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PHYSICAL AND MATHEMATICAL MODELING OF SPINEL SETTLING IN HIGH-LEVEL WASTE GLASS

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ABSTRACT

A two-dimensional unsteady mathematical model of particle settling in high-viscosity liquid is presented. To verify this model by laboratory experiments, the settling of monodisperse alumina particles in a model liquid was observed. The alumina particles had similar sizes as spinel crystals, the viscosity of the model liquid was close to that of a high-level waste glass, and the density difference between alumina and the model liquid was also close to the density difference between spinel and molten glass. The field equations of the mathematical model were solved for the initial and boundary conditions corresponding to the laboratory arrangement. The comparison of the mathematical description with laboratory experiments is the first step in the verification procedure of the model for spinel settling during high-level waste vitrification.

INTRODUCTION

The formation of insoluble phases in glass is an important problem of the vitrification of high-level waste (HLW). Spinel particles precipitated from molten glass below the liquidus temperature settle on the bottom of the melting space and may block the pour spot. To reduce the risk of melter failure due to spinel settling, the requirement has been imposed on HLW glass that its liquidus temperature is below the minimum temperature of the glass melter (estimated as 1050°C). This restriction substantially increases the process and disposal cost due to the limited waste fraction loaded to the glass. One possibility for reducing the cost of HLW vitrification is to increase waste loading to an extent, at which the amount of accumulated spinel phase does not interfere with melter operation. An assessment of spinel accumulation during melter operation demands detailed knowledge of spinel behavior in glass including thermodynamics, kinetic, hydrodynamics and rheology of the spinel-glass system.

The rate of spinel settling and accumulation on the melter bottom can be estimated using mathematical modeling. The experimental results by LaMont and Hrma [1] have shown that spinel crystals behave as a swarm rather than individual particles. The description of settling by the Stokes law appears inadequate. The aim of this work is to formulate a two-dimensional mathematical model of insoluble particle settling in high-viscosity liquid and to verify this model with laboratory experiments.

EXPERIMENTAL PART

The settling rate of monodisperse alumina particles that had an average size of 10 and 100 μm in model liquid was observed during laboratory experiments. The model liquid was prepared by using the glycerin and the citric acid [2] mixed at the ratio to adjust the viscosity of the final liquid at the laboratory temperature (20°C) to the same value as that of HLW glass, just above the liquidus temperature. The density of the prepared liquid was 1300 kg/m^3. The density of alumina, 4000 kg/m^3, ensures that the density difference in the studied system is close to that between spinel and molten glass.

The viscosity of the alumina-particle suspension in the model liquid was measured by Hoppler's viscometer. The suspension of 10-μm particles up to the concentration of particles in the liquid, c = 100 kg/m^3, did not exhibit a measurable contribution to the viscosity of the pure model liquid, which was 2.5 Pa·s at 20°C. Larger alumina particles, 100 μm, increased the viscosity of the suspension (up to c = 100 kg/m^3) as follows

$$\eta_{\text{SUSPENSION}} = \eta_{\text{PURE LIQUID}} + b\,c \qquad (1)$$

where b = 0.0127 m^2/s. Equation (1) was incorporated into the mathematical model.

The experimental arrangement is shown in Figure 1. The observation cell consisted of two cubic parts having a dimension of 5 × 5 × 5 cm. In the beginning of the experiment, the bottom cell was filled with pure model liquid and covered by a thin sheet. Then the upper (bottomless) part was placed above the bottom cell and filled with the colored hand-mixed suspension of alumina particles. The sheet separating both liquids was slowly pulled up and the shift of the originally plane interface produced by the settling of alumina particles was monitored by a videocamera.

Figure 2 shows an example of experiments. As can be seen, the particles did not settle individually; rather, the suspension plunged into the pure liquid below. The asymmetrical shape of the plume is caused by concentration non-uniformity of the suspension at the original interface. The shift of the settling front as a function of particle size and concentration was observed. The average velocities of the settling front, obtained by fitting straight lines to data, were used for the verification of the mathematical model (see Figure 3).

The sedimentation rate of alumina particles (80 μm, c = 100 kg/m^3) was measured as the thickness of the settled layer at the bottom of the cell. Figure 4 (see Section 4) compares experimental values with those calculated by the mathematical model.

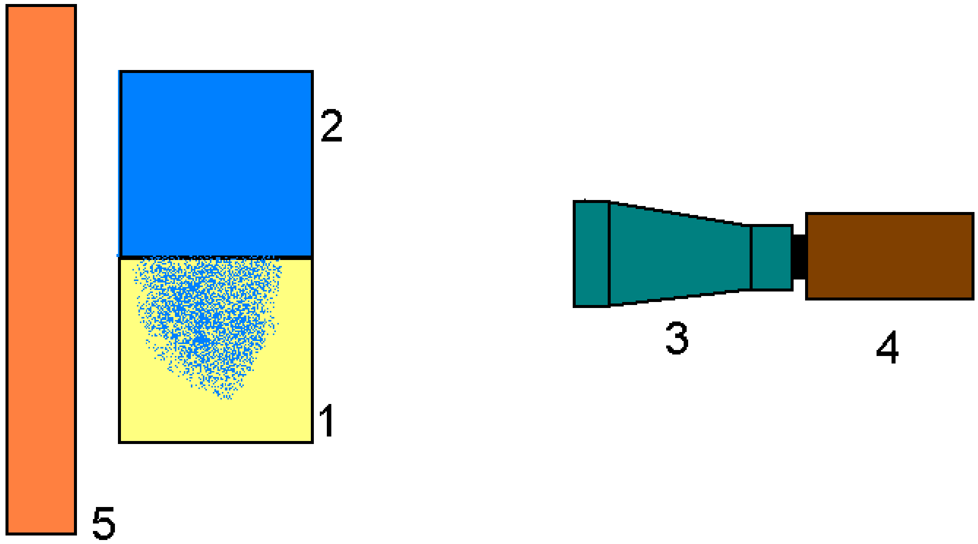
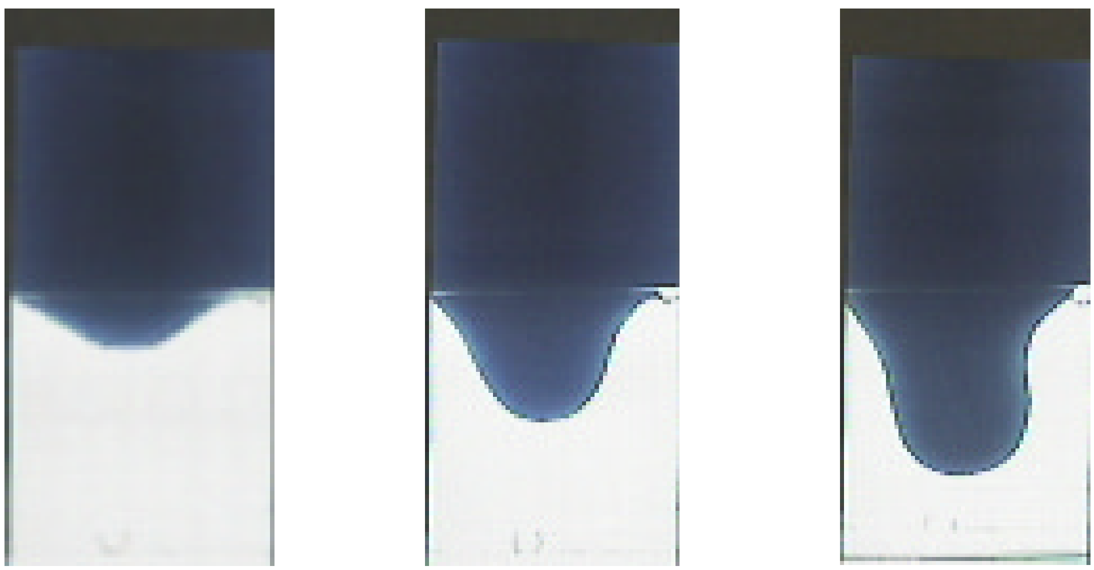


Figure 1. The scheme of the observation method: 1 - bottom cell with pure model liquid, 2 -upper cell with alumina suspension, 3 – objective, 4 – videocamera, 5 – illuminative box



2 minutes 4 minutes 6 minutes

Figure 2. Motion of the interface between the liquid with suspended alumina particles (100µm, c = 12.5 kg/m^3) and the pure liquid.

MATHEMATICAL MODEL

The mathematical description of the problem is based on the theory of continua. We postulate that the presence of solid particles increases the average density of the liquid. The buoyancy force acting on the unit volume of the suspension is given by

$$F_v = g\,c\left(1 - \frac{\rho_l}{\rho_s}\right) \tag{2}$$

where g is the acceleration due to gravity, c is the concentration of particles, and ρ_s and ρ_l are the density of solid particles and the liquid, respectively.

The settling rate of solid particles is predicted by Stokes law:

$$v_{STOKES} = \frac{(\rho_s - \rho_l)\,g\,d^2}{18\,\rho_l\,\nu} \tag{3}$$

where d is the particle diameter and ν is the kinematic viscosity of the liquid.

The mathematical model formulated for a two-dimensional case involves the momentum balance, continuity equation and the balance of solid particles:

$$\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial \mathrm{x}} + v\frac{\partial u}{\partial \mathrm{y}} = \nu\left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2}\right) - \frac{1}{\rho_l}\frac{\partial P}{\partial x} \tag{4}$$

$$\frac{\partial v}{\partial t} + u\frac{\partial v}{\partial x} + v\frac{\partial v}{\partial y} = \nu\left(\frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2}\right) - \frac{1}{\rho_l}\frac{\partial P}{\partial y} - \frac{1}{\rho_l}c\,g\left(1 - \frac{\rho_l}{\rho_s}\right) \tag{5}$$

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \tag{6}$$

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2}\right) - \left[u\frac{\partial c}{\partial x} + \left(v - \frac{(\rho_s - \rho_l)\,g\,d^2}{18\,\rho_l\,\nu}\right)\frac{\partial c}{\partial y}\right] \tag{7}$$

Here u and v denote velocity components, P is the pressure, D is the diffusion coefficient representing Brown's motion of particles, and x and y are the horizontal and vertical coordinates.

The set of field Equations (4–7) was solved numerically using the method of control volumes for the initial and boundary conditions corresponding to the laboratory arrangement.

NUMERICAL RESULTS AND DISCUSSION

The experimental system with a heavy fluid initially placed above a lighter liquid in a gravitational field (as described in Section 2) produces the Rayleigh-Taylor instability [3–5]. The variability of the shape of the settling front (the interface between the falling suspension and the pure liquid) indicates that hand mixing of the suspension did not ensure a perfectly uniform dispersion of particles. In addition, the particle concentration at the initial interface probably fluctuates, affecting the horizontal position of the settling front.

To estimate the effect of experimental fluctuations, the additional initial condition was incorporated in the model. In the first set of calculations, the particle concentration in the middle of the cell was increased by 5, 10 and 20% in the middle of the cell. The corresponding time development of the settling front was almost independent of the value of the fluctuation.

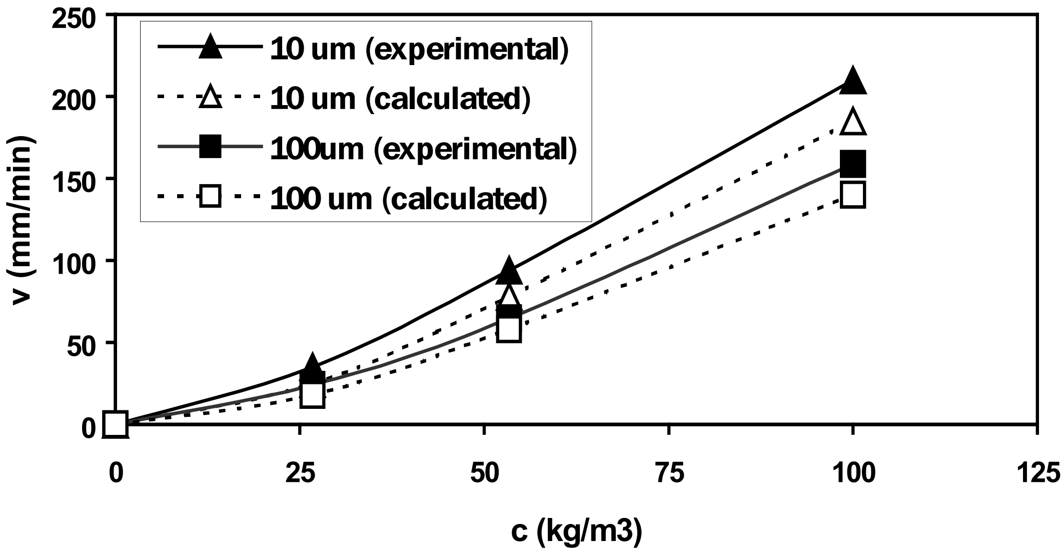


Figure 3. The comparison of experimental and calculated values of the settling front velocity.

The following set of calculations was performed to verify the model's ability to predict the influence of particle size and concentration on the settling-front velocity using 10% fluctuation in the middle of the cell. Figure 3 compares experimental and calculated results. The differences between the experimental and calculated the settling front velocities were within 15%.

Figure 4 demonstrates experimental verification of particle accumulation in the bottom of the cell simulated by the model. The calculated growth rate of settlings is about 1.5 times larger than the experimental value.

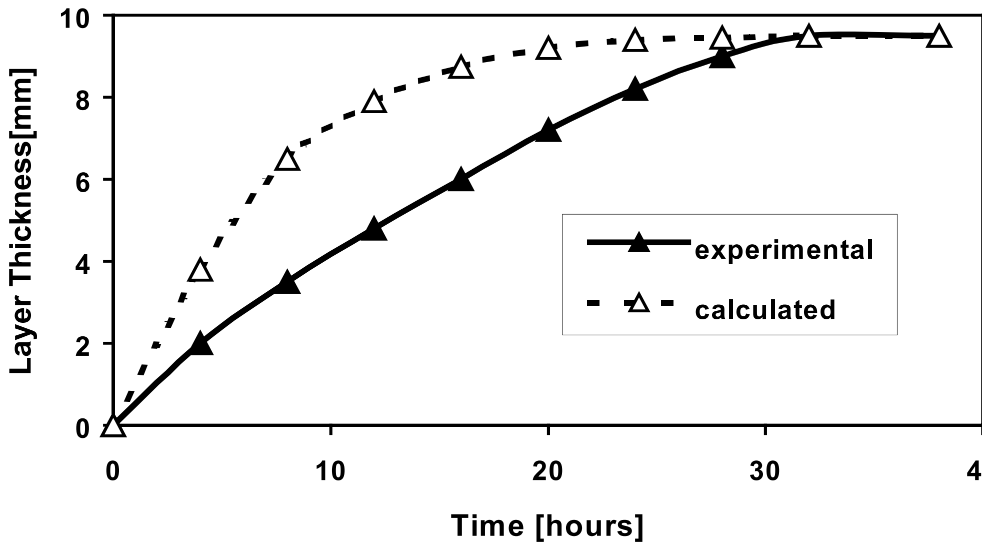


Figure 4. The experimental and calculated sedimentation rate of alumina particles (80 µm, c = 100 kg/m^3).

CONCLUSION

The two-dimensional unsteady mathematical model for settling of insoluble particles in a high-viscosity liquid is the first step toward predicting spinel accumulation in the HLW melter. The movement of suspension falling into a pure liquid was evoked by particle concentration fluctuation higher than 10% in the center of the initial suspension-pure liquid interface. The mathematical model with input data, including initial concentration distribution, liquid/suspension density, and viscosity, was verified by the laboratory measurement of the effect of particle size and concentration on the settling velocity. Experimental and calculated values were in acceptable agreement with differences up to 15%. In addition, the model simulates the accumulation of particles in the cell bottom. The sedimentation rate predicted by the model was about 1.5 times higher than the experimental value.

Future work will focus on the three-dimensional simulation of spinel accumulation, including the effect of thermal convection and the experimental data of spinel formation and dissolution in HLW glass melt.

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