

Determination of the Melting Mechanism of Granular Powders for Vitrification by Argon-Oxygen Induction Thermal Plasmas

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Abstract: - In this paper the melting mechanism of granular raw powders by argon-oxygen induction thermal plasma (ITP) for vitrification has been described from both numerical and experimental point of views. A 2-dimensional LTE (local thermodynamic equilibrium) model for argon-oxygen induction thermal plasma has been developed to simulate the flow and temperature fields within the plasma torch for various flow conditions of carrier gas taking into account the effects of species diffusion. The model included a metal tube inserted into the torch for powder and carrier gas injection. Simulated results revealed that the higher the flow-rate of carrier gas, the lower the temperature profiles around the centerline of the torch, and the higher the axial plasma velocity. The average particle size of quenched powders became smaller and the softening temperature decreased with higher flow-rate of carrier gas, because of shorter relaxation time and lower temperature respectively. The numerical results well explained the experimental findings and provided a roadmap for the parameter optimization in the melting process.

Key-Words: - Induction thermal plasma, Granular powder, Carrier gas, Relaxation time, Heat transfer.

1 Introduction

Thermal plasmas, source of very high enthalpy, high chemical reactivity offer rapid evaporation rate, steep temperature gradients, wide area and high growth rate, and an attractive and chemically nonspecific route for the synthesis of raw powders. Thus, thermal plasma technology has been found extensive industrial applications for material and environmental processes [1-3]. Radio frequency (RF) induction thermal plasma (ITP) reactors offer several advantages such as high purity due to absence of electrode, large volume of plasma for processing, low plasma velocity and wide pressure range. The high temperature of ITP leads to short evaporation time which translates into relatively small torch with high throughput. A good review of the advancements in the applications of plasmas at different stages of glass production has been narrated by Bessmertnyi [4]. Nowadays, it is a burning issue to reduce the energy consumption and time in melting the granular raw powders for glass production in industrial level. The traditional technologies used for powder melting in glass industries are inefficient, time consuming and not

environment friendly. As the usage of glass has been increasing to meet the demand of modern architecture of buildings and automobile industries, thus, it is crucial how to increase the glass production keeping the energy consumption minimum and environment benign atmosphere. In this circumstance, ITP technology offers the distinct advantage of providing essentially one-step processes and short time avoiding the multitude of steps and long time required in melting the raw powders in glass industries.

In this paper, the feasibility of melting raw granular powders for vitrification in argon-oxygen thermal plasma system has been demonstrated both numerically and experimentally. We shall discuss about the influence of carrier gas flow-rate on the temperature and flow fields, particle temperature history, and composition of the quenched powders collected at the reaction chamber of the ITP reactor.

2 Modeling

The schematic torch geometry is presented in Fig.1. Table 1 summarizes the torch dimensions and

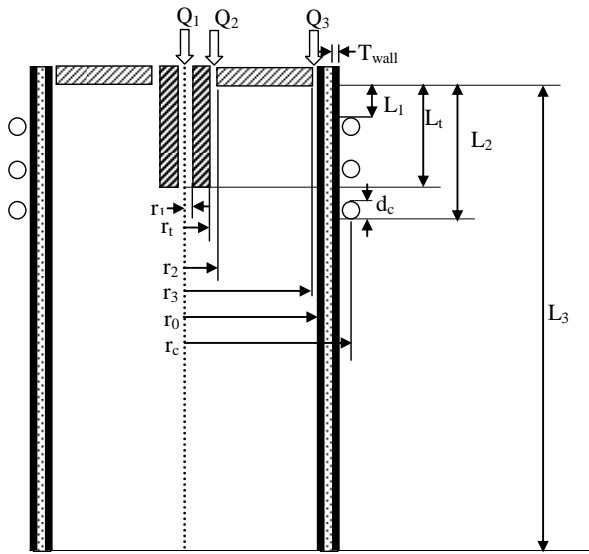


Fig.1 Schematic geometry of ITP torch

operating conditions. The experimental discharge was sustained at 20 kW plug power; thus, assuming 50% overall efficiency of the reactor, the plasma power was set to 10 kW. In this model, the conservation equations along with the vector potential form of Maxwell's equations [5] have been solved simultaneously with some reasonable assumptions and boundary conditions including a metal tube inserted into the plasma torch for carrier gas and powder injection.

2.1 Conservation Equations

The following 2-dimension conservation equations have been solved simultaneously along with the vector potential form of Maxwell's electromagnetic equations.

Continuity Equation:

$$\nabla \cdot (\rho \mathbf{u}) = 0 \quad (1)$$

Momentum Equation:

Axial momentum:

$$\rho \mathbf{u} \cdot \nabla u = -\frac{\partial p}{\partial z} + \nabla \cdot \mu \nabla u + \frac{1}{r} \frac{\partial}{\partial r} \left(r \mu \frac{\partial v}{\partial z} \right) + \frac{\partial}{\partial z} \left(\mu \frac{\partial u}{\partial z} \right) + F_z \quad (2)$$

Radial momentum:

$$\rho \mathbf{u} \cdot \nabla v = -\frac{\partial p}{\partial r} + \nabla \cdot \mu \nabla v + \frac{1}{r} \frac{\partial}{\partial r} \left(\mu r \frac{\partial v}{\partial r} \right) + \frac{\partial}{\partial z} \left(\mu \frac{\partial u}{\partial r} \right) - \frac{2\mu v}{r^2} + \rho \frac{w^2}{r} + F_r \quad (3)$$

Theta momentum:

$$\rho \mathbf{u} \cdot \nabla w = \nabla \cdot \mu \nabla w - \frac{\rho v w}{r} - \frac{1}{r} \frac{\partial (\mu w)}{\partial r} + \mu \frac{\partial}{\partial r} \left(\frac{w}{r} \right) \quad (4)$$

Energy Equation:

$$\rho \mathbf{u} \cdot \nabla h = \nabla \cdot \left(\frac{\kappa}{C_p} \nabla h \right) + \dot{P} - \dot{R} \quad (5)$$

Species Mass Diffusion Equation:

$$\rho \mathbf{u} \cdot \nabla Y_i = \nabla \cdot (\rho D_i^m \nabla Y_i) \quad (6)$$

Maxwell's Equation:

$$\frac{\partial^2 \mathbf{A}_\theta}{\partial z^2} + \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \mathbf{A}_\theta}{\partial r} \right) - \frac{\mathbf{A}_\theta}{r^2} = j \mu_0 \sigma \omega \mathbf{A}_\theta \quad (7)$$

Source Terms:

$$F_r = \frac{1}{2} \mu_0 \sigma \text{Real} [\mathbf{E}_\theta \mathbf{H}_z^*]$$

$$F_z = \frac{1}{2} \mu_0 \sigma \text{Real} [\mathbf{E}_\theta \mathbf{H}_r^*]$$

$$\dot{P} = \frac{1}{2} \sigma [\mathbf{E}_\theta \mathbf{E}_\theta^*]$$

$$\dot{R} = \text{Volumetric radiation loss}$$

Where, ∇ : gradient operator in cylindrical coordinates, ρ : mass density, σ : electrical conductivity, κ : thermal conductivity, C_p : specific heat at constant pressure, \mathbf{u} : velocity vector, D_i^m : effective diffusion coefficient of species i , Y_i : mass fraction of species i , \mathbf{A}_θ : vector potential, h : enthalpy, \mathbf{E}_θ : electric field, \mathbf{H} : magnetic field, μ : viscosity, ω : angular frequency ($2\pi f$), μ_0 : permeability of free space ($4\pi \times 10^{-7}$ H/m), j : complex vector ($\sqrt{-1}$).

Table 1: Torch dimensions & operating conditions

Torch length (L_3)	190 mm
Distance to initial coil position (L_1)	19 mm
Distance to end of coil position (L_2)	65 mm
Length of injection tube (L_t)	52 mm
Coil diameter (d_c)	5 mm
Wall thickness of quartz tube (T_{wall})	1.5 mm
Torch radius (r_0)	22.5 mm
Coil radius (r_c)	32 mm
Inner radius of injection tube (r_1)	1 mm
Outer radius of injection tube (r_t)	4.5 mm
Outer radius of inner slot (r_2)	6.5 mm
Inner radius of outer slot (r_3)	21.5 mm
Plug power	20 kW
Plasma power	10 kW
Working frequency	4 MHz
Working pressure	101.3 kPa
Flow rate of carrier gas (Q_1)	3 ~ 9 l/min of Argon
Flow rate of plasma gas (Q_2)	2 l/min of Argon
Flow rate of sheath gas (Q_3)	22 l/min Argon and 2 l/min Oxygen

2.2 Assumptions

The present computation was concerned only within the plasma torch. The discharge was supposed to be sustained in a cylindrical water-cooled quartz tube. Argon gas was injected as carrier gas through a nozzle tube and as well as plasma gas. The sheath gas composed of argon and oxygen and was injected with a swirl to stabilize the plasma and protect the quartz tube from overheating. It was assumed that plasma flow was steady, 2-dimensional, axi-symmetric, laminar and incompressible. The plasma was in the state of LTE and optically thin. In this model it was assumed that plasma consisted of electron and five heavy species O, O⁺, O₂, Ar and Ar⁺. Furthermore, viscous dissipation and displacement currents were negligible and each coil turn was assumed to be planar.

2.3 Boundary Conditions

The boundary conditions for the mass, momentum, energy and species mass conservation equations were: at the inlet, gas temperature was set to 300 K and uniform velocity profiles were assumed based on the given flow rates; on the axis of symmetry, the symmetry conditions were imposed; on the walls, the no-slip condition was assumed; the outer wall temperature was set to 350 K; and, at the exit, axial gradients were set equal to zero. The inserted tube

was assumed water cooled at 300 K. On the tube wall, the velocity was set to zero.

2.4 Computational Procedure and Thermophysical Properties

The conservation equations, which are listed in previous section, were solved numerically using the SIMPLER algorithm of Patankar [6]. The algorithm was based on a control-volume finite-difference scheme for solving the transport equations of incompressible fluids. Calculations were performed for a 44 (in radial direction) by 93 (in axial direction) non-uniform grid system.

Thermodynamic and transport properties of argon and oxygen gases required for the simulation included mass density, specific heat at constant pressure, viscosity, electrical and thermal conductivity and radiative loss coefficient. The transport properties, which are function of temperature, were calculated under LTE conditions using Chapman-Enskog first approximation to Boltzmann equation [7]. The effective diffusion coefficient of species was calculated based on the following equations:

$$D_i^m = \frac{(1 - Y_i)}{\sum_{j \neq i, j=1}^v \frac{x_j}{D_{ij}}} \quad (8)$$

$$D_{ij} = 2.628 \times 10^{-2} \sqrt{\frac{(M_i + M_j)}{2M_i M_j}} \frac{T^{1.5}}{\bar{\Omega}_{ij}^{(1.1)}} \quad (9)$$

Where x_i and Y_i are mole fraction and mass fraction respectively, D_{ij} is the binary diffusion coefficient between species i and j , M_i and M_j are the molecular weights of species i and j respectively, and $\bar{\Omega}_{ij}^{(1.1)}$ is the collision integral between species i and j . The ambipolar diffusion coefficient for ions can be approximated as $D_a = D_{ion} (1 + T_e/T_{ion})$. As the thermal equilibrium condition i.e $T_h = T_e = T_{ion}$ was applied thus, $D_a \cong 2D_{ion}$.

2.5 Simulated Results and Discussion

Simulation has been performed for the torch shown in Fig.1 and the operating conditions described in Table 1 to investigate the carrier gas flow-rate effects on temperature and velocity fields. Fig.2 depicts the predicted temperature contour within the torch. It is observed that at increased carrier gas flow-rate, the temperature sharply decreases along the centerline and also the high temperature zones (below 8000 K) shrink axially. This is attributed from the large heat transfer to the cold carrier gas injected through the inserted nozzle tube. The high temperature (6000 K to 11000 K) is sufficient enough to evaporate the injected granular powders in the torch within very short time. Fig.3 shows the plasma temperature along the centerline ($r=0$) for different carrier gas flow-rate. It is found that plasma

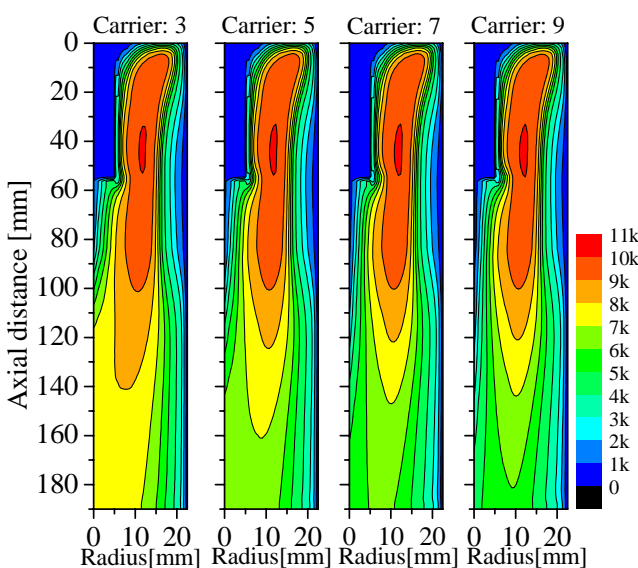


Fig.2 Effects of carrier gas flow-rate on the temperature contours

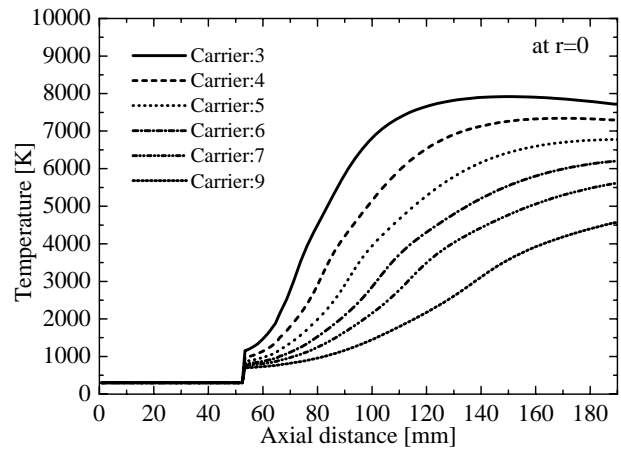


Fig.3 Impact of carrier gas flow-rate on the centerline ($r=0$) temperature

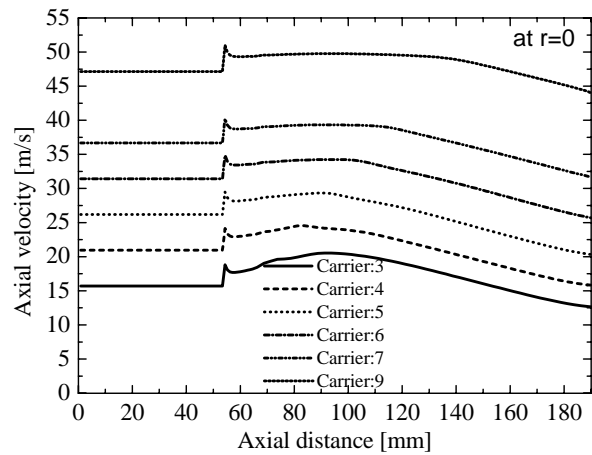


Fig.4 Impact of carrier gas flow-rate on the axial plasma velocity at the centerline ($r=0$)

temperature increases with the distance from the nozzle exit, attains the maximum value at the downstream of the torch. It is evident from Fig.3 that carrier gas flow-rate strongly affects the temperature around the centerline of the torch. Fig.4 reveals that the axial plasma velocity along the centerline ($r=0$) increases with the increase of carrier gas flow-rate because of large momentum of excess carrier gas. Maximum velocity is attained below the coil region due to the effect of Lorentz force. Due to the increased momentum, the large carrier gas flow-rate in turn, enhances the flow of melted powders downward to the substrate.

3 Particle Temperature

Using the temperature profiles presented in the previous section, the particle temperature with distance from the nozzle tube was predicted by solving the equation of energy balance considering

heat transfer to particles due to plasma flow. For a spherical particle with symmetric boundary conditions, the heat transfer into the particle is described by the conduction equation, i.e.,

$$\rho C_{pw} \frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \kappa \frac{\partial T}{\partial r} \right) \quad (10)$$

According to Lee *et al* [8], the Nusselt number correlation for heat transfer between plasma and particles can be expressed as:

$$N_u = \left(2 + 0.6 \text{Re}^{1/2} \text{Pr}^{1/3} \right) \times \left(\frac{\rho_\infty \mu_\infty}{\rho_w \mu_w} \right)^{0.6} \left(\frac{C_{p\infty}}{C_{pw}} \right)^{0.38} \quad (11)$$

Where, r : radial distance from the center of the particle, k : Boltzmann's constant, Re : Reynolds number, Pr : Prandtl number, and ρ , C_{pw} and κ are the density, specific heat, and thermal conductivity of the particle respectively.

Using the boundary conditions described in Ref. [8], the particle temperature was calculated with the distance from the nozzle exit. The properties of feed powders are enlisted in Table.2. The diameter of feed powder was set to 58 μm . As the powder is melted and evaporated, particle's axial velocity reaches very close to plasma velocity. The heat transfer to the particles mainly depends on the plasma temperature, and the higher the plasma temperature the larger the heat transfer to the particles. The particle temperature with distance from the nozzle exit is shown in Fig.5 for different flow-rate of carrier gas. It is evident from the figure

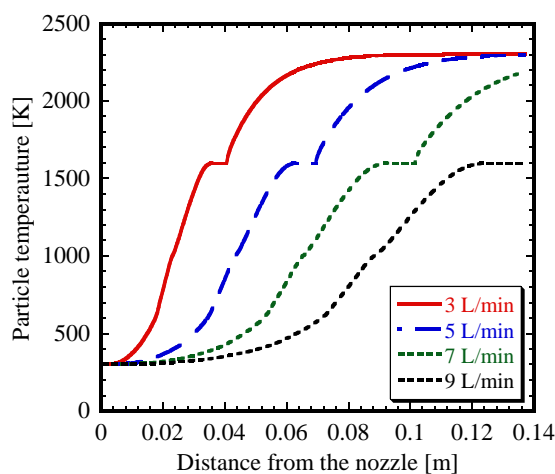


Fig.5 Particle temperature profiles with the distance from the nozzle exit at $r=0$

Table 2: Properties of feed powders

Properties	
Vitrification temperature	1672 K
Vaporization temperature	2500 K
Void fraction	80%
Effective thermal conductivity	0.1 W/m-K
Mass density	2300 Kg/m ³
Specific heat	2200 J/Kg-K

that with the distance from the nozzle exit, larger flow-rate of carrier gas reduces the heat transfer to the particles, which causes lower particle temperature at higher flow-rate of carrier gas. It is also remarkable that at a flow-rate of 9 l/min, the heat transfer to particles is so small that the particle temperature is even less than melting point.

4 Experimental

4.1 Setup and Measurement Details

The experimental setup consists of a power supply unit (4 MHz, 20 kW), an induction plasma torch, and an attached reaction chamber below the torch. The plasma torch consists of a water-cooled quartz tube (45 mm inner diameter and 190 mm length), surrounded by a water-cooled three-turn induction coil which couples electromagnetic (EM) energy to the plasma at a frequency of 4.0 MHz. The plug power was 20 kW. Argon gas was introduced as the carrier gas (3 to 9 l/min) and the plasma gas (2 l/min). The sheath gas was composed of argon (22 l/min) and oxygen (2 l/min). The sheath gas, was injected with an induced swirling motion from the outer slots, protected the inner surface of the quartz tube from overheating.

The granular powders of raw material were prepared by the spray-drying method from the reagents of Na_2CO_3 , CaCO_3 and SiO_2 for the soda-lime-silica glass with the composition of Na_2O :16, CaO :10 and SiO_2 :74 in wt%. Na_2O mainly reduces the melting temperature of raw powders. The raw materials with about 58 μm in diameter was injected into the thermal plasma at a rate of 5 g/min by powder feeder and quenched glass powders were collected on a water-cooled substrate at 340 mm down to the nozzle. Other experimental conditions are the same as those listed in Table 1.

The micrograph and size distribution of the particles was carried out by scanning electron microscope (SEM) on JSM5310 (JEOL). The structures of the collected powders were determined by X-ray diffractometry (XRD). XRD was carried out on Miniflex (Rigaku) with Cu K_α radiation at 30 kV and

15 mA. The data were collected in the 2θ range 3-90° with a step size of 0.02° and a scanning speed of 4°/min. The thermogravimetric differential thermal analysis (TG-DTA), for thermal analysis was performed with thermogravimetry on TG8120 (Rigaku); the measured temperature in the range of 20-1400°C, at the rate of 10°C/min. The composition of the powders was analyzed by ICP-8100 (SHINADZU).

4.2 Experimental Results

As described in section 2.5, the simulated results revealed that the large flow-rate of carrier gas decreased the plasma temperature and increased the plasma and particle axial velocity which, in turn, decreased the relaxation/residence time. Fig.6 describes the effects of carrier gas flow-rate on the softening temperature and content of Na₂O in the quenched powders. The lower plasma temperature and shorter relaxation time, caused by the larger carrier gas flow-rate, resulted the lower evaporation rate of Na₂O, which in turn gave rise to the composition of Na₂O. Larger composition of Na₂O in quenched powders means smaller heat transfer to particles. Thus, the softening temperature, which was affected by the composition of Na₂O in the powders, decreased at larger carrier gas flow-rate. These experimental results are consistent with the theoretical predictions described in Fig.5.

5 Conclusions

A 2-dimensional LTE model was developed to

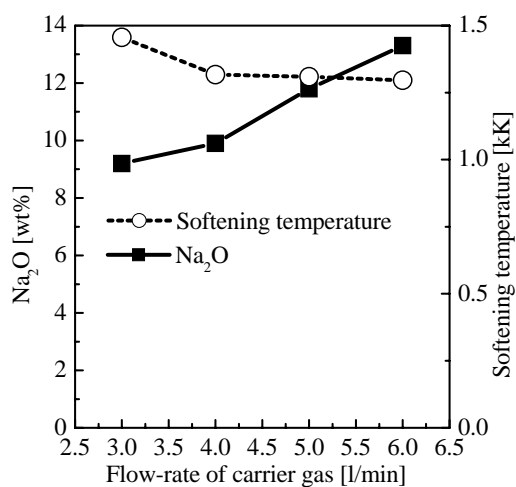


Fig.6 Distributions of Softening temperature and Na₂O contents of the quenched powders with the flow-rate of carrier gas

simulate the temperature and velocity profiles of argon-oxygen ITP including a metal tube inserted into the plasma torch for injecting the carrier gas and granular raw powders. Theoretically, it was found that the larger flow-rate of carrier gas caused to decrease the temperature and increase the velocity profiles around the centerline of the torch, which in turn, affected the composition of quenched powders and throughput. Experimental analysis revealed that with increased flow-rate of carrier gas, the softening temperature and heat transfer to particles decreased, as a result, the composition of Na₂O in the quenched powder increased. The experimental findings were well explained by the theoretical results. Even if, we did not consider the effects of the particle loading on the temperature and velocity profiles that will be the subject of the future work.

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