ON THE PREDICTIVE ANALYSIS OF FINAL QUALITY OF COMPOSITE PARTS: GAP BETWEEN AVAILABLE MODELS AND INDUSTRIALS NEEDS

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Abstract
Composite parts manufacturing is characterized by the diversity of processes and by the complexity of material transformations occurring along the process chain. Considering the RTM-process as an illustrative example of industrial application for composites, some challenges for the reliable virtual design of a robust process can be identified.

A deep understanding of the ongoing phenomenon and the capability to predict them can be seen as an enabler for the improvement of part and process quality, but also as a leverage for accelerated development and for cost reduction.

The present paper deals with the assessment of well-established predictive models for composites with respect to physics and to implementation. Furthermore, it also highlights the gaps between expectations from industry and the model output.

The paper resumes the common modelling assumptions for thermo-mechanical analysis adapted to the RTM-process first, followed by a discussion of typical software-driven limitations. The models are discussed with respect to the conditions for material characterization, to the derivation of applicable constitutive laws, and finally with respect to the handling of boundary conditions. The proper formulation of final set of equations taking into account the coupling of the involved physics field is also emphasized.

Dedicated simulation examples performed with representative computation tools are finally discussed to illustrate the gaps between current prediction capabilities and physical observations. The last part of the contribution is dedicated to the short survey of progresses on the field of process monitoring. The focus is set on the meaning of some recent advances for the review of some critical model assumptions.

1. Introduction

In the case of the resin transfer molding (RTM) process for production of composite parts, liquid resin is injected into a dry fibrous preform in a closed mold. After end of injection, resin is cured under temperature. Currently this process is used for production of high-performance but generally small-scaled composite parts and allows high manufacturing rates with very good part quality. Main application fields of this technique are automotive and aerospace industry.

Thermo-mechanical analysis of the process is essential for cost effective and fast development of materials, processes and tools. Especially for thick parts, it is mandatory to achieve required inner part and surface quality. During curing and cooling of the resin, resin shrinkage and residual stresses occur which affects strength, geometrical shape and outer quality of the part. In literature, several models as well as numerical and experimental investigations of RTM, vacuum assisted resin infusion, prepreg and pultrusion processes can be found [1]–[11]. On the basis of the analysis of pultrusion process from Baran [5], a typical procedure for calculation of strain and stress is shown. Temperature field of tool and composite part during the process is calculated using the heat equation. Rate of reaction and curing degree is calculated using semi empirical equations like nth order equation, Kamal Sourour or similar approaches [10], [12], [13]. Here the Arrhenius equation describes the temperature dependence of reaction velocity k and one or more exponential variables describe the order of reaction according to actual degree of cure. If needed, an additional diffusion term is used for more accurate prediction of degree of cure after vitrification [9]. Di Benedetto equation is the commonly used approach for calculation of glass transition temperature T_g in dependence of degree of cure α. For calculation of resin modulus, CHILE model is used [9]. Here the modulus is constant at liquid and cured resin. During vitrification resin modulus depends on temperature difference to actual T_g. Thermal and chemical strains and stresses, as well as moduli and Possion´s ratio of the composite are calculated using model of Bogetti and Gillespie [4].

The commonly used and presented approaches for thermos-mechanical analysis exhibit needs of further investigation for reliable application to industrial needs. One point are the rules of mixture for determination of combined material properties of fiber and matrix system [14]. Baran and Chachad use the mass fraction method for determination of heat capacity and thermal conductivities while pure material is used for measuring those values [5], [15].

Typically, thermos-mechanical studies refer to smaller scaled RTM or pultrusion parts [5], [16]. Scalability of the models, especially for larger and thicker parts is not examined in detail. In aerospace industry, actual requirements show the need for large and complex parts, which imply predictive capabilities to resize traditional manufacturing processes on a reliable way. In other words, the virtual capability to anticipate and to reduce the risk of rescaling existing solutions is clearly needed.

McHugh shows in his study about measurement of specific heat capacity the influence of curing degree on this parameter [17]. In most thermos-mechanical analysis of curing of composite parts, such the analysis of pultrusion process [5], the effective physical properties like density, specific heat and thermal conductivities are measured at fully cured resin samples. The change of material properties according to curing degree is not considered. Influence of this parameter and comparison of different approaches have to be evaluated [18]. Currently, curing in industrial processes is controlled by determined temperature profiles for monitoring reasons. Here reliable models for a time –dependent degree of cure driven process are important. Using a state variable as unique identifier of the effective material properties at each point of the part subjected to predictive analysis ensure a proper behavior according to the local thermal history.

In this study challenges of tooling design for large scaled RTM parts are discussed. Based on thermos-mechanical analysis of pultrusion process [5] influences of variation of material parameters while curing are investigated. Impact on heat contribution and exothermic reaction is calculated and compared when changing heat capacity and thermal conductivities of the materials.

2. Governing Equations and Model Implementation

Implementation of governing equations to investigate influence of curing degrees to material properties and process parameters is shown at the example of a pultrusion process. The used model is already examined by different authors [5], [7], [19]–[21]. Thus, extensive data basis and comparable studies are available. This model is chosen because there are various conformities with the RTM process of large scaled parts in aerospace industry. In both cases, relative thick parts are cured with a 180°C epoxy based thermoset matrix system. Also, manufacturing occurs in a thick metallic tooling, which has to be included to mechanical and heat simulation.

Abaqus finite element (FEM) simulation environment is used for conducting thermo-mechanical analysis of the curing process of composite materials. Based on Abaqus standard thermal-displacement analyses, user subroutines are implemented for calculation of glass transition temperature T_g, curing degree, and internal heat generation. Also calculation of thermal and chemical strain, as well as stiffness matrix for determination of stresses are implemented manually.

Figure 1 shows the used 3D model for calculation of Temperature field, degree of cure, resin modulus and glass transition Temperature. Stress and strains are modeled in a 2D model which represents the cross-section of the beam [5]. The moving of the die through the part is set as a part displacement with constant velocity U of 20 cm/min. Quadratic mesh with element CPE4T and size of 5-10mm in x_1 and x_2 and 15mm in x_3 direction is used.

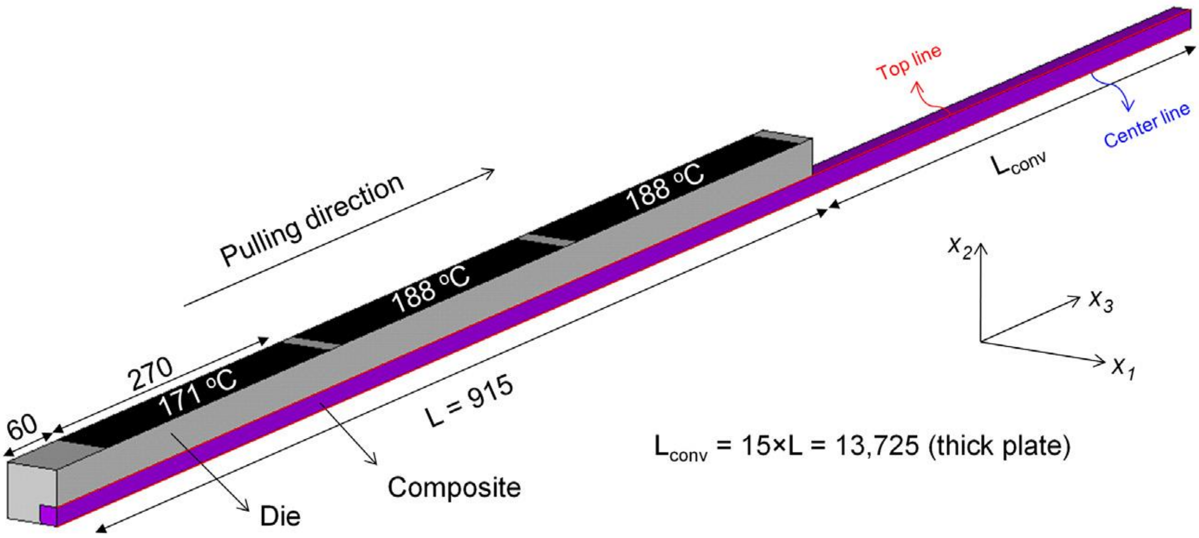

Figure 1. 3D Abaqus model of pultrusion process representing one quarter of the original setup with tooling (grey) and composite die (purple) [5]

The transient energy equations are solved by Abaqus standard module in both, the tool and the die according to Eqs. (1). The heating sources with constant temperatures of 171°C and 188°C are set as boundary conditions as shown in Figure 1. At the outer surfaces of die and tool a convective cooling to environment with ambient temperature of 27°C is applied.

$$\rho_d Cp_d \frac{\partial T}{\partial t} = k_{x1,d} \frac{\delta^2 T}{\delta x_1^2} + k_{x2,d} \frac{\delta^2 T}{\delta x_2^2} + k_{x3,d} \frac{\delta^2 T}{\delta x_3^2} \quad (1)$$

User subroutine “user defined field” (USDFLD) is implemented for calculation of main field variables. Runge-Kutta method allows solving of the differential equation of curing degree α which is shown in Eqs. (2) [5], [13], [22]. Nth order reaction with Arrhenius equation for calculation of velocity constant and implementation of diffusion part for consideration of curing after vitrification is implemented [4].

$$R_r(\alpha) = \frac{d\alpha}{dt} = \frac{1}{H_{tr}} \frac{dH(t)}{dt} = K_0\, e^{-\frac{E}{RT}} (1-\alpha)^n \frac{1}{1 + e^{C(\alpha - (\alpha_{C0} + \alpha_{CT} T))}} \tag{2}$$

Here K_0 is the pre-exponential constant, E activation energy and R the universal gas constant. n is the reaction order. At the diffusion part of the equation C is a fitting constant. α_{C0} represents the critical degree of cure at a temperature of T=0K and α_{CT} is the increase of curing degree related to temperature. The parameters of the semi-empirical model are obtained by fitting of curves from measurements using dynamic scanning calometry (DSC).

Glass transition temperature is calculated using Di Benedetto equation as expressed in Eq. (3) [23]. The material constants T_{g0} and $T_{g\infty}$ are glass transition temperatures of matrix system at uncured and fully cured state. λ is a fitting constant obtained from DSC measurements.

$$\frac{T_g - T_{g0}}{T_{g\infty} - T_{g0}} = \frac{\lambda\alpha}{1 - (1-\lambda)\alpha} \tag{3}$$

Resin modulus E_r is assumed to be isotropic and calculated using CHILE model [9]. Here a liner influence of cure degree is coupled with temperature influenced. It is expressed with Eqs. (4) and (5). E_r^0 and E_r^∞ are moduli of cured and uncured resin, whereby E_r^0 is assumed as $E_r^\infty/1000$ [1], [8], [11]–[13]. T_{C1} and T_{C2} are the temperature of beginning and end of glass transition.

$$T^* = T_g - T \tag{4}$$

$$E_r = \begin{cases} E_r^0 & T^* < T_{C1} \\ E_r^0 + \frac{T^* - T_{C1}}{T_{C2} - T_{C1}} (E_r^\infty - E_r^0) & T_{C1} \le T^* \le T_{C2} \\ E_r^\infty & T^* > T_{C2} \end{cases} \tag{5}$$

Exothermic behavior of resin q is calculated using subroutine “HETVAL” with Eqs. (6) as function of total heat of reaction of resin H_{tr}, which is determined with differential scanning calometry (DSC), and change of cure Δα. V_f is the fibre volume content of the laminate [5], [15].

$$q = (1 - V_f)\rho_r * H_{tr} * \frac{d\alpha}{dt} \tag{6}$$

For application of existing models to larger parts, it has to be examined where new effects become visible and where assumptions are not sufficiently accurate anymore. Possible influences of curing degree to material properties shall be investigated in this work. Especially for isothermal RTM processes, thermal behavior and resin curing has to be calculated coupled to resin flow simulation.

For industrial applications, the simulation software has to be improved regarding ergonomics and efficiency. Especially the implementation of user defined models is laboriously and prone to errors. Here a modular setup would fit to the needs. Also, handling of large models has to be adapted. E.g. a coupling of 3D and 2D models is not possible for complex parts.

3. Application of models for Tool design

In composite molding processes, shape and design of the tooling define large parts of the process. Some important functionalities of the hard tool among others are to shape the semi-finished product, to provide a constant cavity despite the applied vacuum or during the injection step (at higher pressure), and finally to support the customized positioning of inlets and outlets as well as resin distribution channels.

RTM tooling for producing small scaled RTM parts are used in various applications e.g. for automotive and aerospace industry. The requirements for those tools are relatively well studied. Tools for producing larger parts show new characteristics, failure potentials and challenges, which are quite unknown up to now. Proper handling of thermal expansion of the tooling material shows a mean effect on part quality. Whilst a 1m steel die has an expansion of 1.6mm when heating from 20 to 180°C, for larger tools e.g. 10m, the resulting thermal dilatation is 16mm. In addition, in many cases because of large flow length at large scaled parts injection temperature of resin has to be below curing temperature and isothermal processes are not possible anymore. Here, various tool shapes at different temperatures while equipping, injection and curing are difficult to handle.

Thick and large-scaled tools normally have to be produced in components which are welded or glued. This complicates the numerical prediction of thermal expansions as the built-concept results into a kind of geometrical anisotropy. Also, distribution of residual stresses is not well examined up to now.

Heat conductance and specific heat capacities have mayor influences on heating time and required energy. A homogenous heating of the mold is essential for an effective, controllable curing and for minimizing residual stresses. If one side of the part has very high surface requirements, one side of the mold has to have a slightly higher temperature. Here an accurate temperature control, regulated by curing degree has to be realized. For large tools, the weight of the tooling gets important for heating and handling. In many applications, the required stiffness of the mold is realized though the increase of wall thickness; this leads to the manufactury of heavy solutions, with inherent low thermal performance Therefore, thinner tools with a support structure are used often. Here a deformation of tool under acting clamping forces of the press and when opening the tool has to be avoided or considered in mould design.

Surface interaction between part and tooling is not well investigated until now. Especially friction while thermal and chemical shrinkage of part and tool as well as heat conduction between the surfaces is important for heat and stress calculation. At large scaled structural components because of larger surfaces, those parameters are more important.

For purposes of predictive design and sizing of an industrial production, i.e. application of the models to real parts, the wrong estimation of effective material properties resulting from different approaches for mixing rules has to be considered. Especially for large-scaled parts, accurate prediction of temperature field and influence of anisotropic material form the basis for all further calculations [18], [24].

4. Numerical analysis of the process

In state of the art calculations of thermo-mechanical process, material properties e.g. heat capacity, thermal conductance and densities of both fiber and resin are assumed constant over the whole process. In this study, thermo-mechanical calculations are made for investigation of influence of varying material properties on the process.

McHugh [17] shows influence of degree of cure on heat capacity of a composite material. Normally those values are measured with Macro TGA. Here small, fully cured resin samples are used. The change of heat capacity from uncured to fully cured material is changes between 17-24%. Thermal conductivities of fiber and resin treated as mixed material also can differ according to curing degree of the matrix system. Here Johnston shows a deviation of up to 30% [9].

Based on the thermo-mechanical modeling of pultrusion process which is well examined in different studies [5], [6], influence of a possible change of the materials properties heat capacity and thermal conductance is evaluated. Equation and calculation approach presented in chapter 2 and in study of Baran [5] are used. Table 1 shows conducted studies and properties for lumped materials calculated using a mixing approach. For the baseline models values of comparison papers are used [5].

Table 1. Material parameters for thermo-mechanical studies

Study	ρ_{Lumped} [kg/m³]	ρ_{resin} [kg/m³]	Cp [J/kg*K]	k_{11}/k_{22} [W/m*K]	k_{33} [W/m*K]
Original Model [5]	2090.7	1260	797.27	0.5592	0.9053
Cp+15%	2090.7	1260	916.8605	0.5592	0.9053
Cp-15%	2090.7	1260	677.6795	0.5592	0.9053
k+15%	2090.7	1260	797.27	0.64308	1.041095
k-15%	2090.7	1260	797.27	0.47532	0.769505

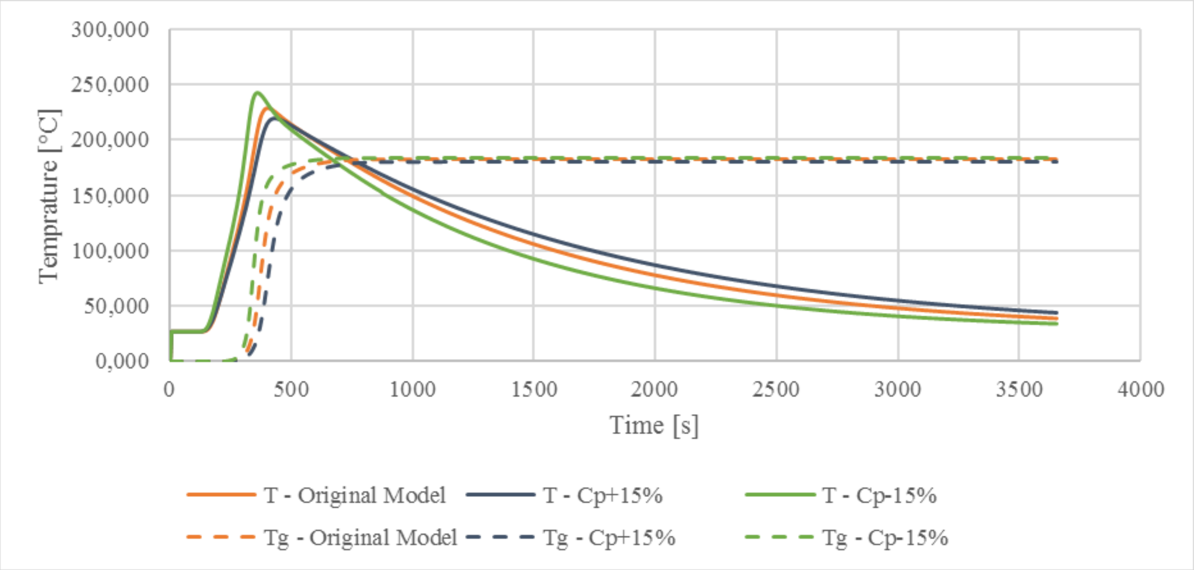

Figure 2. Calculated progression of temperature T and glass transition temperature Tg while pultrusion process with differing material properties

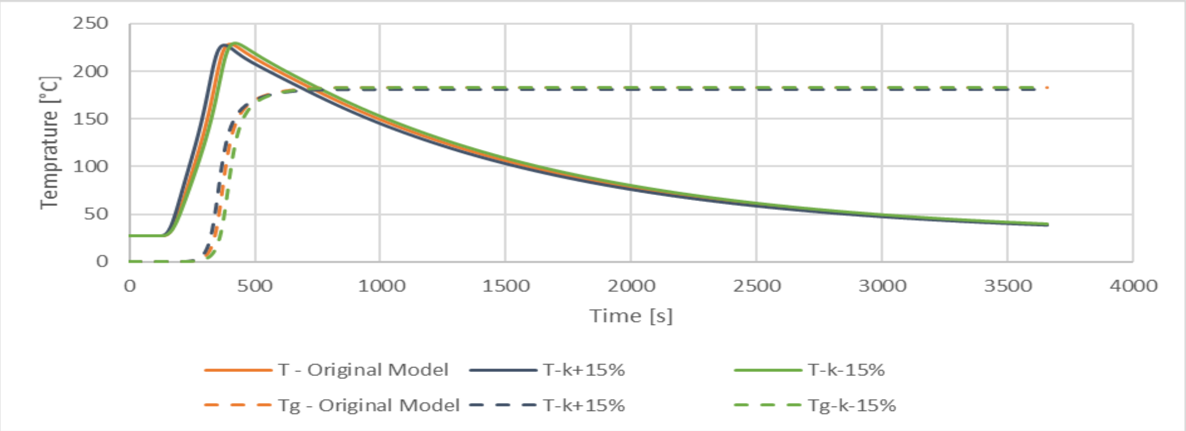

Figure 3. Temperature profile T and glass transition temperature Tg while pultrusion process with varying heat conductance

Figure 2 shows progression of temperature and glass transition temperature in the center of the die. Depending of the heat capacity the temperature peak caused by exothermal reaction differs from 219°C to 243°C. A higher heat capacity leads to a lower exothermal reaction and to a slower cooling of the part after leaving the tool. Temperature change also affects the glass transition temperature. Here a difference of 4°C is detected.

For investigation of influences of varying conductance on the curing process k_{11}, k_{22} perpendicular to fibers and k_{33} in fiber direction have been varied by 15% as shown in Table 1. Figure 3 shows a small impact on the process. The difference of maximum temperature is 2°C. At lower conductance temperature peak reaches center of the part 2 minutes later compared to higher conductance.

5. Conclusions

The study shows the consideration of tooling requirements for producing large scaled composite parts with resin transfer molding process. Here actual models and assumptions are not sufficient. Further investigations are required for effective and reliable numerical implementation of calculating thermo-mechanical behavior. Also, simulation environments have to be improved for fitting industrial needs. Common assumptions like mixing rules and dependency of material parameters on degree of cures have to be considered.

With FEM analysis, influence of differing material parameters on thermochemical curing behavior of an epoxy resin at the example of a pultrusion process is shown. Especially a change in the heat capacity affects temperature field, time and strength of exothermal reaction and cooling speed.

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