INTEGRATING THE SIMULATION OF FLOW AND STRESS DEVELOPMENT DURING PROCESSING OF THERMOSET MATRIX COMPOSITES

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SUMMARY

Current finite element based models for the processing of thermoset composites deal with either flow (pre-gelation) or stress development (post-gelation), and there is currently no means of rigorously combining the two. We present a framework to address this issue, with the ultimate goal of predicting the geometry and stress state of the final cured part, whilst including the effect of pre-gelation resin flow on local geometrical features and fibre volume fraction.

Keywords: Composites, Processing, Resin flow, Finite Element Simulation, Residual stress

INTRODUCTION

The common approach to the process modeling of thermoset matrix composites is to divide the analysis into two distinct stages: pre-gelation and post-gelation. During the first stage, flow of resin through the fibre-bed plays a major role in deformation of the part as the viscosity of uncured resin drops significantly due to melting when the temperature is increased. In order to capture this behaviour, a 2-D finite element (FE) model for porous flow-compaction is commonly introduced based on Darcy's law [1,2]. Once the curing of resin begins, it undergoes a gradual evolution from a viscous fluid to a viscoelastic material while developing solid-like properties along the way until the molecules form a cross-linked structure when the resin gels. At this point, resin flow becomes insignificant as the resin matrix has formed a solid network, and the flow simulation terminates.

The second stage of modeling deals with the behaviour after gelation of resin, where the aim is to predict the final geometry and stress state of the composite laminate [3,4]. The properties of the resin are dependent on its degree of cure, and evolve throughout the analysis. Appropriate micromechanics equations are typically utilized to arrive at the moduli of the fibre-reinforced composite at various stages of cure.

The motivation for the current work is to integrate these two stages involved in process modeling in a unified model. This enables one to track and analyse the resin flow behaviour and stress development in the composite seamlessly throughout the curing process. In order to achieve this goal, a numerical model is developed based on a 2-D flow-compaction FE representation. For this purpose, a Q_2P_{-1} finite element with quadratic approximation of kinematic degrees of freedom (system displacement and relative resin velocity) and linear and discontinuous (across neighbouring elements) representation of resin pressure is developed (Figure 1). However, to be able to model the response at the other extreme of the process (i.e. composite behaviour after gelation of the resin) using the proposed element, a generalized definition for the effective stress of the fibre-bed must be introduced so that it includes the elastic contributions of the resin in shear. The moduli related to this generalized form of fibre-bed stress represent the actual moduli of the fibre-bed when the flow is significant while in the post-gelation regime they represent the moduli of the transversely isotropic composite material. To obtain the properties of the generalized fibre-bed, appropriate micromechanics equations are invoked for the interaction of the fibres with the surrounding resin.

FLOW-COMPACTION IN COMPOSITE PROCESSING

Governing Equations

The governing equations for flow and deformation of porous media involves three distinct equations including the momentum balance equation of the fluid phase, the momentum balance equation of the two-phase system, and the mass conservation equation. In the literature relevant to the modeling of flow phenomena in composites manufacturing, it is very common to assume that the solid and fluid phases are incompressible. The system of governing equations may then be written in the form

$$\begin{cases} \dot{u}_{i,i} + v_{i,i} = 0 \\ -P_{,i} - \mu S_{ij}^{-1} v_j = 0 \\ \sigma'_{ij,j} - P_{,i} = 0 \end{cases} \tag{1}$$

where the effect of body forces on the response is neglected. u, v, and P are the main variables of the above differential equations. u is the displacement of the solid structure which is representative of the displacement field of the system. v is the relative velocity field of the fluid phase, while P represents the pressure of the fluid phase throughout the system, and shear stresses in the fluid phase are assumed to be negligible. μ is the viscosity of the fluid phase, and S is the matrix of permeability of the porous solid structure. σ' represents the effective stress in the porous structure. The usual approach in FE representation of multi-phase media is to substitute the momentum balance equation of the fluid phase into the mass conservation equation [5] that leads to

$$\begin{cases} \dot{u}_{i,i} - \left(S_{ij} P_{,j} / \mu\right)_{,i} = 0 \\ \sigma'_{ij,j} - P_{,i} = 0 \end{cases} \tag{2}$$

As a result, the relative velocity of the fluid is eliminated from the governing equations and the pressure undergoes a second-order differentiation in space. In the present work, we do not neglect the contribution of shear components of stress in the fluid phase to the response of the system. This will help in the generalization of the formulation to model the response of the composite with gelled resin, as the solid resin can carry considerable amounts of shear stress. Tucker & Dessenberger [6] used the volume averaging method to arrive at the governing equations of flow through a stationary fibre-bed. Using the volume averaging method for the case of deformable porous structure, we arrive at a more general form of the governing equations

$$\begin{cases} \dot{u}_{i,i} + v_{i,i} = 0 \\ -\phi P_{,i} + \tau_{m\,ij,j} - \phi\mu S_{ij}^{-1} v_j = 0 \\ \sigma'_{ij,j} + \tau_{m\,ij,j} - P_{,i} = 0 \end{cases} \tag{3}$$

where τ_m represents the shear stress in the fluid phase and ϕ is the volume fraction of the fluid phase or porosity. A 2D plane strain Q_2P_{-1} finite element is developed based on the governing equations in (3). This element is a bi-quadratic isoparametric element with 9 nodes for the system displacement and relative velocity of the fluid phase. As depicted in Figure 1, three internal nodes are assigned to the pressure of the fluid phase, therefore enabling every element to represent its internal pressure distribution as a linear surface. For simplicity, we will refer to this element as the 9-3 element.

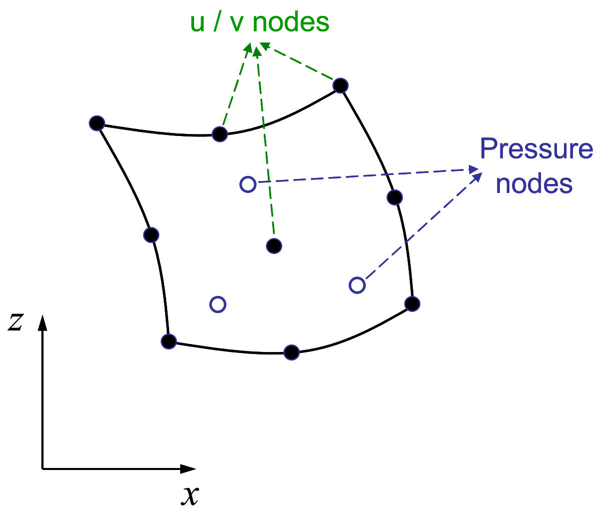

Figure 1, Schematic representation of the proposed finite element.

Compaction of unidirectional angle laminates

Hubert et al. [1] performed simulations involving resin flow and compaction of angle laminates. Their FE model involved a bilinear 2D element developed based on Eqs. (2) with 4 nodes for displacement and pressure degrees of freedom (DOFs), i.e. a 4-4 element. They carried out a comprehensive parametric study of the numerical response and studied the effect of various constitutive properties on the compaction behaviour of the laminates. Hubert and Poursartip [7] also performed an experimental investigation on the compaction of unidirectional angle laminates on convex and concave tools. The samples were made of two different materials including AS4/3501-6 and AS4/8552. The numerical simulations were conducted on examples that closely represented the geometry of the actual specimens used in the experiments. Here, the 9-3 element is used to model the same problem with the same material properties assumed in Hubert's work to compare the results from the two approaches. Figure 2 shows the geometry and boundary conditions of the problem on a convex tool. Two different FE meshes are presented in Figure 3 to study the convergence of the predicted response. The two points where the normal displacements of the laminate are compared (at the corner and at the mid-point of the flat section) are shown in the figure. Table 1 presents the properties of the resin and the fibre-bed for AS4/8552 unidirectional laminate. These values are adopted from references [1] and [7].

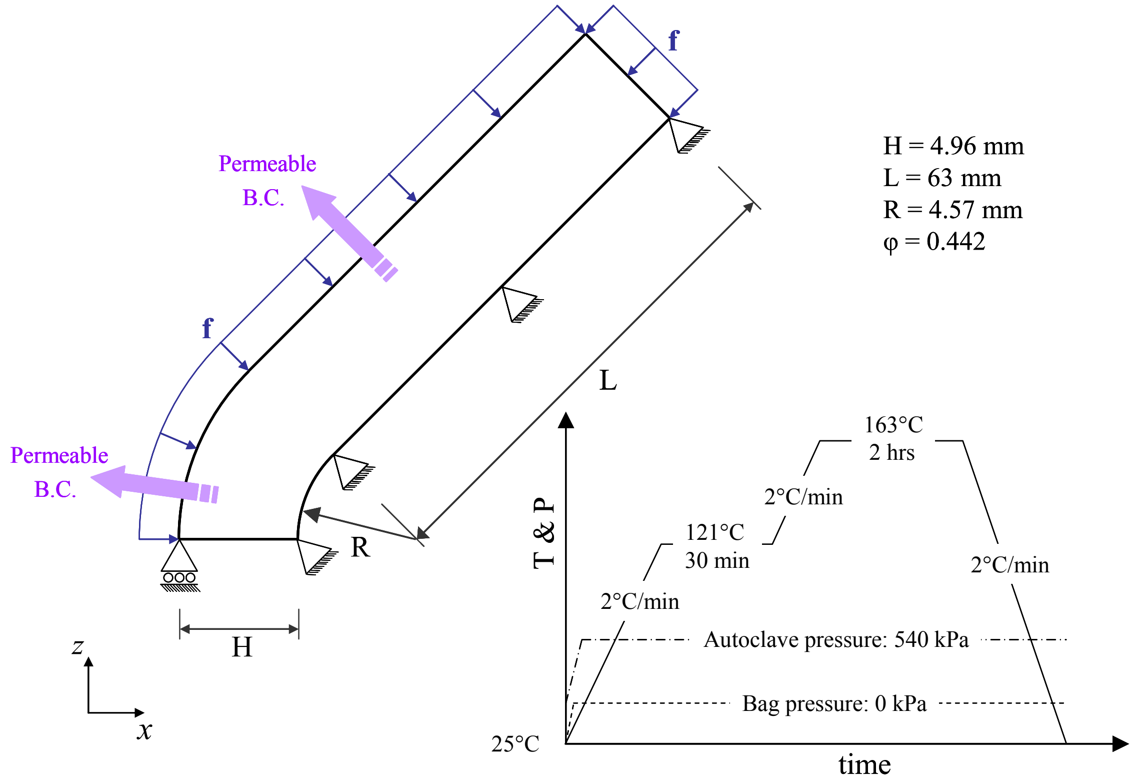

Figure 2, Geometry, BC, and processing cycle used for compaction of a unidirectional angle laminate on a convex tool

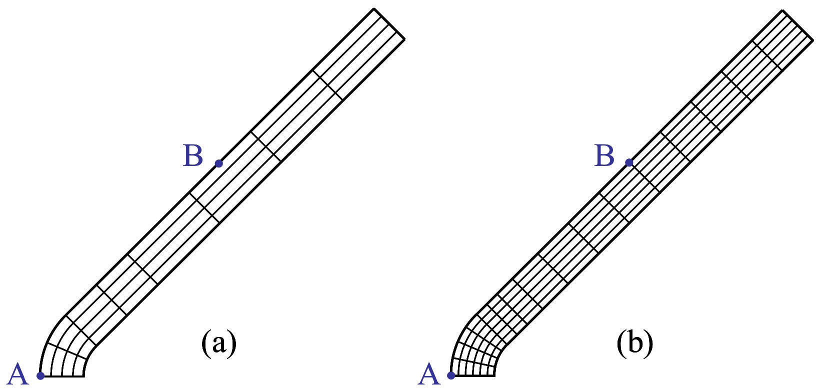

Figure 3, (a) 8×4 mesh, and (b) 16×6 mesh of the angle laminate (half-geometry)

Table 1, Resin and fibre-bed properties for the AS4/8552 angle laminate

Resin degree of cure	Resin viscosity	Fibre-bed permeability	Fibre-bed elastic properties
$\frac{d\alpha}{dt}=\frac{Ae^{-E_a/RT}\alpha^m(1-\alpha)^n}{1+e^{C(\alpha-(\alpha_{C0}+\alpha_{CT}T))}}$ $E_a = 66500\ J/mol$ $A = 1.53\times10^5 s^{-1}$ $m = 0.813$ $n = 2.74$ $C = 43.1$ $\alpha_{C0} = 5.48\times10^{-3}$ $\alpha_{CT} = -0.19/°C$	$\mu = A_\mu \exp(E_\mu/RT)\left(\frac{\alpha_g}{\alpha_g-\alpha}\right)^{A+B\alpha}$ $A_\mu = 3.45\times10^{-10}\ Pa.s$ $E_\mu = 76536\ J/mol$ $A = 3.8$ $B = 2.5$ $\alpha_g = 0.47$	$S_1=\frac{r_f^{\,2}}{4k}\frac{(1-V_f)^3}{V_f^{\,2}}$ $S_2=\frac{r_f^{\,2}}{4k'}\frac{\left(\sqrt{V'_a/V_f}-1\right)^3}{(V'_a/V_f-1)}$ $V_f = 1-\phi$ $V'_a = 0.68$ $k' = 0.2$ $r_f = 4\mu m$	$\underline{\underline{D}} = \begin{bmatrix} 100\ GPa & 0 & 0 \\ & E_3(\varepsilon_3) & 0 \\ Symm\ . & & 0.5\,MPa \end{bmatrix}$ $E_3 = \begin{cases} 2.08MPa & -0.035 < \varepsilon_3 \le 0 \\ 8.65MPa & -0.079 < \varepsilon_3 \le -0.035 \\ 22MPa & -0.1 < \varepsilon_3 \le -0.079 \\ 62.1MPa & \varepsilon_3 \le -0.1 \\ 6.6MPa & \varepsilon_3 > 0 \end{cases}$

The normal displacements at the corner (point A) and the mid-point of the flat section (point B) of the laminate are compared to those obtained by Hubert et al {1,7]. Figure 4 shows the time-history of the normal displacement at the top surface of the AS4/8552 [0] unidirectional angle laminate on a convex tool predicted by the two approaches. A very good correlation is obtained between the current predictions and those obtained by Hubert et al for the displacements at both locations thus verifying the capability of the proposed element to model flow-compaction phenomena in composites processing involving practical geometries.

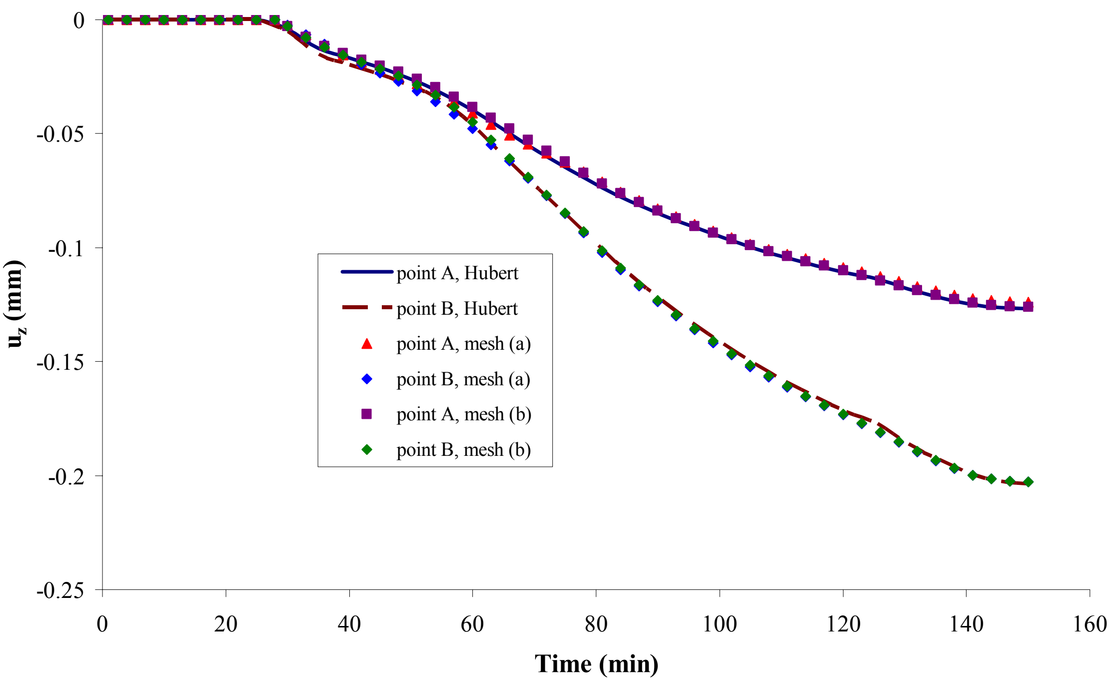

Figure 4, Time-histories of normal displacements at two different locations in a unidirectional AS4/8552 angle laminate with [0°] fibres on a convex tool (Figs. 2, 3)

MODELING THE RESPONSE OF CURED COMPOSITE

An important aspect of this work is to implement the modifications required in the two-phase element so that it can model the different stages of the cure of the material from a fluid resin to a completely cured polymer matrix. As verified earlier, it is naturally expected of the element to successfully model the initial stages of the process consisting of resin flow and compaction of the sample. For the purpose of simplicity, the formulation presented in this section is based on the assumption that the composite material is isotropic. However, the approach is also applicable to the case of transversely isotropic materials.

In order for the two-phase element to successfully model the cured composite, a few critical implementations are required. The assumption of incompressible phases is not relevant to the analysis of cured composites as materials as a cured resin has a bulk modulus that is in the same order of magnitude as its shear modulus. Therefore, compressibility of the phases needs to be considered in the governing equations implemented in the finite element scheme. The common approach in the FE analysis of flow and deformation in porous media toward accounting for compressibility is to obtain the volumetric strain rates of each phase and substitute the summation in the mass conservation equation. For instance, Lewis & Schrefler [5] present the following equation as the mass conservation of a porous system with compressible fluid and solid grains

$$b\dot{u}_{i,i} + v_{i,i} + \left(\frac{b-\phi}{K_f} + \frac{\phi}{K_m} \right) \frac{\partial P}{\partial t} = 0 \tag{4}$$

to be substituted as the first equation of (1) or (3). K_m, and K_f are the bulk moduli of the fluid phase and solid grains (fibres) respectively. b is the biot coefficient defined by

$$b = 1 - \frac{K_{fb}}{K_f} \tag{5}$$

K_{fb} is the bulk modulus of the porous solid structure (fibre-bed in our case). Here, for the method to be consistent with the analysis of solid composite materials, the bulk modulus of the system is defined using the same micromechanics approach taken for the cured composite. The total volumetric strain rate is then obtained using the system's bulk modulus and substituted into the mass conservation equation to have

$$\dot{u}_{i,i} + v_{i,i} - \dot{\varepsilon}_{vs} = 0 \tag{6}$$

The total volumetric strain rate may be written as

$$\dot{\varepsilon}_{vs} = \frac{1}{K_c} \frac{\partial}{\partial t} \left(\frac{tr\, \boldsymbol{\sigma}_s}{\delta_{ii}} \right) \tag{7}$$

where $\boldsymbol{\sigma}_S$ is the total stress of the two-phase system, and K_c is the bulk modulus of the system obtained from the micromechanics formulation of choice for the moduli of the cured composite. As a result of the above modifications in mass conservation equation, a small error is introduced in the flow-compaction response of the system. However, the error is introduced only in the component pertaining to the change of volume of the components. This has a very small and typically neglected effect in the total flow-compaction of porous systems, and therefore the discrepancy is deemed to be negligible.

With the assumption of compressibility, the governing equations in (3) are slightly modified and rewritten in the form of

$$\begin{cases} \dot{u}_{i,i} + v_{i,i} - \dot{\varepsilon}_{vs} = 0 \\ -\phi P_{,i} + \tau_{m\,ij,j} - \phi\mu S_{ij}^{-1} v_j = 0 \\ \sigma''_{ij,j} + \tau_{m\,ij,j} - bP_{,i} = 0 \end{cases} \tag{8}$$

where $\tau_{m\,ij}$ are the components of the deviatoric stress tensor in the matrix phase, and σ''_{ij} represent the components of the Biot effective stress tensor of the porous structure. In the case of resin flow in the processing of composite materials, the term involving the deviatoric stress of the matrix is negligible compared to last term of the second equation in (8) which represents the Darcy interaction force between the two phases. Based on the total equilibrium equation in the 3rd equation of (8) the total stress of the two-phase system may be defined as

$$\sigma_{s\,ij} = \sigma''_{ij} + \tau_{m\,ij} - bP\delta_{ij} \tag{9}$$

Let us define the “s-p” stress (stress of the system excluding the pressure of the matrix phase) as

$$\sigma_{s-p\,ij} = \sigma_{s\,ij} + bP\delta_{ij} = \sigma''_{ij} + \tau_{m\,ij} \tag{10}$$

Combining the deviatoric components of matrix stress with the stress components of the porous structures paves the way toward a formulation for the porous two-phase system that is consistent with the micromechanics representation of choice for the eventually solid composite material. Considering the above points, we may rewrite (8) as

$$\begin{cases} \dot{u}_{i,i} + v_{i,i} - \dot{\varepsilon}_{vs} = 0 \\ -P_{,i} - \mu S_{ij}^{-1} v_j = 0 \\ \sigma_{s-p\,ij,j} - bP_{,i} = 0 \end{cases} \tag{11}$$

The modulus pertaining to the bulk behaviour of the “s-p” system is set to be equal to the modulus of the fibre-bed throughout the whole process modeling. The shear modulus of the “s-p” system is obtained by an additive combination of the shear modulus of the porous structure (pertaining to σ''_{ij}) with the shear modulus of the composite material (obtained from the micromechanics equation of choice).

$$\begin{aligned} K_{s-p} &= K_{fb} \\ G_{s-p} &= G_{fb} + G_c \end{aligned} \tag{12}$$

The above definitions of moduli are consistent with the second definition of the "s-p" stress in (10). When the resin can be considered a viscous fluid, the "s-p" shear modulus is practically equal to that of the fibre-bed as a negligible value for the shear modulus of the resin leads to a negligible value for the shear modulus of the composite (as the load-sharing rule is of a parallel nature, the smaller shear modulus dominates the shear behaviour of the composite). For the case of cured resin, the shear modulus of resin is quite large and therefore the shear modulus obtained from the micromechanics of the composite dominates the value of "s-p" modulus. The bulk behaviour of the system is correctly represented by the mass conservation equation throughout the analysis.

The 9-3 element developed in this work is based on the governing equations in (11) that may be characterized as a u-v-p formulation. According to the very common u-p approach in FE treatment of porous media, the governing equations in (11) may be written as

$$\begin{cases} \dot{u}_{i,i} - \left(S_{ij} P_{,j} / \mu\right)_{,i} - \dot{\varepsilon}_{vs} = 0 \\ \sigma_{s-p\, ij,j} - bP_{,i} = 0 \end{cases} \tag{13}$$

after substituting the fluid phase equilibrium equation into the mass conservation equation. A 9-4 element based on the above u-p formulation is also developed in this work for the sake of comparison. This element is geometrically very similar to 9-3 element with 9 nodes for displacement degrees of freedom and 4 corner nodes for the fluid pressure. The 9-4 element should be considered an element very close to Taylor-Hood element (that could be named 8-4 by the naming system of this work) which is extensively used in FE modeling of multi-phase systems and also incompressible fluids. In the u-p formulation pressure undergoes second order differentiation in space, and therefore needs to be introduced as an essential BC of the system at the boundaries. In the u-v-p formulation, there is no need to specify pressure degrees of freedom at the boundary of the system, and any applied pressure is considered in the traction vectors applied to the system.

Rectangular sample under pressure gradient

Figure 5 depicts a schematic representation of a rectangular elastic solid composite sample under two different magnitudes of normal pressure loading on either side.

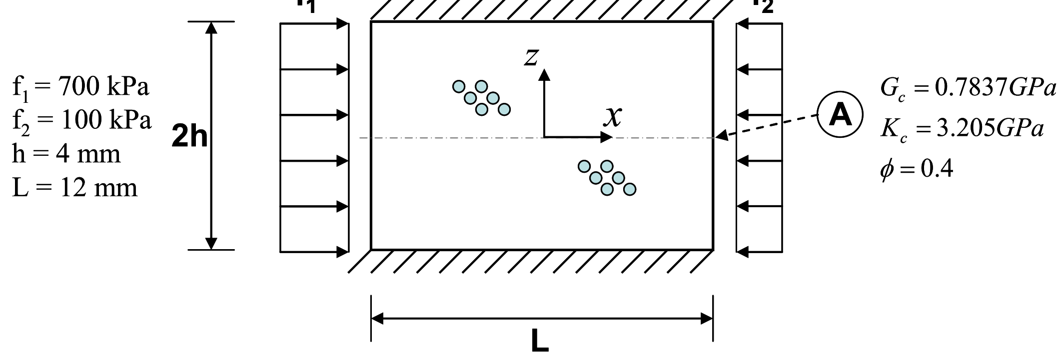

Figure 5, Composite sample under pressure gradient

Assuming isotropic properties, a mesh of 9-3 elements is used to model half of the sample due to symmetry, and the displacement profile of the right-hand-side of sample is presented in **Error! Reference source not found.**. The ratio of the bulk modulus of fibre-bed to the bulk modulus of composite is assumed to be 0.01. The viscosity of the resin is assumed to be a high value at 7×10^{7} Pa.s to reduce the flow of resin to negligible values in the time scale of interest, and the permeability of the fibre-bed is set to 1×10^{-14} m^{2} (a value in the range of typical permeability values for thermosetting composites. The 9-4 element based on the u-p formulation is also used to predict the response of the system under permeable and impermeable boundary conditions on the sides. **Error! Reference source not found.** also presents the response of the 9-4 element under the two different B.C. which shows a good agreement to 9-3 element under impermeable B.C. but is not a good match under assumed permeable conditions.

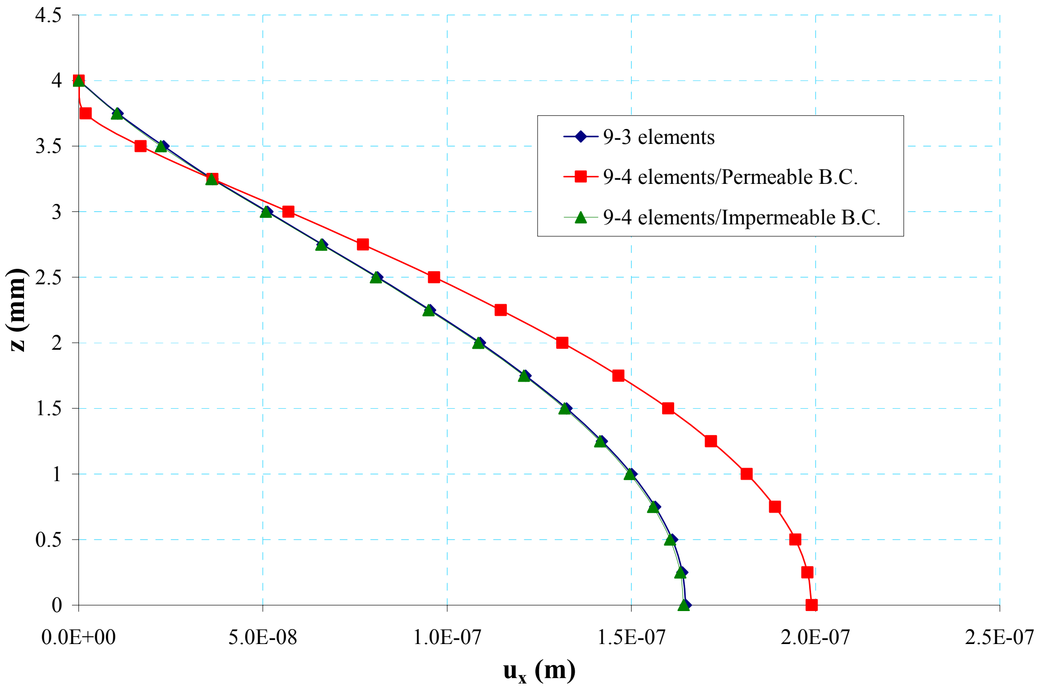

Figure 6, Comparison of displacement profiles for 9-3 and 9-4 elements, obtained using a 12×8 mesh

The problem was also modeled in ABAQUS by an 8-noded solid 2D element based on a plane strain formulation. Various meshes from 6×4 to 192×128 were modeled, and the horizontal displacement of the system at point A was obtained. The values converge to 1.649×10^{-7} m. In the case of using 12×8 9-3 elements, the obtained displacement for point A is 1.648×10^{-7} m, proving the capability of this approach in the prediction of the elastic response of the cured material.

In the case of 9-4 elements with permeable BC, pressure DOFs are forced to take pre-determined values and that leads to a considerable error in the displacement response of the system. It is evident that for the case of 9-4 elements, the displacement response of the system is dependent on the assumed BC. It is very desirable for the response of the solid and cured composite to be independent of the BC considered for the flow of resin during the initial stages of cure. Therefore, the 9-3 element has a clear advantage over the 9-4 element in this regard.

CONCLUSIONS

A 9-3 element based on a u-v-p formulation is developed and verified for the purpose of modeling the flow-compaction response of the thermosetting composite laminates during autoclave processing. An approach is presented to model the stress response of cured isotropic composites using the developed element toward the ultimate goal of integration of modeling the processing of thermosetting composites from the flow-compaction response through to the stress response in a seamless fashion. Through an example, the capability of the proposed two-phase element in the prediction of the elastic response of a cured isotropic composite sample is demonstrated.

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