

# ECCM21

02-05 July 2024 | Nantes - France

## Proceedings of the 21<sup>st</sup> European Conference on Composite Materials



**Vol 5**



## Manufacturing

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**Published by:**

The European Society for Composite Materials (ESCM) and the Ecole Centrale de Nantes.

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These Proceedings have an ISBN (owned by the Publisher) and a DOI (owned by the Ecole Centrale de Nantes).

ISBN: 978-2-912985-01-9

DOI: [10.60691/yj56-np80](https://doi.org/10.60691/yj56-np80)

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## Editorial

Each volume gathers contributions on specific topics:

- Vol 1. Industrial applications**
- Vol 2. Material science**
- Vol 3. Material and Structural Behavior – Simulation & Testing**
- Vol 4. Experimental techniques**
- Vol 5. Manufacturing**
- Vol 6. Multifunctional and smart composites**
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**This collection** contains the proceedings of the 21st European Conference on Composite Materials (ECCM21), held in Nantes, France, July 2-5, 2024. ECCM21 is the 21st in a series of conferences organized every two years by the members of the European Society of Composite Materials (ESCM). As some of the papers in this collection show, this conference reaches far beyond the borders of Europe.

The ECCM21 conference was organized by the Nantes Université and the Ecole Centrale de Nantes, with the support of the Research Institute in Civil and Mechanical Engineering (GeM).

Nantes, the birthplace of the novelist Jules Verne, is at the heart of this edition, as are the imagination and vision that accompany the development of composite materials. They are embodied in the work of numerous participants from the academic world, but also of the many industrialists who are making a major contribution to the development of composite materials. Industry is well represented, reflecting the strong presence of composites in many application areas.

With a total of 1,064 oral and poster presentations and over 1,300 participants, the 4-day event enabled fruitful exchanges on all aspects of composites. The topics that traditionally attracted the most contributions were fracture and damage, multiscale modeling, durability, aging, process modeling and simulation and additive manufacturing.

However, the issues of energy and environmental transition, and more generally the sustainability of composite solutions, logically appear in this issue as important contextual elements guiding the work being carried out. This includes bio-sourced composites, material recycling and reuse of parts, the environmental impact of solutions, etc.

We appreciated the high level of research presented at the conference and the quality of the submissions, some of which are included in this collection. We hope that all those interested in the progress of European composites research in 2024 will find in this publication sources of inspiration and answers to their questions.

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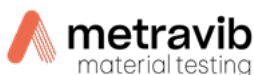


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# ANISOTROPIC WARPAGE PREDICTION OF INJECTION MOLDED PARTS WITH PHENOLIC MATRIX

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**Keywords:** Short Fiber Reinforced Phenolic, Injection Molding Simulation, Warpage Analyses, CHILE-Model

## Abstract

Injection Molding is one of the most important processes to manufacture short fiber reinforced composites. During mold filling the fibers orientate depending on the flow field. The final fiber orientation influences the thermo-mechanical behavior of the part. During the holding stage, the matrix solidifies from fluid to solid, having crucial impact on the mechanical attributes and causing thermal and chemical shrinkage. The combination of these effects leads to residual stresses and warpage of the injection molded part, which may lead to dysfunctionality and waste production.

One strong tool to minimize warpage in an early stage of part design is process simulation. Based on a virtual prediction, tool correction cycles, dysfunctional parts and therefore costs and production energy can be reduced. However, such prediction models need adequate material and process modeling, accounting for the anisotropic and thermo-visco-elastic material behavior. This work presents an approach to model warpage of short fiber reinforced phenolic parts, by a combination of a CHILE-approach for the matrix and orientation averaging of mean field homogenized properties to consider the fiber orientation. Fiber orientation, temperature and curing field are determined in a preceding mold filling simulation.

## 1. Introduction

In a changing economy, where reduction of production energy, waste and carbon emissions becomes more and more important, it is crucial to produce parts cost- and energy-efficient, with most efficient material use. This applies especially for short fiber reinforced polymer composites, showing a high lightweight potential with simultaneous possibility to be produced at low temperatures (compared to metal alloys) and therefore reduce the energy effort in production and use phase.

To reduce cost and production energy, simulation is an important tool. When simulating fiber reinforced polymers (FRPs), it is important to consider information of the manufacturing process like fiber orientation, material state and temperature field. The fiber orientation introduces anisotropic material behavior on macro-scale, having crucial impact on warpage. The importance of using process information is shown by Kärger et al. [1] for continuous reinforced polymers as well as by Görthofer et al. [2] and Meyer et al. [3] for sheet molding compound. Despite that, a warpage analysis also requires a matrix model, being able to capture the significant changes, which the matrix undergoes during solidification. Hence, the matrix model should at least be a function of temperature and degree of cure, or a fully thermo-viscoelastic approach [4]. Unfortunately, such thermo-viscoelastic approaches require



a high amount of computational effort and needed experimental data, over a wide range of temperatures and frequencies. Therefore, the so-called cure hardening instantaneously linear elastic (CHILE) approaches [5] represent a compromise of needed data and model accuracy. Bernath [4] showed that CHILE-approaches are capable to model the behavior of continuous FRPs for holding, curing and cooling stage, especially since the material is heated up and cooled down in a monotonic way. Furthermore, Bernath [4] names the ejection forces and demolding procedure to have crucial impact on the part's warpage, in addition to the named process phases.

A state of the art process simulation only considers orientation tensors to compare the fiber orientation field as presented by Advani and Tucker [6]. Therefore, a complete and unequivocal reconstruction of the micro-state is not possible. On macro-scale, the anisotropic material behavior and therefore the influence of fibers is considered by homogenization approaches for matrix and fibers. According to [6] the information provided by the second- and forth-order orientation tensor is sufficient for modeling the anisotropic material behavior for non-bending fibers.

To model the residual stresses and warpage of short fiber reinforced phenolics during holding, ejection and cooling, this work presents a combination of a CHILE-approach for matrix modeling with orientation averaging of mean field homogenized properties to consider fiber-induced anisotropy. Additionally, chemical and thermal shrinkage are considered.

## 2. Material modeling

### 2.1. CHILE-model for mechanical behavior

To capture polymer phenomena like creeping and relaxing over a wide range of temperatures and frequencies, a fully thermo-visco-elastic approach would be necessary, calculating the mechanical stress  $\sigma_{ij}$  at time  $t$  by

$$\sigma_{ij} = \int_0^t G_{ijkl}(t - \xi) \frac{d\varepsilon_{kl}(\xi)}{d\xi} d\xi, \quad (1)$$

where  $\varepsilon_{kl}$  is the strain tensor,  $\xi$  represents the time integration variable and  $G_{ijkl}$  the relaxation modulus tensor, which can be approximated with a Prony series for example.

In a CHILE-approach, the material history is neglected and the mechanical stress is given by

$$\sigma_{ij} = \int_0^t C_{ijkl}(T(\xi), c(\xi)) \frac{d\varepsilon_{kl}(\xi)}{d\xi} d\xi, \quad (2)$$

with  $T$  being the temperature and  $c$  the degree of cure. The elastic stiffness  $C_{ijkl}$  is defined as

$$C_{ijkl} = \begin{cases} C_{ijkl}^{cr}, & \Delta T_1 \\ C_{ijkl}^{cr} + \frac{T_g(c) - T - T_{c1}}{T_{c2} - T_{c1}} (C_{ijkl}^{cg} - C_{ijkl}^{cr}), & \Delta T_2. \\ C_{ijkl}^{cg}, & \Delta T_3 \end{cases} \quad (3)$$

Here  $T_g(c)$  is the glass transition temperature, depending on the actual state of cure. Furthermore,  $\Delta T_1 \in T_g(c) - T < T_{c1}$ ,  $\Delta T_3 \in T_g(c) - T > T_{c2}$  and  $\Delta T_1 \leq \Delta T_2 \leq \Delta T_3$ , where  $T_{c1}$  and  $T_{c2}$  represent material specific parameters.  $C_{ijkl}^{cr}$  and  $C_{ijkl}^{cg}$  are the instantaneous elastic moduli in pure rubbery and pure glassy state. This approach can be extended by defining  $C_{ijkl}^{cr}$  and  $C_{ijkl}^{cg}$  as function of temperature and degree of cure, which is not applied within this work due to a lack of data.

One disadvantage of the CHILE-approach is that no frozen-in stresses are released when rising over  $T_g$ , since the material history is neglected. However, due to the thermal history of the process route, this aspect is uncritical for the regarded process and material combination [4].

## 2.2. Modeling of curing kinetics

The curing kinetics are modeled with the Kamal-Sourour approach [7], determining the change of degree of cure by

$$\frac{dc}{dt} = (K_1 + K_2 c^m)(1 - c)^n, \quad (4)$$

with

$$K_{1,2} = A_{1,2} \cdot \exp\left(\frac{-E_{1,2}}{R_g \cdot T}\right), \quad (5)$$

where  $A_1, A_2, m$  and  $n$  are material specific parameters,  $E_1$  and  $E_2$  are activation energies and  $R_g$  represents the universal gas constant.

The glass transition temperature is depending on the degree of cure and calculated according to DiBenedetto [8] by

$$T_g = T_{g,0} + \frac{(T_{g,\infty} - T_{g,0})\kappa_{Tg}c}{1 - (1 - \kappa_{Tg})c}, \quad (6)$$

with  $T_{g,0}$  and  $T_{g,\infty}$  as  $T_g$  corresponding to  $c = 0$  and  $c = 1$  and  $\kappa_{Tg}$  representing a material-specific modeling parameter.

## 2.3. Homogenization and orientation averaging

The elastic moduli for rubbery and glassy state  $E_{ijkl}^{cr}$  and  $E_{ijkl}^{cg}$  introduced in Section 2.1 represent the homogenized values of matrix and fibers. The values are determined according to Tandon and Weng [9] similar to the work Görthofer et al. [2].

The thermal conductivity is determined for a transversal isotropic material consisting of fibers and matrix in a first step. According to Clayton [10] the thermal conductivity in fiber direction  $\lambda_{\parallel}$  is given by

$$\lambda_{\parallel} = \Phi_f \lambda_f + (1 - \Phi_f) \lambda_M, \quad (7)$$

with  $\Phi_f$  being the fiber volume fraction and  $\lambda_f$  and  $\lambda_M$  being the thermal conductivity of fibers and matrix. Accordingly, the thermal conductivity perpendicular to fiber direction  $\lambda_{\perp}$  is given by

$$\lambda_{\perp} = \frac{\lambda_M}{4} \left( \sqrt{(1 - \Phi_f)^2 \left(\frac{\lambda_f}{\lambda_M} - 1\right)^2 + 4 \frac{\lambda_f}{\lambda_M} - (1 - \Phi_f) \left(\frac{\lambda_f}{\lambda_M} - 1\right)} \right)^2. \quad (8)$$

The thermal expansion is also defined in fiber direction  $\vartheta_{\parallel}^{th}$  and perpendicular  $\vartheta_{\perp}^{th}$  according to Schapery [11] by

$$\vartheta_{\parallel}^{th} = \frac{\Phi_f E_f \vartheta_{th,f} + (1 - \Phi_f) E_M \vartheta_{th,M}}{\Phi_f E_f + (1 - \Phi_f) E_M}, \quad (9)$$

and

$$\vartheta_{\perp}^{th} = (1 + \nu_f) \Phi_f \vartheta_{th,f} + (1 + \nu_M) \Phi_M \vartheta_{th,M} - \overline{\vartheta_{th}} \bar{\nu}, \quad (10)$$

with  $E_f, \vartheta_{th,f}, E_M$  and  $\vartheta_{th,M}$  being elastic modulus and thermal expansion of fibers and matrix,  $\nu_f$  and  $\nu_M$  are the poisson ratios of fibers and matrix.  $\overline{\vartheta_{th}}$  and  $\bar{\nu}$  represent the volume averaged thermal expansion coefficient and poisson ratio.

At this point, the elastic moduli, thermal expansion and conductivity are defined for a transverse isotropic homogenized fiber matrix compound. However, this is not valid for most orientation

states of short fiber reinforced polymers. Therefore, these values are further orientation averaged, like presented by Advani and Tucker [6], with information provided by the orientation tensor.

The specific heat capacity and chemical shrinkage are not orientation-dependent and therefore determined by simple volume averaging. The chemical shrinkage of the fibers is zero in this case.

### 3. Simulation model and results

#### 3.1. Geometry, process conditions and material

The regarded geometry is a rectangular plate with 480 mm length, 190 mm width and 3 mm height. The material is injected via a 185 mm long sprue, with start a diameter of 9 mm and an end diameter of 15.5 mm, positioned in the center of the plate.

The analysis contains the process steps holding, curing, ejection and out-of-tool cooling. Temperature, curing field and fiber orientation are determined in a preceded mold filling simulation (see Section 3.2). Two different cases are simulated for the curing time, being 10 s and 60 s. Furthermore, both curing durations are simulated with and without ejection step, resulting in four different simulations. The ejection step is approximated by a displacement of 20 mm of the plate's corners in positive  $x_3$ -direction between end of curing and start of out-of-tool cooling for a duration of 3 s. During the 40 s holding and following curing, the tool temperature is assumed to be constant 170 °C, realized by a Dirichlet boundary condition and displacement of all surface nodes is disabled. For the ejection and out-of-mold cooling, with a duration of 6000 s, a convective boundary condition is applied at the surface, approximating a heat exchange with the environment, having constant 20 °C and a heat transfer coefficient of 15 W/(m<sup>2</sup>K). During ejection and out-of-mold cooling the nodes at the starting surface of the sprue are fixed, to prevent a movement of the complete part.

The simulated material is a 55 %-weight short glass fiber filled phenolic compound. The fibers are assumed to have identical length and an aspect ratio of 15. The warpage analysis is performed within *Simulia Abaqus 2018* and the Material modeling is realized in a UMAT Subroutine [12].

#### 3.2. Preceded mold filling simulation and data transfer

The information about fiber orientation as well as temperature and curing field are determined in a preceded process simulation. The simulation model and process conditions are described in [13].

Since the mold filling simulation and warpage analysis are performed on different meshes, the fields of temperature, curing and fiber orientation must be mapped, to make the data usable on the warpage analysis mesh. MPCCI MapLib [14,15] is used to map these data, which is also successfully applied in [2,16] for orientation mapping in context of SMC material.

According to the orientation averaging, each different orientation tensor would create a different orthotropic material behavior. This would result in a number of materials equal to the number of elements, coming along with high numerical effort. Hence, the orientation tensors are spectrally decomposed such that the eigenvectors can be used as local coordinate systems and the eigenvalues describe the degree of anisotropy. The eigenvalues are clustered with a k-means algorithm in the eigensystem down to 30 clusters corresponding to one representative orthotropic material for each cluster [12].

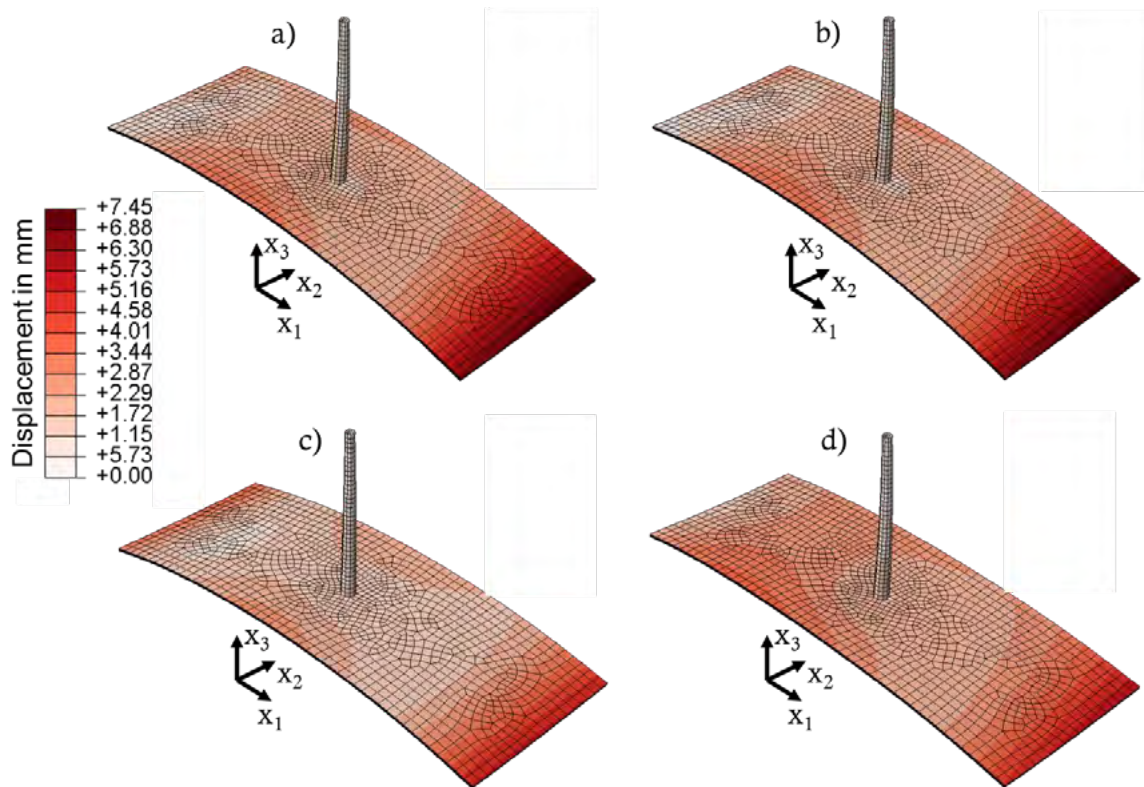
#### 3.3. Results of the warpage analysis

The results of the four different simulations are shown in Fig. 1. In general, the plate deforms to a convex shell with a maximum displacement of nearly 7.5 mm in case of 60 s curing.

However, due to lack of material and experimental data, the absolute values are not validated. The displacement after 60 s curing is identical for the simulation with and without ejection step (Fig. 1 b) and a)). After 60 s curing time, the degree of cure is  $c > 0.75$  in the whole domain and even  $c > 0.9$  in the plate. Hence it is  $T_g > 190$  °C and the matrix behavior is constant and identical in the complete part (cf. Eq. (3)). Consequently, the ejection deformation has no influence on the warpage, due to the elastic behavior. The warpage is a purely result of residual stresses due to thermal and chemical shrinkage in combination with anisotropy.

If only 10 s curing time are applied, the results with and without ejection step differ from each other, since the material behavior is not linear elastic during the ejection. For an ejection after 10 s curing it is  $T_g < 170$  °C in most regions of the part, so the material behavior different within the part and partially depending on  $T$  and  $T_g$ . The material behavior is changing during the ejection deformation, resulting in higher deformations in Fig. 1 d) compared to Fig. 1 c).

A shorter curing time results in less frozen in stresses at the point of ejection and therefore the deformations are lower in the simulation, since the material is still in the rubbery state, where less residual stresses built up. This would not be the case in reality, since effects like movement of the part or gravity, which would act over a longer period of time, would have higher influence on the less cured part with entropy elastic behavior.



**Figure 1.** Simulated displacement of the plate after holding, curing, ejection and cooling. The displacements are scaled 5 times to visualize the convex shell. a) 60 s curing without ejection step, b) 60 s curing with ejection step, c) 10 s curing without ejection step, d) 10 s curing with ejection step [12]

In summary, the CHILE-approach in combination with anisotropic material modeling is able to capture residual stresses and warpage with respect to process parameters, fiber orientation,

curing and temperature for injection molded parts with short fiber reinforcement in a meaningful way.

#### 4. Conclusion

A combination of CHILE-approach for matrix behavior with anisotropy due to fiber orientation to model the warpage of injection molded short fiber reinforced phenolic parts was presented. The process steps holding, curing, ejection and out-of-tool cooling have been simulated. Besides the mechanical behavior, also thermal conductivity and expansion are modeled in an anisotropic way. A preceded mold filling simulation provided the information about fiber orientation, temperature and curing field, which is mapped to be used within the warpage analysis. Four different simulations with different curing times and ejection deformations are compared, showing that the approach is able to capture the influence of the process conditions. This represents a first step for accurate warpage analysis of fiber reinforced injection molded parts. A more complex matrix modeling, like a path-dependent or fully visco-elastic approach, would be meaningful to capture the complex behavior, especially during the solidification process. The fiber length has crucial impact on the material behavior and fiber length distributions should therefore be recognized in the homogenization. Furthermore, the heat exchange between the part and tool is of great importance and should be modeled with a high level of detail, which also includes the eventual peeling of the part from the tool wall due to shrinkage.

#### Acknowledgments

We thank the ‘Ministerium für Wissenschaft, Forschung und Kunst Baden-Württemberg’ and the ‘Ministerium für Wirtschaft, Arbeit und Wohnungsbau Baden-Württemberg’ for the financial support of the ‘Profilregion’, which initialized the method development within this work, as well as the German Research Foundation for funding the project ‘MeproSi’ (project number: 464119659), where the work has been continued with focus on the process induced fiber orientation.

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02-05 July 2024 | Nantes - France

## Volume 5 **Manufacturing**



ISBN: 978-2-912985-01-9

DOI: 10.60691/yj56-np80