A Two-Phase Integrated Flow-Stress Process Model for Polymeric Composite Materials

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ABSTRACT

Process modelling of polymeric composite materials is usually carried out in a numerical framework consisting of several independent sub-models. Using this sequential approach does not capture the interplay between different phenomena involved during processing despite the fact that they occur concurrently. In this paper, a two-phase integrated flow-stress model is developed in order to overcome the drawbacks of the decoupled approach. Porous flow and stress development formulations are integrated into a unified modelling framework that captures both effects in a simultaneous and seamless fashion. In this new methodology, no restriction is imposed on the compressibility of the two phases in keeping with an ongoing effort at UBC to extend the model to account for the presence of all three-phases, including gas, liquid, and solid. The governing equations are developed for the general case when the properties of the resin phase evolve during curing from an unsolidified fluid to a fully solid material. The consistency of the constitutive equations is maintained at the two extremes of the curing process, namely, the early stages of the process when the fluid phase behaves as a liquid and the final stage corresponding to a cured and fully solid composite material. Adopting a u-v-P (solid displacement-fluid velocity-pressure) finite element formulation in a 2D plane strain finite element code developed in MATLAB, the governing equations are solved using an implicit time integration scheme that considers material and geometric nonlinearities. Case studies are presented to evaluate the performance of the code during the whole curing process as well as at the two ends of the material behaviour regime. Comparisons are made with the predicted results from the decoupled stress model alone in order to show the significance of using the integrated model to capture the temporal and spatial variation of resin flow on the process outcome.

1 INTRODUCTION

Predicting the behaviour of fibre-reinforced polymeric composite materials through an accurate process modelling is highly desirable due to the high production cost and substantial risk involved in their manufacturing. A precise model that considers various and evolving properties of composite materials' constituents during their manufacturing is invaluable. To achieve the desired outcome, a process model needs to take into account all main phenomena involved in the processing, such as resin flow, stress developments, chemical hardening, etc.,. However, complicated interconnection of these diverse phenomena makes the development of such models very challenging. Moreover, a proper process model has to consider all such complexity in an efficient manner in order to be utilized in modelling the manufacturing process of large-scale composite structures.

Composite materials processing is usually modelled using different independent sub-modules in a so-called "integrated sub-model" framework [1-5]. Several modules such as cure kinetics, thermal and heat transfer analysis, resin flow, stress development, etc., are carried out sequentially. In this approach, the outcome at the end of each module, such as the resulting deformed geometry and volume fraction of constituents, is used as input for the subsequent modules while the interaction between the various phenomena is not captured. Hubert et al. [5] developed a flow model for processing of composite materials at early stages of processing when the resin behaves in a liquid-like manner. In this flow model the constituents of the composite material were assumed to be incompressible. Johnston et al. [4,6] focused on the stress development module. Based on these flow and stress models, a multi-physics 2D plane strain finite element code, called COMPRO, was developed for process modelling of polymeric composites. Later, Arafath [7] introduced the COMPRO Component Architecture (CCA) in the commercial finite element software, ABAQUS. In the sub-model approach, the overlap between the resin flow and stress build-up is ignored. A major drawback of this approach is that it cannot consider the possibility of resin gelation occurring at various spatial locations in the composite structure at different instants of time.

In this work, an Integrated Flow-Stress (IFS) model for processing of composite materials is presented. To illustrate the main concepts of the integrated approach and in order to simplify the formulation, the composite material is assumed to be isotropic in the two-dimensional plane of interest, therefore making it applicable to the transverse plane of a unidirectional laminate. Considering the Biot's effective stress formulation [8,9] for early stages of processing when the resin behaves as a liquid as well as using classical solid mechanics for the other extreme when the resin is fully solidified, the composite processing can be modelled in a seamless fashion with no restriction placed on the compressibility of its constituents. The capability of the developed framework on processing of a composite material undergoing curing is investigated and it is demonstrated that the model can cope just as easily with the early stages of the process as it does with the final stages when the composite system behaves as a solid.

2 METHODOLOGY

2.1 Governing Equations

The two-phase system considers a compressible fluid phase (which can solidify during the process) flowing through a porous medium (fibre-bed) referred to as the solid skeleton. Three conservation equations govern the response of the system at all stages of the process whether the solidifying fluid phase behaves as an unsolidified fluid (liquid-like behaviour) or when it solidifies to form a homogenized solid composite material. They include: *i*) mass conservation of the system, *ii*) momentum conservation of the matrix phase, and *iii*) momentum conservation (equilibrium) of the system. These equations can be written as follows:

$$
\begin{aligned}
&\dot{u}_{i,i} + v_{i,i} + \frac{\varphi}{\rho_F}\dot{\rho}_F + \frac{(1-\varphi)}{\rho_S}\dot{\rho}_S = 0 \\
&P_{,i} + \mu\, S_{ij}^{-1}\, v_j = 0 \\
&\sigma_{SK\,ij,j} - b\, P_{,i} = 0
\end{aligned}
\qquad (1)
$$

In the system of equations (1), the dummy indices i and j=1,2,3 denote Cartesian coordinate directions and coma and overdot denote spatial and time derivatives, respectively, ρ is density, and φ is the volume fraction of the fluid phase. The subscripts F, S, and SK denote fluid (e.g. liquid or gas), solid (e.g. solid grains in soil mechanics or fibres in composite materials), solid skeleton (e.g. fibre-bed in this case), respectively. The basic variables of the model are, u the displacement of the system (associated with the solid structure), v the volume-averaged relative velocity of the fluid phase (i.e. flow velocity), and P the fluid pore pressure. Also, in the Darcy's law incorporated in the model as momentum equation of the system, μ is the fluid viscosity and S_{ij} is the permeability tensor of the solid skeleton. The last equation represents equilibrium of the composite system where σ_{SK} is identical to the so-called effective stress of the skeleton when Biot's coefficient b is unity. Biot's coefficient, b, which takes on a value between zero and unity, represents deformability of the porous media and determines the share of pore pressure contributing to the overall load carrying capacity. For isotropic materials, Biot's coefficient is expressed in terms of the bulk moduli of the skeleton and the solid phase, K_{SK} and K_S, as follows:

$$b = 1 - K_{SK}/K_S \tag{2}$$

For effective incorporation of all the diverse properties and behaviours of composite materials during processing into a unified model and accounting for a continuously hardening (solidifying) fluid phase, a new scalar quantity, λ, termed 'solidification factor' is introduced by Niaki et al. [10,11]. This parameter is a measure of the solidification of the fluid phase and varies between zero and unity, $0 \le \lambda \le 1$. The solidification factor could be a function of any state variable used in process modelling, such as degree of cure (χ), temperature (T), viscosity (μ), etc., depending on the type of material, type of process, and so forth. As shown schematically in Figure 1, $\lambda = 0$ corresponds to the unsolidified fluid. The fluid phase (resin or gas, as a special case of fluid), solid phase (fibres), and solid skeleton (fibre-bed) are incorporated in the model according to the Biot's poroelastic model [8,9]. At the other extreme when $\lambda = 1$ corresponds to the fully solidified resin, there is no distinct solid and solid skeleton phases as everything is considered homogenized (smeared) into a continuum solid composite material. In the intermediate regime (when $0 < \lambda < 1$), the fluid phase is partially solidified and has a combination of fluid- and solid-like behaviour. While the material undergoes curing, the fluid phase/solid grains gradually become more and more part of the composite material (through contributing to the solid skeleton) and less and less a pure fluid/solid. Therefore, the properties of the solid/solid skeleton changes from the fibre/fibre-bed properties at one extreme to the fully homogenized composite material at the other extreme as a function of λ. In a functional form:

$$\mathbf{D}_S = f(\mathbf{D}_f, \mathbf{D}_c, \lambda) \ , \ \mathbf{D}_{SK} = f'(\mathbf{D}_{fb}, \mathbf{D}_c, \lambda) \ , \ \alpha_{SK}^{th} = g(\alpha_{fb}^{th}, \alpha_c^{th}, \lambda) \ , \ \alpha_{SK}^{cs} = g'(\alpha_c^{cs}, \lambda) \tag{3}$$

where subscripts f, c and fb denote fibre, composite, and fibre-bed, respectively, $\mathbf{D}$ is the stiffness matrix of the material, and α_{SK}^{th} and α_{SK}^{cs} are coefficients of thermal expansion/shrinkage and cure shrinkage, respectively. When the fluid phase evolves from an unsolidified liquid-like material to a solidified matrix phase forming a solid composite, it can be shown that [10]:

$$\begin{aligned} -\frac{\varphi}{\rho_F}\dot{\rho}_F - \frac{(1-\varphi)}{\rho_S}\dot{\rho}_S & \\ = (1-b)\dot{u}_{i,i} - \left(\frac{\varphi}{K_F} + \frac{b-\varphi}{K_S}\right)\dot{P} + \varphi(1-\lambda)\delta_{ij}\dot{\varepsilon}^f_{F\,ij} + (1-\varphi)(1-\lambda)\delta_{ij}\dot{\varepsilon}^f_{S\,ij} & \\ + \frac{1}{3K_S}\delta_{ij}D_{SK\,ijkl}\dot{\varepsilon}^f_{SK\,kl} & \end{aligned} \tag{4}$$

where the superscript f is used to denote the free component of strain, ε, or strain rate quantity to which it is attached, D_{ijkl} is stiffness tensor, and δ_{ij} is the Kronecker delta (or identity tensor). In a solidifying material as a result of variation of material properties due to solidification as described in Eq. (2) (i.e. changes in both K_{SK} and K_S), the magnitude of $\dot{b}$ can be significant. Conceptually, as the resin material undergoes solidification and hence contributes to load (stress) carrying capacity of the system, a portion of the pressure ($\dot{b}\,P$) in the unsolidified fluid should be incrementally added to the stress carried by the solid skeleton. This additional stress can be regarded as a residual stress induced in the material due to transformation of the pressure bearing fluid into a homogenized solid composite material. In this regard, $\dot{\sigma}_{SK}$ is related to the skeleton strain through its elastic constitutive stiffness tensor, as follows:

$$\dot{\sigma}_{SK_{ij}} = D_{SK_{ijkl}} \dot{\varepsilon}^{\sigma}_{SK_{kl}} + \dot{b}\, P \delta_{ij} \tag{5}$$

where $\dot{\varepsilon}^{\sigma}_{SK}$ is the mechanical strain rate of the skeleton. In contrast to the sequential sub-model approach, the proposed methodology allows for the transfer of the pressure carried by the liquid resin to the solid composite material in a seamless fashion.

2.2 Finite Element Implementation

The governing differential equations of the system are converted into their weak form using the Galerkin method. The system displacement vector, u_i, relative velocity vector of the fluid phase, v_i, and fluid pore pressure, P, are discretized spatially within a low order bi-linear isoparametric element with 4 corner nodes for the system displacements and relative velocities of the fluid phase, $\bar{\mathbf{u}}$ and $\bar{\mathbf{v}}$, respectively, as degrees of freedom and only one internal central node assigned to the fluid pressure $\bar{\mathbf{P}}$ (the so-called 4-1 element as shown in Figure 2). An incremental form of a system of algebraic equations is obtained using the backward Euler scheme for temporal integration combined with the Newton's nonlinear solution method. The details of the iterative solution procedure and all matrix and vector quantities can be found in [10].

3 NUMERICAL EXAMPLE

The FE matrix equations have been coded into a MATLAB software developed in-house. The composite material is assumed to be a unidirectional laminate with fibre orientations perpendicular to the domain of interest (2D plane strain condition). In this example, a composite system undergoes curing under different boundary conditions shown in Figure 3. The fibres and resin are taken to be AS4 carbon fibre and 3501-6 epoxy resin with initial volume fraction of $\varphi_0 = 0.42$ with properties presented in [12,13]. The applied temperature cycle is also shown in Figure 4 along with the predicted viscosity (μ) and degree of cure (χ) where the cure kinetics and viscosity models for 3501-6 resin are adopted from Lee et al. [14] and Hubert et al. [5], respectively. The evolution of the solidification factor (λ) is also shown in the figure. As mentioned previously, different functions can be used for expressing the solidification factor in terms of the state variables as well as solid and solid skeleton properties. The solidification factor is assumed to be a piece-wise linear function of the degree of cure as follows:

$$\begin{aligned} &\lambda = 0 && \text{for } \chi < \chi_a \\ &\lambda = \frac{\chi - \chi_a}{\chi_b - \chi_a} && \text{for } \chi_a \le \chi \le \chi_b \\ &\lambda = 1 && \text{for } \chi_b < \chi \end{aligned} \tag{6}$$

The demarcation points χ_a and χ_b are chosen to be 0.4 and 0.6 in this example. The solid and solid-skeleton properties are also assumed to be linearly dependent on the solidification factor:

$$\begin{aligned} K_S &= K_f + (K_c - K_f)\lambda \\ K_{SK} &= K_{fb} + (K_c - K_{fb})\lambda \\ G_{SK} &= G_{fb} + (G_c - G_{fb})\lambda \\ \alpha^{th}_{SK} &= \alpha^{th}_{fb} + (\alpha^{th}_c - \alpha^{th}_{fb})\lambda \\ \alpha^{cs}_{SK} &= \alpha^{cs}_c \lambda \end{aligned} \tag{7}$$

The permeability and fibre-bed elasticity models data are summarized in [10] which are taken from [15] and [5,16], respectively, where constants are adopted from Hubert [17]. The mechanical properties of the composite material, K_c, G_c, α_c^{th} and α_c^{cs}, are calculated during the simulation using a micromechanics model adopted by Johnston et al. [6] and Bogetti and Gillespie [3]. For this example, the fluid phase follows the pseudo-viscoelastic (PVE) formulation [18] with relaxation time and weight factors taken from [19].

The results of these IFS simulations are presented in Figure 5 along with results from the stress model (developed by Zobeiry et al. [18]). The evolution of resin volume fraction using the IFS model is shown in Figure 5(a) . The stress model has no means of capturing the continuous changes in volume fraction of the resin for all boundary conditions considered which stems from ignoring the resin flow in such an approach. As expected for stresses shown in Figure 5(b), the stresses obtained from both the IFS and stress models are in perfect agreement for the impermeable boundary conditions (red lines in Figure 5(b)). For permeable and constrained boundary condition (blue lines), at the early stages of the process, the resin flows out of the system thus relieving some of the developed pressure as it undergoes expansion due to temperature rise. While the resin is still in its pre-gelation state and flows out, no stresses develop in the system. This behaviour is well captured by the IFS model. Stresses only start to develop after gelation of the resin. This observation explains the notable difference in the final predictions of residual stresses in the system between the IFS model and the stress model for permeable top surface. However, for the permeable top surface the stress model yields the same results as in the impermeable case which further highlights the errors involved when flow is ignored in the process simulation. For the vertical strains under the free top surface condition (Figure 5(c)), again the stress model erroneously predicts the same strains for both the permeable and impermeable top surface conditions. In contrast, the IFS model predicts a considerably different value of strain for the case of permeable top surface.

4 CONCLUSION

A methodology is briefly presented in this paper to integrate the simulation of resin flow and stress development into a unified modelling framework for processing of composite materials. The model is developed for a two-phase material system comprising of solid material (fibres) forming a solid skeleton phase (fibre-bed) and a fluid phase (e.g. resin). The governing equations are developed for the general case when the resin phase evolves during curing from an unsolidified fluid to a fully solid material while maintaining the consistency of the constitutive formulations with those used for each of the two extremes. Adopting a u-v-P finite element formulation, the governing equations are implemented in a MATLAB code and solved using an implicit solution method that considers both material and geometric nonlinearities. The performance of the code is evaluated through a numerical case study. The predictive capability of the integrated flow-stress (IFS) model and the significance of considering both the flow and stress build-up simultaneously on the final deformation and state of stress in the composite material was demonstrated through comparison to the stress model. Detailed analysis of the predicted results shows the potential of the current two-phase IFS model to be extended to the more general IFS model applicable to composite materials with more general orthotropic properties.

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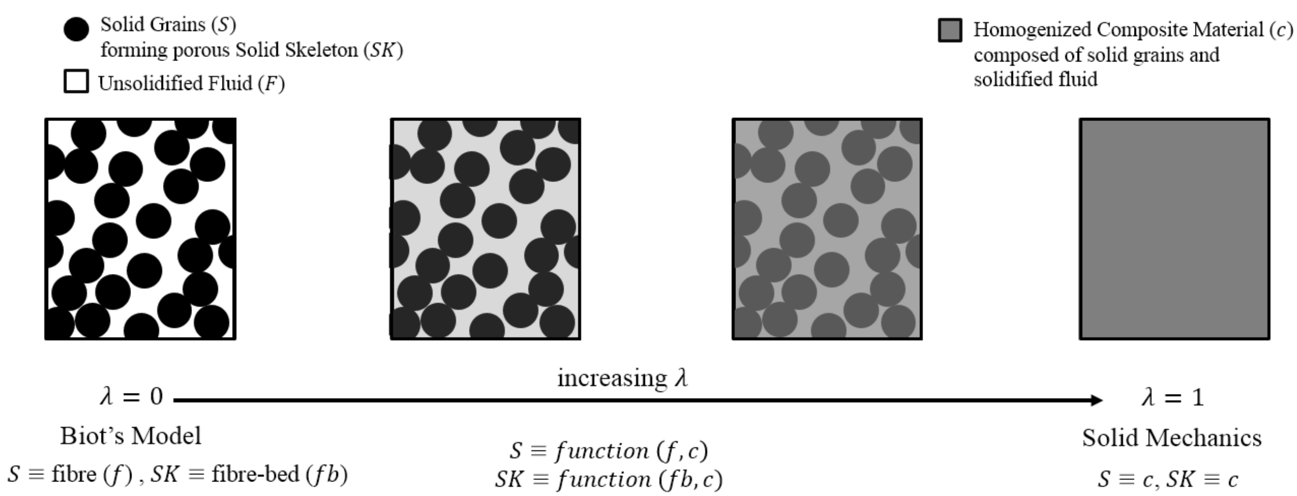

Figure 1. Evolution of the properties of the solid/solid skeleton and fluid phases with increasing solidity of the fluid phase

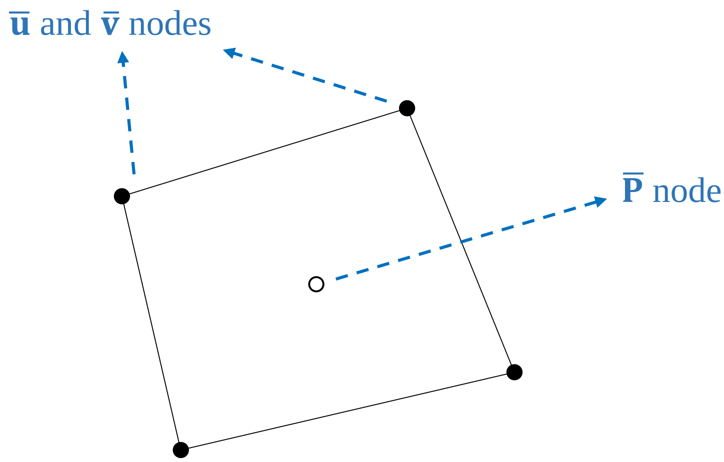

Figure 2. Schematic representation of 4-1 element

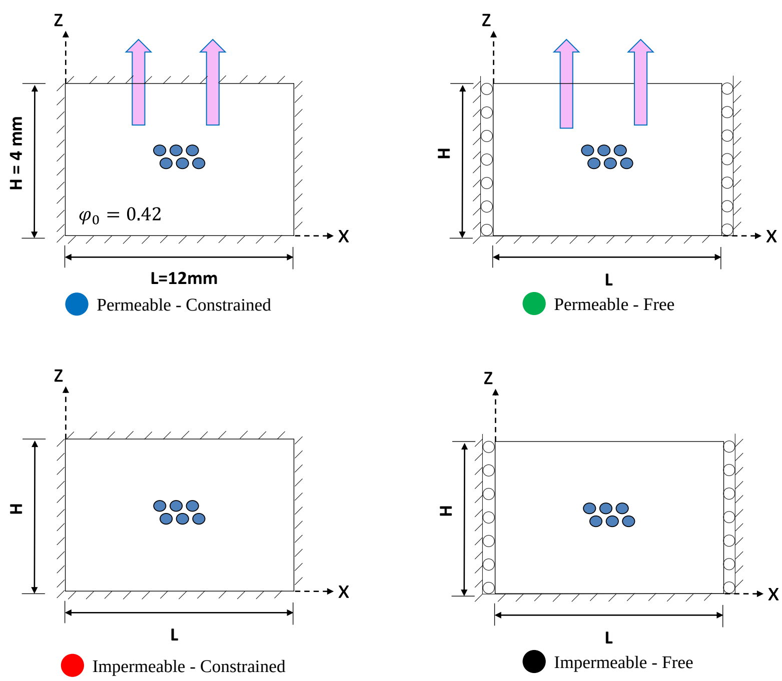

Figure 3. Geometry and different boundary conditions of sample undergoing curing

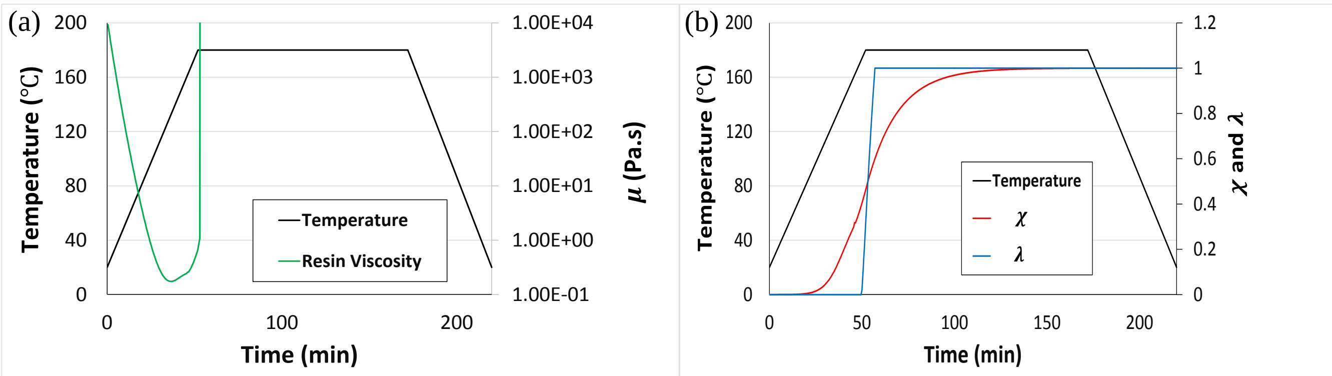

Figure 4. (a) time-history of applied temperature and (b) predicted viscosity, degree of cure, and solidification factor of material

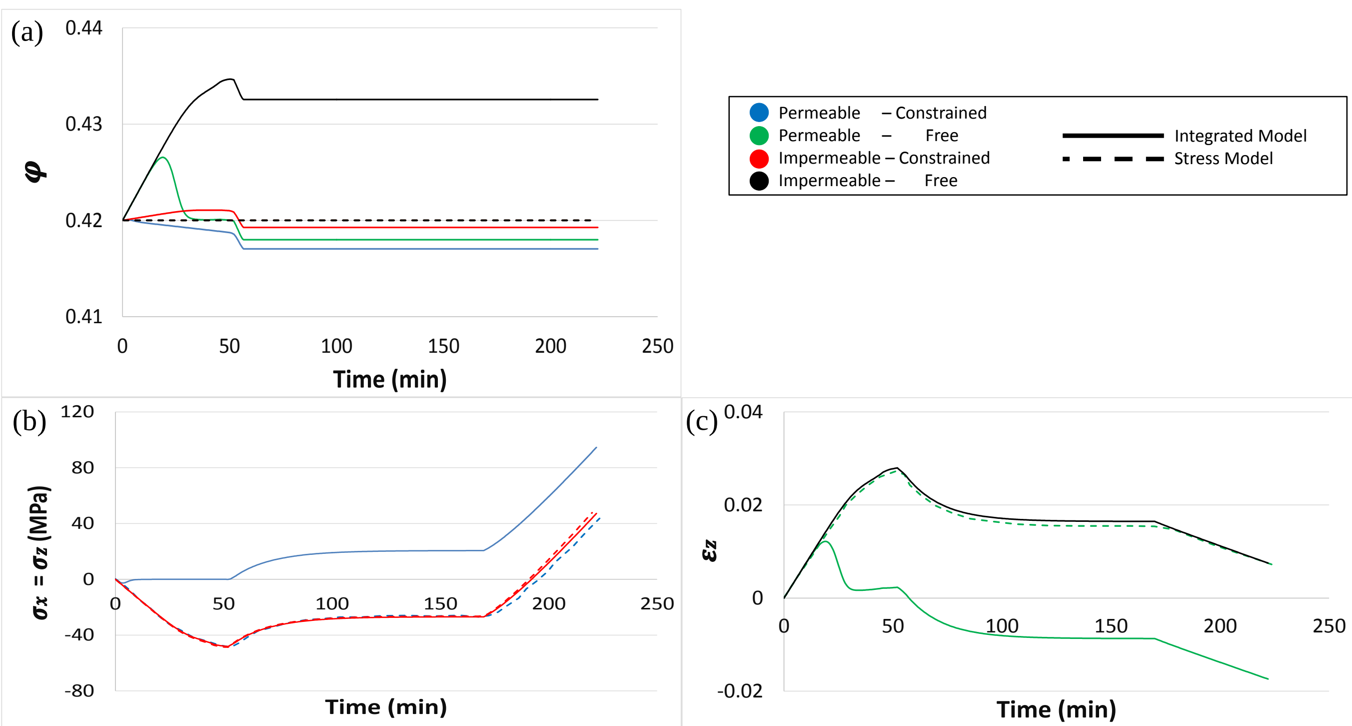

Figure 5. Predicted time-histories of the: (a) resin volume fraction, (b) stresses, and (c) total strain in the vertical direction ε_z