Modeling Void Drainage with Thin Film Dynamics

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Abstract: Voids in composite materials can lead to degraded structural performance. The following is a study of voids or bubbles in uncured viscous polymer resin during composites processing. The goal is to determine if voids can successfully migrate towards vacuum pathways, coalesce with the pathways, and escape. Inherent to the coalescence process is the drainage and rupture of the resin thin film formed between voids and the resin free surface. COMSOL Multiphysics 4.2 + Microfluidics module is employed for modeling. The model consists of a single spherical void in a cylindrical axisymmetric two-phase domain of resin and air. Results suggest that resin thin film drainage can be successfully modeled as an exponential decay consistent with experimental results. Thin film rupture modeling is limited due to mesh dependency issues. Also, void dynamics is strongly dependent on void body force and surface tension effects as characterized by the Bond number (Bo).

Keywords: Voids, Bubbles, Coalescence, Thin Film Drainage, Thin Film Rupture

1. Introduction

One of the methods to fabricate composites is from prepregs. A prepreg is like a tape with unidirectional continuous fibers partially covered with a polymeric resin. The prepregs are stacked in the desired sequence on top of a tool using a pressure roller. The pressure roller redistributes the resin and partially consolidates the stacked layers. The composite at this stage will have some void regions that have no resin or fibers. The stacked sequence is subjected to a vacuum to remove the air and water vapor from these prepregs so it is important that there are pathways to extract these voids before the prepregs are fully consolidated in an oven to fabricate a void free composite. Voids in polymer matrix composite materials can compromise structural performance. The following is a study of voids or bubbles in uncured viscous polymer resin as it is being processed to form composites. The goal is to determine if voids can successfully migrate through fibrous porous media towards vacuum pathways, coalesce with the pathways, and escape under processing conditions. Precursor to the coalescence process is the drainage and rupture of the resin thin film formed between voids within the resin in the proximity of the resin free surface. Figure 1 describes a simplified model schematically. Note the presence of air evacuation and flow due to applied vacuum and resin flow near the embedded void. It will be important to establish how these flows induce the movement of the void which in turn needs to rupture through the resin interface before it merges with the air being evacuated by the applied vacuum. The ability of the void to break through the resin surface will be a function of the resin thin film dynamics as the void approaches the air-resin interface. For this work, the scope is focused on the establishment of resin thin film dynamics under the influence of body forces and surface tension effects without the presence of fibers and is modeled using COMSOL.

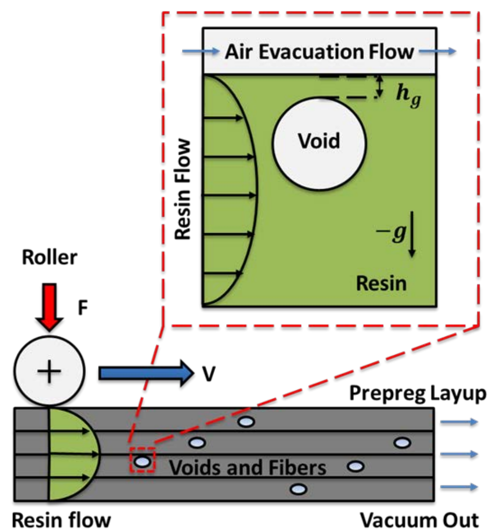

Figure 1. Void migration during composite processing. External pressure is applied to encourage void migration. Resin thin films of thickness h_g are formed between voids and vacuum pathway free surfaces.

2. Theory

The rising bubble through viscous fluid problem has been studied in depth for many years. This problem is encountered in a wide range of industrial processes. Some examples are the production of foams, industrial gases, and chemicals. Of particular interest is the bubble coalescence dynamics between adjacent bubble and a fluid free surface. As noted by Janssen and Anderson (2011), the coalescence process of two moving drops (or bubbles) or a single drop (or bubble) with a free surface can be divided into three stages: (1) approach, (2) drainage, and (3) rupture.

2.1 Void Approach

The approach of a void or bubble towards another bubble or a free surface can be modeled with conventional Navier-Stokes equations. The Stokes equation is a simplification of the Navier-Stokes equations that are applicable for the transport of small bubbles in viscous fluids with negligible fluid inertia (i.e. viscous polymer resin). It can be written with the incompressibility as:

$$\nabla p = \mu \nabla^2 \boldsymbol{u} + \boldsymbol{f}$$

$$\nabla \cdot \boldsymbol{u} = \boldsymbol{0}$$

Here, ∇p is the pressure gradient, μ is the dynamic viscosity, $\boldsymbol{u}$ is the velocity field, and $\boldsymbol{f}$ are applied body forces. The Stokes equation is applicable when the Reynolds number is small. An important dimensionless number for understanding the rising bubble problem is the Bond number (Bo) and is defined by Pigeonneau and Sellier (2011) as:

$$Bo = \frac{\rho g a^2}{3\gamma}$$

Here, ρ is the density of the fluid, g is the acceleration due to gravity, a is the bubble radius, and γ is the surface tension of the interface. Note the Bond number can be used to measure the importance of surface tension versus body forces. This will be important for establishing the effectiveness of a void's approach towards a free surface.

2.2 Void Drainage

When the bubble arrives close to another bubble or free surface, the fluid in between the interfaces begins to drain away reducing the film thickness. There are many approaches to model the dynamics of this type of thin film drainage. One particular model noted by Debrégeas et al. (1998) has been experimentally observed for an air bubble thin film in PDMS fluid approaching the free surface:

$$h(t) = h_0 \exp(-t/\tau)$$

$$\frac{1}{\tau} = \frac{\rho g a}{\mu}$$

Here, h is the film thickness of the bubble/free surface interface, h_0 is the initial film thickness, t is time, τ is the characteristic time, and μ is the fluid viscosity. Other more complicated models have been presented by Howell (1999) and others. Key to this work will be the comparison of this model with implemented numerical model.

2.3 Void Rupture

As the void or bubble drainage occurs, the influence of molecular interactions become more apparent. It is noted by Janssen and Anderson (2011) and Debrégeas et al. (1998) that a bubble will spontaneously rupture when the thin film thickness h approaches 70 [nm]. At the nanoscale, long range van der Waals forces act to rapidly enhance the drainage process. In addition, as the film thickness reduces it causes instability in the thin film surface which leads to film rupture liberating the void. It was noted by Debrégeas et al. (1998) experimentally that the growth velocity of a small rupture of a bubble film in viscous fluid scales as follows:

$$V \sim C\gamma/\eta$$

Here, V is the film rupture growth velocity, γ is the surface tension of the interface, and C is a constant. It is difficult to numerically model the film rupture process due to the large length scale difference between bubble radius (millimeters) and the film thickness as it reduces to nanometers. In this work, we seek to establish general trends of void approach and drainage such that the void rupture time can be extrapolated. We hypothesize that if voids can arrive and coalesce at vacuum pathways, they will escape the layup and not cause structural issues upon resin curing. It is of interest to estimate the time scales that voids take to migrate towards vacuum pathways and initiate drainage.

3. Methods

Level set methods are popular for compressible and incompressible flow. The interface is represented by the zero contour of a signed distance function, called the level set function. The movement of the interface is governed by a differential equation for the level set function. Level set methods automatically deal with topological changes and can provide high-order accuracy. Figure 2 displays the Level Set Method schematically. It depicts areas that represent fluid interfaces that are tracked by the plotted level set function (right) with respect to the flat x-y plane (left). As time elapses, the interface is found by advecting the level set variable (Φ) in time, which is translated to the x-y plane as a moving interface. One can imagine that this process is significantly less difficult to compute versus direct parameterization of the interface in time. A reinitialization process is necessary in order to maintain the level-set function as a signed distance function. One of the primary drawbacks of level set methods is that they are not conservative. For incompressible two-phase flow, loss or gain of mass is possible, which is not physical. The advection process is governed by a differential equation and needs to be implemented by a (weighted) essentially non-oscillatory method as noted by Shu and Osher (1988). The interface is identified by zero level (i.e. $\Phi = 0$) and the level set function is continuous near the interface. COMSOL uses $\Phi = 0$ and $\Phi = 1$ to differentiate between fluids. Primary difficulty in solving interface advection comes from the fact that the level set equation is of highly hyperbolic nature. (i.e. no dissipation mechanism or an extremely convection dominant problem). Attention must be given when advecting the function to mitigate these issues and improve interface stability.

Olsson and Kriess (2005) proposed an alternative level set function, together with an advection scheme, resulting in conservation of area bounded by the interface. Note the velocity field is assumed to be divergence free. A smeared out Heaviside function is employed as the level set function. Over the interface, it varies smoothly from zero to one. The advection of the level set function is performed using an intermediate step. This keeps the shape and width of the profile across the interface constant. In addition, the level set function is smooth, which makes it possible to extend derivatives to higher order. Olsson and Kriess (2005) describe in detail the methodology that overcomes some of the difficulties encountered with the usual methods that have been employed in this work.

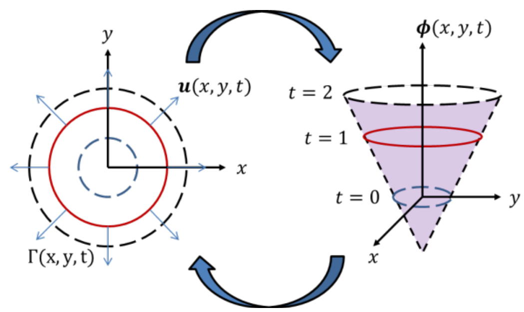

Figure 2. Level Set Method (LSM) schematic for two-phase fluid systems.

4. Governing Equations

The governing equations for the incompressible two phase flow with interface tracking include the conventional Navier-Stokes and continuity equations with the addition of the level set variable for the interface advection equation. Those equations are written respectively with the following:

$$\boldsymbol{\nabla} \cdot \boldsymbol{u} = \mathbf{0}$$

$$\mathbf{u}_t + (\boldsymbol{u} \cdot \nabla)\boldsymbol{u} = -\frac{\nabla p}{\rho} + \frac{1}{\rho Re}\nabla \cdot \left(\mu(\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T)\right) + \frac{1}{Fr^2}\boldsymbol{e}_g + \frac{1}{\rho We}\tilde{F}_{sv}$$

$$\Phi_t + \mathbf{u} \cdot \nabla\Phi = 0$$

Here, $Re = \rho_{ref} u_{ref} l_{ref} / \mu_{ref}$ is the Reynolds number, $Fr = u_{ref} / \sqrt{l_{ref} g}$ is the Froude number, $We = \rho_{ref} u_{ref}^2 l_{ref} / \gamma$ is the Weber number and $\tilde{F}_{sv}$ is a nondimensionalized surface tension contribution (i.e. $\tilde{F}_{sv} = \boldsymbol{F}_{sv}/\gamma$). Note that the surface tension per interfacial area, i.e. interfacial stress, at a point $\boldsymbol{x}_I$ on the interface is defined by the following:

$$\boldsymbol{F}_{sa}(\boldsymbol{x}_I) = \sigma\kappa(\boldsymbol{x}_I)\hat{n}(\boldsymbol{x}_I)$$

Thus, one can resolve this as a body force at any point $\boldsymbol{x}$ as follows,

$$\boldsymbol{F}_{sv}(\boldsymbol{x}_I) = \sigma\left(-\nabla \cdot \frac{\nabla\Phi}{|\nabla\Phi|}\right)\nabla\Phi$$

This formulation will results in the same total force as $\boldsymbol{F}_{sa}(\boldsymbol{x}_I)$, but distributed over a finite interface width. This is necessary to maintain robust numerical computation in the level-set method. To make the density and viscosity vary smoothly over the interface, the following relationships can be applied for fluids 1 and 2:

$$\rho = \rho_1 + (\rho_2 - \rho_1) \cdot \Phi$$

$$\mu = \mu_1 + (\mu_2 - \mu_1) \cdot \Phi$$

5. Numerical Model

COMSOL Multiphysics 4.2 + Microfluidics module is employed for numerical solution. The model consists of a single spherical void in a cylindrical axisymmetric two-phase domain of resin and air. Figure 3 and Table 1 displays the simplified model schematically with baseline parameters. COMSOL was used to solve the transient problem with initialization of the laminar two-phase flow with the Level Set Method. Of interest are the interface evolution between resin and air in time, in which the influence of interfacial tension between resin and void and the body force (buoyancy) is accounted for. The air domain is modeled as a fictitious fluid with the viscosity and density being 100 times smaller than those of the resin to avoid large difference in magnitude in the final assembled stiffness matrix, but at the same time to address the differences in the physical behavior of the air from that of the resin. Meshing was performed by the default triangular meshing algorithm available in COMSOL Multiphysics 4.2 with an average mesh size of h_{avg} = 6.25E-6 [m].

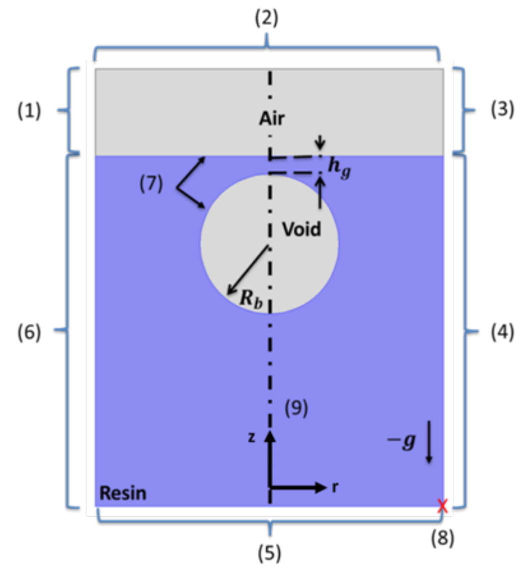

Figure 3. Axisymmetric baseline model setup with parameters defined in Table 1.

Table 1: Axisymmetric 2D baseline parameters

Parameters	Values	
Domain width	1.00 mm	
Domain height	1.25 mm	
Air domain thickness	0.25 mm	
Void radius, R_b	0.20 mm	
Thin film thickness, h_g	0.05 mm	
Interfacial tension, σ	0.03 N/m	
Level set reinitialization, γ	0.001 m/s	
Phase	**Density, ρ [kg/m^3]**	**Viscosity, η [Pa-s]**
Resin	1000	10
Air	10	0.1
Edge ID	**Boundary Condition**	
(1)-(6)	No slip wall	
(7)	Initial air-resin interface	
(8)	Pressure point constraint	
(9)	Axis of Symmetry	

6. Results and Discussion

The key resulting parameter is the change in resin film thickness h_g versus time. Figure 4 displays a plot on the log scale of non-dimensional film thickness versus non-dimensional drainage time as a function of the Bond number (Bo). These trends are of similar form to the numerical results presented by Pigeonneau and Sellier (2011) for a similar rising bubble problem with a boundary-integral technique. The resin film thickness is non-dimensionalized with respect to the initial void radius (i.e. $h^* = h/2a$). The drainage time scales (i.e. $t^* = tga/6\nu$) with ν as the kinematic viscosity. Note that Bo = 0.0 represents the analytical solution of a void of either very small size bubble or very high surface tension. The analytical solution is given by Pigeonneau and Sellier (2011) as the following implicit equation for $h(t)$:

$$\left(\gamma_E + \frac{1}{2}\right) h + h \ln h = \left(\gamma_E + \frac{1}{2}\right) h_0 + h_0 \ln h_0 - t$$

Here, $\gamma_E \approx 0.57721$ or the Euler constant. In general, increasing Bo leads to longer film drainage time between Bo = 0.0 to Bo = 1.0. The linear trend on the log-linear scaled plot implies an exponentially decaying interface film thickness. This trend was observed experimentally in [1] for gas bubbles in a viscous fluid. From this trend, one can extrapolate the rate of decay and determine how much time until the drainage reaches tens of nanometer scales. At this scale, one can expect the interface film to rupture allowing the void to escape.

When the solution is completed, the bubble is observed to rupture into the free surface; however, we remark that this rupturing is not physical, but due to numerical artifacts. Since the mesh of this problem is on micron scale and the coalescence is on nanometer scale as noted by Janssen and Anderson (2011), there exists a mesh dependency issue. To explore this, a mesh refinement study was performed. Results are shown in Figure 5.

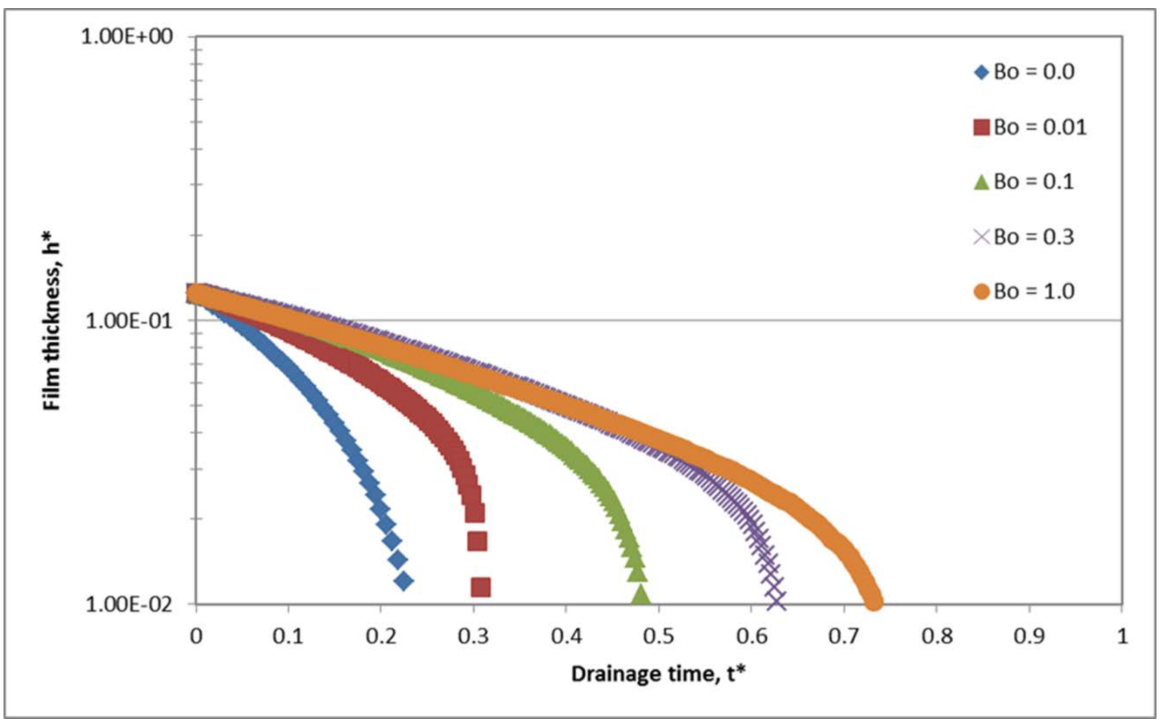

Figure 4. Reduction in interface film thickness as a function of time.

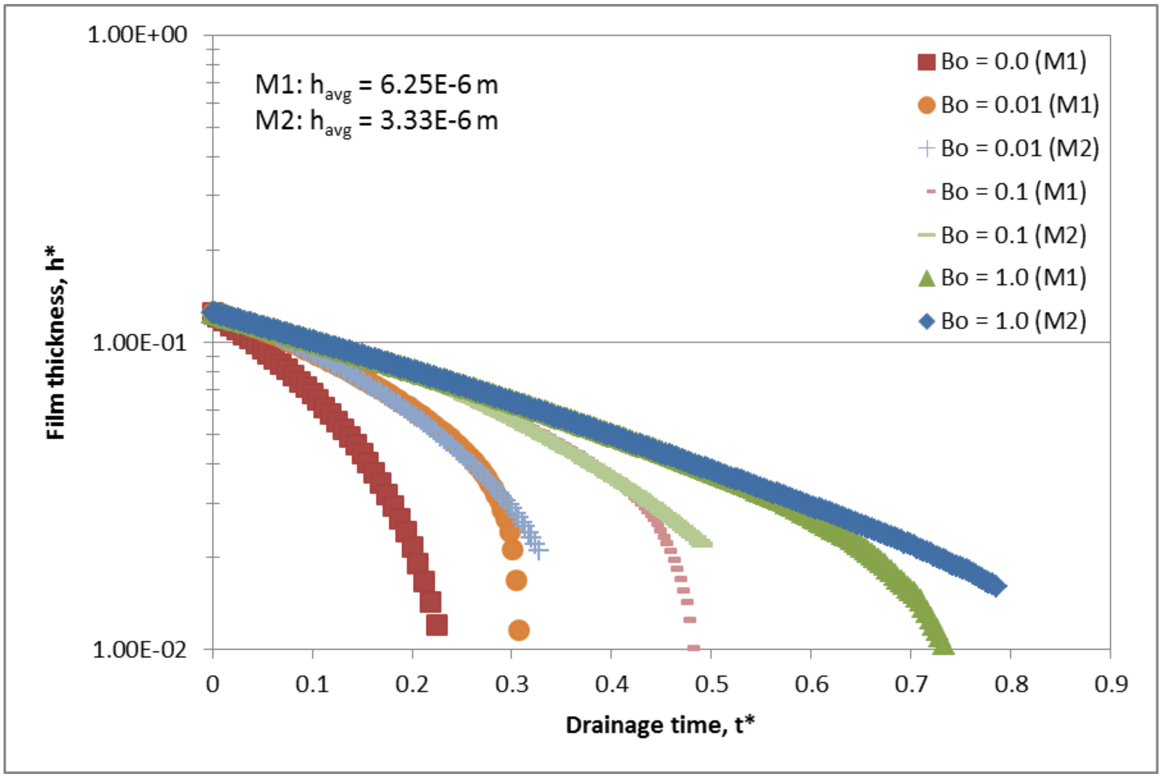

Figure 5. Resin thin film thickness mesh dependence.

In Figure 5, the drainage curves are generated with two different mesh size called M1 and M2. M1 has an average element size of 6.25E-6 [m] and M2 has a smaller average element size of 3.33E-6 [m]. The results show that for both meshes, the linear region representing the exponential drainage of the interface film is formed; however, the rupture times (i.e. where the plots cross the x-axis) are different. The trends from M2 suggest a slower drainage time compared to M1. This is because is the film due to drainage is thinner than the thickness of the film elements. When this happens, the Level Set Method breaks down in accuracy, leading to instability and artificial bubble rupture. Key is that the exponential drainage constant in the linear region can be extracted and one can predict a general time scale for void rupture based on processing conditions.

7. Conclusions

Void dynamics were found to be strongly dependent on void body force and surface tension effects as characterized by Bo. Results suggest that resin film drainage at the interface with the bubble can be successfully modeled as an exponential decay, though film rupture modeling with the level-set method is limited due to mesh dependency issues attributed to the fact that results are suspect once the film becomes thinner than the film element size. Knowledge of film drainage information can provide valuable insight into void removal efficiency. Implications of this work can be applied to many fields where gas bubble migration through viscous fluids is of interest (i.e. oil & gas industry, biomedical engineering, MEMS, etc.).

8. References

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