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On the physical relevance of power law-based equations to describe the compaction behaviour of resin infused fibrous materials

Jonathan P.-H. Belnoue*, Mario A. Valverde, Maria Onoufriou, Xiaochuan (Ric) Sun, Dmitry S. Ivanov, Stephen R. Hallett

Bristol Composite Institute (ACCIS), University of Bristol, BSB 1TR, Bristol, United Kingdom

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ABSTRACT

The mechanical performance of structural parts (e.g. a 20 m+ aircraft wing) made from pre-impregnated (prepreg) fibre reinforced polymers (FRP) are intimately linked to the material's meso-scale (sub-mm length) and micro-scale (μm length), which in turns depends upon the way the fibre mat impregnated with resin has been processed. Particularly important is the way the resin flows through the fibre bed, as that influences the level of porosity and the orientation of the fibres in the final part by controlling the stack's change of thickness during manufacture. In this paper, the applicability of a recently proposed phenomenological model, able to predict the thickness evolution of a wide class of prepreg stacks under processing conditions, is investigated. One of the model's main advantages is that, unlike other flow-compaction models, it can capture the transition between squeezing and bleeding flow. The model's significance for a variety of reinforcements (i.e. UD carbon, UD glass and carbon fibre plain weave) and resin systems (i.e. first and second generation of toughened thermoset and thermoplastic resins) is demonstrated. A new physics-based interpretation of the material parameters is proposed. This gives rise to an analytical expression relating the thickness evolution with time to the applied temperature and pressure cycles, the viscosity of the resin, and meso- and micro- geometrical characteristics of the reinforcements. The contribution paves the way towards a better understanding and control of the variability and increased digitalisation of the design and manufacturing processes of composite structures.

1. Introduction

1.1. Motivation

The recent push for reduction in carbon emissions has fuelled an unprecedented demand for fibre reinforced polymers (FRPs). Once limited to small-series and high-end aerospace and Formula 1 parts, composite materials are becoming widely used in fields such as the mainstream automotive, energy and construction sectors. In addition to their well-known high stiffness to weight ratio, FRPs also present the advantage to resist environmental factors such as corrosion and fatigue well and have good thermal and electrical insulation properties. However, the design and manufacture of composite structures has historically been a manual, time-consuming and costly process and the cost-effectiveness and competitiveness of FRP parts is an ongoing issue. There are two main limitations for industry to be able to meet its targeted part number and production rate over the next decade and to continue to grow at the current pace:

1. Greater integration of the manufacturing constraints is needed at the design stage as, too often, composite parts are designed based on structural performance only and then found to be un-manufacturable when sent to the workshop. A tedious and costly process of iterative modifications of both the structural design and the manufacturing method is then triggered in order to find the best compromise between manufacturability and structural performance.
2. A greater shift towards automation is required, as the current process remains very manual, especially as hand layup still allows to reach greater quality for geometrically complex parts [1].

The many different ways a composite can be manufactured (i.e. thermo-stamping, thermoforming, resin transfer moulding (RTM), vacuum assisted resin transfer moulding (VARTM), autoclave curing, vacuum-bag-only processes, etc.) cover a large range of temperature (i.e. room temperature to above 400 °C) and applied loading rate (i.e. instantaneous to 0.01 bars per second) [2] and there is also a large variety of resins (i.e. thermosets, thermoplastics, etc.) and fibres types (i.e. carbon, glass, etc.) and internal architecture (unidirectional, non-woven, and textile - woven, braided, stitched of various patterns, etc.) that can be combined. As a result, simulating the manufacturing of a composite

* Corresponding author.

E-mail address: jonathan.belnoue@bristol.ac.uk (J.P.-H. Belnoue).

is an extremely complex task. This, partly, explains why virtual process chains for composites have only recently started to appear in academia for the forming, followed by infusion of dry textiles [3], prepreg technology [4] and also sheet moulding compound [5]. Because each of the processes described above involve fundamentally different physics, the tools to predict their outcome have, historically, been developed in isolation. This is a further obstacle to the deployment of design for manufacture practices in industry as this makes it very tedious to compare the effect of different manufacturing processes on a part micro- and meso-scale architecture (and hence its structural performance). Further, challenges have been brought by the recent rapid escalation of the use of automated processes such as Automated Tape Laying, Automated Fibre Placement (AFP) or Fibre Patch Placement (FPP) as they use material in a very different format. AFP tapes are typically 6-mm wide whilst prepreg sheets laid-up by hand are typically 300 mm. In those conditions, the assumption made in traditional composites manufacturing simulations that size effects are negligible is often not valid anymore. This was discussed, in the case of thermoplastics, as early as the 1990's by Wang and Gutowski [6]. More recently Nixon-Pearson et al. [7] performed an experimental study that allowed to relate the lateral expansion of toughened thermoset prepreg tapes to the processing conditions and Hautefeuille et al. [8] conducted a similar study where the tape expansion was measured in-situ. Meso-scale modelling of prepreg compaction performed by Sorba et al. [9] allowed deeper understanding of the phenomenon, in particular with respect to the role of the interfaces.

Consolidation is a key mechanism to achieve a high-quality part, regardless of the process involved. It controls the structural properties of the final part as it influences the level of porosity and fibre path defect generation. A comprehensive review of void formation mechanisms in prepreps is provided by Centea et al. [10] and May et al. [11] performed a benchmark exercise of permeability measurements in dry textiles that highlighted the link between the compaction state of the preform and the ease with which it can be impregnated. Farhang and Fernlund [12] more specifically studied the relation between porosity and thickness variation (i.e. compaction) in no-bleed prepreg laminates. A second important aspect of compaction is its link to part thickness conformance and fibre path defect formation. After having originally concentrated in understanding the phenomena at play [13], research on the influence of compaction on part quality has, in recent years, focused on the modelling and predictive aspects. For example, Levy and Hubert [14] proposed an analytical approach that can predict thickness deviation from a targeted value that may arise in geometrically complex parts and Šimáček and Advani [15] developed a numerical approach to predict part thickness variation and porosity levels in corner laminates. Although important contributions, these two approaches are limited by the complexity of parts that they can handle. Recent work [16] from some of the authors of the present contribution, looked more specifically at the link between compaction and fibre path defects and demonstrated that a part's geometric complexity can be tackled within a finite element framework. Very recent work by the same authors [17] allowed to remove issues pertaining to the computational cost of such an approach, allowing simulations to run in tens of minutes rather than weeks, as in the case of the original formulation.

Traditionally, low viscosity systems are modelled using percolation theories (Darcy's law) combined with structural response of interacting fibres [18] whilst when the viscosity of the resin is high, shear flow theories for incompressible materials [19] are used. In percolation (bleeding) flow, the assumption is made that the application of a consolidation pressure on a fibre mat impregnated with a viscous resin results in the expulsion of the resin out of the reinforcements static in-plane and deformation of the reinforcement out-of-plane. Shear flow (also referred to as squeezing flow) theories, on the other hand, assume that the fibres are convected with the resin under the effect of the viscous drag force [20]. To decrease the processing time, new manufacturing techniques tend to use very high pressure and pressure rates. Moreover,

new material formulations where thermoplastic and thermoset resins are combined, which aim to improve the toughness and the resistance to delamination of conventional systems, have also appeared in the last 20 years or so. Recent separate studies from Nixon-Pearson et al. [7] and Hautefeuille et al. [8] have shown that in these cases, a transition between flow modes can be observed. This is a problem from a modelling point of view as the equations underpinning bleeding and squeezing flow respectively are difficult to combine, with percolation-driven compaction theories assuming the additive decomposition of pressure into strain-dependant (i.e. non-linear elastic) and strain rate-dependant (i.e. viscous) terms, and shear flow theories predicting the multiplicative superposition of an elastic and a viscous terms at the laminate level.

1.2. The defgen protocol model

Based on extensive experimental characterisation [7], Belnoue et al. [21] proposed a phenomenological model that considers the transition between squeezing and bleeding flow. Building on the work by Kelly [22] who, revisiting previous experimental data [23], suggested that multiplicative superposition of strain and strain rate functions could also be used to represent percolation flow, they postulated that bleeding can be described as squeezing longitudinally (rather than transversely) to the fibres making the formulation of a unified model for squeezing and bleeding flow much easier. For each flow mode, an equation relating pressure ($p = -\sigma$), to the strain (ϵ) and its rate ($\dot{\epsilon}$) in the compaction direction was derived based on geometrical considerations at both the micro- (i.e. the fibre scale) and the meso-scale (i.e. the ply scale). A criterion for the transition between the flow modes was derived based on the assumption that fibres' convection is impeded once the shear deformation at ply edges has reached a critical level. At the meso-scale, this gave rise to the existence of a compaction limit [24] passed which the fibres are so tightly packed that the melt cannot compact anymore. The model was then implemented as a 3D hyper-viscoelastic material in a user material subroutine for the Finite Element (FE) package Abaqus/Standard and used to study the impact of consolidation on fibre path defect generation for a wide range of industrially relevant geometries such as corner and tapered laminates [16] or parts with embedded AFP gaps and overlaps [25]. Good predictions for the thickness evolution of prepreg stack and consolidation-driven defects were made. The whole workflow associated with what is termed the DefGen ProToCoL (**Processing Tool for Composite Laminates**) is illustrated in Fig. 1.

As shown in [9], in order to capture the non-homogeneous distribution of strain within an elementary tape/ply from which the size-effect mentioned above originate, modelling the consolidation behaviour of fibre reinforced fluids within a FE framework should involve a sub-element resolution through the thickness of a ply. As a result, simulations of the compaction of a volume of material of only a few mm³ can take several hours. As illustrated in Fig. 2, the DefGen ProToCoL framework is based on a more pragmatic approach that allows the use of only 1 element through the thickness of a ply. The 1D semi-analytical model, on which the framework is based, was derived from the equation proposed in [26] for the viscous flow (i.e. Stokes flow) of highly anisotropic materials squeezed between two rigid platens where a no-slip condition is assumed. As illustrated in Fig. 2 (see also [21]), the model's formulation starts with the rearrangement of Rogers's equation to express the apparent viscosity of a unit cell (i.e. η_{micro}) where the squeezing of a Newtonian fluid (of viscosity η_{resin}) between two square fibres is considered. The size, d , of the idealised square fibres is expressed as $d = \sqrt{\pi}R$ where R is the fibres' diameter. Then, Rogers's relation is used, at a ply level, to formulate an expression for the apparent viscosity, η_{meso} , of a volume of prepreg of viscosity η_{micro} , initial thickness t , initial width (i.e. transversely to the fibres) w and initial length (i.e. along the fibres) l . The obtained expression does not account for the diminution of the apparent viscosity of the prepreg with the applied strain rate - also referred to as shear thinning - that is well documented in the literature [27]. Considering that there is no analytical solution for the Stokes flow

Fig. 1. DefGen ProToCoL workflow - 1. The fibre reinforced fluid compaction response is characterise as described in [7]. 2. Material parameters are extracted following the procedure described in [21]. 3. Simulations of the auto-clave consolidation of composites parts are performed as described by [16, 25].

Fig. 2. Schematic representation of the main assumption behind the DefGen model. The model in the centre of the figure that illustrates inhomogeneous strain distribution within a ply is taken from [9].

of a non-Newtonian two-scale fluid and that shear thinning is known to be related to the reorganisation and interaction of the fibres, the final expression for the apparent viscosity of the volume of prepreg considered is obtained by weighting η_{meso} with a power-law function (i.e. $\eta_{rate} = e^b(-\dot{\epsilon})^a$) so that when $\epsilon \leq \epsilon^l$, the apparent viscosity, η_{app} , of the prepreg is expressed as in eq. (1) whilst for $\epsilon > \epsilon^l$, eq. (2) is used.

$$\eta_{app}(\epsilon, \dot{\epsilon}) = 4 \left(\frac{w}{t} \right)^2 \sqrt{\chi_l} e^{-3\epsilon} k \left(\left(\frac{k}{\sqrt{\chi_f} \cdot \exp(\epsilon) - k} \right)^2 + 3 \right) e^{\bar{b}_{squeeze}} (-\dot{\epsilon})^a \quad (1)$$

$$\eta_{app}(\epsilon, \dot{\epsilon}) = 4 \left(\frac{w}{t} \right)^2 \chi_l e^{-2\epsilon} k \left(\left(\frac{k}{\sqrt{\chi_f} \cdot \exp(\epsilon) - k} \right)^2 \right) \left(\frac{l}{d} \right)^2 e^{\bar{b}_{bleed}} (-\dot{\epsilon})^a \quad (2)$$

In eqs. (1-2), $\epsilon^l = -\ln(\sqrt{\frac{2}{3}} \frac{t}{w} \tan(\gamma_{lock}) + 1)$ is the strain at locking, e.g. critical level of shearing at ply edges, and $\chi_l = e^{-2\epsilon^l} = \frac{V_f^{max}}{V_f^0} \chi_f$. In these expressions, V_f^0 is the initial fibre volume fraction, V_f^{max} is the maximum

fibre volume fraction and $\gamma_{lock} = \arcsin(\frac{4A_0 - \sin 2\alpha}{1 - \cos 2\alpha})$ is the critical shear angle (i.e. the maximum shear angle in the tape at locking). Following considerations on fibre rearrangements, it can be shown (see [21]) that

$\alpha = \arcsin(\frac{A_0}{A_f} \sin \alpha_f)$ where $\alpha_f = \frac{A_0}{A_0 + A_f} \arctan(\frac{R}{4A_f})$, $A_0 = \frac{\pi R^2}{2V_f^0}$ and $A_f = \frac{\pi R^2}{2V_f^{max}}$. Practically, at a given processing/test temperature, the model only needs 4 material parameters:

- k that is the size of the (square) fibres normalised by the size of the unit cell. k can be assimilated to a “1-dimensional” fibre volume fraction and written as $k = \frac{d}{l_{UC}}$ using the same notations as on Fig. 2;
- a that is related to the flow behaviour index n (i.e. $a = n - 1$) and controls whether the fluid is shear-thinning ($n < 1$), Newtonian ($n = 1$) or dilatant ($n > 1$);
- and, finally, b which increases as the melt becomes more viscous (i.e. flow consistency factor). b takes different values under squeezing ($b_{squeeze}$) and bleeding (b_{bleed}) hence the 4 parameters in total. In order to write the equations in a concise way the notation $\bar{b} = b + \ln \eta_{resin}$ is adopted.

For a given load cycle, the ordinary differential equations presented in eqs. (1) and (2) can be numerically solved in terms of the through-thickness strain which allows to predict the evolution with time of the thickness of the ply stack.

1.3. The contribution of this paper

So far, a clear limitation of the DefGen PRoToCoL model is the lack of an analytical expression that links the model's bespoke material parameters to known physical quantities (such as the resin viscosity or the fibre content) for which established measurement techniques exist and that have available historical data that could be used directly to feed the simulations (essentially removing the need for step 1 and 2 on the workflow illustrated in Fig. 1). In the present contribution, the model's ability to capture the behaviour of prepreg made from different resin system (i.e. 1st and 3rd generation toughened thermoset and thermoplastic systems) and fibres type and architecture (i.e. UD and woven carbon, UD glass) is demonstrated. For each studied case, material parameters can be extracted, and the model can accurately predict how the samples' thickness evolves following the application of temperature and pressure cycles. Contrasting the evolution of the parameters with temperature with the type of the constituents in the prepreg they describe allows a physics-based reformulation of the model. Analytical expressions linking the material parameters to the resin viscosity, the initial porosity of the fibre bed and the fibre radius are established. Following Chandler [28], the flow consistency factor is reinterpreted as a function of an energy barrier that controls the flow transition. This way, the artificial change of flow direction hypothesised in the original model is made redundant. The newly derived expressions open a number of exciting prospects in the context of manufacturing 4.0 of composites [29]. For example, they make it possible to design the prepreg content to reduce defects occurrence or to adapt the processing cycle based on the known/measured volume fraction and porosity of the fibre bed.

2. The defgen protocol testing procedure

2.1. Materials

Although the DefGen ProToCoL model was originally derived for toughened thermosetting unidirectional (UD) prepreg, over the last few years, it has been used at the Bristol Composites Institute (ACCIS) to predict the behaviour under compaction of a much wider range of resins and fibrous reinforcements combinations. In all these cases, the model allowed to accurately describe the thickness evolution of fibrous reinforcement impregnated with resin subjected to a range of temperatures, pressures and pressure rates. This can either be explained by some real underlying physics behind the model's equations that were not completely grasped in the original formulation or, on the contrary, by an exaggeration of the physical significance of the model and the ability of the power law to fit well with any scenario [30]. In this paper, the analysis of the dependencies between the extracted bespoke parameters of the DefGen model and known geometrical and physical characteristic of the different components of the system supports the arguments that the first of these scenarios should be favoured.

In addition to the original 2nd and 3rd generation UD carbon-fibre/toughened thermosets systems (i.e. IM7/8552 [31] and IMA/M21 [32] both from Hexcel®) used in the original publications [7, 21], data from tests on 3 supplementary systems are presented and analysed:

- A 3rd generation toughened thermoset system (M21) with 2/2 twill weave carbon fibres (IMA) from Hexcel® [32] referred to as "M21 woven" in the manuscript.
- A 1st generation toughened thermosets system with UD glass reinforcement (E-Glass/913) from Hexcel® [33] referred to as "913 glass" in the manuscript.
- A UD carbon system with thermoplastic resin (TC1100 Cetex® PPS) from Toray Advanced Composites referred to as "PPS" in the manuscript.

These provide a wide range of resin viscosities, fibre types and architecture that is also consistent with the variety of material types found in the composite industry.

2.2. Testing procedure

The studied materials were all tested following the procedure described in [7] (see top left corner of Fig. 1). Specimens were laid-up in a cruciform configuration with fibre layers alternatively orientated at 0° and 90°. All the samples were 16 plies thick no matter the thickness of the plies (which varies from one material to another) and the in-plane dimensions of each of the plies was 30 mm × 50 mm. These were tested using an Instron 8801 universal testing machine (as illustrated in Fig. 1) with temperature-controlled hot plates transferring the load and temperature to the samples. The specimens were loaded under a ramp-dwell regime where the fast application of load is followed by long dwell/creep intervals (the idea being that this allows to cover a larger space of rate-pressure-thickness within one test and hence, ensures, a more reliable fit). Each sample was tested at a constant temperature throughout the loading. Specimens made from thermoset-based prepreg were loaded to 240 N in 1200s. The ramp-dwell program included five 240 s steps with an incremental load of 40 N, starting at 80 N. The loading rate at each step was approximately 0.1 MPa/s. Testing temperature varied from between 30 °C and 120 °C. For the PPS thermoplastic system, the testing program was altered to reflect better the conditions to which these types of materials are subjected in a real manufacturing. The specimen dimensions were changed to 15 mm × 30 mm in plane and 12 plies through thickness. Hence, all the applied loads were multiplied by 50 in comparison to those applied to the thermoset based systems and the spectrum of temperature explored varied between 260 °C and 320 °C with 10 °C increments.

2.3. Results

In all cases, the model was shown to be able to fit the experimental data well and, as illustrated in Fig. 3, parameters could be extracted as can be judged by the linear trends and collapse to a master-curve at different temperature on the considered graphs. The set of parameters extracted at each tested temperature and for each material considered are reported in Tables 1–5. Some interesting remarks can be drawn from the graphs presented in Fig. 3. Firstly, the model predicts that the 913-based system (see Fig. 2a/) that has the most porous structure (i.e. the diameter of the glass fibres is greater than for the carbon fibres and the inter-fibre channel are therefore bigger) displays minimal squeezing. Similarly, the woven structure of the M21 woven system prevents large lateral expansion of the fibre bed. This also results in a reduced level of squeezing as illustrated in Fig. 2b/. On the other hand, whilst it would be expected that the thermoplastic system (see Fig. 2c/) should display significantly more squeezing than bleeding flow, this is not distinctly apparent in the above figure. In fact, it is predicted to only bleed at temperatures below the melting point of 285 °C. This is attributed to the fact that, at these temperatures, the compaction plateau is reached very quickly which may lead to the model mispredicting the flow behaviour. Some bleeding is also predicted at high temperatures, but this is more in line with experimental observations that shows that contrary to what is normally assumed, bleeding also occurs in thermoplastic-based systems [34]. These remarks help building confidence in the validity of the assumptions underlying the model's formulation, which were summarised in Section 1.2. Secondly, the evolution of the parameter k with temperature that was observed in the original set of data for toughened UD prepreps IM7-8552 and IMA-M21 (see Fig. 1) still holds for the newly characterised systems. Given that k is only supposed to account for the size of the inter-fibre channels prior to compaction, one may rightly wonder why that is the case and what physical phenomena not accounted for in the present model formulation is responsible for this? Lastly, we shall also draw the reader's attention to the fact that in every parameter extraction procedure (see Fig. 3), the straight lines corresponding to bleeding and squeezing respectively seem to consistently be simply shifted by $\ln((\frac{L}{d})^2)$. As one of the main differences between

Fig. 3. Parameters for the DefGen model can be extracted by varying k until all the experimental data points converge to a single straight line in a log-log diagram where $\eta_{rate} = e^b(-\dot{\epsilon})^a$ (see eqs. (1-2)) is plotted as a function of the strain rate in the compaction direction (i.e. $|\dot{\epsilon}|$). Parameters a and b , then, directly follow as fitting constants of the obtained lines. Figures a/, b/ and c/ represent such a diagram for 913 glass, M21 woven and the PPS materials, respectively. Each colour corresponds to a test at a given temperature.

Table 1

Compaction parameters extracted from isothermal ramp-dwell tests for UD IMA-M21 prepreg [21]. Viscosities were taken from [35].

T (°C)	30	40	50	60	70	80	90
k	0.940	0.864	0.816	0.810	0.806	0.806	0.806
a	-0.926	-0.864	-0.793	-0.754	-0.764	-0.754	-0.759
$b_{squeezing}$	-15.991	-13.020	-12.660	-12.627	-12.624	-12.624	-12.624
$b_{bleeding}$	-32.718	-29.747	-29.387	-29.353	-29.350	-29.350	-29.350
η_{resin} (Pa.s)	34,195.93	9327.346	2784.744	909.732	326.377	129.464	57.180

Table 2

Compaction parameters extracted from isothermal ramp-dwell tests for UD IM7-8552 prepreg [21]. Viscosities were taken from [36].

T (°C)	30	40	50	60	70	80	90
k	0.949	0.919	0.884	0.850	0.824	0.806	0.795
a	-0.971	-0.953	-0.941	-0.926	-0.893	-0.859	-0.821
$b_{squeezing}$	-17.51	-16.01	-15.12	-14.58	-14.27	-14.08	-13.96
$b_{bleeding}$	-32.85	-31.35	-30.46	-29.92	-29.61	-29.42	-29.3
η_{resin} (Pa.s)	11,799.67	3932.335	1664.947	682.777	284.502	118.540	48.612

Table 3

Compaction parameters extracted from isothermal ramp-dwell tests for CF/PPS. The viscosities have been taken from a fit to manufacturer data proposed in [37].

T (°C)	260	280	290	300	310	320
k	0.921	0.834	0.664	0.662	0.652	0.638
a	-0.921	-0.875	-0.779	-0.757	-0.736	-0.727
$b_{squeezing}$	-16.71	-14.79	-12.35	-12.43	-12.52	-12.54
$b_{bleeding}$	-31.0	-28.16	-27.38	-27.98	-27.61	-27.96
η_{resin} (Pa.s)	982.695	615.630	493.380	398.473	324.189	265.596

Table 4

Compaction parameters extracted from isothermal ramp-dwell tests for IMA-M21 prepreg with plain weave fibre architecture. Viscosities were taken from [35].

T (°C)	30	40	50	60	70	80	90
k	0.8	0.74	0.68	0.66	0.66	0.66	0.66
a	-0.920	-0.951	-0.679	-0.739	-0.770	-0.762	-0.762
$b_{\text{squeezing}}$	-22.857	-21.157	-19.897	-19.077	-18.697	-18.690	-19.050
b_{bleeding}	-39.583	-37.883	-36.623	-35.803	-35.423	-35.416	-35.776
η_{resin} (Pa.s)	34,195.93	9327.346	2784.744	909.732	326.377	129.464	57.180

Table 5

Compaction parameters extracted from isothermal ramp-dwell tests for a UD prepreg HexPly 913 with E-glass fibres. Viscosities were taken from the manufacturer datasheet [33].

T (°C)	30	60	90	120
k	0.776	0.720	0.681	0.676
a	-0.917	-0.904	-0.793	-0.750
$b_{\text{squeezing}}$	-23.012	-21.422	-19.546	-17.342
b_{bleeding}	-39.266	-37.676	-35.800	-35.700
η_{resin} (Pa.s)	1615.088	413.160	36.140	6.415

eq. (1) and eq. (2) is the presence of the term $(\frac{l}{d})^2$, this suggest that the hypothesis on the change of the flow direction after locking may have been exaggerated.

3. Physics-based re-interpretation of the defgen protocol model

In this section, the two last remarks raised in Section 2.3 are used as a basis to reformulate and reinterpret the DefGen ProToCoL model. As shown in the rest of the paper, this allows to give a real physical meaning to the parameters used. A recent publication by [28] showed that the traditional expression for a power-law fluid can be rewritten as:

$$\eta = K \dot{\gamma}^{n-1} = \eta_0 e^{\left(-\frac{\Delta E_1}{RT} + \frac{\Delta S_1}{R}\right)} \left(\dot{\gamma}\right)^{\frac{m}{RT} - \frac{q}{R}} \quad (3)$$

where η is the viscosity of the fluid, K is the flow consistency factor, n is the flow behaviour index and $\dot{\gamma}$ is the shear rate. One of the main disadvantages of the power-law fluid formulation is that whilst $\dot{\gamma}$ is unambiguously defined in s^{-1} and n is dimensionless, the dimension of K (i.e. $Pa.s^n$) depends on the value of n . In Chandler's formulation, on the other hand, each parameter has a defined dimension that does not change with the value of the other material constants which makes a material characterisation procedure more robust. $R = 8.314 Jmol^{-1}K^{-1}$ is the gas constant, T is the absolute temperature expressed in K , $\dot{\gamma}$ is a dimensionless shear rate magnitude and η_0 is a viscosity term expressed in $Pa.s$. ΔE_1 and m are both expressed in $Jmol^{-1}$ whilst, ΔS_1 and q are in $Jmol^{-1}K^{-1}$.

Chandler's relation (i.e. the last part of eq. (3)) was derived from combining the Stokes-Einstein equation (which was first derived by Einstein in his PhD thesis and relates the diffusion coefficient of a "Stokes" particle undergoing a Brownian motion in a quiescent fluid at uniform temperature to the viscosity of the fluid) with the conventional expression of a power-law fluid, as displayed in the first part of eq. (3). The derivation involves the definition of an enthalpy, ΔH , which is an energy barrier required to overcome forces between flow units such as frictional and electrostatic forces and an activation entropy, ΔS , which corresponds to the energy that needs to be provided to the system in order to maintain a certain structural order that is different to the one in the fluid at rest. In the case of a Newtonian fluid or a fluid at rest, ΔS is null. The quantities ΔE_1 , ΔS_1 , m and q in eq. (3), follow from the expressions of ΔH and ΔS respectively:

$$\Delta H(\dot{\gamma}') = m \ln(\dot{\gamma}') + \Delta E_1 \quad (4)$$

$$\Delta S(\dot{\gamma}') = q \ln(\dot{\gamma}') + \Delta S_1 \quad (5)$$

where ΔE_1 and ΔS_1 are the values of ΔH and ΔS determined at the deformation rate of $1 s^{-1}$.

In the context of the DefGen ProToCoL model, eq. (3) can be compared to eqs. (1) and (2) and used to write:

$$b = b_{\text{squeezing}} = b_{\text{bleeding}} + 2 \ln\left(\frac{l}{d}\right) = -\frac{\Delta E_1}{RT} + \frac{\Delta S_1}{R} = -\frac{\Delta F_1}{RT} \quad (6)$$

$$a = \frac{m}{R} \frac{1}{T} - \frac{q}{R} \quad (7)$$

where ΔF_1 is an Helmholtz free energy at a deformation rate of $1 s^{-1}$. ΔF_1 acts as an energy barrier that controls the ability of the resin to flow through a fibre network of a given initial porosity. The dependence of both b and a with $\frac{1}{T}$ implies that each of these parameters can theoretically be determined from the knowledge of the other. This agrees well with [38] observation that, for material systems IM7/8552 and IMA/M21, experimentally extracted values of the two parameters are almost perfectly correlated (i.e. correlation coefficient of 0.95). In eq. (6), the remark made at the end of section 2.4 on the irrelevance of changing the flow direction upon locking is used to reduce the amount of material parameters needed by the model. This is also justified by the fact that b now identifies to an energy barrier that controls the flow of resin. In essence, this definition equates to the loosening of the assumed coupling between fibre bed locking and the transition between squeezing and bleeding flow, with the later no longer being considered to trigger the former. The relation of the equations of the DefGen ProToCoL model to Chandler's equation not only allows for these parameters to be directly quantified from the underlying physics (as opposed to just fitting constants), it also provides understanding of how these parameters evolve with temperature. This is used in the next section of the manuscript where the comparison of parameters extracted for different resins and fibres types and architecture combinations is used to understand how k , b and a depend on the known properties of the resin and the fibre bed. Lastly, it is worth remarking that, contrary to the more general case described by Chandler in eq. (5), ΔS , which characterises structural ordering, does not depend on $\dot{\gamma}'$, as locking (i.e. maximum structural ordering) is only a function of geometrical characteristics of the ply and unit cell. Consequently, $q = 0$.

The physics-based reinterpretation of the model proposed allows to replace eqs. (1) and (2) with:

$$\eta_{app}(\epsilon, \dot{\epsilon}) = 4 \left(\frac{w}{t}\right)^2 \sqrt{\chi_l} e^{-3\epsilon} k \left(\left(\frac{k}{\sqrt{\chi_f} \cdot \exp(\epsilon) - k} \right)^2 + 3 \right) \eta_{resin} e^{b\epsilon} \epsilon'^a \quad (8)$$

$$\eta_{app}(\epsilon, \dot{\epsilon}) = 4 \left(\frac{w}{t}\right)^2 \chi_l e^{-2\epsilon} k \left(\left(\frac{k}{\sqrt{\chi_f} \cdot \exp(\epsilon) - k} \right)^2 \right) \eta_{resin} e^{b\epsilon} \epsilon'^a \quad (9)$$

where, similarly to $\dot{\gamma}'$, ϵ' is a dimensionless strain rate magnitude.

The fibre reinforced fluid is expressed as a general power-law fluid where the flow consistency factor is multiplicatively split into a term inherited from shear flow theories (in red) and a term reminiscent of percolation flow theories (in green). Hence, it is shown later in the paper that b (that is now defined as an energy barrier controlling the ability

Table 6

Summary of the differences, in units, between the original formulation of the DefGen ProToCoL model and the newly introduced physics-based reformulation.

	Original formulation	New formulation
w : ply width	mm	Mmm
t : ply thickness	mm	mm
l : ply length	mm	–
d : $\sqrt{\pi R}$ where R is the fibres' diameter	mm	–
η_{resin} : resin viscosity	Pa.s	Pa.s
χ_l : stretch in compaction direction at locking	dimensionless	dimensionless
χ_f : stretch in compaction direction on the compaction plateau	dimensionless	dimensionless
k : normalised (average) size of the inter-fibre channels	dimensionless	dimensionless
b : $-\frac{\Delta f_l}{RT}$ (linked to the flow consistency factor in the original formulation)	$\ln(s^d)$	dimensionless
a : $\frac{m}{RT}$ (flow behaviour index –1 in the original formulation)	dimensionless	dimensionless
ϵ : Hencky strain in the compaction direction	dimensionless	dimensionless
$\dot{\epsilon}$: rate of Hencky strain in the compaction direction	s^{-1}	–
ϵ' : strain rate magnitude in the compaction direction	–	dimensionless

of the resin to flow through the fibre network) depends on the fibre-bed porosity and the fibre radius which is reminiscent of permeability calculations using the Kozeny-Carman equation. The differences remaining between the equations before (i.e. eq. (8)) and after (i.e. eq. (9)) locking come from assumption made on the material compressibility (and resulting width expansion) under the 2 regimes. Practically, passed locking, $\epsilon \approx \epsilon^l$ and thus $\chi_l e^{-2\epsilon} \approx \sqrt{\chi_l} e^{-3\epsilon}$. Similarly, the “+3” term between the brackets in eq. (8) disappears in eq. (9) by assuming $l \gg d$ and could just be reintroduced. Hence, although, χ_l still appears in the equation, the flow transition from the original model is essentially removed as, in the case of a 1D analysis, eqs. (8) and (9) could be merged into one. The slight changes of units introduced by the model reformulation are summarised in Table 6.

4. Discussion

4.1. Apparent volume fraction

Another concept from physical chemistry that can be applied for the apparent viscosity of a fibre reinforced fluid is the notion of Stokes radius (also called hydrodynamic radius for a polymer) which expresses the apparent radius of a molecule which diffuses within the fluid it is part of. As it moves, the molecule can drag other fluid units so that its apparent radius increases. Therefore, the hydrodynamic radius is not only a function of the radius of the molecule but also of the fluid mobility (that is the inverse of the viscosity). A similar notion can be used in relation to the DefGen ProToCoL model. Hence, for high viscosity resins the apparent and actual fibres' radii will differ. This will result in the inter-fibres channels being smaller and the effective fibre volume fraction greater than their nominal values. Whether the Stokes radius, $r_{apparent}$, is greater than the fibre radius r_{fibre} or not, the size of a unit cell, l_{UC} , stays the same. $x = r_{apparent} - r_{fibre}$ cannot exceed a certain value, x_{max} , that is dictated by the maximum fibre packing. The aim of the present contribution is to analyse how the material parameters of the DefGen ProToCoL model changes with a change of fibre type, fibre bed architecture and/or resin type. In the original manuscript where the DefGen ProToCoL model was formulated [21], it was experimentally found that k evolves with temperature which contradicts its prescribed meaning of purely geometrical volume fraction related parameter. In the light of the considerations made in this paragraph, it can be suggested that the effective fibre radius may evolve indeed as it depends on resin viscosity which is itself dependant on temperature.

The high degree of contrast between the fibre types, fibre architectures and resin types of the prepreps studied, suggests that comparison of the evolution of their “raw” k values directly with the viscosity of the resin would not provide significant insight into the relationship between the parameter k and resin viscosity. It is proposed here, instead, to construct a dimensionless normalised fibre coating thickness, $\frac{x}{x_{max}}$,

Table 7

Summary of the parameters used to plot Fig. 4.

	r_{fibre} (μm)	k_{max}	k_{min}	$T_{ph. trans.}$ ($^{\circ}C$)	$\eta_{ph. trans.}$ (Pa.s)
M21	3.5	0.952	0.70	35	21,761.640
8552	3.5	0.952	0.70	45	2798.642
PPS	3.5	0.952	0.70	285	550.579
913 glass	6.0	0.952	0.67	30	26,607.763
M21 woven	3.5	0.791	0.61	35	21,761.640

that accounts for the differences in fibre types and architecture and a normalised viscosity, $\frac{\eta_{resin}}{\eta_{ph. trans.}}$, that compares the resin viscosity η_{resin} to the viscosity of the same resin at the temperature at which a large viscosity drop is observed (i.e. solid to liquid transition), $\eta_{ph. trans.}$. For thermoplastics, $\eta_{ph. trans.}$, directly identifies to the viscosity at the melting temperature. The use of the normalised viscosity allows to correct for the differences in viscosity magnitudes and range exhibited by the different resin systems used.

Using the various considerations made here, the normalised fibre coating thickness, can be expressed as the quotient of the fibre coating thickness, x , and the fibre radius, r_{fibre} . Hence, calling k_{max} the value of k at high resin viscosity (i.e. when the apparent volume fraction corresponds to that at maximum fibre packing) and k_{min} the value of k at low resin viscosity, leads to the relation:

$$l_{uc} = \frac{r_{fibre}}{k_{min}} = \frac{r_{fibre} + x}{k} = \frac{r_{fibre} + x_{max}}{k_{max}} \quad (10)$$

This then results in:

$$\frac{x}{x_{max}} = \frac{kl_{UC} - r_{fibre}}{k_{max}l_{UC} - r_{fibre}} = \frac{k \frac{r_{fibre}}{k_{min}} - r_{fibre}}{k_{max} \frac{r_{fibre}}{k_{min}} - r_{fibre}} = \frac{\frac{k}{k_{min}} - 1}{\frac{k_{max}}{k_{min}} - 1} \quad (11)$$

Remarking that $\frac{k}{k_{min}} = 1 + \frac{x}{r_{fibre}}$, we can finally write:

$$\frac{x}{x_{max}} = \frac{1}{\left(\frac{k_{max}}{k_{min}} - 1\right)} \frac{x}{r_{fibre}} \quad (12)$$

$\frac{x}{x_{max}}$ and $\frac{\eta_{resin}}{\eta_{ph. trans.}}$ have been calculated for the 5 prepreg systems considered using the parameters summarised in Table 7 that have been measured/extracted from experimental data (r_{fibre} , k_{min}), taken from manufacturers data sheets ($T_{ph. trans.}$, $\eta_{ph. trans.}$) or from reasonable assumptions (k_{max}). Hence, it was assumed that the fibre distribution transversely to the fibres is isotropic so that k_{max} can be determined from $k_{max} = \sqrt{V_{fmp}}$ (with V_{fmp} being the fibre volume fraction at maximum packing) and $k_{min} = \sqrt{V_f^0}$. Hence, the theoretical maximum volume fraction in a UD ply is 90.7% assuming hexagonal close packed fibre arrangement. Similarly, the maximum volume fraction of the woven

Fig. 4. Master curve controlling the evolution of k with viscosity/temperature.

system was estimated to be 65% [39]. It is worth noting that although woven materials have very different micro-mechanical behaviour to prepreg made of UD reinforcements (they exhibit dual-scale flow mechanisms where the porosity is high at some locations and low elsewhere), the DefGen model does not capture this level of complexity. The material porosity characterised by the parameter k is taken in a homogenised sense. The weave type, however, comes into play when considering the maximum possible packing characterised by the parameter k_{max} discussed here.

In Fig. 4, the $\frac{x}{k_{max} - k_{min}}$ values derived from the values of k extracted from the compaction tests of the 5 prepreg systems under consideration are plotted as a function of $\frac{\eta_{resin}}{\eta_{ph.trans.}}$. This reveals the existence of a master curve around which most of the data points agglomerate, with the most notable error being for the PPS system. This curve can be approximated by a hyperbolic tangent function that is plotted as a solid black line in Fig. 4. The hyperbolic tangent function varies between -1 and 1 and is often used to describe transition phenomena and is therefore well suited to the context described here. This provides a way to estimate the value of k based on the known evolution of the resin viscosity with temperature and the structural characteristic of the fibre bed. Hence, the assumption of an isotropic distribution of the fibres in the transverse plane allows to re-write eq. (12) as:

$$\frac{1}{\left(\sqrt{\frac{V_{fmp}}{V_f^0}} - 1\right) r_{fibre}} \frac{x}{k_{max} - k_{min}} = \tanh\left(\frac{\eta_{resin}}{\eta_{ph.trans.}} + 0.25\right)$$

and we finally have:

$$k = \sqrt{V_f^0} \left(1 - \tanh\left(\frac{\eta_{resin}}{\eta_{ph.trans.}} + 0.25\right)\right) + \sqrt{V_{fmp}} \tanh\left(\frac{\eta_{resin}}{\eta_{ph.trans.}} + 0.25\right) \quad (13)$$

It is worth noting that the observed divergence of the PPS points from the overall trend may be explained by a likely underestimation of $\eta_{ph.trans.}$ in this case. Hence the viscosity of the PPS used was derived from a fit to experimental data presented in [37] and that was developed over a range of temperatures that were all higher than the melting point.

4.2. Flow transition

In the same way, the parameter $\bar{b}$ can be determined using the development of Section 3, the evolution of the resin viscosity with temperature and some considerations of the fibres' microstructural arrangement prior to compaction. This section shows how a master curve for the evolution of $\bar{b}$ with temperature can be constructed. The different steps followed are summarised in Fig. 5.

The first step is to follow eq. (6) and plot the evolution of $\bar{b}$ (see eqs. (1-2)) as a function of $\frac{1}{T}$. Similarly to what was done in Section 4.1 with the viscosity, resin independent curves are built by using the relative temperature, $\frac{T}{T_{ph.trans.}}$, instead of T (interestingly, this gives rise to an expression that is similar to some relations controlling diffusion creep in metals [40]). The obtained curves are presented in Fig. 5. a/. For all 5 material systems, $\bar{b}$ is shown to evolve linearly with $\frac{1}{T}$ which validates the interpretations made in Section 3. Moreover, the use of the relative temperature shows that the data points of the 3 systems made from UD carbon fibre tend to converge to the same linear relation.

It is worth noting that when $\frac{T}{T_{ph.trans.}} < 1$ (i.e. when the prepreps behave more like a solid rather than a liquid), the data points tend to diverge from their characteristic straight lines. That characteristic was used to help determine the values of $T_{ph.trans.}$ presented in Table 7. It was discussed in Section 4.1 that for $\frac{T}{T_{ph.trans.}} < 1$, $r_{apparent} > r_{fibre}$ and as the curve for UD glass reinforcements is different from the UD carbon fibre curve, it appears that $\bar{b}$ (and by extension ΔF_1) depends on r_{fibre} . This agrees well with the open literature on bleeding flow [18] to which ΔF_1 relates as it controls the ability of resin to diffuse within the prepreg. Numerical simulations of Stokes flow of resin through fibre-beds made of fibres of different radii and varied level of porosity/volume fraction [41] suggested that ΔF_1 should be a function of the ratio $\frac{\epsilon_0^2}{(1-\epsilon_0)} r_{fibre}$ (where $\epsilon_0 = 1 - V_f^0$ is the porosity) that characterises the tightness of the fibre-bed. Based on this, the magnitudes of the gradient $(-\frac{\Delta E_1}{RT})$ and ordinate at origin $(\frac{\Delta S_1}{R})$ of the curves (see Fig. 5a/) characteristic of the 3 different fibre beds considered are plotted as a function of $\frac{\epsilon_0^2}{(1-\epsilon_0)} r_{fibre}$ in Fig. 5b/ and c/ respectively. These graphs reveal the existence of the linear evolution of ΔF_1 with $\frac{\epsilon_0^2}{(1-\epsilon_0)} r_{fibre}$. As would be expected $\Delta F_1 = 0$ when $r_{fibre} = 0$ i.e. a block of pure resin subjected to a pressure p will naturally flow. As illustrated in Fig. 5d/, it follows that the curve $\frac{\bar{b} - \ln(\eta_{resin})}{\frac{\epsilon_0^2}{(1-\epsilon_0)} r_{fibre}} = f\left(\frac{T}{T_{ph.trans.}}\right)$ is a master curve that can be used to

determine the value of $\bar{b}$ of any fibre reinforced resin melt of known fibre volume fraction, fibre radius and variation of the respective resin viscosity with temperature. As illustrated in Fig. 5d/, replacing r_{fibre} by $r_{apparent}$ (see Section 4.1) removes the non-linearity observed at relative temperature lower than 1 thus leading to:

$$\frac{\bar{b} - \ln(\eta_{resin})}{\frac{\epsilon_0^2}{(1-\epsilon_0)} r_{apparent}} = -16\,628 \frac{T_{ph.trans.}}{T} + 12\,471 \quad (14)$$

where 16 628 and 12 471 are the gradients of the curves in Fig. 5b/ and c/ respectively.

4.3. Shear thinning

The last material constant that needs to be analysed is the parameter a which controls the non-Newtonian behaviour of the prepreg. Hence for $-1 < a < 0$ shear thinning is observed whilst if $a > 0$ the fibre reinforced melt exhibits a shear thickening behaviour. In all cases studied, the values of a very clearly indicate that the viscosity of the prepreg decreases with the applied strain rate (i.e. shear thinning). Based on eq. (7), all the extracted values of a were plotted as a function of $\frac{T_{ph.trans.}}{T}$ in Fig. 6. Although the conformity to the theory is not as convincing as for the other two parameters, the data points broadly converge towards the straight line (drawn as a solid black line in Fig. 6) described by eq. (7) and where $q = 0$ (see Section 3) and $\frac{m}{RT_{ph.trans.}} = -0.9$ are used. However, the following considerations need to be taken into account:

1. The value of a is very sensitive to the other parameters, which leads to large variabilities of the (average) values reported in Fig. 6.
2. Matveev et al. [38] performed a sensitivity analysis of how a affects the predicted final thicknesses. This revealed a low influence of this parameter compared to the other parameters k and b .

Fig. 6. Variation of the shear thinning parameter with temperature.

4.4. And dry fibres?

Automated Dry Fibre Placement (ADFP) is an alternative to the more widely used AFP process, where dry fibres tapes (rather than pre-impregnated material) are deposited on a mould using a poly-articulated robot (see Fig. 7a/), thus additively building the structure of the part on a layer by layer basis. The manufacture is then completed by the infusion, in an oven rather than an autoclave, of the dry fibre preform

Fig. 5. Construction of the master curve controlling the evolution of b with temperature.

with resin. The method has significant potential for producing composite parts at lower cost and increased productivity [42]. However, before the method can be developed for industrial use, further work is first required to provide fundamental understanding of the material [43]. Particularly important is the compaction behaviour of the dry tow [44]. This determines the quality of the infusion and the final structural properties of the part through the volume fraction. As a side remark, it is interesting to note that a compromise needs to be found between infusibility, which is increased at low fibre volume fractions, and mechanical performance that improves with higher fibre content. As power law functions are often used to describe the compaction response of dry reinforcement [45], this suggests that the DefGen ProToCoL model could well be adapted to depict the through-thickness compressive behaviour of ADFP dry fibre tows.

Consequently, this hypothesis is now explored for the case of Dry-Fibre AFP. Dry fibre tapes, HiTape produced by Hexcel® [46], were tested using a similar experimental program and setup to that described in Section 2.2. The collected data (mainly thickness evolution with time of cruciform specimens subjected to a predefined load cycle at a given temperature) were used to explore the feasibility of the DefGen ProToCoL model to describe the compressive behaviour of these raw materials during processing and to confirm if the formulation developed for prepreg compaction, described in the previous sections, can be extended to (predominately) dry materials. The tapes consist of 12k UD carbon tows with a thermoplastic veil (binder) on each surface (that can be activated at 180 °C to increase tack during the layup process). The total amount of binder in the tape is of approximately 3%. As each tow is only 6.5 mm-wide it was not possible to keep the same specimen configuration (i.e. 30 mm in plane dimension) as in Section 2.1. As illustrated

Fig. 7. a/ AFP machine (National Composites Centre, UK) taken from [42], b/ Compaction test set-up, c/ Representative thickness evolution (averaged over 4 samples) of cruciform cross-ply specimens made from dry fibre Hi-Tape material and subjected to a ramp-dwell loading program.

Table 8

Final thickness of samples made from dry fibre AFP tapes HiTape compacted under isothermal conditions and ramp-dwell loading schedule.

Temperature (°C)	Average (mm)	CoV
Room Temperature	1.38	0.04
150	1.43	0.08
260	1.34	0.07

Table 9

Compaction parameters extracted from isothermal ramp-dwell tests for Dry fibre AFP tapes HiTape.

Temperature (°C)	30	150	260
k	0.80	0.820	0.817
a	-0.95	-0.95	-0.95
$b_{\text{squeezing}}$	-10.69	-10.47	-10.46
b_{bleeding}	-24.70	-24.48	-24.47

in Fig. 7b/, CP specimens with in-plane dimensions matching the width of a tow were manufactured and, this time, 8 plies through the thickness. To keep stresses generated within the samples in the same order of magnitude as in Section 2.1 whilst applying loads that are higher than the load cell sensitivity, 4 samples were tested simultaneously and the average thickness variation over those 4 samples was measured (rather than the individual thickness in each of the tested samples). The ramp-dwell load program included five 240 s steps with an incremental load of 20 N, starting at 60 N. The experiments were performed in a furnace (to have full control on the applied temperature) fixed to an Instron MJ 6272 equipped with a 1 kN load-cell and aluminium plates through which the load was transferred to the samples. Three sets of experiments were performed at room temperature, 150 °C and 260 °C respectively to ensure that the binder has a minimal effect. At each temperature, three repeats were performed. The results revealed only a small effect of temperature with average final thickness consistently close to 1.39 mm at the three temperatures. However, large coefficients of variation (CoV)

between tests repeats (see Table 8) was observed with measured final thicknesses comprised between 1.21 mm and 1.65 mm.

Fig. 7c/ illustrates a representative (test performed at 150 °C) thickness evolution curve extracted from these experiments (red solid line) superimposed to its fit using the DefGen ProToCoL procedure (black dashed line). As the shape of the curve is very similar to those of the prepregs studied in the rest of the manuscript, it can be concluded that the model provides a good fit of the experimental data. The samples initial thicknesses (2.35 mm on average, again with a large range of variability) were evaluated measuring the final distance between the aluminium compaction plates and the overall crosshead displacement of the top plate. Together with the nominal fibre density and the nominal areal weight of the tows this allowed for approximation of the initial volume fraction as 56% (after debulking). Inputting these numbers into the DefGen ProToCoL model (as originally formulated) allowed for the extraction of the parameters reported in Table 9.

With only 3% in weight of resin within the material, the question of the relevance of eqs. (13), (14) and (7) is raised as it is difficult to articulate what $T_{ph.trans.}$ and η_{resin} correspond to for a dry fibrous

material. This is reinforced by the inspection of the values reported in Table 9 that show no evolution of the parameters with temperature, which contradicts the relations derived in eqs (13), (7) and (14). However, a constant value of k with temperature is in fact compatible within the framework developed in this paper, as in the absence of resin we must have $x = r_{\text{apparent}} - r_{\text{fibre}} = 0$. Secondly, the dependence of a and b with temperature contained in eqs. (7) and (14) can be neutralised if we set $T_{\text{ph. trans.}} = T$ for all T . Combining this assumption with eq. (7) gives rise to $a = -0.9$ (which is very close to the experimental value given in Table 9). The only question remaining then is what value of η_{resin} should be used in eq. (10). Intuitively we know that this value should be “very small” (i.e. it should correspond to the effective viscosity of air with a small percentage of thermoplastic resin). However, because it is used within a logarithm function it can impact the value of b greatly as $\ln(0) = -\infty$. Instead of inputting parameters into eq. (14) to verify how close predictions are from the experimentally extracted parameters, the validity of the equation for dry fibre AFP tapes is checked by trying to determine η_{resin} from the experimentally extracted value of $\bar{b}$. For values of V_f^0 estimated from the measured sample thickness (i.e. 56%), or derived from the value of k extracted experimentally (i.e. $V_f^0 = k^2 = 64\%$), $\eta_{\text{resin}} = 1 \text{ MPa.s}$ or $\eta_{\text{resin}} = 1.45e^{-2} \text{ MPa.s}$, respectively. The 2 orders of magnitude between the two estimations illustrate the high sensitivity to the input introduced by the logarithmic function that was mentioned above. Although these values should be taken with great care, they are, as expected, relatively low (in particular for $V_f^0 = 64\%$) and this suggests that eq. (14) might still hold for dry fibrous materials. Overall, the mechanics of dry fibre tapes within the AFP process is much more complex than just the compaction, with hysteresis phenomena under multi-stage compaction and spring-back of the material when the pressure applied by the roller is released [44]. However, the preliminary results presented in this section suggest that some adaptation of the DefGen ProToCoL model is an option worth pursuing to describe the behaviour of dry fibre tows during the AFP process.

5. Conclusion

In the present contribution, the validity of a model developed for the compaction of UD toughened thermoset prepregs to a much wider class of fibre impregnated materials was explored. It was shown that the model performs well in describing the behaviour under compaction of prepreg systems made from a relatively wide range of resins (i.e. 1st, 2nd and 3rd generation toughened thermoset, thermoplastic etc.), fibre type (carbon and glass fibre) and fibre architecture (UD and woven materials). One of the drawbacks of the original model formulation was the phenomenological nature of the material parameters used. Analysing how these parameters change with the type of prepreg resulted in a much deeper physical insight. Expressions relating the parameters' values to known quantities such as resin viscosity and fibre volume fraction were developed. In the last section of the paper the validity of the model to describe the compressive behaviour of dry fibre AFP tapes was also explored.

Material variabilities can significantly affect composite manufacturing outcomes. The thickness differences observed, here, between nominally identical samples subjected to the same thermo-mechanical loading illustrates well the issues that this variability can cause for industries where dimensional tolerances are tight (e.g. the assembly of different parts of an aircraft). The equations developed will be useful to bring predictions relating to part manufacture into the early stages of design, thus helping in the material and process selection. On a more academic front, they can also help better understanding of how the observed material variabilities propagate in the parts, which will ultimately help in selection of more robust process cycles. Beyond composite manufacturing simulations the present work can also find applications in the world of biomechanics (see [47] for example).

Declaration of competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

- [1] Elkington M, Bloom D, Ward C, Chatzimichali A, Potter K. Hand layup: understanding the manual process. *Advanced Manufacturing: Polymer & Composites Science* 2015;1:138–51.
- [2] Advani SG. *Flow and rheology in polymer composites manufacturing*. Amsterdam: Elsevier; 1994.
- [3] Aridhi A, Arfaoui M, Mabrouki T, Naouar N, Denis Y, Zarroug M, Boisse P. Textile composite structural analysis taking into account the forming process. *Composites Part B: Engineering* 2019;166:773–84.
- [4] Varkonyi B, Belnoue JP-H, Kratz J, Hallett SR. Predicting consolidation-induced wrinkles and their effects on composites structural performance. *International Journal of Material Forming* 2019.
- [5] Göthofer J, Meyer N, Pallicity TD, Schöttl L, Trauth A, Schemmann M, Hohberg M, Pinter P, Elsner P, Henning F, Hrymak A, Seelig T, Weidenmann K, Kärger L, Böhle T. Virtual process chain of sheet molding compound: development, validation and perspectives. *Composites Part B: Engineering* 2019;169:133–47.
- [6] Wang EL, Gutowski TG. Laps and gaps in thermoplastic composites processing. *Composites Manufacturing* 1991;2:69–78.
- [7] Nixon-Pearson OJ, Belnoue JP-H, Ivanov DS, Potter KD, Hallett SR. An experimental investigation of the consolidation behaviour of uncured prepregs under processing conditions. *J Compos Mater* 2016;51:1911–24.
- [8] Hautefeuille A, Comas-Cardona S, Binetruy C. Mechanical signature and full-field measurement of flow-induced large in-plane deformation of fibrous reinforcements in composite processing. *Composites Part A: Applied Science and Manufacturing* 2019;118:213–22.
- [9] Sorba G, Binetruy C, Leygue A, Comas-Cardona S. Squeeze flow in heterogeneous unidirectional discontinuous viscous prepreg laminates: experimental measurement and 3D modeling. *Composites Part A: Applied Science and Manufacturing* 2017;103:196–207.
- [10] Centea T, Grunenfelder LK, Nutt SR. A review of out-of-autoclave prepregs – Material properties, process phenomena, and manufacturing considerations. *Composites Part A: Applied Science and Manufacturing* 2015;70:132–54.
- [11] May D, Aktas A, Advani SG, Berg DC, Endruweit A, Fauster E, Lomov SV, Long A, Mitschang P, Abaimov S, Abliz D, Akhatov I, Ali MA, Allen TD, Bickerton S, Bodaghi M, Caglar B, Caglar H, Chiminelli A, Correia N, Cosson B, Danzi M, Dittmann J, Ermanni P, Francucci G, George A, Grishaev V, Hancioglu M, Kabachi MA, Kind K, Deléglise-Lagardère M, Laspalas M, Lebedev OV, Lizaranzu M, Liotier PJ, Middendorf P, Morán J, Park CH, Pipes RB, Pucci MF, Raynal J, Rodriguez ES, Schledjewski R, Schubnel R, Sharp N, Sims G, Sozer EM, Sousa P, Thomas J, Umer R, Wijaya W, Willenbacher B, Yong A, Zarembo S, Ziegmann G. In-plane permeability characterization of engineering textiles based on radial flow experiments: a benchmark exercise. *Composites Part A: Applied Science and Manufacturing* 2019;121:100–14.
- [12] Farhang L, Fernlund G. Void and porosity characterization of uncured and partially cured prepregs. *J Compos Mater* 2016;50:937–48.
- [13] Hubert P, Poursartip A. Aspects of the Compaction of Composite Angle Laminates: an Experimental Investigation. *J Compos Mater* 2001;35:2–26.
- [14] Levy A, Hubert P. Vacuum-bagged composite laminate forming processes: predicting thickness deviation in complex shapes. *Composites Part A: Applied Science and Manufacturing* 2019;126:105568.
- [15] Šimáček P, Advani SG. A continuum approach for consolidation modeling in composites processing. *Compos Sci Technol* 2020;186:107892.
- [16] Belnoue JP-H, Nixon-Pearson OJ, Thompson AJ, Ivanov DS, Potter KD, Hallett SR. Consolidation-Driven Defect Generation in Thick Composite Parts. *Journal of Manufacturing Science and Engineering* 2018;140 071006-071015.
- [17] Belnoue JP-H, Hallett SR. A rapid multi-scale design tool for the prediction of wrinkle defect formation in composite components. *Mater Des* 2020;187:108388.
- [18] Hubert P, Poursartip A. A Review of Flow and Compaction Modelling Relevant to Thermoset Matrix Laminate Processing. *Journal of Reinforced Plastics and Composites* 1998;17:286–318.
- [19] Engmann J, Servais C, Burbidge AS. Squeeze flow theory and applications to rheometry: a review. *J Nonnewton Fluid Mech* 2005;132:1–27.

- [20] Harrison P, Lin H, Ubbink M, Akkerman R, Van De Haar K, Long AC. Characterising and modelling tool-ply friction of viscous textile composites, in: ICCM International Conferences on Composite Materials; 2007.
- [21] Belnoue JP-H, Nixon-Pearson OJ, Ivanov D, Hallett SR. A novel hyper-viscoelastic model for consolidation of toughened prepregs under processing conditions. *Mechanics of Materials* 2016;97:118–34.
- [22] Kelly PA. A viscoelastic model for the compaction of fibrous materials. *The Journal of The Textile Institute* 2011;102:689–99.
- [23] Kelly PA, Umer R, Bickerton S. Viscoelastic response of dry and wet fibrous materials during infusion processes. *Composites Part A: Applied Science and Manufacturing* 2006;37:868–73.
- [24] Servais C, Michaud V, Manson J-AE. The packing stress of impregnated fiber mats. *Polym Compos* 2001;22:298–311.
- [25] Belnoue JP-H, Mesogitis T, Nixon-Pearson OJ, Kratz J, Ivanov DS, Partridge IK, Potter KD, Hallett SR. Understanding and predicting defect formation in automated fibre placement pre-preg laminates. *Composites Part A: Applied Science and Manufacturing* 2017;102:196–206.
- [26] Rogers TG. Squeezing flow of fibre-reinforced viscous fluids. *J Eng Math* 1989;23:81–9.
- [27] Moon C-K, Um Y-S, Lee J-O, Cho H-H, Park C-H. Development of thermoplastic prepreg by the solution-bond method. *J Appl Polym Sci* 1993;47:185–8.
- [28] Chandler HD. A physical basis for non-Newtonian power-law viscosity. *Soft Mater* 2019;17:137–42.
- [29] Hermann T, Miene S, Henke T, Schelte A, Kelly PA, Bickerton S. Investigating flow mechanics within a novel nondestructive injectability measurement for fibre preforms. in 2017.
- [30] Kruckenberg T, Paton R. Compaction of dry and lubricated performs, in: seventh International Conference on Flow Processes in Composite Materials (FPCM7). Newark DE, USA 2004:425–36.
- [31] Hexcel, HexPly® 8552, Epoxy matrix (180 °C/356°F curing matrix), in, Hexcel Corporation, https://www.hexcel.com/user_area/content_media/raw/HexPly_8552_eu_DataSheet.pdf, 2016.
- [32] Hexcel, HexPly® M21, 180 °C (350°F) curing epoxy matrix, in, Hexcel Corporation, https://www.hexcel.com/user_area/content_media/raw/HexPly_M21_global_DataSheet.pdf, 2015.
- [33] Hexcel, HexPly® 913, 125 °C curing epoxy matrix, in, Hexcel Corporation, https://www.hexcel.com/user_area/content_media/raw/HexPly_913_eu_DataSheet.pdf, 2016.
- [34] Valverde MA, Kupfer R, Kawashita LF, Gude M, Hallett SR. An experimental study on preform evolution during thermoplastic composite overmoulding, in: Twenty-second International Conference on Composite Materials (ICCM22), Melbourne, Australia; 2019.
- [35] Lukaszewicz D. Considerations for automated layup of carbon-fibre thermoset preimpregnates, in: aerospace engineering. University of Bristol; 2011.
- [36] D. Van Ee, A. Poursartip, HexPly 8552, Material Properties Database for use with COMPRO CCA and Raven, in, 2009.
- [37] ten Thije RHW, Akkerman R, van der Meer L, Ubbink MP. Tool-ply friction in thermoplastic composite forming. *International Journal of Material Forming* 2008;1:953–6.
- [38] Matveev MY, Belnoue JP-H, Nixon-Pearson OJ, Ivanov DS, Long AC, Hallett SR, Jones IA. A numerical study of variability in the manufacturing process of thick composite parts. *Compos Struct* 2019;208:23–32.
- [39] Thompson AJ, El Said B, Ivanov D, Belnoue JP-H, Hallett SR. High fidelity modelling of the compression behaviour of 2D woven fabrics. *Int J Solids Struct* 2018;154:104–13.
- [40] Courtney TH. Mechanical behavior of materials: second edition. Waveland Press; 2005.
- [41] Skartsis L, Khomami B, Kardos JL. Resin flow through fiber beds during composite manufacturing processes. Part II: numerical and experimental studies of newtonian flow through ideal and actual fiber beds. *Polymer Engineering & Science* 1992;32:231–9.
- [42] Veldenz L, Di Francesco M, Giddings P, Kim BC, Potter K. Material selection for automated dry fiber placement using the analytical hierarchy process. *Advanced Manufacturing: Polymer & Composites Science* 2018;4:83–96.
- [43] Liu Y-N, Yuan C, Liu C, Pan J, Dong Q. Study on the resin infusion process based on automated fiber placement fabricated dry fiber preform. *Sci Rep* 2019;9:7440.
- [44] Lectez A-S, El Azzouzi K, Binetruy C, Comas-Cardona S, Verron E, Lebrun J-M. Three-dimensional mechanical properties of dry carbon fiber tows subjected to cyclic compressive loading. *J Compos Mater* 2018;52:2661–77.
- [45] Somashekar AA, Bickerton S, Bhattacharyya D. Exploring the non-elastic compression deformation of dry glass fibre reinforcements. *Compos Sci Technol* 2007;67:183–200.
- [46] Hexcel, HiTape Reinforcements, in, Hexcel, https://www.hexcel.com/user_area/content_media/raw/HiTape.pdf, 2016.
- [47] Federico S, Herzog W. On the permeability of fibre-reinforced porous materials. *Int J Solids Struct* 2008;45:2160–72.