1 Introduction to the investigation of isothermal processes in the production of large-scaled composite structures

1.1 Introduction to the topic and objective of the thesis

In composite manufacturing, thermoset polymer matrix and high-strength fibres can be combined to produce lightweight structural parts. These materials are intensively used in the mobility sector, i.e. in the aerospace, naval and automotive industries. Potential applications include large composite shells (wings, fuselage), high-volume components such as stiffeners (Stringers, frames, cleats), and tanks (hydrogen tanks). Various manufacturing processes such as resin transfer moulding (RTM), pultrusion and prepregs are used for producing composite components [44, 65, 12].

The increasing demand for higher production rates of new, complex, and large high-performance composite parts brings new challenges concerning virtual capabilities of predicting process-induced distortion and deriving the proper design of the curing tool, as well as sizing the process parameters and heating means accordingly. In various applications, high thermal stability is desired and achieved with the matrix system's corresponding glass transition temperature T_g. To achieve this, in many cases, a curing step is performed with a temperature range from 120 °C to 220 °C. High curing temperatures can lead to residual stresses induced by thermal shrinkage and a chemical shrinkage caused by the crosslinking of the polymers. Here, the mismatch of coefficients of thermal expansion (CTEs) between fibre and resin and the anisotropy of the material cause the inner stresses while the laminate shows a different behaviour to a single ply. Because of this behaviour, stacking sequence, thickness ratio, effective heating and cooling rates, and the extent of mismatch of CTEs between fibre and resin, can have a considerable influence on the development of stresses. Other factors can be the interaction with the production tool and the demoulding process of the parts from the mould.

In this thesis, the focus is set on dry-fibre processes, also called liquid composite moulding (LCM)-processes. Here, a fibrous preform is positioned in a formgiving mould and enclosed in a cavity that can be rigid or partly flexible. In an infiltration step, liquid resin is infused or injected into the preform and cured afterwards. Common LCM-processes are vacuum assisted resin infusion (VARI) and RTM. Figure 1 compares the typical non-isothermal temperature cycle for an LCM-process and an adapted process at isothermal temperatures, which is the investigation topic of this work. In the processes currently used for producing large-scale structural composite parts in the aviation industry, in most cases, the preforms are positioned in the mould at room temperature. Afterwards, the mould is heated to the infiltration temperature where the resin has a viscosity sufficiently low and a working life sufficiently long to saturate the preform fully. In the following, the components are heated to the curing temperature, held for a curing time, and cooled down afterwards. At or near room temperature, the parts are demoulded, which means the cured composite parts are separated from the production mould. So-called fully isothermal processes are considered for the cost-effective production of composite parts, especially at high rates. Here, the demoulding is done directly after finishing the curing cycle at or near the curing temperature. There is no cooling step of the material before demoulding. The isothermal approach has several advantages:

- The time for heating and cooling the part from the loading and demoulding temperatures to the higher curing temperatures can be shortened or completely avoided. By this, the parts can be produced at higher production rates.
- Energy savings due to avoidance of actively cooling the part once the curing dwell is completed, and re-run a heating step for the next part. Furthermore, the suppression of the cited steps leads to a reduction of process cycle times.
- Most metals have a significantly higher CTE compared to the carbon-fibre-reinforced polymer (CFRP)-material. In this case, during the cooling step, the mould shrinks onto the parts, which leads to higher forces in the demoulding process and can damage the part depending on the geometry. At a component length of 10 m and a temperature delta of 160 K e.g., the thermal contraction in the cooling step of a steel tool is around 20 mm and 40 mm for the case of an aluminium tool. In this, a CFRP part contracts by only roughly 0.5 mm. Therefore, tooling materials have to be used in many cases with a CTE similar to the one of the carbon

fibre compound as special metal alloys such as INVAR, or CFRP. Both have disadvantages, such as the availability of the material, high prices, a high specific heat capacity, low durability and complex processing. At an isothermal process, the CTE does not influence stresses and geometry. Therefore, a larger variety of materials can be used for the production tools, which allows higher geometrical flexibility, the realization of larger geometries, especially when closed moulds are used, and reduction of the costs of the moulds.

As presented in Figure 1, the isothermal production of composite materials leads to new challenges in the adapted process steps of demoulding and cooling of the components and opens new research topics. The following two main differences are addressed in this thesis:

1. The demoulding of the part at elevated temperatures brings new process requirements. Because of the limited accessibility of a hot part and tool, automation of this step is often required. This can be done in various ways, such as applying vibration, suction or shocks. One option to overcome the retention forces between part and mould is to induce a short, transient mechanical load in the form of a shock by a controlled low-velocity impact. During the demoulding, the part is held at a high temperature which leads to slightly lower performance and unknown load states which is typically not part of the design processes of composite parts.
2. In the baseline process, where the temperature of the part is decreased to room temperature in a closed tool, in most cases, a widely homogeneous, controlled cooling is achieved. Using an isothermal process, which includes demoulding at high temperatures, the hot part is released directly into a significantly colder environment, leading to a fast, inhomogeneous, uncontrolled cooling conditions of the parts.

1.2 Structure of the thesis

In this Section, the structure of the thesis is shown. The literature research, investigations, experiments and finite element method (FEM) calculations are

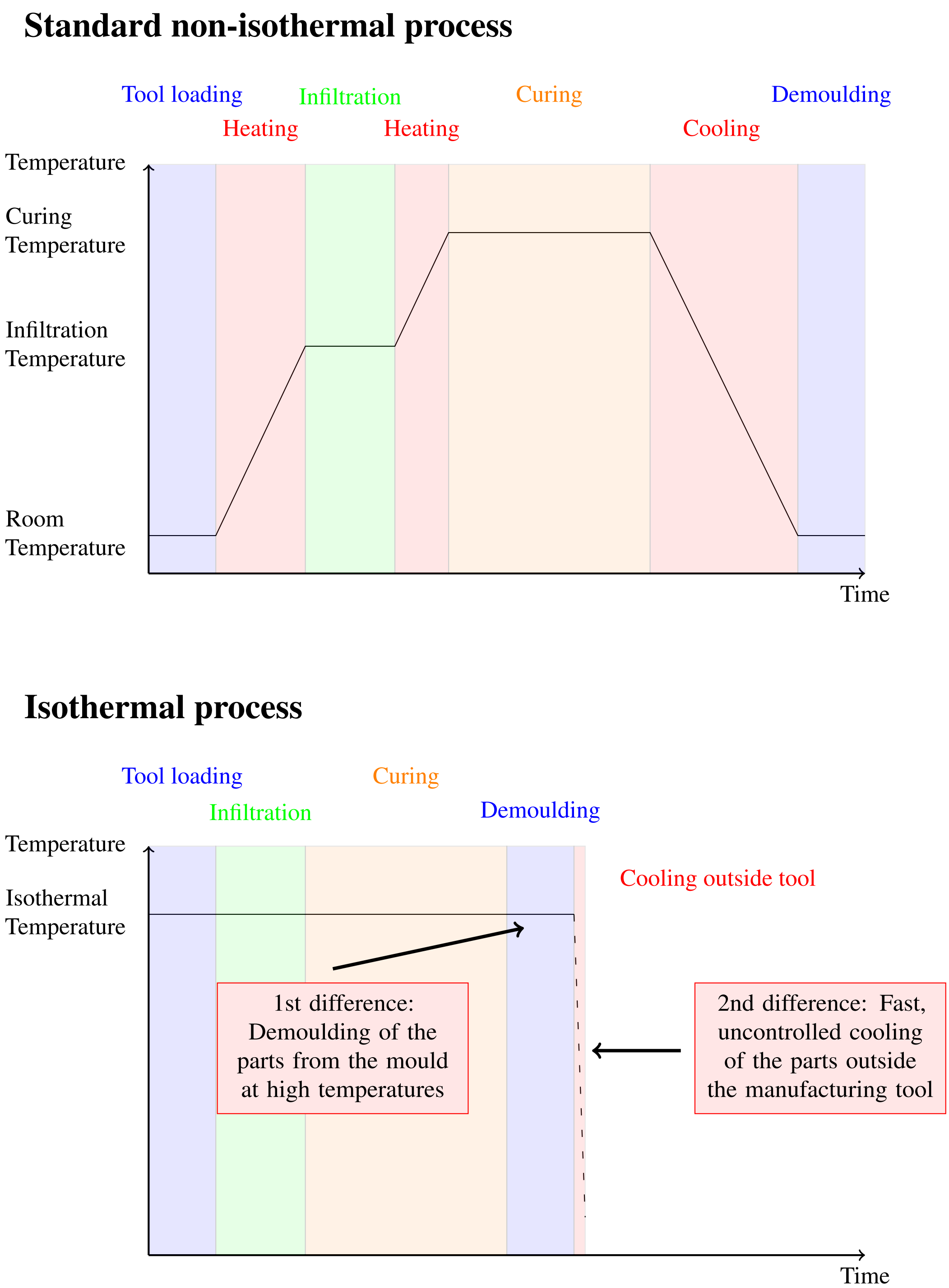

Figure 1: Manufacturing processes using an isothermal temperature cycle in comparison with classical temperature cycles. The two main differences are the investigation topics of this thesis.

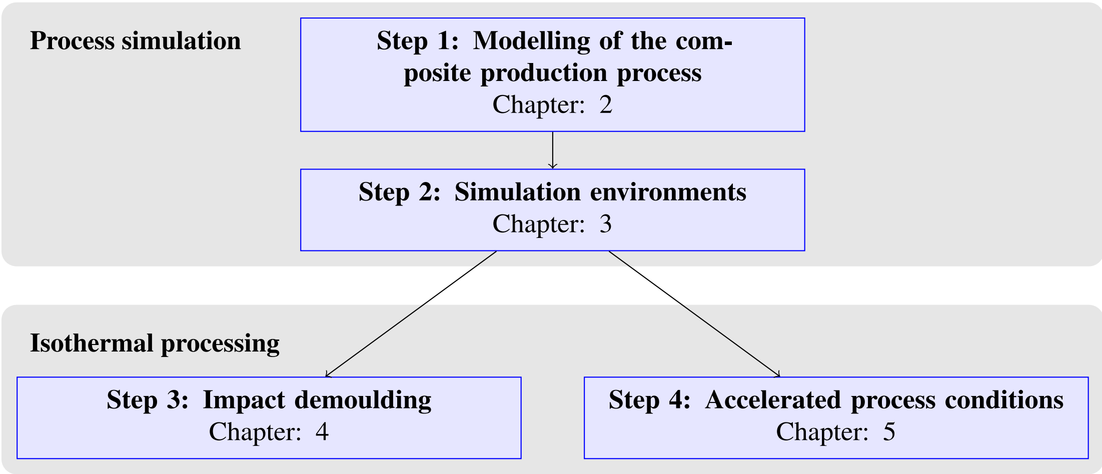

Figure 2: Structure and Chapters of the Thesis

linked to address the academic aspects of the topics exposed in Chapter 1. Figure 2 shows the main sections of the work.

The investigations in this thesis are made with FEM-calculations, manufacturing experiments and laboratory tests. The used theories and the state of the art are reviewed in Chapter 2 to understand the physics behind the manufacturing process and implement the mathematical equations.

A consistent framework covering the heat transfer, the degree of cure-dependent mechanical behaviour, and the cooling/demoulding step are proposed in Chapter 3 as a basis for the subsequent investigations. The content of this Chapter is shown in Figure 3.

To ensure the proper implementation of the models and in order to be able to extend them, a state-of-the-art model for the thermo-chemical calculation of stresses and strains is implemented in the typically used FEM simulation environment ABAQUS (Section 3.2). The same model has also been implemented in the more general and flexible environment COMSOL Multiphysics (Section 3.3), which allows the extension of existing models to fulfil current industrial and scientific needs, thus providing the technical basis for combining failure criteria with modelling process stresses and strains during manufacturing. While different failure initiation criteria are considered, the Puck model is chosen and implemented in the COMSOL Multiphysics simulation environment. The handling of the respective simulation environments is discussed in Section 3.4. Section 3.5 covers the mathematical classification. In particular, the equation

Step 2: Simulation environments Chapter: 3

FEM Calculation
- 3.2
- Implementation of a literature model in Abaqus

FEM Calculation
- Chapter: 3.3
- Implementation of the literature model in Comsol Multiphysics

Study
- Chapter: 3.5
- Mathematical classification of the used equations and implementation to finite-element-environments

Conference Paper
- Comsol Conference 2018
- Title: Homogenization of Fiber Composite Material Properties: an Adaptive Multiphysics Implementation

Conference Paper
- ECCM18
- Title: On the predictive analysis of final quality of composite parts: Gap between available models and industrial needs

Figure 3: Structure of the Chapter simulation environments

types of the models used for the calculations in this thesis are discussed. Moreover, the solving algorithms of the partially differential equations are compared. The material data for the failure initiation are evaluated as a function of the temperature of the matrix system. Therefore, mechanical tests are conducted using neat resin specimens. Further material data are derived from literature values and shown in Section 2.8. A particular focus is placed on analysing the dependency of the parameters on the degree of cure and temperature.

One of the main challenges of implementing an isothermal manufacturing approach for LCM-processes is to demould the parts from the production tools at elevated temperatures. The investigations are shown in Chapter 4, and the structure of this Chapter is shown in Figure 4.

In Section 4.2, the influence of demoulding forces on the cooling approach of the process is demonstrated by way of example to explain the need and advantage of demoulding near the curing temperature.

One possible way of removing the component from the hot curing tool is to apply a short, transient mechanical load to overcome the retention forces. A low-velocity impact can apply this load, e.g. a weight dropped on a part or tool or a shock induced by a hammer. In Section 4.4, simulation approaches are developed to virtually assess the impact force needed for this demoulding process. This allows the dimensioning of the energy input and the definition

Step 3: Impact demoulding Chapter: 4

FEM Calculation
- Chapter: 4.2
- Comparison of the forces occurring during demoulding at different temperatures

FEM Calculation
- Chapter: 4.4
- Dynamic simulation of the transient dynamic response of the structure at demoulding using an explicit FEM approach

Experimental investigation
- Chapter: 4.6
- Dynamic response of the structure at demoulding measured with laser triangulation

FEM Calculation
- Chapter: 4.7
- Calculation of failure initiation due to demoulding impact using an implicit FEM approach and Puck failure initiation

Experimental investigation
- Chapter: 4.7.2
- Failure initiation due to demoulding impact

Conference Paper
- Conference: SAMPE 2022
- Title: Analysis of transient response and failure initiation by impact demolding of composite parts

Scientific novelty of this Chapter:
- Calculation of the transient-dynamic response of the structure by an impact force
- Contact condition with damage criteria for the interface between part and tool for the FEM calculation of the demoulding process
- Combination of the impact calculation with a 3D puck inter-fibre failure criteria

Figure 4: The structure of the Chapter treating the demoulding of composite parts from the production tool by an impact force

of the properties and geometries of an impactor. FEM simulation models are developed to investigate the transient dynamic response of tool and part during a demoulding procedure induced by an intended low-velocity impact. Here, the software Abaqus Explicit is used, which is based on an explicit solver. To calculate the demoulding process, the implementation of an adhesive contact condition between the composite part and the production tool with a damage criteria is investigated.

In Section 4.7, simulation approaches are developed to assess the criticality of the demoulding step for failure initiation. Here, a three-dimensional implementation of the Puck inter-fibre failure criteria is used with an implicit solver in the simulation environment Abaqus 2021 with user subroutines. In addition to the calculations, experimental trials were performed to investigate the validity of the simulations. Section 4.3 explains the use case and experimental setup.

In Chapter 5, the influence of the accelerated cooling of the components to ambient temperature after demoulding at or near curing temperature on their structural integrity is investigated. The structure of the Chapter is shown in Figure 5.

Step 4: Accelerated process conditions Chapter: 5

Characterization of material properties
- Chapter: 5.2
- Temperature-dependent strength values of cured neat resin

Experimental investigation
- Chapter: 5.3
- Accelerated cooling of thick-walled laminates, infusion process

FEM Calculation
- Chapter: 5.2
- Meso-scale model
- Strains and stresses during accelerated cooling of RTM components, Puck damage initiation.

Experimental investigation
- Chapter: 5.4
- Material properties during accelerated cooling of RTM components

FEM Calculation
- Chapter: 5.3
- Full-scale model
- Strains and stresses during accelerated cooling including cure-induced stresses and strains, Puck damage initiation.

Journal Paper
- Journal of Composite Materials
- Titel: Assessment of structural integrity of thermoset composite parts subjected to accelerated cooling during the manufacturing process using the Puck-criteria for failure initiation

Scientific novelty of this Chapter:
- Application of an incremental linear-elastic model with the Puck inter-fibre failure criterion to evaluate the criticality of accelerated cooling of the composite part

Figure 5: Structure of the Chapter addressing the criticality of failure initiation during rapid cooling of composite components after demoulding

In addition to the demoulding process itself, this cooling process represents the second main challenge for isothermal processing, which is a key focus of the scientific investigations in this thesis, as shown in Section 1. Based on the simulation framework and input data proposed in Chapter 3, the influence of accelerated cooling conditions during manufacturing on the structural integrity of the composite parts is investigated. Initially, the temperature field, stresses, and failure initiation markers are calculated for different cooling conditions in a small-scale geometry considering only the cooling phase (Section 5.2). This study aims to demonstrate the criticality of rapidly cooling the material from curing temperature to room temperature. In particular, it shall be analysed whether damage has to be expected due to stresses caused by inhomogeneous temperature distribution through the layers of the material. The material parameters for the thermal model, the mechanical model, and the failure initiation are obtained from mechanical tests and values found in the literature (Section 2.8). The results obtained in the calculations are verified using manufacturing trials and mechanical tests: using the VARI process, damage occurrence and the deformation of thick composite parts with symmetric and non-symmetric stacking after a rapid cool-down is investigated in Section 5.3. The trials were complemented by simulations involving the full-scale geometry and end-to-end curing process (chemical and thermal-induced stresses and strains). Section 5.4 describes the manufacturing of RTM parts with different cooling scenarios. The material strength of the produced parts was analysed and compared.

2 Thermochemical and mechanical modelling of the composite production process

This Chapter describes the scientific and mathematical basis of the models used in this thesis, the mathematical implementation, and common approaches for mechanical constitutive models. This includes all aspects of the multiphysics simulation: heat transfer, calculation of stresses and strains, and failure initiation. Current research's state of the art is shown based on literature research. This Chapter provides the theoretical basis for the following investigations. Figure 6 shows the position of this Chapter in the context of the work.

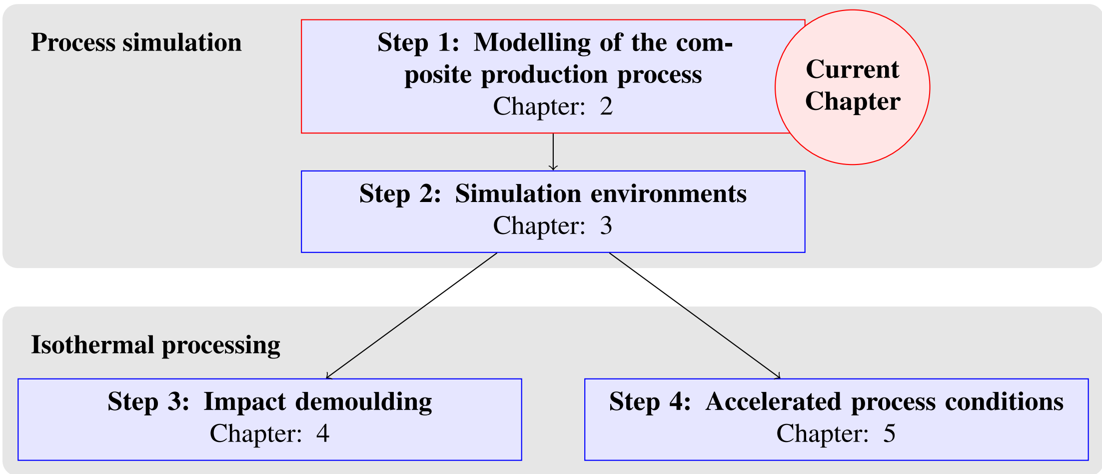

Figure 6: Position of Chapter 2 in the structure of the thesis

2.1 Interaction of the calculation models

Figure 7 illustrates the different elements of the calculation: the thermal calculation of the part and, in many cases, its production environment, e.g., the curing tools, form the basis of the simulation. The degree of cure of the resin is calculated simultaneously to predict the exothermic heating of the resin, which has an impact on the thermal model. The glass transition temperature and resin Young's modulus are calculated using models, including dependency on the degree of cure and temperature. A homogenisation procedure is used to obtain the resulting values of the matrix's changing material properties and the fiber's properties, which are assumed to be constant. Chemical shrinkage and thermal