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Axisymmetric Marangoni convection in microencapsulation

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Abstract

Spherical shells used as laser targets in inertial confinement fusion (ICF) experiments are made by microencapsulation. In one phase of manufacturing, the spherical shells contain a solvent (fluorobenzene (FB)) and a solute (polystyrene (PAMS)) in a water–FB environment. Evaporation of the FB results in the desired hardened plastic hollow spherical shells, 1–2 mm in diameter. Perfect sphericity is demanded for efficient fusion ignition and the observed surface roughness maybe driven by Marangoni instabilities due to surface tension dependence on the FB concentration (buoyant forces are negligible in this micro-scale problem). Here we model this drying process and compute nonlinear, time-dependent, axisymmetric, variable viscosity, infinite Schmidt number solutocapillary convection in the shells. Comparison with results from linear theory and available experiments are made.

1. Introduction

Successful ICF experiments require high and spherically uniform laser energies which are absorbed by spherical fuel capsules made of a plasma polymer [1]. The compression of the fuel must be uniform and perturbations on the inner capsule wall would grow during implosion due to Rayleigh–Taylor instabilities. The resulting mixing of the polymer and fuel degrades the fusion ignition. Manufacturing of 1 mm fuel targets [2] has been achieved by microencapsulation (see Fig. 1) to produce hollow PAMS mandrels. These mandrels are then coated with the plasma polymer. Heat treatment decomposes the PAMS which diffuses through the plasma polymer coating leaving behind the desired target hollow shells. McQuillan [3] has demonstrated that Marangoni instabilities driven by surface tension dependence of the FB concentration are the cause of outer surface deformations and deviation from sphericity observed with 2 mm shells. McQuillan and Takagi [4] were able to prevent the formation of surface ripples in 1–2 mm mandrels by manipulating the prevailing Marangoni numbers in the experiments. These Marangoni instabilities can cause more and serious bumps during the manufacturing of planned larger spherical targets. Hence, it is essential to study the hydrodynamics of the convective patterns which develop in the spherical shells during the drying process.

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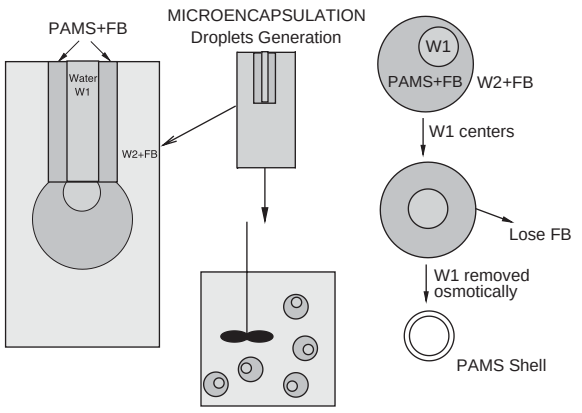

Fig. 1. Sketch of microencapsulation. Flows through the capillaries of the generator produce droplets of water, surrounded by a mixture of FB and PAMS, and suspended in an aqueous solution. The FB is removed by evaporation over several hours/days leaving a cured solid PAMS shell. The water droplet is then later removed by osmosis into ethanol.

The pattern of motion realized in a convectively unstable system with spherical symmetry can be considered without reference to the physical details of the system. Numerous theories have been developed over the years to study the heat transfer in the mantles of terrestrial planetary interiors which occurs by convection [5]. Busse [6] studied the patterns of nonlinear convection in a homogeneous fluid contained between two concentric spherical boundaries assuming a spherically symmetric gravity force and distribution of heat sources. Nonlinear axisymmetric convective motions of self-gravitating, infinite Prandtl number fluids in spheres and spherical shells for different modes of heating were modeled by Zebib et al. [7]. A theoretical study of the linear and weakly nonlinear variable viscosity convection in spherical shells with an infinite Prandtl number fluid and two modes of heating was also performed by Zebib [8]. Three-dimensional steady thermal convection of an infinite Prandtl number, Boussinesq fluid with temperature-dependent viscosity was examined by Ratcliff et al. [9]. Linear stability analysis of the Marangoni mechanism in spherical shells during microencapsulation was considered by Subramanian et al. [10,11]. In the present paper, we study nonlinear axisymmetric convection in the shells assuming infinite Schmidt number.

2. Mathematical model

We consider a spherical shell of initial thickness $L_r = R_{2_0}^* - R_1^*$, where R_1^* and $R_{2_0}^*$ are the shell's inner and initial outer radii, respectively. The aspect ratio of the shell is $\eta = R_1^*/R_{2_0}^* < 1$ (all starred quantities are dimensional). The inner boundary is assumed stress free and impermeable, while nonlinear boundary conditions are prescribed at the moving outer boundary. The shell contains a mixture of a solvent and a solute with concentrations C^* and $(1-C^*)$, respectively. The ambient is a mixture of the solvent and water into which the solvent is evaporating. Thus, there is a net mass flux across the receding outer surface.

The physical quantities are nondimensionalized with respect to ρ_r, μ_r, ν_r, D_r, L_r, $t_r = L_r^2/D_r$, C_r, D_r/L_r, $P_r = \mu_r/t_r$ for density, dynamic viscosity, kinematic viscosity, mass diffusivity, length, time, concentration, velocity, and pressure, respectively. We assume linear variation of interfacial tension σ^* with concentration C^* according to $\sigma^* = \sigma_r - \gamma(C^* - C_r)$, where subscript r designates a reference state. The Capillary number $Ca = \Delta\sigma/\bar{\sigma} = (\bar{\sigma} - \sigma_r)/\bar{\sigma} = \gamma C_r/\bar{\sigma}$. Here, $\bar{\sigma} = \sigma_r + \gamma C_r$ and $\gamma = -\mathrm{d}\sigma^*/\mathrm{d}C^*$. Relevant nondimensional quantities are: the mass transfer Biot number $Bi = KL_r/\rho_r D_r$ based on an assumed mass transfer coefficient K which is taken as constant, the Reynolds number $Re = \gamma C_r L_r/\mu_r \nu_r$ and the Marangoni number $Ma = Re\,Sc$, where the Schmidt number $Sc = \nu_r/D_r$ is about 10^6 and is assumed infinite in our nonlinear model.

In the limit of $Ca \to 0$ considered here, it was shown [11] that the $O(1)$ outer surface is a perfect sphere $r_2 = r_{2d}(t)$ determined by the diffusive state, while deviations from sphericity are $O(Ca)$. The diffusive state is the solution of the nonlinear dimensionless system:

$$\frac{\partial C}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial C}{\partial r}\right) \tag{1}$$

with initial condition $C(r, 0) = 1$. The boundary conditions derived from conservation of the solute and solvent are

$$\frac{\partial C}{\partial r}(r_1, t) = 0 \tag{2}$$