

Liquid Composite Molding reproducibility in real world production of fiber reinforced polymeric composites: A review of challenges and solutions

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Liquid Composite Molding (LCM) suffers from multiple factors that contribute to pronounced uncertainty of process characteristics. This results in compromised reproducibility which is associated to high scrap or the unpredictable behavior of approved parts. However, LCM is still attractive for Fiber Reinforced Polymeric Composites (FRPC) production due to its economic advantage (i.e. in relation to Autoclave), the capability of some of its variants to produce high performance parts and its potential for process optimization. This review analyzes each uncertainty with respect to its origins and its impact in part or process, based on a combination of past literature and original numerical results. The possible methods to counteract uncertainties are critically discussed, with an eye on both the scientific and feasibility (technical/ economical) aspects. The overall aim is to provide to future LCM implementations a roadmap of the most critical challenges and solutions regarding the establishment of a reproducible process.

Keywords: fiber reinforced polymeric composites; composites manufacturing; liquid composite molding; defects; uncertainties; process monitoring and control; permeability; cost estimation;

1. Introduction

Liquid composite molding (LCM) is a family of manufacturing methods for fiber reinforced polymeric composites (FRPC). It includes Resin Transfer Molding (RTM), Wet Compression Molding, Resin Infusion under Flexible Tooling (RIFT), Structural Reaction Injection Molding (SRIM) and their respective variants¹. The highest performance composites (e.g. biomedical, aerospace and defense) are generally manufactured by Autoclave using pre-impregnated preforms (prepregs). LCM methods dominate the manufacture of high quality parts (parts with characteristics that conform with or are close to high performance standards with low to average cost²). Although a

critical objective for LCM methods, high reproducibilityⁱ, has been reached in certain cases, (i.e. RTM), there is recent evidence that there is room for improvement with respect to reproducibility and accurate processing³. Failure to improve, constrains penetration of LCM products in the high-quality composites market and it is therefore of interest to address the issue of reproducibility in LCM. The first step in this direction is the identification of the origins of variations. Poor reproducibility originates from uncertaintiesⁱⁱ of the process inputs which, in turn, may lead to a pronounced rejection rate that requires either reworking or scrapping with obvious implications to production costs and environmental burdens⁴. Past work with a substantial relation to reproducibility are quite limited; Mesogitis et al.⁵ analyze the sources of uncertainty in FRPC manufacturing with a focus on their association to process simulations. Potter et al.⁶ summarize and discuss a vast range of defects that potentially emerge on composites manufacturing, some of which are associated to reproducibility in LCM. The current study will focus on understanding and counteracting the barriers of reproducibility in LCM, with the aim to establish a roadmap for future implementations. In section 2 the uncertainties are categorized and analyzed. In section 3, possible methods to counteract uncertainties are presented and critically discussed. Section 4

ⁱ Reproducibility: The closeness of agreement between independent test results obtained under reproducibility conditions, i.e. conditions where test results are obtained with the same method on the same or similar test items in different laboratories, by different operators, using different equipment, in different locations, or on different days.

ⁱⁱ Uncertainty: A range of values within which it is estimated that the true value of a quantity of interest lies. It is typically aimed to form the narrowest possible range that encompasses all possible sources of error, including the intrinsic randomness of the system, inaccuracy due to a lack of knowledge, deficiencies of equipment, etc.

includes a realistic estimation of the potential economic benefit of a dominant uncertainty counteracting approach.

2. Uncertainties of process inputs and their impact on the process or part characteristics

Uncertainties in any of the LCM process inputs (materials, geometry, conditions, etc.) may limit reproducibility (i.e. in a process that is not robust enough). The best way to classify uncertainties is by their impact on the final product quality. In that context, uncertainties can be categorized in four uncertainty types:

- (i) Fiber (filament and tow) displacement and preform deformations
- (ii) Variations in the chemical composition and purity of the polymeric matrix
- (iii) Inappropriate geometrical characteristics of the part-tool interface
- (iv) Variations in the curing temperature

Each of the above categories includes uncertainties that emerge in various stages of FRPC production (pre-existing in supplied constitutive materials, preparation, forming, filling and curing). The purpose of this section is to analyze and decrypt this complex system of uncertainties with respect to their description, their type, the production stage they emerge in and their impact in production. A summary of these attributes is presented in Table 1 while they will be described in detail in the paragraphs that follow.

2.1. Fiber displacement and preform deformations

The characteristic of the preform, to be highly susceptible to fiber displacement (i.e. tow misalignment) or other deformations, is a significant source of uncertainties in LCM. Uncertainties in this category will impact mainly the filling stage as fiber displacement generally causes uneven distribution of permeability (a measure of the ability of the

Table 1. The description, type and stage of impact of expected uncertainties per production stage.

		Description of uncertainty	Type of Uncertainty	Main stage of impact	
Production stage of uncertainty formation	Supply	Inherent textile defects	Fiber displacement and preform deformations	Forming, Filling	
		Inherent batch-to-batch resin variation	Variations in the chemical composition and purity of the polymeric matrix	Filling, Curing	
	Preparation	Preforming defects	Fiber displacement and preform deformations	Forming, Filling	
		Matrix mixing defects	Variations in the chemical composition and purity of the polymeric matrix	Filling, Curing	
		Matrix aging		Filling, Curing	
	Forming	Edge effects (Race tracking)	Inappropriate geometrical characteristics of the part-tool interface	Filling	
		Draping defects	Fiber displacement and preform deformations	Filling	
		Nesting			
	Filling	Reinforcement wash-out		Filling, Curing	
		Tool deflection	Inappropriate geometrical characteristics of the part-tool interface	Filling, Curing	
		Deficiencies of injection unit (poor maintenance, poor calibration, mixing errors)	Variations in the chemical composition and purity of the polymeric matrix	Filling, Curing	
		Curing	Variations of the environmental temperature	Variations in the curing temperature	Filling, Curing
			Equipment deficiencies (i.e. cool circuit problem)		Curing

fibrous preform to facilitate fluidic flow in its interior) in the preform. The non-repeatable locality of permeability fluctuations, can under certain conditions undermine the stability and quality of the filling process (i.e. low vacuum quality with an impact on compaction behavior) and thus can result in resin-rich and clustered fiber volumes in the final product. Such trends make the part prone to in-service structural defects^{7,8} and lead to early fracture and failure.

2.1.1. Inherent textile imperfections

The manufacturing of reinforcement textiles suffers from imperfections of the final product such as incorrect fiber orientation, missing yarns, gaps, cuts and other⁹. Such imperfections pre-exist in the materials provided in the FRPC industries. An example of such imperfections in non-crimped fabrics (NCF) are the openings and channels because of the stitches. The stitches and openings themselves are design-in features that cannot be considered as defects, but under certain circumstances they can facilitate the formation of in-process defects (i.e. preferential flow paths). Lomov¹⁰ studied their formation and his indicative finding is that in a $0^\circ/-45^\circ/90^\circ/45^\circ$ carbon NCF preform, ~25% of stitches create a 0.6mm wide opening between the tows. Similar effects have been found in woven fabrics where there is a higher tow misalignment tendency (associated to the lack of stitches) and pinholes are created at tow intersections. Vanaerschot et al.¹¹ measured (image processing), modelled and characterized statistically such tendencies. The various defects generated by the textile fabrication may lead to the formation of random flow channels. Drapier et al.¹² investigated the effect of stitching density of NCF in the transverse permeability. They found that permeability increases linearly with stitch density due to the openings created by the stitches. The variance in permeability measured by their experiments (~20%) was partly attributed to the openings. Yun et al.¹³ used different distribution media with various

permeabilities in order to investigate its effect on flow and void content. They found that the final void content increases with distribution media permeability increase: flow paths through pinholes at the tow intersections of woven fabrics, are created more easily with higher distribution media permeability. The more intense the pinhole flows, the less the uniformity of the flow front, which is essentially a void-generating condition.

2.1.2. Defects induced by preforming

In order to generate the preform, the textile needs to be cut and stacked in the desired orientations. These activities unavoidably contribute to fiber displacement (i.e. unintendedⁱⁱⁱ shear or loss of edge tows) as they involve extensive mechanical (i.e. cutting knife) and/or human handling¹⁴. Possible layer stitching performed at the preforming stage can have similar effects with the design-in stitches of the textiles discussed in 2.1.1. Rieber and Mitschang¹⁵ applied various stitching patterns in glass fiber twill weave preforms and investigated their effect in the in-plane permeability. Their key result is that the lower the stitching seam distance the more the effective permeability is reduced. Additionally, in any case where a compression mechanism conforms the preform to the cavity shape (either by an off-line frame or by the tool itself in-line) there is the risk of further unintended shear or other defects (fiber buckling, fiber wrinkling, yarn slippage etc.) due to draping on the cavity shape. There are known impacts of the above in production: Edge tow loss caused by cutting and handling contributes to edge effects (§ 2.3.2) or unintended shear which are both associated to random local permeability

ⁱⁱⁱ The term ‘unintended’ is used to discriminate from shear induced by the cavity geometry; the latter constitutes a characteristic of the reinforcing structure and cannot be considered as a defect.

variation. Shear in general (intended or not) has been extensively studied with respect to its influence on local permeability. Endruweit & Ermanni¹⁶ indicated that a preform with a fiber volume content (FVC) of 51%, based on 2×2 twill weave glass fabric becomes less permeable with shear. Similar results were elsewhere verified: Aranda et al.¹⁷ reported that under constant cavity height conditions, the in-plane permeabilities of a 0°/90° carbon NCF-based preform dropped non-linearly as shear angle increased above 20°. The formation of defects such as fiber buckling, fiber wrinkling and yarn slippage has been associated to the existence of excessive shear deformation forces¹⁸. Chen et al.¹⁹ studied the formation of such defects for bi-axial NCF with a pillar stitch, formed over a hemisphere tool. They concluded that forming to the hemispherical geometry induced in-plane and out-of-plane wrinkling with wrinkling strains (the compressive strain along each primary fiber direction) in the range of -0.03 to -0.3. Lee et al.²⁰ also investigated deformations caused to NCFs by hemispherical forming. They observed in-plane and out-of-plane buckling which they managed to quantify using an image processing method and proposed keeping the preform under tension during forming (Blank Holder Force) as a method to minimize these deformations. Ouagne et al.²¹ found that tetrahedron forming of woven flax preforms causes out-of-plane buckling on some faces and edges with buckling height in the scale of ~1mm. Additionally they observed that applying too much tension to the preform during forming in order to prevent buckling and wrinkling may cause the weft to slip from the warp yarns (yarn slippage) thus creating relatively big gaps that compromise the reinforcing structure.

2.1.3. Nesting

When the preform is compacted (i.e. top tool closing) the tows may shift into the

unoccupied space between two adjacent tows (nesting) depending on the compaction pressure, weaving type, layer number and orientation²². An immediate result is the reduction in the compaction levels per individual preform thickness. Yousaf et al.²³ found that the thickness of a 6-layer woven preform with nesting under typical compaction is 12% lower than the thickness at the same compaction without nesting. The tow displacement induced by nesting changes randomly local permeability and flow patterns. Jiang et al.²⁴ found that the in-plane permeability parallel to the fiber direction of a two-layer unidirectional fabric decreased as much as $\sim 3/4$ of a scale of magnitude due to nesting blocking the flow channels that initially existed between the tows. Hoes et al.²⁵ focused on the permeability scatter for woven fiberglass mats and by a systematic elimination of other factors (handling, weaving structure, intrinsic material variability, etc.) concluded that nesting is the main contributor to permeability scatter.

2.1.4. Reinforcement wash-out

Reinforcement wash-out is the preform deformation induced during filling by the fluid itself due to the combined effect of the clamping force and excessive injection pressure. Typically it results in tow displacement close to the inlet (where injection pressure or flow velocity is maximum)²⁶ and resin-rich volumes. Kaynak & Kas²⁷ observed fiber waviness in the vent region and a through displacement at the bottom layer of the inlet region after injecting unidirectionally for their RTM case study. Richardson & Zhang²⁸ quantified the wash-out (distance from initial position) for non-woven hemp reinforcement for a variety of pressures. Fiber wash-out is formed during filling early enough to influence the remainder of the filling process by facilitating flow through channel formation. Although often the region close to the inlet is affected the most, the extent and shape of preform deformation may vary even for the same conditions. Konstantopoulos et al.²⁹ while investigating the effect of preform thickness on the

unsaturated transverse permeability, found that high thickness preforms are more prone to reinforcement wash-out compared to low thickness preforms with the same FVC due to their higher compressibility.

2.2. Variations in the chemical composition and purity of the polymeric matrix

In this section the appearance of variations to the chemical composition of nominally identical polymeric materials will be discussed. Uncertainties in this category may affect both filling and curing as the variation in chemical composition has an impact on fluid viscosity and the kinetics of the curing reaction. Impacts of these uncertainties in production include uneven curing characteristics and untimely (premature or delayed) termination of the process, both of which influence the quality of the final part; they have been associated to structural integrity threats such as void formation³⁰, residual stresses³¹ and degradation of the polymeric matrix³².

2.2.1. Inherent batch-to-batch resin variation

Polymeric resins are manufactured in batch quantities. For instance, Unsaturated Polyester Resins typically used in thermoset-based FRPC, are manufactured by allowing a batch quantity of the base chemical constituents to react for several hours, extracting the unblended resin and finally blending the resin with styrene. The broad range of molecular weights of the unblended resin and the variance in the stoichiometric ratio of the styrene during blending result in differences in the chemical description of different batches of the final product³³. According to³⁴, since such differences can have significant impact in FRPC manufacturing, it is essential to test the resin as delivered at least with simple tests such as density test, melt flow index test and heat deflection test. There is currently no previous work that studies the impact of uncertainties of this type in production.

2.2.2. Defects induced by mixing

The most critical step in preparing the polymeric matrix is mixing the constituents (resin, curing agent, catalyst, etc.). While one-component matrices are already mixed when supplied, two-component matrices are mixed by the FRPC manufacturer. In the second case, there are two options: manual and automatic mixing. Manual mixing typically involves weighing, pouring and stirring, with all stages subject to significant human errors. Although automatic mixing is more accurate for such operations, it requires regular and thorough calibration as well as maintenance activities. Experience shows that injection units that support automatic mixing can often be subject to poor maintenance which in turn leads to impurities in the matrix or other injection difficulties due to cured resin residue. Errors originating from manual or automatic mixing are largely random in type and intensity and can impact the process significantly. Nunez et al.³⁵ found that deviation in the curing agent ratio by just 1 part per hundred (phr) (from 34 to 35 phr) that was added to a DEGBA epoxy resin, lead to a decrease in the enthalpy of the reaction by ~23 J/g and the glass transition temperature by 4°C. Decreases of these key properties in that range are operationally significant as they cause the decrease of curing rate due to reduced exothermic heat flux³⁶. The study of Pandiyan Kuppusamy & Neogi³⁷ shows that the combined deviation of the catalyst and accelerator that were added into an ambient-curing polyester resin from 1 to 2 phr, results in an increase of the peak exotherm temperature by 8°C and an increase of the rate of temperature rise by 5.47°C/min. The combined effect of these increases is associated by the authors to a significant rise in the curing rate.

2.2.3. Aging

Polymeric resins suffer from a gradual deterioration of their properties, referred to as

aging. Aging is a broad term that includes different types of aging (physical, chemical, hydrothermal) that may occur at different stages of the lifetime of the polymer (i.e. prior, during or post curing)³⁸. Focusing on uncured resin, aging occurs during shelf life (unopened product) or storage life (opened product). Shelf and storage life are in general lower in 1-component matrices due to the fact that the theoretically unreactive curing agent that they contain, in fact presents slow reactivity. Hamerton et al.³⁹ showed that a non-latent 1-component matrix loses up to ~50% the enthalpy of the reaction after shelf life of 70 days in ambient conditions which is evidence of shelf curing. Hakala et al.⁴⁰ found that when an epoxy-based 1-component matrix is stored at 20°C for 16 days, cures by ~20%. The resins of 2-component matrices suffer mainly from thermo-chemical and photo-chemical aging due to the surrounding environmental conditions. Sands et al.⁴¹ showed that an epoxy resin which is free from curing agent and catalyst, took 6 months in a dark environment with temperature < 25°C before reaching the threshold of unacceptable degradation. The uncertainty in production emerges from using resins of different aging from production cycle to production cycle or even mixtures of resins of different aging in the same production cycle.

2.3. Inappropriate geometrical characteristics of the part-tool interface

The interface between part and tool is an area sensitive to the formation of preferential flow paths (a region with higher flow velocity) that result in filling defects (i.e. dry spots). The locality and size of such flow paths depend on non-random factors (i.e. the textile structure or the FVC of the preform) and on random ones (i.e. the cutting quality, placement, or preform deformations that are present) which contribute to the overall poor reproducibility. The effects of flow channeling include resin-rich and clustered fiber volumes that lead to structural defect formation as well as deviation from the desired geometrical description (i.e. higher part thickness)^{42,43} that may lead to post-

processing or scrap. It must be noted that the specific uncertainty category is highly dependent on design features (i.e. tool/ preform design) and additionally past work is based on equipment/ setups intended for academic research. In that sense the quantified results presented here are not representative of actual production.

2.3.1. Edge effects

Between the preform edges and the cavity walls a preferential flow path is created mainly because of imperfect positioning of the preform (i.e. gaps between preform and tool). The terms “edge effects” or “race tracking” are used in literature to describe the above. Young et al.⁴⁴ observed that the edge flow during unidirectional filling of woven fiberglass mats precedes non-edge flow by values in the scale of 100 mm, depending on the number of layers (7 to 9) as well as the gap size (1 to 3 mm). Lawrence et al.⁴⁵ observed the flow of unidirectional filling of woven fiberglass mats. They found that the race tracking strength (ratio of permeability of edge flow to permeability of bulk flow) varied in the range of ~4 to ~27 for the warp direction and ~2 to ~34 for the weft direction, depending on the placement quality. The significant flow velocity differences that edge effects cause at different regions of the preform may result in filling imperfections (i.e. dry spots). The intensity and locality (i.e. which edge exactly) of the problem is rather random as it depends highly in the cutting quality and placement of each individual preform. Devillard et al.⁴⁶ studied the flow in unidirectional filling of a cavity with an L-shaped rubber insert. They determined the average race tracking strength per cavity region for woven fiberglass mats and found that the measurements showed significant standard deviation (as high as ~95%) depending on cutting and placement quality.

2.3.2. Tool deflection

Ideally, the compaction mechanism (top tool closing, vacuum bag, etc.) in LCM methods achieves the targeted part thickness uniformly. Realistic setups however include many scenarios where this is not the case: Outward pressures (injection pressure or compaction pressure) may overcome the local forming force and deform the tool

(flexible, semi-rigid or rigid), thus affecting local cavity thickness (tool deflection). Robinson & Kosmatka⁴⁷ investigated the VARTM process for 24 mm thick laminates utilizing a flow distribution layer. They found that peripheral injection caused deflection and thickness increase by ~2.5 mm close to the inlet. Maclaren et al.⁴⁸ while experimenting with light RTM found that radial injection at 130kPa in a tool with a 6 mm thick polycarbonate (semi-rigid) top half, caused the nominally 4mm thick cavity to deflect above the inlet by 1.03mm. However, since the specific injection system is volume-controlled, injection pressure is largely defined by characteristics of the preform (compressibility, FVC, defects). For instance, the suitable injection pressure (and consequently the deflection potential) rises with preform FVC⁴⁹ due to the inverse proportionality between preform permeability and FVC. Additionally, when the tool is semi-rigid or flexible, flow channels are formed not only within the reinforcing structure (reinforcement wash-out) but also between the preform and tool plates (tool deflection). In production, tool deflection causes variations in filling time and introduces the need for a waiting time post injection where the tool will relax and regain the intended dimensions (post-filling time). Timms et al.⁵⁰ found that tool deflection appears to have a strong impact on post-filling time; under flexible tooling with peripheral injection it can reach almost triple value than that of semi-rigid tooling (RTM light).

2.4. Variations in temperature

Curing is a thermally activated and controlled chemical reaction: The heating profile defines the curing behavior and affects curing attributes (curing time, cure-induced defects, etc.) which are important for production efficiency and costs. Uncertainties in this category can impact heavily the curing stage. Various temperature discrepancies (exotherm reaction, difference between material and heater temperatures, temperature

gradients, etc.) are known in LCM but only the non-repeatable ones will be discussed here (i.e. excessive exotherm reaction due to high thickness parts is a repeatable defect and therefore does not affect reproducibility). Such discrepancies impact production and part quality similarly to uncertainties of §2.2: uneven curing or untimely process termination, leading to void/ distortion formation and matrix degradation.

2.4.1. Variations of the environmental temperature

The simplest example of non-reproducible heating is when the part is left to cure at ambient temperature. This is typical for large structures (turbine blades, boat hulls, etc.) where heating by other sources is unpractical⁵¹. Typical times for resins to cure at ambient temperature are ~2-3 days⁵². This time interval may include day-night temperature fluctuations while mean temperatures will obviously vary with the seasons. Resin manufacturers' data sheets assume a curing cycle time based on an average ambient temperature (~25 °C). Indicative curing cases were simulated by the authors of this paper (Figure 1) to provide insight of the problem in production, as it has not been investigated elsewhere. The in-house simulation software "CureSim" was used. The simulations correspond to neat polymer curing of the system Epikote RIMR 135/Epikure 1366. The Prout-Tompkins kinetic model used for the calculations is considered highly compatible with the specific material. Thermal analysis, the kinetic model and the determination of all required kinetic parameters for this material are described in detail in a previous publication of the authors⁵³. Case A involves curing under constant ambient temperature of 23°C, as proposed by the manufacturer. Case B considers a triangular variation of temperature (representative of the temperature fluctuation at the 21st of June for a location at 45° North latitude⁵⁴: the parallel mid-way between Equator and North Pole that crosses central Europe, USA, etc.) over a 24 hour period. Cure to 97% took 410.4 min longer for Case B.

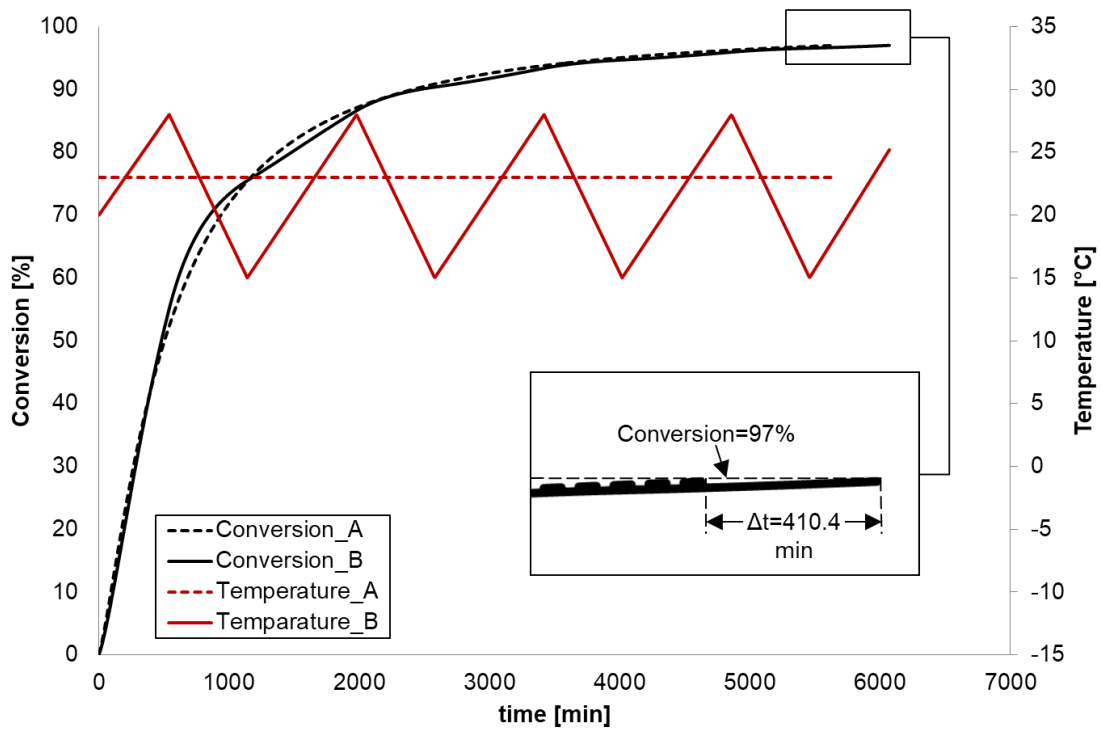


Figure 1. Curing simulation for case A: constant room temperature and Case B: room temperature with day-night fluctuation at the 21st of June for a location at 45° North latitude (Material: Epikote RIMR 135/ Epikure 1366).

2.4.2. Deficiencies of the heating system

Commercial heating units share minimum operational standards and generally provide the targeted temperature. However, the success in reaching and maintaining the temperature is not dependent solely on the heating units as these may be only one of the components of the heating system. For instance, a heating system based on hot liquid (water or oil), typically used in RTM, combines a heating unit, a temperature controller, a complex hydraulic circuit comprising of hot and cool lines, a heat exchange unit, a pump and possibly a liquid treatment unit (in case of water). In mass production, the heating system may be designed to support more than one heating unit to allow multiple simultaneous heating operations. The maximum number of simultaneous operations depends on the heat capacity of the cool line, the latter being defined by various factors

(heat exchanger size, pump size, flow volume, circuit length etc.) which are currently not standardized. A simplified description of the heating unit operation is that cool water is provided to the unit as input, and hot water is the unit output directed to the tool. The cool and hot lines are discrete circuits whose water never mixes. The role of the cool line is to allow continuous regulation of the temperature in the hot line (i.e. suppress overshoots) by heat exchange. When a single cool line supports an excessive number of heating units, there is a higher risk of poor regulation (i.e. if temperature in the cool line increases, heat transfer flow between cool and hot lines decreases. See Figure 2). In such a scenario, the output temperature of each heating unit would present ripple around the target temperature and the ripple amplitude would be proportional to the temperature rise in the cool line⁵⁵. From all the above, it is evident that the circumstantial number of concurrent heating operations defines the heating quality of an LCM process. Exemplary curing simulations were executed to provide indicative quantification of the problem in production (Figure 3). The software tool, model and materials described in the previous paragraph were used. Case C is curing under constant temperature at 70°C (as the manufacturers proposed for heated operations). Case D considers a realistic ripple around the target temperature of 70°C. Cure to 97% in Case C was reached 19.4 min later than Case D.

3. Work to minimize the negative impact of input uncertainties

3.1. In the development stage: Improvement of predictability

For the minimization of the uncertainties discussed in §2, experts often address the development stage (where the product and process are designed). The aim here is to make smart design choices which counteract uncertainties. The development stage precedes the hardware realization and besides prior experience and best practice,

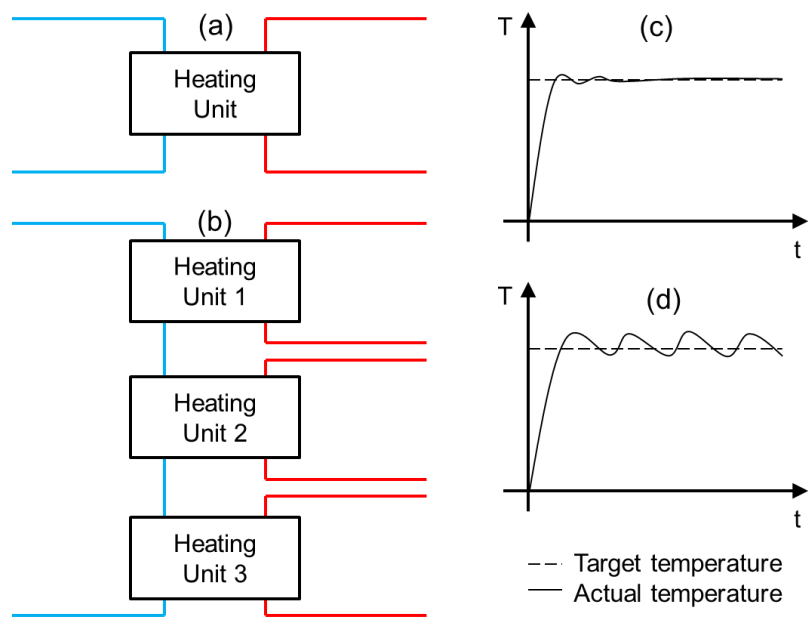


Figure 2. The cool (blue) and hot (red) line circuits of (a) a single heating unit and (b) multiple heating units; (c) and (d) represent the temperature regulation per unit respectively.

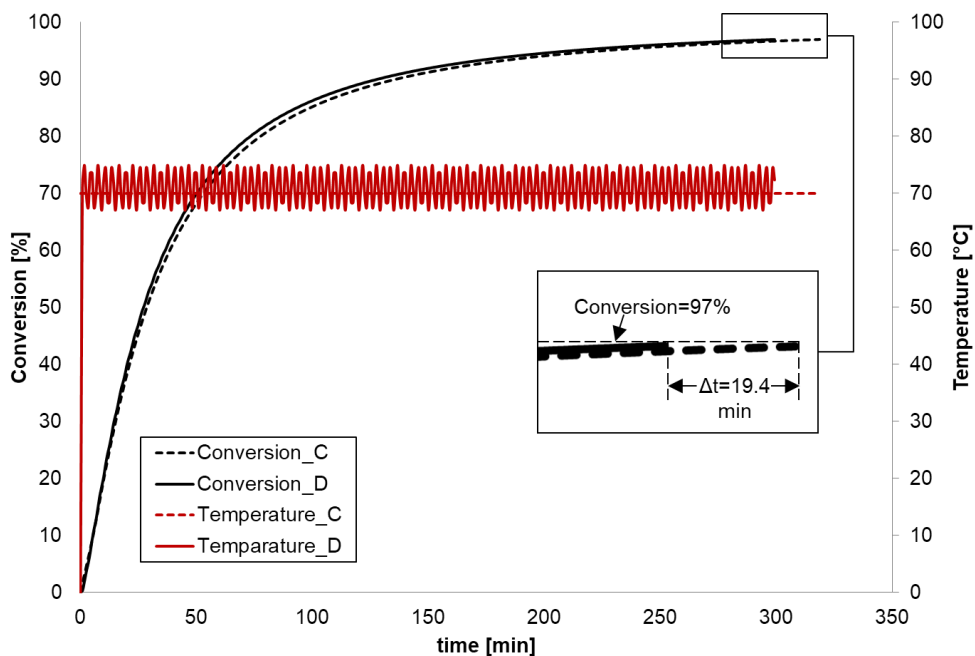


Figure 3. Curing simulation for case C: constant temperature of 70°C and Case D: temperature with ripple around 70°C (Material: Epikote RIMR 135/ Epikure 1366).

attempts to optimize design are also based on predictions of the behavior of the system (i.e. simulations). The benefit of this approach is that many different case studies can be examined easily (in software level). The drawback is that the predictions provided by the simulation software may have questionable agreement with reality. Possible reasons for this discrepancy include modelling unable to capture the actual phenomena in full extent and simulation inputs that differ from their corresponding actual values.

Indicatively, Swery et al.⁵⁶ showed that the preform permeability, a simulation input affecting heavily the results, is overestimated by a factor of two when model-derived instead of experimentally determined. However, in order to benefit the system-to-be, design choices made in development must be based on accurate predictions. This raises the issue of the predictability^{iv} of the system. Since a simulation can only be as good as its inputs, predictability depends highly on providing accurate simulation inputs. The aim in the following sections is to capture the state-of-the-art in determining and understanding the most influential simulation inputs, mathematical models and material characteristics.

3.1.1. Model base used in simulations

The core of a simulation designed for LCM processes is the internal mathematical model that processes the other inputs (conditions, geometry, materials) in order to provide a prediction for filling or curing behavior. The base for (component-level, not textile level) flow modeling is normally Darcy's law⁵⁷ which leads to different solutions depending on the filling (and solving) direction.

^{iv} Predictability: The closeness of agreement between predicting mechanisms (i.e. simulations) and reality.

Models covering unidirectional⁵⁸ or radial flow⁵⁹ have been proposed and perfected over the years. Such models consider mainly in-plane flow in unsaturated media (dry preforms) while they can account easily for different injection modes (constant pressure or constant flow rate) with minor adjustments. The above provide the mathematical basis for current simulation algorithms. Current research on flow modeling modifies this basis in order to account for known discrepancies such as race tracking⁶⁰, the different filling behavior in saturated and unsaturated media⁶¹, flow through the thickness direction⁶², dual scale permeability (total fabric permeability that differs from permeability within the tows)⁶³, hydrodynamic compaction modeling⁶⁴ and inappropriate assumptions made in the derivation of established models such as the assumption of negligible difference between flow initiated from a circular inlet (real world condition) and an elliptical inlet (model demand)⁶⁵.

Past work on cure modeling is often based on the Kamal-Sourour model⁶⁶; a combination of previously developed models (the n^{th} order and autocatalytic) which over the years has been found to agree with the curing kinetics of many polymers used in FRPC^{67,68}. Current research on cure modeling includes modifications of this model in order to account for diffusion occurring in the curing process^{69,70} and methods to account for realistic curing conditions⁵³.

3.1.2. Material characteristics used in simulations

The basic equations for flow and cure modeling discussed above contain material-dependent parameters. Specifically, the flow model base is heavily influenced by permeability (K), a direction-dependent characteristic represented mathematically at the three dimensions by a second grade tensor. The importance of permeability led to significant research activity on the topic of permeability determination. The central demand in this research area is to focus on the material characteristics and their

influence on permeability. In that context, the effect of different preform characteristics (layer orientation, thickness, fiber volume content, tow structure/size, etc.) on permeability^{25,71,72} as well as the effect of the permeant⁷³ have been studied while different inter-university benchmark comparisons of permeability results (round robin studies) have been conducted⁷⁴⁻⁷⁶. The equipment to determine permeability (permeameter) currently is not standardized and its commercial availability is limited. Permeameters have been developed mostly by different research groups and differ significantly. Results show significant variance (~20-30%) in permeability for the same test cases, caused primarily by the inherent textile/ preform uncertainties and secondarily by different hardware/ procedures of permeameters⁷⁷. This result has triggered research that focuses on permeameter characteristics and their procedures: dipoles of permeameter characteristics have been investigated and compared^{60,62,78-81}, such as saturated or unsaturated permeability determination, continuous (i.e. camera) or discrete (i.e. point sensors) flow detection method, radial or unidirectional filling and analytical (i.e. Darcy-based) or inverse (i.e. optimization algorithm) calculation. The above comparisons have not yet led to definitive answers: each case has pros and cons that have not been quantified holistically.

On the other hand, the cure model base is heavily influenced by the kinetic parameters it contains (i.e. activation energy and pre-exponential factor) that characterize the polymeric matrix. These can only be quantified on the basis of thermal analysis such as Differential Scanning Calorimetry (DSC). Different methods have been developed in the past to process DSC measurements and derive reliable values for the kinetic parameters, namely the Kissinger method⁸², Isoconversional methods⁸³ and the Fitting methods⁸⁴. The comparison of the above methods for given case studies has also been a topic of investigation⁸⁵.

3.2. In the production stage: Improvement of repeatability or adaptability

The minimization of uncertainties is alternatively pursued by intervening in production itself. As discussed in § 2, many uncertainties are caused by activities of the production process (preform deformation during preforming, inappropriate mixing of the polymeric matrix, etc.). An area of investigation is therefore the optimization of uncertainty-generating production activities such that they become less uncertain and more repeatable (work on repeatability^v). An alternative intervention in production is to focus on building uncertainty-immune systems (instead of minimizing uncertainties). This can be achieved by process monitoring and control which enables the system to recognize and possibly respond appropriately and timely to the formation of defects (work on adaptability^{vi}).

3.2.1. Repeatability

As discussed in §2.1.1, the constituent materials of FRPC may suffer from inherent defects. A method to ensure repeatability is the inspection of the constituent materials (prior to their usage in production) that will enable the use of batches with characteristics in acceptable ranges^{9,34}. Secondly, variations and deviations arise in production due to poor maintenance or poor calibration of equipment. In FRPC production, certain hardware units (i.e. injection and heating unit) need intensive

^v Repeatability: The closeness of agreement between independent test results obtained under repeatability conditions, i.e. conditions where test results are obtained with the same method on identical test items in the same laboratory by the same operator using the same equipment and within short intervals of time.

^{vi} Adaptability: A measure of the ability of the system to self-regulate on given conditions such that the system outcome is in better agreement with the desired characteristics, as compared to the case where it does not self-regulate.

maintenance due to their exposure to abrasive materials and extreme conditions. For instance, mixing problems that initiate from poor maintenance of the injection unit have already been discussed in §2.2.2. This highlights that maintaining and calibrating are important operations that need to be performed regularly despite them interrupting production. Thirdly, human handling associated to some uncertainties (§2.1.2) can be substituted by robotic handling which shows potential to achieve higher accuracy (minimal defects) and faster working rhythm. Automated and robotic technologies that have found applications in composites manufacturing (cutting, stacking, placing, etc.) have been reviewed by Fauster et al.⁸⁶. Finally, textile defects are largely associated to the textile inherent tendency to deform (i.e. easy tow movement). To minimize this behavior, textiles often contain tackifiers or binder materials which are activated under certain conditions and facilitate processes like preforming, draping and handling⁸⁷.

3.2.2. Adaptability

Process monitoring and control dedicated to FRPC production has been developing continuously over the last two decades. The vast range of different technologies, their benefits and limitations have been previously reviewed^{88,89}. Monitoring and control concepts can be applied in the filling or curing stage. Filling monitoring and control addresses all uncertainties associated to the reinforcement (§ 2.1) or the interface between tool and reinforcement (§ 2.3) collectively. The central idea is the detection of the flow front in key positions of the tool and the exploitation of the matrix arrival information to modify (manually or automatically) filling parameters (activate/deactivate vents and inlets, change the injection pressure or flow rate, etc.)^{90,91}. The automatic modification of filling parameters is achieved in these works by optimization algorithms that identify the optimal course of action in real-time.

Cure monitoring and control addresses the uncertainties associated to the polymeric matrix (§ 2.2) or the thermal conditions (§ 2.4) collectively. The concept here is based on detecting continuously the degree of cure on one or more areas of the curing part and use this information to modify parameters associated to curing (i.e. identify and initiate the end of the process)^{92,93}. In this case, heavy automatic modifications of the process parameters are on a premature stage (i.e. in relation to industrial readiness or compliance with relevant standards). Possible results include cycle reduction and prevention of over-curing which is associated to matrix degradation and stress concentrations that lead to distortions.

3.3. Risks of uncertainty minimization approaches

Work on predictability, repeatability or adaptability as described above is not risk free. Errors in the results may originate from inherent material uncertainties, model or equipment deficiencies and from the unintended influence of the selected approach on the materials.

(a) Inherent material uncertainties: Material uncertainties will emerge in any experimental process, including material characterization that is a prerequisite of work on predictability (i.e. determination of permeability or kinetic parameters). For instance, the typical 20-30% scatter of permeability that appears in permeability studies will have a corresponding impact on simulation reliability. The apparent paradox is that work on predictability suffers from one of the uncertainties it is trying to minimize (inherent to defects). However, the difference is that the specific problem in permeameters appears in a far more controlled environment where it can be properly investigated.

(b) Model deficiencies: Modelling suffers from model simplifications: the established flow models are solutions of Darcy's Law with certain simplifying assumptions such as constant viscosity (the initiation of curing starts only after filling is

complete), the absence of void content, and more⁶⁵. A much discussed simplification in established cure modelling is that they do not account for diffusion⁶⁹ while different model approaches for the determination of kinetic parameters are still investigated⁸⁵. Meanwhile, translation of raw measurements from sensors to curing degree is in many cases on a rather early stage partially due to the fact that the measurement may be affected not only by curing but also other phenomena. For instance, an ultrasound measurement contains information for both curing and void content⁹⁴.

(c) Diversity of equipment and their respective procedures: Work on predictability relies on the construction of permeameters. Repeatability introduces pre-production inspection equipment as well as automation/ robotics in-production. Finally, adaptability introduces sensing systems that should be integrated in the production system. What all of the above equipment has in common is that the equipment itself and the procedure it uses, are not standardized for use in FRPC manufacturing. Current applications are custom fits to specific production setups whose principles may vary among different providers. This raises the question which equipment and procedure produces the best results for a given case. Moreover, customized solutions unavoidably include higher risk (risk of malfunction or other non-intended behavior) as compared to standardized equipment exactly because they are unprecedented.

(d) Influence on the material properties: Work on repeatability and adaptability may affect the materials under production. Inspection of dry textiles prior to production includes additional handling and therefore increases the total risk of tow misalignment and preform deformation. Moreover, the use of tackifier or binder materials in the textiles have been found to affect permeability and flow^{95,96}. Also, sensor integration in the tool or part which is essential in most sensor applications may have a negative impact. Sensing elements within the part constitute a foreign body and a threat to

structural integrity when their geometry is not close to the one of reinforcement fibers⁹⁷. Integrating the sensor in the tool may under certain conditions disrupt the thermal field close to the mold-mounted sensor thus creating thermal and curing gradients⁹⁸.

4. Investigation of the economic potential of process monitoring and control

There is an important point to consider when discussing about the cost implications of poor reproducibility and of approaches to minimize it. Poor reproducibility caused by uncertainties results in cost increase (i.e. scrap cost) and even if work on uncertainty minimization (i.e. process monitoring and control) was error free (which as discussed in §3.3 is not the case), it still produces additional cost burdens than need to be less than the prevented ones in order for it to make sense. This section will examine the potential cost benefits and burdens of Process Monitoring and Control (PMC).

4.1. Process monitoring and control procedure

Process monitoring can result in two different cost saving scenarios: The first case leads to cost benefits by detecting scrap earlier in the production line: in the filling or curing step instead of later Non-Destructive Inspection (NDI). As such, all costs of resources associated to steps between the early scrap detection and NDI can be avoided. As this type of PMC takes no other action it will henceforth be referred to as **passive PMC**.

The second case leads to cost benefits by controlling the process conditions such that a defect is minimized (and therefore some scrap may be avoided). As this type of PMC directly counteracts the formation of a defect, it will henceforth be referred to as **active PMC**.

4.2. RTM case studies

Based on a typical aerospace RTM production chain, 3 case studies for PMC were investigated:

- Case Study V1: Typical RTM process with no PMC. Here, scrap can only emerge in NDI (Scrap NDI V1).
- Case Study V2: RTM with passive PMC. Here scrap can emerge in curing (Scrap Cure V2) or NDI (Scrap NDI V2) but the total scrap rate is equal the one of Case Study V1.
- Case Study V3: RTM with active PMC. Here, scrap can emerge in curing (Scrap Cure V3) or NDI (Scrap NDI V3) but due to control there is a decrease in the total scrap as compared to Case Study V1.

The manufacturing cost estimations were performed using a self-developed cost tool, ALPHA, which was described elsewhere in detail^{99,100}.

4.2.1. Quantification of the financial influence of PMC

In order to quantify the cost efficiency of PMC in the Case Studies V2 and V3 compared to Case Study V1, the following parameters are defined:

- Monitoring efficiency: The ratio of number of detected unacceptable defects to total unacceptable defect number per part. It is assumed that each unacceptable defect detection corresponds to reality (the particular defect indeed exists).
- Control efficiency: The ratio of number of initially unacceptable defects that through control became acceptable to the total number of controlled defects per part (successful or not).
- Normalized part cost: The ratio of cost per part with PMC to cost without PMC.

To demonstrate holistically the influence of monitoring and control efficiency (0% to 100%) the normalized part cost for an assumed total scrap rate of 20% (with no PMC) was calculated. Scrap rate typically is not that high when rework/ repair is involved but it was chosen in this case due to additional limitations of reworking in the aerospace

industry as compared to other industries. Nevertheless the effect of other scrap rates is also discussed below.

The resulting surface represents the cost saving potential of PMC with respect to the system capability to detect and/or control part quality (Figure 4.a).

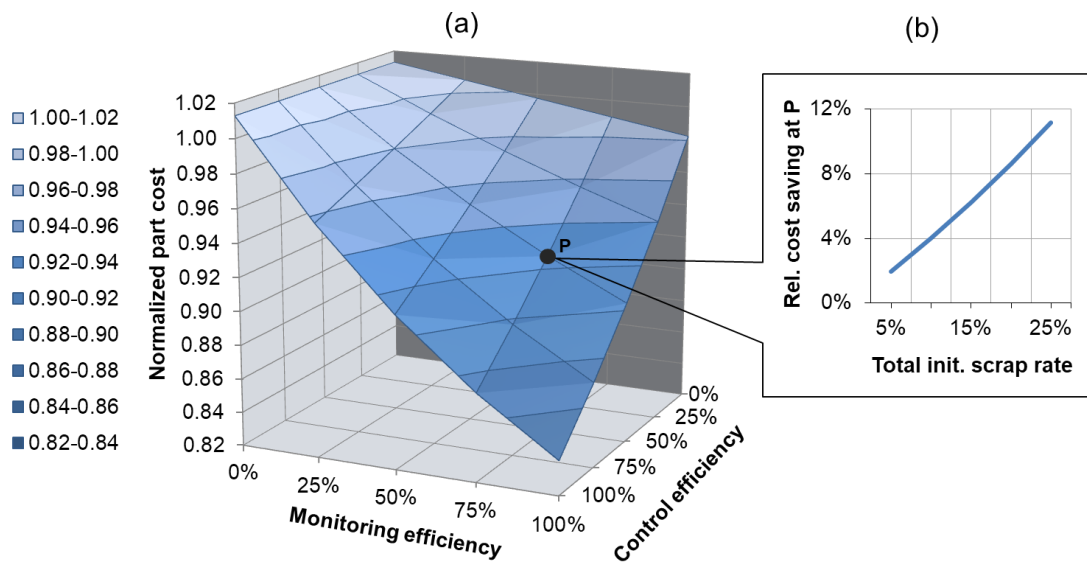


Figure 4. (a) PMC performance surface for total initial scrap rate of 20%. At the exemplary point P, 75% of the unacceptable defects are detected by the monitoring system and 50% of them can be compensated by the control system. (b) Relative cost savings at point P for other scrap rates.

The surface can be interpreted as follows: Case Study V01 corresponds to cost efficiency of 1 and is not part of the surface as it does not include PMC at all. The line of zero monitoring effectiveness of the surface corresponds to cost efficiency slightly above 1. This means that there is a small cost burden caused by PMC equipment. The line of zero control effectiveness of the surface corresponds to Case Study V02 where there is only passive PMC. At monitoring efficiency of 100% (the detection of all scrap at the curing stage and no scrap at NDI) the financial efficiency is ~0.98 (small benefit) and is caused by avoiding manufacturing steps that would normally follow before the

scrap detection. Case Study V03 is the rest of the surface. At the random point P on the surface seen in Figure 4, 75% of defects detected and 50% of these detected defects become acceptable (a realistic case considering that not all defects can be detected or reversed by control), the costs will drop to ~0.92. In the absolute best case scenario for Case Study V03 when all defects are prevented by control, the part cost is ~84% of the cost without PMC. However, this cost reduction of about 16% corresponds specifically to the initial scrap rate of 20%. An analysis for other initial scrap rates reveals an increase of the cost reduction due to PMC at point P (75% monitoring and 50% control efficiency) with scrap rate increase (Figure 4.b).

The above estimation confirms the two main possibilities for cost reduction that result from PMC. First, it is possible to implement monitoring just to detect scrap in-line (during curing or filling) instead of NDI (Case Study V2). The benefit for such a system depends highly on the costs of implementing monitoring, its efficiency in detecting scrap and the economic distance to the original point of scrap detection. The second possibility is to reduce the scrap rate by smart process control based on in-line sensor data (Case Study V3). This approach has large leverage in reducing manufacturing costs and especially in composite industry where high raw material costs and laborious operations (i.e. extensive preforming and process preparation steps) in combination with low recycling possibilities make scrap parts highly costly.

5. Conclusion

This paper makes the following advancements in understanding and enhancing the reproducibility of LCM processes:

- The factors contributing to poor reproducibility were categorized, isolated and described based on the most current knowledge.

- The impact of the above factors on production was quantified based on past work. In certain cases where relevant past work did not exist the impact was quantified by original numerical results (§ 2.4.1, 2.4.2).
- The available concepts and approaches to minimize the factors contributing to poor reproducibility were analyzed with respect to their capabilities and limitations.
- A cost estimation was performed for a typical industrial-ready RTM line and the impact of process monitoring and control on the production cost per part was quantified for a wide range of process monitoring and control capabilities.

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