



# Modeling analysis on the silica glass synthesis in a hydrogen diffusion flame



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## ABSTRACT

Silica glass ingot synthesis by flame hydrolysis deposition (FHD) is an important approach to obtain high-purity synthetic silica glass with high refractive index homogeneity. Using the precursor of silicon tetrachloride ( $\text{SiCl}_4$ ), the silica ingot ( $\text{SiO}_2$ ) can be synthesized in the oxy-hydrogen diffusion flame through a set of kinetic reactions and subsequent cooling. The homogeneity of the synthetic silica glass is influenced by the temperature/diameter equality of silica droplets and the residue of radicals (e.g. OH) in the synthetic silica. In this study, the mixing system of oxy-hydrogen diffusion flame and silica droplets in a furnace were modeled by a Euler–Lagrange multi-phase model, where the interphase exchange of mass, momentum and energy between the gas phase molecules and liquid phase silica droplets during the formation and transportation of silica droplets were included. The silica droplets were tracked by a Lagrangian formulation that includes the discrete phase inertia, hydrodynamic drag, the force of gravity and the dispersion due to turbulent eddies. The molecular gas phase reactions are described by a simple kinetic mechanism involving the major species during the formation of molecular  $\text{SiO}_2$ , with kinetic and thermodynamic parameters taken from the literature when available.

The effect of initial equivalence ratio on the flame structure was analyzed in the study, where the high-temperature region is uniform over the glass ingot and the OH residue is limited at unity equivalence ratio. The probability distributions of silica droplet diameter and temperature were analyzed by employing a developed droplet growth model, where the condensation rate is controlled by the in situ  $\text{SiO}_2$  vapor concentration and the local flow-condition-determined mass transfer rate. The maximum diameter of silica droplets in the furnace is  $1.39 \times 10^{-4}$  m, and the percentage of silica droplets decreases with the increasing of diameter. Two high probability regions were observed for the droplet temperature distribution, the temperature below 1500 K accounts for 40% of the total droplet number, and the temperature range between 3500 and 4500 K accounts for 58%. The droplet diameter on the ingot cap distributes in the range from  $4 \times 10^{-5}$  to  $9 \times 10^{-5}$  m with approximately probability of 85%. The distribution of droplet temperature on the ingot cap is much uniform, with more than half (67%) of the droplets has the temperature between 3100 and 3200 K.

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## 1. Introduction

Flame hydrolysis deposition (FHD) using silicon tetrachloride ( $\text{SiCl}_4$ ) and oxy-hydrogen flame is an important approach to obtain high-purity synthetic silica glass with high refractive index homogeneity [1,2], which is crucially affected by the size, morphology and chemical composition of the formed silica droplets [3]. FHD method followed by a rapid consolidation at 1380 °C in vacuum ambiance has been shown to be capable of fabricating thick silica glass film up to 37  $\mu\text{m}$  with a high deposition rate of 8  $\mu\text{m}/\text{min}$ , which has a rather uniform refractive index  $1.4456 \pm 0.0005$  in

the wavelength 1500–1600 nm and a small absorption coefficient less than the magnitude of  $10^{-7}$  in the wavelength 1300–1600 nm [4]. FHD, also called flame aerosol synthesis is a process involving combustion and aerosol science and engineering. Detailed diagnostic analysis of the process with the concurrent chemistry, transport and particle dynamics would inevitably advance the understanding and technical control of the manufacture of fumed silica. However, experimental diagnosis on the droplet existing multiphase flame is mainly limited by the short residence times of chemical reaction and particle growth, as well as the high process temperature [3]. Since the formation and growth of silica droplets are highly coupled with the temperature and species distribution in the flame, interaction between the gas

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phase and the dispersed droplet phase should be included in the modeling to systematically investigate the effect of flame configuration on silica homogeneity.

The interacting effects of flame structure and silica formation have been experimentally and numerically investigated in the literature with different focuses. The thermophoretic transport of silica in chemical vapor deposition (CVD) processes was modeled but neglecting the counteraction of silica particles to the flame structure, where the uniformity and magnitude of the deposition are shown to be sensitive to the distance between the burner and the deposition target [5]. Temperature measurements in a silica-laden vapor axial deposition (VAD) flame by spontaneous Raman spectroscopy showed that the addition of silicon tetrachloride ( $\text{SiCl}_4$ ) produces substantial changes in flame structure, i.e. rise of the thermal front [6]. Another spatially resolved temperature and OH radical concentration measurements in coflow oxy-hydrogen diffusion flames by coherent anti-Stokes Raman spectroscopy (CARS) and planar laser-induced fluorescence (PLIF) show that in the non-reacting zone temperature decreases due to the addition of  $\text{SiCl}_4$ , whereas in the silica formation zone flame temperature increases due to the exothermal hydrolysis and oxidation of  $\text{SiCl}_4$  while the OH concentration decreases dramatically due to the reaction with HCl and the equilibrium shifts caused by the consumption of  $\text{O}_2$  and  $\text{H}_2\text{O}$  in  $\text{SiCl}_4$  related reactions [7]. The effect of silica formation on flame temperature and OH concentration was also experimentally and numerically observed in counterflow oxy-hydrogen flames [8], where the modeling incorporated detailed mechanism with  $\text{H}_2/\text{O}_2$  reactions, silica generating reactions and chlorinated species reactions. Based on a modified kinetics mechanism for the combustion of  $\text{SiCl}_4$  in an  $\text{H}_2/\text{O}_2/\text{Ar}$  flame, the effect of  $\text{SiCl}_4$  addition on the flame temperature and OH concentration was modeled and compared with the measurements by Laser-induced fluorescence (LIF) [9], where the underprediction of OH concentration is expected to be improved by considering particle formation and surface chemistry in the future study. Equilibrium analysis [9] also suggested that the flame temperature rise at  $\text{SiCl}_4$  addition is mainly attributed to the condensation of silicon oxides since the heat release in the combustion of  $\text{SiCl}_4$  is much small compared to that in the reaction between  $\text{H}_2$  and  $\text{O}_2$ . A simple kinetic mechanism describing the formation of silica starting from the  $\text{SiCl}_4$  combustion in high-temperature oxy-hydrogen flames were developed to reveal the impact of reaction channels on the overall reaction, where the reactivity of  $\text{SiCl}_4$  towards  $\text{H}_2\text{O}$  is highly favored over its dissociation [10]. The deposition rate of synthetic silica glass in CVD processes is influenced by the available  $\text{O}_2$  or  $\text{H}_2\text{O}$  amounts for oxidation or hydrolysis to completely convert  $\text{SiCl}_4$  to spherical silica particles, however, whose size and microscopic morphology are observed independent of the carrier gas [11].

To improve the purity of synthetic silica glass, the turbulent combustion field in the furnace was modeled with the silica droplets involved. The effect of equivalence ratio on the turbulent flame structure was examined by employing a detailed mechanism of  $\text{H}_2\text{-O}_2$  oxidation and  $\text{SiCl}_4$  hydrolysis. The growth and transportation of silica droplets in the oxy-hydrogen diffusion flames in the furnace were modeled by considering the mass, momentum and energy exchanges between the gas and dispersed phases. The modeling results were analyzed to numerically suggest more preferable furnace configurations for silica synthesis.

## 2. Numerical model

### 2.1. Turbulent oxy-hydrogen flame modeling

The turbulent non-premixed oxy-hydrogen flame were modeled by Large Eddy Simulation (LES), where large eddies are

resolved directly and small eddies are modeled. The Favre mean equations solved for mass, momentum, species and total enthalpy are given as,

$$\frac{\partial \bar{\rho}}{\partial t} + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j) = 0 \quad (1)$$

$$\frac{\partial \bar{\rho} \tilde{u}_i}{\partial t} + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j \tilde{u}_i) = -\frac{\partial \bar{p}}{\partial x_i} + \frac{\partial}{\partial x_j} \left( \mu_{\text{eff}} \left( \frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} - \frac{2}{3} \frac{\partial \tilde{u}_k}{\partial x_k} \delta_{ij} \right) \right) + \bar{\rho} g_i \quad (2)$$

$$\frac{\partial \bar{\rho} \tilde{Y}_\alpha}{\partial t} + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j \tilde{Y}_\alpha) = \frac{\partial}{\partial x_j} \left( \left( \bar{\rho} D_\alpha + \frac{\mu_t}{\sigma_t} \right) \frac{\partial \tilde{Y}_\alpha}{\partial x_j} \right) + \bar{\omega}_\alpha \quad (3)$$

$$\frac{\partial \bar{\rho} \tilde{H}}{\partial t} + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j \tilde{H}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_t}{Pr_t} \frac{\partial \tilde{H}}{\partial x_j} + \sum_\alpha h_\alpha \left( \bar{\rho} D_\alpha + \frac{\mu_t}{\sigma_t} \right) \frac{\partial \tilde{Y}_\alpha}{\partial x_j} \right) + \bar{\omega}_H \quad (4)$$

where Boussinesq hypothesis has been employed here to parameterize the deviatoric part of the subgrid scale stress and the isotropic part was neglected assuming low Mac number. The turbulent Schmidt number and Prandtl number are both set as 0.85. The turbulent viscosity  $\mu_t$  is given by Smagorinsky–Lilly model,

$$\mu_t = \bar{\rho} \cdot \min(\kappa d, C_s \Delta)^2 \sqrt{2 \tilde{S}_{ij} \tilde{S}_{ij}} \quad (5)$$

where the minimum function determines the mixing length for subgrid scales,  $\kappa$  is von Karman number constant,  $d$  is the distance to the closest wall,  $C_s$  is the Smagorinsky constant,  $\Delta$  is the local grid scale, and  $\tilde{S}_{ij}$  is the viscous strain-rate tensor.

The mean production rate  $\bar{\omega}_\alpha$  of species  $\alpha$  by chemical reaction is calculated by the eddy-dissipation-concept (EDC) model [12] that incorporates detailed chemical mechanism for oxy-hydrogen and silica generating reactions in the turbulent flow. EDC assumes the reactions occur in fine turbulent scales with length fraction  $l^*$  and fine time scale  $\tau^*$  ( $*$  denotes fine-scale quantities),

$$l^* = C_l \left( \frac{\nu \varepsilon}{\kappa^2} \right)^{1/4} \quad (6)$$

$$\tau^* = C_\tau \left( \frac{\nu}{\varepsilon} \right)^{1/2} \quad (7)$$

where constants  $C_\xi = 2.1377$ ,  $C_\tau = 0.4082$ ,  $\nu$  – kinetic viscosity,  $\kappa$  – kinetic energy and  $\varepsilon$  – energy dissipation rate. Treating the fine scale as a constant pressure reactor, reactions proceed over the fine time scale  $\tau^*$  with the initial condition of current species and temperature in the cell, reaching the final state with species mass fraction  $Y_\alpha^*$ . The fine-scale reactions are governed by the Arrhenius rates. The mean production rate for species  $\alpha$  is then calculated as,

$$\bar{\omega}_\alpha = \frac{\bar{\rho} l^{*3}}{\tau^* (1 - l^{*3})} (Y_\alpha^* - Y_\alpha) \quad (8)$$

where  $l^{*3}$  represents the volume fraction of the fine scale. Detailed twenty-step oxy-hydrogen reaction mechanism from CHEMKIN model [13] plus reduced four-step  $\text{SiCl}_4$  hydrolysis mechanism [10] is included in the volumetric reaction model, as listed in Table 1. The 15 involved species are  $\text{H}_2$ ,  $\text{H}$ ,  $\text{O}_2$ ,  $\text{O}$ ,  $\text{OH}$ ,  $\text{HO}_2$ ,  $\text{H}_2\text{O}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{SiCl}_4$ ,  $\text{SiCl}_3\text{OH}$ ,  $\text{SiCl}_2\text{O}$ ,  $\text{SiClOOH}$ ,  $\text{SiO}_2$ ,  $\text{HCl}$  and  $\text{N}_2$ .

The radiative heat transfer, mainly contributed by gas-phase  $\text{H}_2\text{O}$ , is solved by a Discrete Ordinate (DO) model [14], which solves the radiative transfer equation (RTE) for a finite number of discrete solid angles in the global Cartesian system. Each octant of the angular space is sectioned into two uniform extents in the polar and azimuthal angles respectively, and thus RTE in a total of  $8 \times 2 \times 2$  directions are solved in the whole angular space  $4\pi$ . Each control angle is further divided into  $2 \times 2$  pixels to trace the energy incoming or outgoing to the face. Gray gas model is used in the modeling by assuming the scattering coefficient, the scattering phase function, and the refractive index are all independent of wavelength. The absorption coefficient as a function of species concentration, pressure and temperature is calculated by the weighted

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