiments. In order to improve the fluidity of the powders in feeding and melting efficiency, the raw material was granulated into spherical

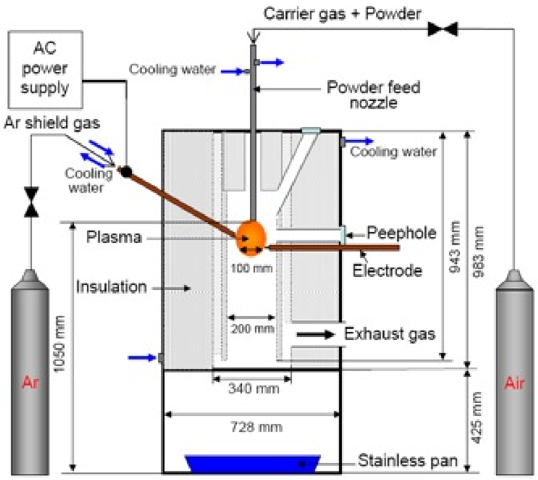

Fig.3 Multi-phase AC arc for in-flight melting glass production.

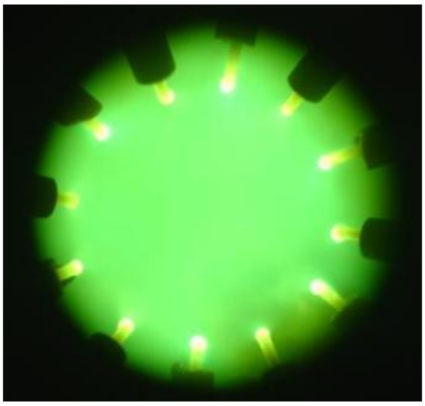

Fig. 4 Multi-phase AC arc with 12 electrodes.

powders by the spray drying method. The average diameters were 51 and 80 µm corresponding to the soda-lime glass and alkali-free glass raw materials, respectively.

**Fig. 3** presents the schematic diagram of the experimental setup which consists of an AC power supply, 12 electrodes, a reaction chamber, a powder feeder, and a gas supply control system. The 12 electrodes configuration was symmetrically distributed at an angle of 30 degree. The 12 electrodes were divided into two layers to increase the plasma volume, upper 6 inclined electrodes and lower 6 horizontal electrodes, and the angle between upper and lower electrodes was 30 degree. The detailed electrical circuit diagram of the transformers for converting from 3-phase AC to 12-phase AC was described by Yao et al. [2] and Matsuura et al. [3]. The electrode material was thoriated tungsten (98% W and 2% $ThO_2$) with 3.2 mm in diameter. Argon (purity 99.99%) was injected around the electrodes to prevent them from oxidation at the flow rate of 60 L/min. The total power was 46 kW; total current 290 A, total voltage 185 V. The discharge voltage and current of each electrode were 35-50 V and 80-100 A, respectively. The diameter of the arc formed by the 12 electrodes was about 100 mm during the stable discharge as shown in **Fig. 4**. The granulated raw material was injected into the arc at the feed rate of 30 g/min with air as the carrier gas by the powder feeder.

The RF thermal plasma was also used as the high-temperature heating source. The typical operating conditions of the RF plasma were as follows: torch power: 10 kW; work frequency: 4 MHz; total pressure: 101.3 kPa; argon plasma gas: 2.0 L/min; sheath gas: 22 L/min (Ar) + 2 L/min ($O_2$); carrier gas: 20 L/min (Ar); feed rate: 30 g/min.

The oxygen burner was used for the comparison with the thermal plasmas as the heating source for the in-flight melting. Propane was used as the fuel. The combustion power of the burner was 38 kW, the flow rate of the propane: 25 L/min, the primary oxygen: 12 L/min, secondary oxygen: 105 L/min. The feed rate and flow rate of carrier gas were same with the 12-phase arc experimental conditions.

The morphology of the particles was performed with scanning electron microscope (SEM) on JSM5310 (JEOL, Japan), and the average diameter was evaluated by the image analysis on SEM photographs. The structures of the powders were determined by X-ray diffractometry (XRD) which was carried out on Miniflex (RIGAKU, Japan) with Cu K$\alpha$ radiation. The contents of $Na_2O$, $B_2O_3$, and total Fe ions in the melted powders were analyzed by inductively coupled plasma (ICP) spectroscopy on PRODIGY (LEEMAN, USA). The in-flight melting behavior of the powders was studied and the results for the different heating sources were compared to provide guidelines for the glass industry.

## 3. Results and discussion

The raw materials of the alkali-free glass powders with different carrier gas flow rates were injected into the multi-phase AC arc at the flow rate of 30 g/min. The cross-section images of the particles were performed with SEM to observe the image of the particle inside as shown in **Fig. 5**. The images show that the particle inside of the raw material has incompact structure with many pores which were caused by the evaporation of water in slurry droplets when the particles were dried. However, the particle inside of completely melted powders has a homogeneous and a compact structure with spherical shape. The bubbles appeared on the particle inside were formed by the decomposed gas from the raw material. Some incompletely melted particles with irregular shape, and the amount of unmelted particles increases along with an increase of the carrier gas flow rate. The average diameter of the melted powders shrinks at least 50% of

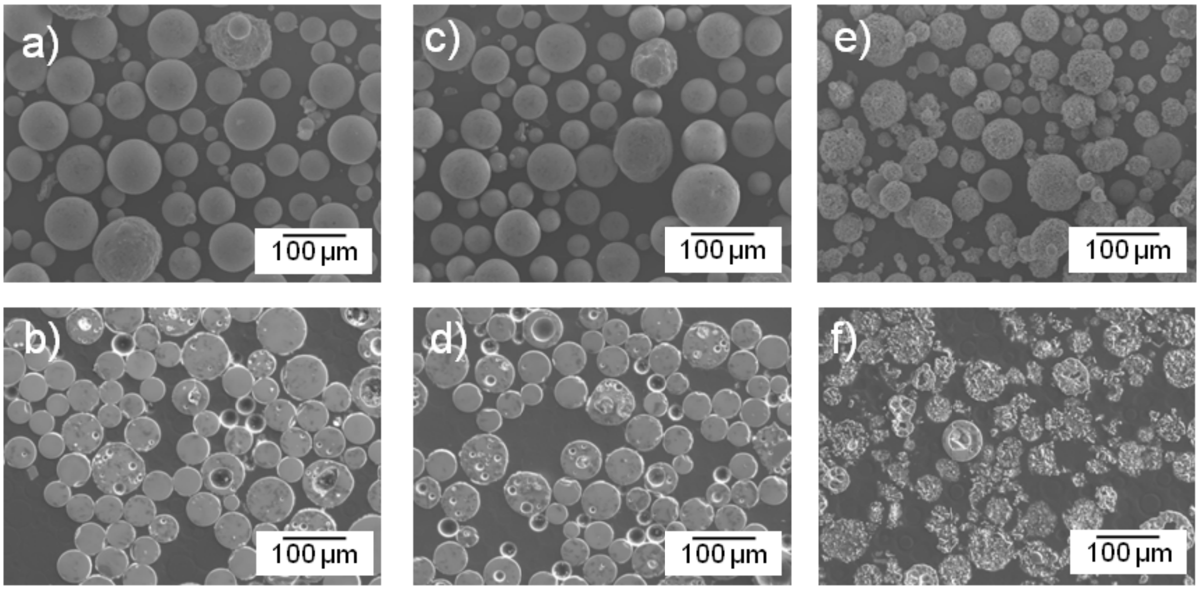

Fig. 5 Alkali-free glass powders after in-flight melting by multi-phase AC arc with different carrier gas flow rates; a,b: 10 L/min, c,d: 20 L/min, e,f:30 L/min.

the original diameter.

The XRD analysis was performed to demonstrate the structures of the powders before and after melting. As seen from **Fig. 6**, the diffractive peaks of $SiO_2$ and $BaCO_3$ clearly represent in XRD pattern of the raw material before being heated. The powders prepared at flow rate of 10 and 20 L/min only appear the $SiO_2$ peaks, but the powders prepared at 30 L/min still contain some $BaCO_3$ peaks. It indicates that $BaCO_3$ and $H_3BO_3$ in the raw material almost decomposed at smaller flow rate of the carrier gas. A decrease of the intensity of $SiO_2$ (101) peak makes it clear that the crystal $SiO_2$ reacted with other compounds becomes diminished at smaller flow rate. Especially for the glass powders at the carrier gas flow rate of 10 L/min, the patterns without any peaks reveals that the powders are the whole amorphous structure with typical glass characteristic which means the reactions among compounds were complete during the in-flight melting.

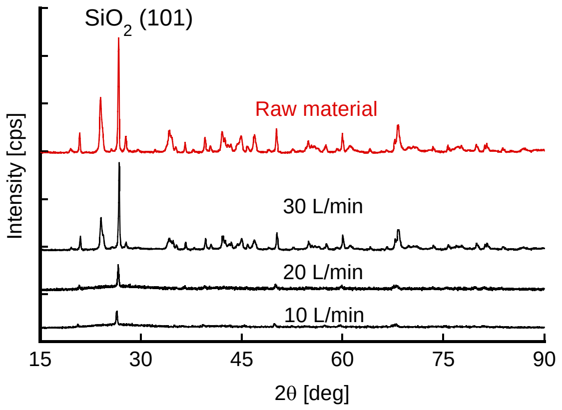

Fig. 6 XRD spectrum of alkali-free glass powders after in-flight melting with different carrier gas flow rates.

The ratio of reacted crystal $SiO_2$ in the quenched powders to that in raw material is used to reflect the vitrification degree of the raw material. It can be measured by the internal standard method with XRD, ZnO as the standard material. The vitrification degree of the quenched powders is shown in **Fig. 7** for the alkali-free and the soda-lime glass powders. The analysis shows that the highest vitrification degree was obtained at the carrier gas flow rate of 10 L/min, but the vitrification decreases at 30 L/min especially for the alkali-free glass powders. There are two main reasons to explain the low

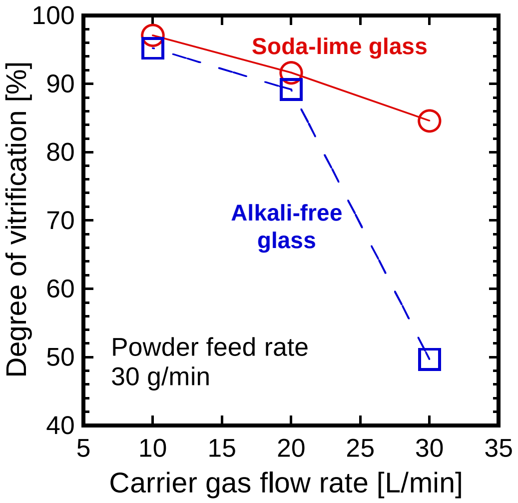

Fig. 7 Effect of carrier gas flow rate on the degree of vitrification of the melted powders.

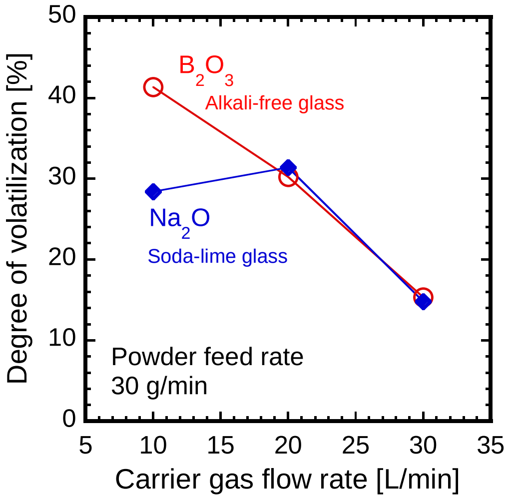

Fig. 8 Effect of carrier gas flow rate on $B_2O_3$ and $Na_2O$ volatilization of the melted powders.

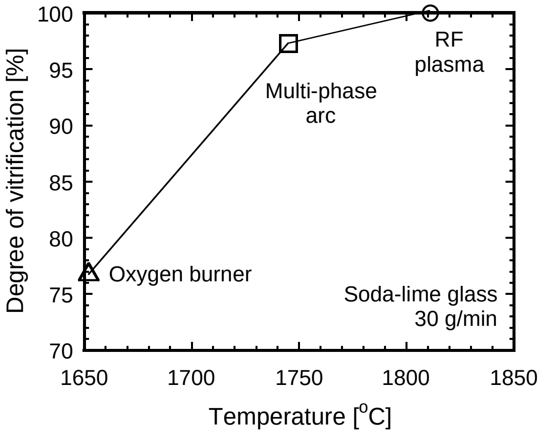

Fig. 9 Relationship between the powder temperature and the degree of vitrification for RF plasma, multi-phase AC arc, and oxygen burner.

degree of vitrification under larger flow rate of the carrier gas. Larger flow rate leads to lower plasma temperature as well as to shorter residence of the powders, then the less energy transferred to the powders during the flight in the plasma. The degree of vitrification of the alkali-free glass is lower than that of the soda-lime glass due to higher viscosity of the alkali-free glass, resulting in slower vitrification. Since the reaction and melting first occur on the surface of the particle, then propagate into the inside. The higher viscosity of molten glass on surface will delay the melting and the removal of decomposed gas inside the particle.

The glass properties like the viscosity and softening point can be changed by controlling the $B_2O_3$ and $Na_2O$ content for the alkali-free glass and the soda-lime glass, respectively. Since $B_2O_3$ and $Na_2O$ are volatile materials, the exposure of these materials to high temperature surroundings will increases the volatilization loss and the cost of glass production. Hence, the evaluation of volatilization degrees of $B_2O_3$ and $Na_2O$ is important to the in-flight melting. The volatilization degrees of $B_2O_3$ and $Na_2O$ in the quenched powders are presented in **Fig. 8** from the estimation of these contents in the powders analyzed by ICP. It is notable that the high vitrification always accompanies with the high volatilization of $B_2O_3$ and $Na_2O$. High temperature of the thermal plasma leads to the high vitrification, and short residence time within the high temperature region can suppress the volatilization of $B_2O_3$ and $Na_2O$. A good balance between the vitrification and volatilization can be obtained by controlling the appropriate conditions.

The heating temperature has a strong influence on the heat transfer to the powders. **Figure 9** shows the temperatures of the powders obtained during the in-flight melting from the measurement of the amounts of total Fe ions and $Fe^{3+}$ [1,4]. The temperatures of the powders heated by the different heating sources are in the order of the RF plasmas, the 12-phase AC arc, and the oxygen burner. Combining the advantages of the high temperature of the plasma and the high thermal-efficiency of the oxygen burner improves the glass quality and increases productivity with minimum energy consumption. The lowest temperature of the burner flame (about 2500°C) leads to the least heat transfer to powders which is responsible for the incompletely melted powders. Compared with the soda-lime glass powders, the alkali-free glass powders are more difficult to melt under the same operating conditions.

## 4. Conclusion

Innovative in-flight melting technology was successfully developed to melt granulated glass raw materials. The high vitrification degree achieved within milliseconds reveals that the new in-flight melting technology can reduce energy consumption and shorten the production cycle.

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## References

[1] Y. Yao, et al., Sci. Technol. Adv. Mater., 9, 025013 (2008).

[2] Y. Yao, et al., Chem. Eng. J., 139, 390 (2008).

[3] T. Matsuura, et al., Thin Solid Films, 515, 4240 (2008).

[4] V.C. Kress, I.S.E. Carmichael, Am. Mineral. 73, 1267 (1988).