In-flight melting of granulated powders in multi-phase arc for glass production

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Abstract: The in-flight melting technology with alternative current (AC) multi-phase arc was developed to save energy and shorten production cycle for glass industry. As the power increases, the amount of crystal SiO_2 and the diameter of quenched powders decrease. The vitrification of alkali-free glass powders is lower than that of soda-lime glass powders. Higher plasma temperature and longer residence time of 12-phase arc improve the vitrification.

Keywords: thermal plasmas, multi-phase arc, in-flight melting, glass production

1. Introduction

The use of thermal plasmas in materials processing is becoming an increasingly active and attractive field for the development of new technology. The potential applications of thermal plasma processing in industries cover a wide range of activities, such as spray coating. the extraction of metals, the remelting and refining of metals or alloys, the synthesis of advanced materials as well as the treatment of toxic and hazardous waste [1-5]. Arc plasma as the energy source with high energy efficiency has been used in the welding and cutting of metals, steelmaking, synthesis of nanoparticles and so on [6]. Three-phase circuits for generating arc plasma can supply high power with no time variation and less ohmic loss of distributing lines, compared with direct current (DC) or two-phase circuits. The valuable feature of an electric power system with more than three phases is that the instantaneous power is completely constant when the power source and its load are balanced [7, 8].

The typical melting system for glass is refractory-lined melting furnace, fired by air and natural gas or oil as fuel, which has been used over 100 years in glass industry [9]. The glass industry is one of the most energy intensive industries, only second to the aluminum industry. Especially, the melting and fining process takes most energy and time of whole technology. In addition, lots of emissions are produced during melting due to the usage of fossil fuel, which brings more environmental pollution.

In this study, an innovative in-flight glass melting technology with multi-phase alternating current (AC) arc was developed to melt granulated raw material for the purpose of energy conservation and environmental protection. Also, the melting behavior of particles in arc was evaluated in the paper.

2. Experimental

The raw materials for soda-lime glass and alkali-free glass were prepared into granulated powders using the spray-drying method, and the mean diameter was 51 μm and 80 μm, respectively. The target composition of soda-lime glass was $16Na_2O$-$10CaO$-$74SiO_2$ (wt%) made by Na_2CO_3, $CaCO_3$ and SiO_2, the composition of alkali-free was $49SiO_2$-$15B_2O_3$-$10Al_2O_3$-$25BaO$-$1Sb_2O_3$ (wt%) prepared by SiO_2, H_3BO_3, Al_2O_3, $BaCO_3$ and Sb_2O_3.

A new type of arc plasma reactor with 12-phase AC discharge has been developed to get stable and continuous arc by the transformers for converting from the 3-phase AC to the 12-phase AC. The input of the three-phase power supply is connected to 200V (60Hz) commercial power lines. The primary coils of transformers are divided into two parts: one is the Δ connection and the other is the Y connection. The turn's ratio of the windings between the primary and secondary coils of the transformer was designed $1/\sqrt{3}$. The 12-phase power supply can be realized by the combination of these circuits, then, each line is connected directly to the corresponding electrode of the reactor, as shown in Ref. [10].

The schematic diagram of the experimental setup is shown in Fig.1. It consists of 12 electrodes, reaction chamber, powder feeder and AC power supply. The configuration of 12 electrodes is symmetrically arranged

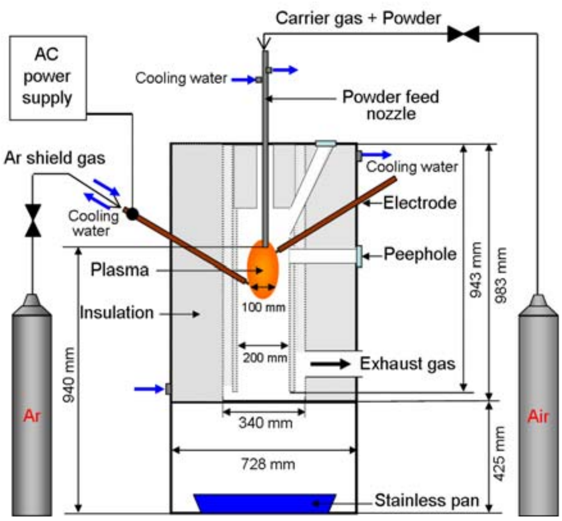

Fig.1 Schematic diagram of the experimental setup

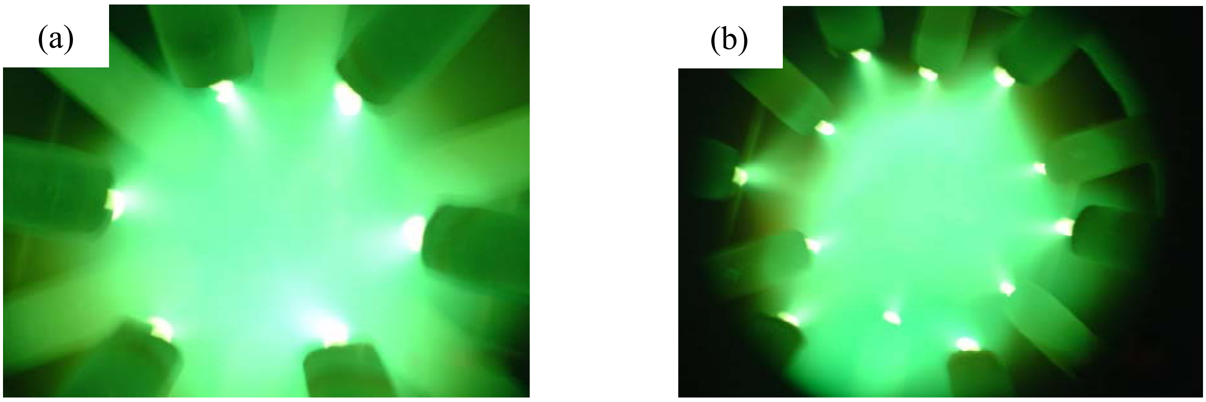

Fig.2 Photographs of multi-phase arc plasma: (a) 6-phase arc; (b) 12-phase arc

by the angle of 30 deg. The 12 electrodes are divided into two layers, upper 6 electrodes and lower 6 electrodes. The electrode material is tungsten (purity 99.9%) with 3 mm in diameter. Water was used to cool the host of electrodes and the nozzle of powder feeder, and argon gas was injected around the electrodes to prevent them from oxidation. The set-up can be operated with 6 electrodes discharge or 12 electrodes discharge. Fig.2 shows the generated arc plasma with 6 electrodes discharge and 12 electrodes discharge. The discharging voltage and its current were 30-45 V and 80-100 A, respectively. The diameter of arc plasma was about 100 mm, and the distance between two layers of electrodes was 15 mm. The initial powders were injected into arc plasma with air carrier gas by the powder feeder. The melted powders were quenched on the stainless steel pan at a distance of 920 mm. The flow rate of carrier gas was 20 l/min, the argon shield gas was 36 l/min.

The structures of the powders were determined by X-ray diffractometry (XRD). XRD was carried out on Miniflex (Rigaku) with CuK_α radiation at 30 kV and 15 mA. The data were collected in the 2θ range 3-90^o with a step size of 0.02^o and a scan speed of 4^o/min. The micrograph and size distribution of particles were performed by scanning electron microscope (SEM) on JSM5310 (JEOL). The composition of quenched powders was analyzed by electron probe microanalysis (EPMA) on JXA-8200 (JEOL).

3. Results

3.1 Six-phase arc

Fig.3 shows the XRD patterns of raw materials and quenched powders with 6-phase arc at a feed rate of 30 g/min. The XRD patterns illustrate that there are no obvious diffractive peaks of carbonates or other compounds besides SiO_2 in the samples of quenched powder, indicating the decompositions of carbonates or other compounds in raw material were mostly complete in the process of in-flight melting. As the input power increases, the peak intensity of SiO_2 in both glass powders decreases. The results indicate that the amount of crystal SiO_2 becomes smaller and the ratio of amorphous structure with the glass characteristics is increased. Vitrification is a process of converting material into a glass-like amorphous solid which is free of any crystalline structure. The vitrification degree is defined as the ratio of reacted SiO_2 in quenched powders to the total SiO_2 in raw material.

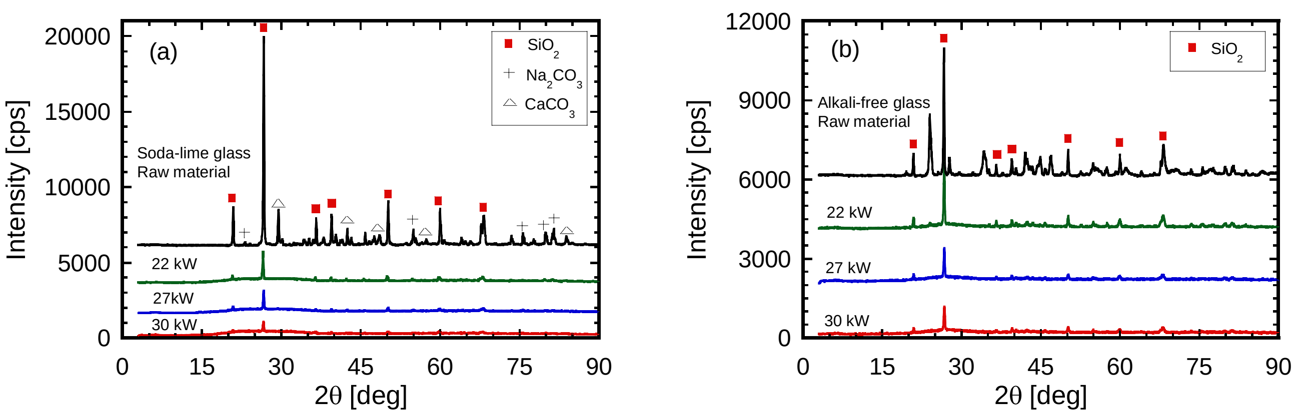

Fig.3 XRD patterns of raw materials and quenched powders heated by 6-phase arc: (a) soda-lime glass powders; (b) alkali-free glass powders

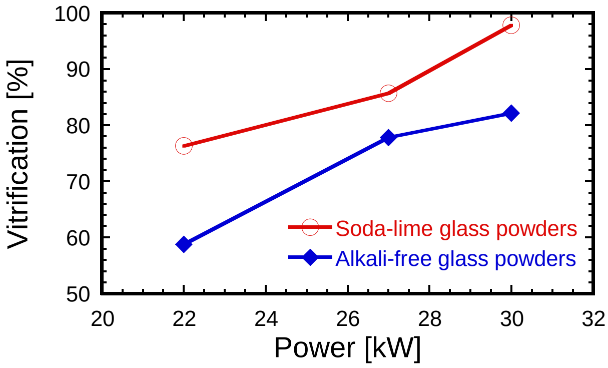

Fig.4 Vitrification of samples heated by 6-phase arc

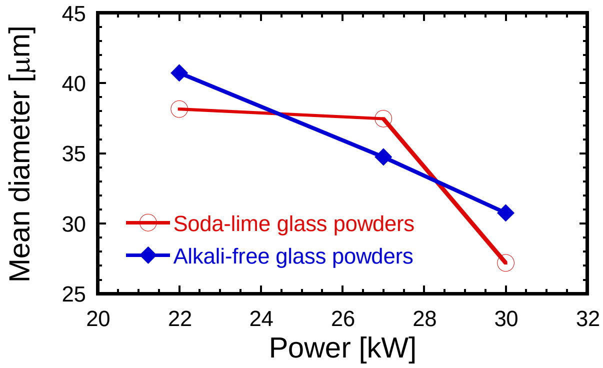

Fig.6 Variation of mean diameters of glass powders

The effect of input power on the vitrification ratio was analyzed by the XRD analysis [11], as shown in Fig.4. As the input power of arc increases, the vitrification ratio of soda-lime glass powders and alkali-free glass powders increases due to higher plasma temperature caused by more energy input. Under the same input power of 30 kW, the vitrification of soda-lime glass powders and alkali-free glass powders is 97.8% and 82.1%, respectively. Comparing the relationship between glass temperature and viscosity, the viscosity of alkali-free glass is higher than that of soda-lime glass under same temperature. High viscosity makes the decomposed CO_2 from carbonates difficult to move to surface quickly in melting process, that decreases the vitrification.

The morphology of raw materials and quenched powders heated by 6-phase arc with 30 kW is shown in Fig.5. It is found that the granulated particles of raw materials had rough surface and porous structure from Fig.5 (a) and (b). The porosity of raw materials of soda-lime glass and alkali-free glass determined by the pycnometer method was 73% and 72%, respectively. The SEM photographs show the particles shrank in diameter and the surface became smooth after heating. Among these particles, all particles of soda-lime glass were smooth and spherical, but some alkali-free glass particles marked by "*A*" were still porous and irregular, indicating these particles melt incompletely during in-flight time in 6-phase arc. The results are consistent with the above vitrification analysis.

The mean diameter of glass powders is shown in Fig.6. With the input power increasing, the mean diameter of quenched powders decreases. Compared with their raw material, the glass particles shrank more than 40% in diameter after heat treatment.

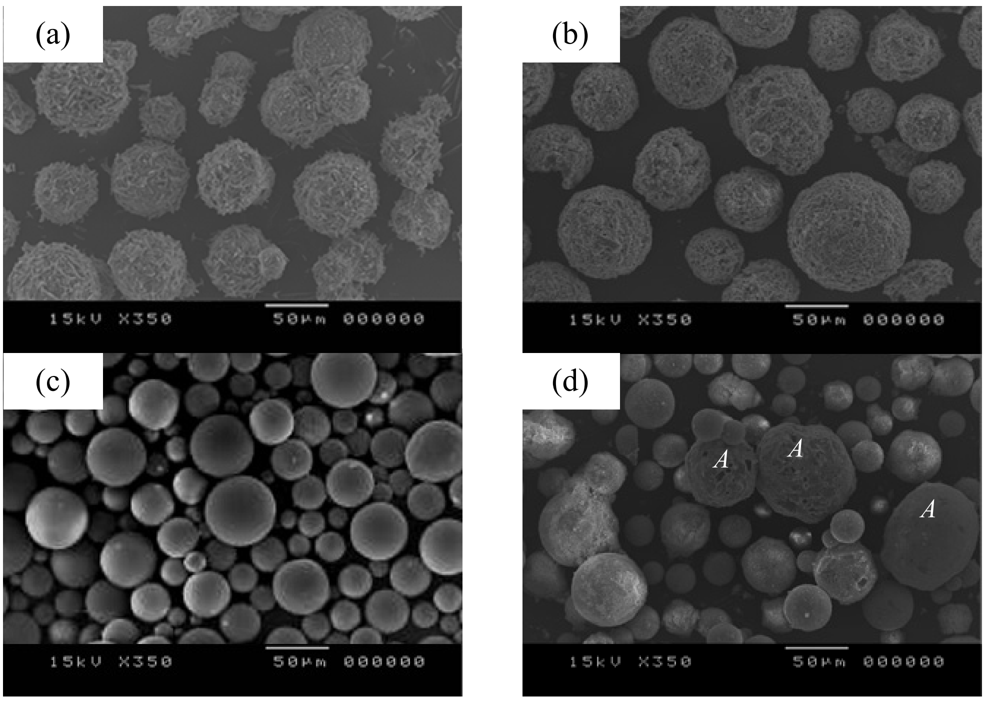

Fig.5 SEM images of raw materials and quenched powders with 30 kW power: (a) soda-lime glass raw material; (b) alkali-free glass raw material; (c) soda-lime glass powders; (d) alkali-free glass powders

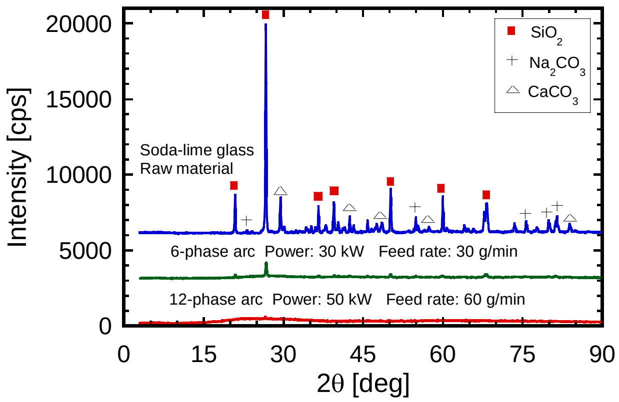

Fig.7 XRD patterns of raw material and quenched powders heated by 12-phase arc

3.2 Twelve-phase arc

The XRD patterns of raw material and quenched powders with 12-phase arc at a feed rate of 60 g/min are shown in Fig.7. The curve of quenched powders shows that there are not any peaks in the pattern, revealing the reactions among compounds were complete during heating. Also, the vitrification analysis suggests the melting ratio attained 100% with these conditions. Compared with 6-phase arc, 12-electrodes discharging obtained the stable arc with higher power and larger volume. Large volume of arc plasma prolongs the residence time of particle flying, and more power inputted to electrodes increases the plasma temperature. Hence, the vitrification of particles treated by 12-phase arc is higher.

The particle size distributions of raw material and quenched powders heated by 12-phase arc are compared, the results are shown in Fig.8. The peak of size distribution of quenched powders is narrower than that of raw material. The average diameter of quenched powders is 25.9 μm, shrank about 50% after heating. It can be concluded that the diameters of quenched powders are related with their respective vitrification ratio. Higher vitrification leads to more shrinkage and smaller size. According to above analysis, the reactions first occur at

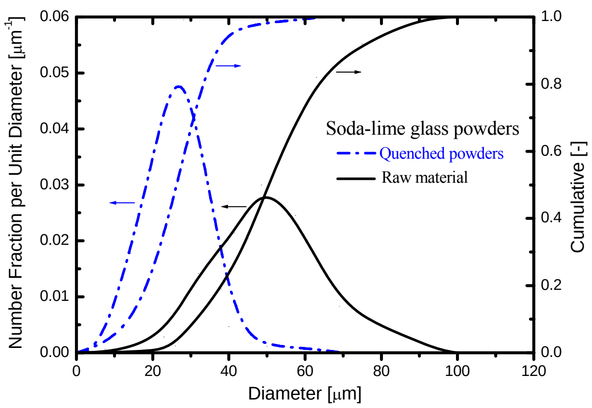

Fig.8 Particle size distributions of raw material and quenched powders heated by 12-phase arc

the outer skin of the particle, then, the zone of reaction move into the inter solid. The reaction model of particle melting in the process of in-flight melting will be investigated next step. The composition analysis shows that the content of Na_2O of quenched powders prepared by 6-phase arc and 12-phase arc is 14.8% and 13.6% corresponding to the input power 30 kW and 36 kW, respectively. More volatilization of Na_2O also reveals higher temperature of particles arrived in 12-phase arc, which is in agreement with above results.

4. Conclusion

The multi-phase alternating current (AC) arc was developed to melt the granulated glass raw material for the purpose of energy conservation and environment protection. Results show that the vitrification increases and the particle diameter decrease with an increase in input power. Compared with the 6-phase arc, the 12-phase arc can obtain higher plasma temperature with larger volume and longer residence time during in-flight heating. The melting of alkali-free glass raw material is more difficult than that of soda-lime glass raw material due to higher viscosity. The particle shrinkage in diameter is related with its vitrification, and higher vitrification leads to more shrinkage of particle.

Acknowledgements

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References:

[1] M. I. Boulos, Pure Appl. Chem., **68**, 1007 (1996).
[2] P. R. Taylor and S. A. Pirzada, Adv. Perform. Mater., **1**, 35 (1994).
[3] J. Heberlein, Pure Appl. Chem., **74**, 327 (2002).
[4] J. Chang, Sci. and Technol. Adv. Mater., **2**, 571 (2001).
[5] T. Watanabe, M. Shigeta and N. Atsuchi, Int. J. Heat Mass Transfer, **49**, 4867 (2006).
[6] B. Ravary, L. Fulcheri, J. A. Bakken, G. Flamant and F. Fabry, Plasma Chem. Plasma Process., **56**, 2123 (2001)
[7] K. Matsumoto, Plasma Sources Sci. Technol., **5**, 245 (1996).
[8] R. J. Cogdell, Foundations of Electrical Engineering, Englewood Cliffs, NJ: Prentice-Hall (1990).
[9] R. Gonterman and M. Weinstein, Am. Ceram. Soc. Bull., **83**, 21 (2004).
[10] T. Matsuura, K. Taniguchi and T. Watanabe, Thin Solid Films, **515**, 4240 (2006)
[11] D. E. Willis, Chromatographia, **5**, 42 (1972).