A NOVEL EXPERIMENTAL APPROACH TO QUANTIFY DUAL-SCALE FLOW IN LIQUID COMPOSITE MOLDING

Benedikt Neitzel[1], Florian Puch[1,2]

[1] Plastics Technology Group, Department of Mechanical Engineering,
Technische Universität Ilmenau, 98693 Ilmenau, Germany, benedikt.neitzel@tu-ilmenau.de

[2] Thüringisches Institut für Textil- und Kunststoff-Forschung e.V., 07407 Rudolstadt, Germany

Keywords: Void formation, Permeability, Liquid composite molding, Dual-scale flow, Fiber reinforced plastics

ABSTRACT

A novel methodology is presented, that uses photo-reactive resin to freeze the flow on demand. Depending on the transparency and thickness of the textile preform, instant crosslinking of photo-reactive functional groups is possible, resulting in a freezing of the flow. Only by spontaneous crosslinking of the complete specimen, both void formation and void transport are frozen in situ. The presented methodology extends the examinable area of microscopic flow phenomena, compared to conventional methods that only allow point-wise measurement of the flow [1].

An experimental setup for the impregnation of glass fiber fabric with photo-curable resin is presented, consisting of an injection mold with a transparent glass cover and ultra violet (UV) light emitters to freeze the flow at different filling levels. The resulting specimen are investigated by microscopy to capture the dual-scale effect of the complete flow front. With the novel methodology, dual-scale flow effects become clearly visible and quantifiable. The results enable conclusions on the flow related formation of voids and allow a comparison of theoretical models with quantified measurements.

1 INTRODUCTION

During impregnation of textile preforms by liquid composite molding (LCM), the processing parameters take major influence on the quality of the produced parts. Due to the negative impact of voids on the mechanical properties of composite structures, void formation is investigated in a multitude of studies [2–4]. Irregularities of resin flow inside the mold may result in void formation, causing negative effects on the mechanical properties such as flexural modulus and flexural strength [5] of the composite.

One major cause of void formation is irregular microscopic flow of resin, impregnating the textile preform. Woven textile structures, typically used for reinforcement of composites, consist of multiple fibrous tows arranged in warp and weft-direction. The individual tows, consisting of a multitude of filaments, show a significantly decreased permeability compared to the channels between the tows.

These permeability differences may result in a so called dual-scale flow effect, with different impregnation rates inside the tows and the channels [6–9]. [Fig. 1]

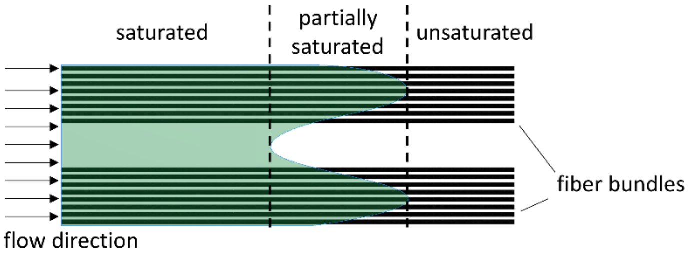

Figure 1: Saturated, partially saturated, and unsaturated regions of the flow front in a unidirectional preform. [9]

The permeability differences result in diverging impregnation velocities in both domains. At low flow velocities, the tows may be impregnated quicker than the cannel, because of capillary forces between the filaments. In this case, air will be entrapped in the channel, creating so-called macro-voids. At high flow velocities, the channel may be filled quicker than the surrounding tows and result in entrapped micro-voids between the filaments. If the flow in both domains is sufficiently balanced, the impregnation is nearly uniform and the entrapped air is minimized. Semi-empirical models are used to characterize the effect and to calculate the resulting void formation [2,10].

The modified capillary number Ca* represents the ratio of viscosity-dependent and capillary-force dependent flow. Dominance of one of the respective flow phenomena results in an emerging dual-scale effect of the flow. The extent of the disproportionality of the modified capillary number influences the extent of the dual-scale velocity differences. The modified capillary number is calculated as in equation (1):

$$Ca^* = \frac{\mu \cdot \bar{u}}{\gamma \cdot cos(\theta)} \quad (1)$$

Where μ is the dynamic resin viscosity, $\bar{u}$ the averaged macroscopic flow velocity, γ the surface tension and θ the contact angle between resin and fibers. While the modified capillary number contains information about the ratio of flow inducing forces, it does not imply immediate information about the uniformity of flow inside a textile preform. To set the modified capillary number in context to the uniformity of flow, the application of GUEROULT'S model of the dual-scale flow ratio can be considered:

$$\frac{\Delta t_t}{\Delta t_c} = \frac{K_c}{K_t} \cdot (1 - \varphi_{FT}) \cdot \left[1 - \frac{F_S \cdot K_c \cdot \varphi_{FT}}{d_{Fi} \cdot (1 - \varphi_{FT}) \cdot L_t \cdot Ca^*} \cdot ln\left(\frac{Ca^* \cdot d_{Fi} \cdot (1 - \varphi_{FT}) \cdot L_t)}{F_S \cdot K_c \cdot \varphi_{FT}} + 1\right)\right] \quad (2)$$

Equation (2) forms a ratio of flow in the tows and the channels between the tows. Δt_t describes the duration needed for resin to fill a given length of a tow in the fabric. Whereas Δt_c describes the duration needed to fill the exact same given length of a channel. Additional to the modified capillary number, this ratio of durations is calculated considering properties of the textile preform, such as the microscopic permeability of the channels K_c and the tows K_t, the fiber volume content inside the tows φ_{FT}, the diameter of the filaments d_{Fi}, the length of a tow L_t and a shape factor F_S which depends on whether the flow is parallel or transverse to the tow direction.

According to the calculation, the flow can be categorized into three different cases (Fig. 2):

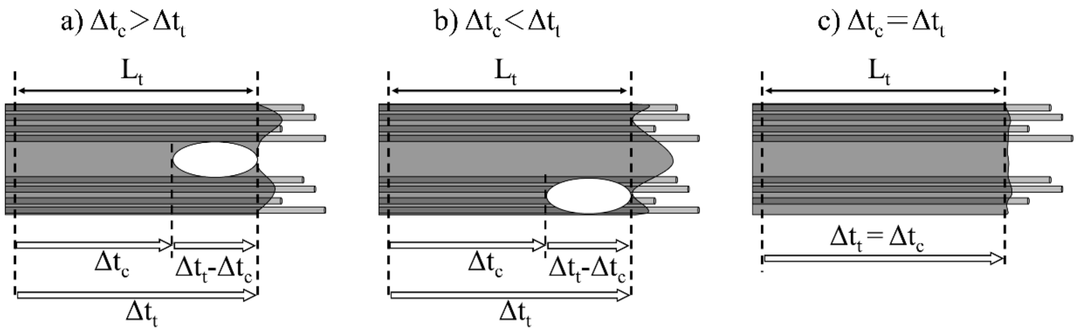

Figure 2: Different durations of dual scale resin flow for a given length. a) Longer filling duration inside channels b) longer filling duration inside tows c) equal filling durations [11]

When the duration ratio is greater than 1, the resin velocity is slower inside the channel and macro-voids may be formed. At a calculated value lower than 1, the resin velocity in the tow is slower and micro-voids may be entrapped. At a ratio of 1, the flow is uniformly distributed and no voids will be formed.

To validate the semiempirical model, a reproductible visualization of the dual-scale flow needs to be accomplished. State of the art for analyzing the dual-scale flow, void formation and void transportation are optical methods [12–14]. Moreover, x-ray, ultrasonic and micro-CT measurements can be used to observe resin flow in closed cavities [2]. Due to the restricted field of view or limited resolution of these conventional methods, only point-wise observation of local flow states is possible [1]. To provide a detailed insight into dual-scale flow, large areas of specimen must be analyzed to receive statistically relevant information about influences of macroscopic flow effects. A novel methodology has been proposed, that uses UV photo-reactive resin to freeze the flow on demand [9]. Depending on the transparency of the textile preform and the thickness of the specimen, instant crosslinking of photo-reactive functional groups of the resin is possible, resulting in a spontaneous freezing of the flow. This approach enables a holistic view on the dual-scale flow inside the complete fabric. However, pressure gradients during crosslinking, may result in partial progression of the flow below the irradiated surface of the specimen. Only in the case of spontaneous crosslinking of the complete molded part, both void formation and void transport are frozen in situ.

2 EXPERIMENTAL SETUP

2.1 Flow front examination

To create specimen with a frozen flow front, a textile preform is placed in an aluminum mold and covered with a glass-plate that is transparent for light in the ultraviolet (UV) range (Fig. 3). The mold is designed with a flash gate to ensure a linear flow. An UV-emitter array consisting of three YG-TGD20-405 LED spotlights from Shenzhen Creality 3D Technology Co, Ltd., Shenzhen, China with a system power of combined 23.4 W and emitted wavelengths of 400 – 405 nm is mounted 70 mm above the mold. When the emitters are switched on at 100% intensity, the flow stops virtually immediately. The textile reinforcement used is a plain weave glass fiber fabric, type 92130 manufactured by Porcher Industries Germany GmbH, Erbach, Germany.

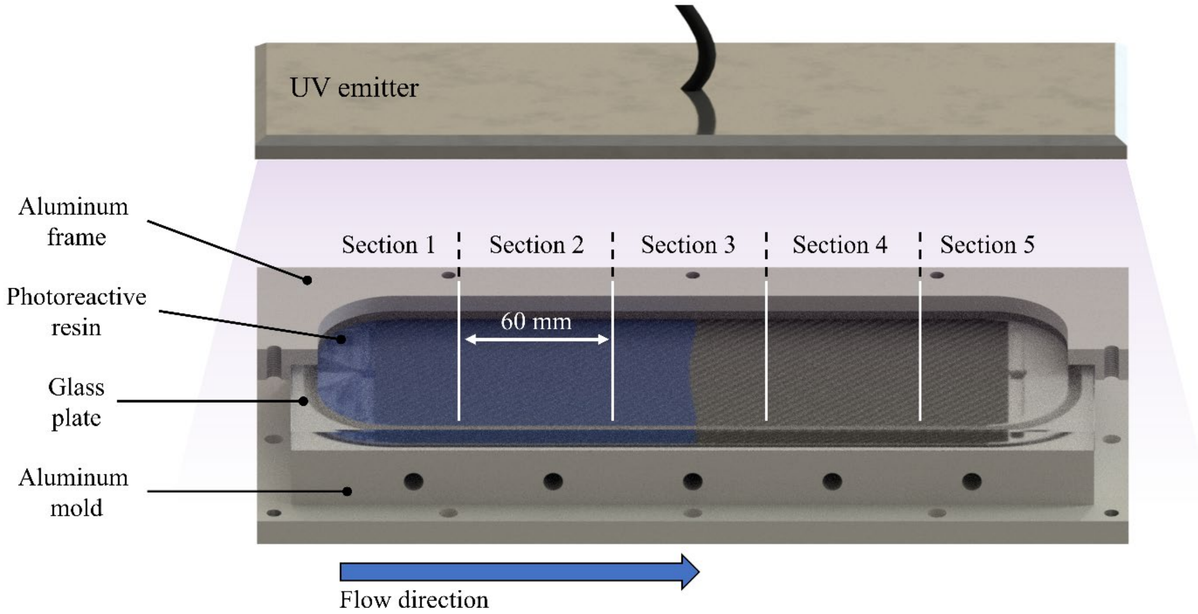

Figure 3: Experimental setup for UV-curable specimen

Markings on the glass every 60 mm of the flow path, enable stopping the flow at varying defined pressure gradients. To achieve uniform compression of the fabric, eight bolts inside an aluminum frame cover are tightened with a defined torque to press the glass plate onto the fabric with a total pressure of 4.4 MPa. Injection pressures from 0.05 MPa to 0.25 MPa were used to fill the cavity. The impregnation was recorded on video, to allow the calculation of the according flow velocities in each section. Resulting cured specimen were optically analyzed using a Keyence VHX-7000 microscope at 100× magnification.

2.1 Depth of cure measurement

The applicability of the given methodology using UV-curable resin, relies on sufficiently fast crosslinking, so that the flow front throughout the complete thickness of the laminate is stopped immediately. If the flow below the irradiated surface progresses, the measurements of the dual-scale effect are inaccurate. To define the limitations of applicable specimen thickness, multiple layers of fabric were stacked in the same mold setup (Fig. 4).

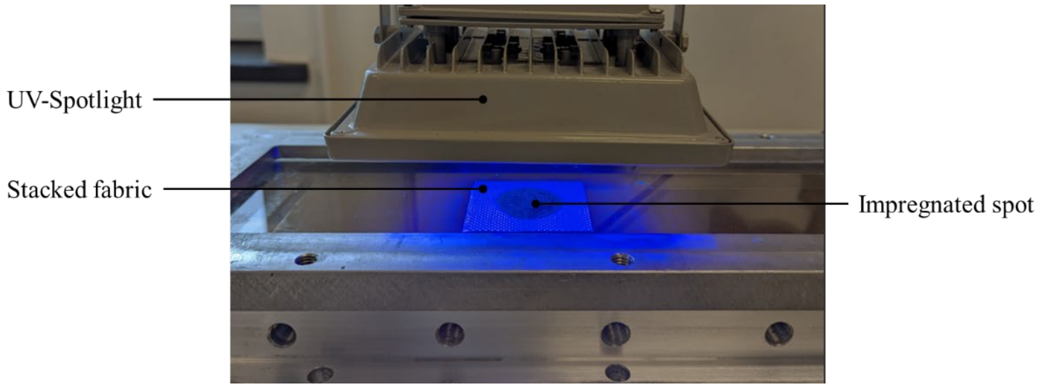

Figure 4: Experimental setup for depth of cure determination

For each layer a drop of 0.5 ml resin was put onto the stack of fabric. A microcontroller was programmed to power the spotlights for defined durations. The duration was increased in steps of 50 ms after each experiment and it was checked how many layers of fabric were cured after the radiation. For each repetition a new stack of fabric with a new batch of resin was used.

3 RESULTS AND DISCUSSION

The application of the methodology for instantly curing one layered specimen, shows clearly distinct edges between impregnated and dry areas (Fig. 5).

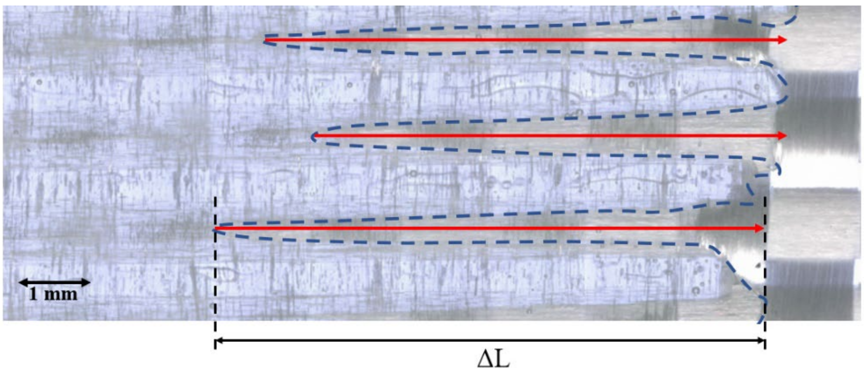

Figure 5: Measurement of the flow path difference ΔL

The optical analysis of the single layer specimen indicates, that the measured difference between the flow front position inside the tows compared to the channels, is nearly constant along the complete mold at constant pressure. With increasing pressure gradients, increasing flow path differences are observed. At rising modified capillary numbers, the flow path difference is increased (Fig. 5). The observations are in good agreement with the coherences of equations (1) and (2). While the measurements of flow path differences at the first segment of the mold are in good agreement to the findings of GUEROULT ET AL., increasing flow distances show noticeable deviations between measurements and the theoretical model. This effect can be attributed to cross flows in the transverse direction of the fibers during impregnation. While the theoretical model is focused only on the local characterization of dual-scale flow, the measurements of the flow path difference are affected by cross flow effects during the complete impregnation process. [9]

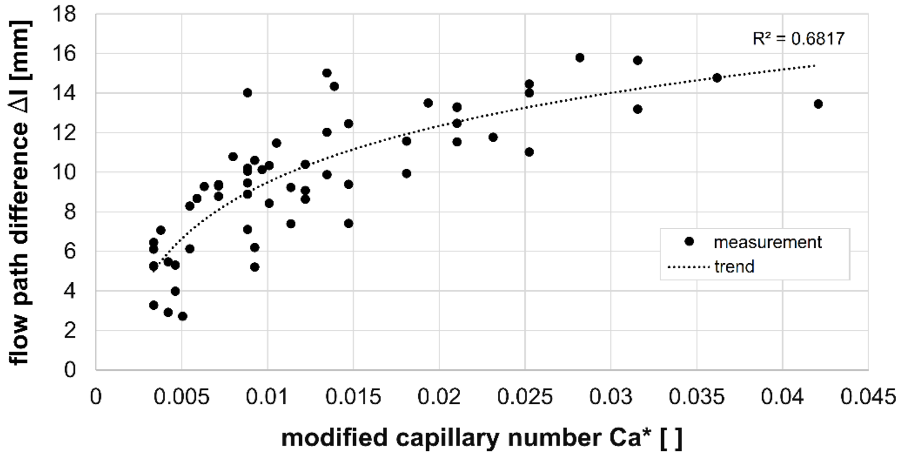

Figure 6: Trend of the flow path difference in coherence with the local modified capillary number. [9]

Radiation of the specimen is applied only to the upper surface of the mold. To evaluate the error of flow progress along the cross-section of the composite, the experimental setup for depth of cure determination (Fig. 4) was developed. At first the influence of room lighting was assessed by observing an impregnated single layer of fabric inside the mold without engaging the UV-emitter. The resin did not pass the point of gelation in a timeframe of 30 minutes, so the artificial lighting inside the laboratory was neglected for further investigations. The first layer of glass fiber was cured to a solid state after a radiation duration of 250 milliseconds. After a total of 450 ms, the first and second layer were solid and inseparably connected. With increasing number of layers, the thickness of the specimen increases by 0.3 mm. Additional to the increasing cross section, the transmission of the UV light is lowered by each layer of fibers. Both effects lead to increasing durations for each added layer to solidify.

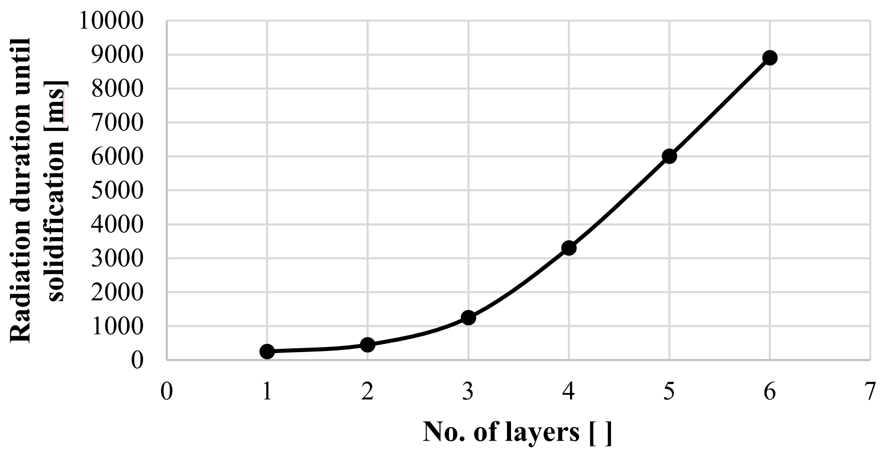

Figure 7: Duration of UV-radiation needed to solidify multiple layers of glass-fibers

While the given experimental setup is suited to assess the duration of crosslinking for each layer to pass the state of sufficient crosslinking and a solid connection of the layers, it is not applicable to determine the exact time when gelation takes place and the flow stops. To evaluate the error of measurement of the flow path differences, the time between the passing of gelation of the surface until the gelation of the bottom of the first single layer must be assessed. With the results of the first layer being cured to a solid state after 250 ms and the second layer being connected after 450 ms, it can be concluded, that the solidification itself from surface to bottom of one layer has to be completed in the timespan of 200 ms. These results are based on the assumption, that additional crosslinking after radiation can be neglected in the experimental setup. The proportional through-thickness progression of the flow front, depending on the depth cure of an exemplary specimen with four layers of glass fiber textile is quantitatively depicted in Fig. 8.

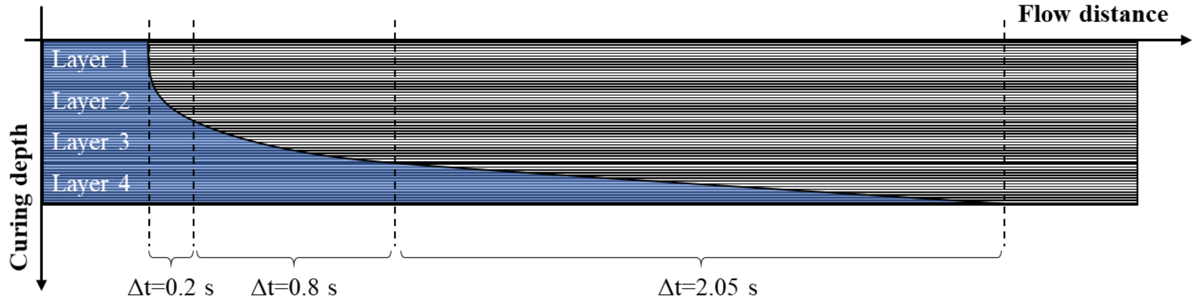

Figure 8: Through-thickness flow progression inside composite with four layers of glass fiber textile reinforcement

The average flow velocity of the entire set of experiments was 0.31 mm/s with a maximum velocity of 1.00 mm/s at the parameter setting of the highest pressure gradient in the first mold section and an injection pressure of 0.25 MPa. This results in an average inaccuracy of 0.07 mm in length measurement and a maximum of 0.20 mm. The relative error of the flow path difference is shown in Table 1.

Experimental point	Flow velocity [mm/s]	Flow path difference [mm]	Inaccuracy of length measurement [mm]	Relative error of flow path difference [%]
Average	0.31	9.69	0.07	1.44
Max. velocity	1.00	14.81	0.20	2.70

Table 1: Average and maximum relative error of flow path determination

4 CONCLUSIONS

With the novel methodology, dual-scale flow effects become clearly visible and quantifiable in a holistic approach. The results enable conclusions on the flow related formation of voids and allow a comparison of theoretical models with quantified measurements. Evidence was provided, that single layer glass fiber reinforced specimen of 0.30 mm thickness are solidified in a sufficiently short timespan to achieve measurements of the dual-scale flow with comparatively small relative errors of up to 2.70%.

With increasing numbers of layers and hence increasing specimen thickness, together with reducing UV transmission rates, the required radiation duration for sufficient crosslinking increases significantly. Depending on the flow velocity of the resin, the applicability of the presented methodology is hence limited to only a small number of reinforcement layers.

However most relevant for the validation and adaption of theoretical models is the case of defined flow in a single layer of textile preforms. With the option of adding a second layer to include layer interactions with still comparably short crosslinking durations, the methodology is highly appropriable for scientific use cases.

REFERENCES

[1] K. M. Pillai, "Modeling the Unsaturated Flow in Liquid Composite Molding Processes: A Review and Some Thoughts," *Journal of Composite Materials*, vol. 38, no. 23, pp. 2097–2118, 2004.

[2] M. Mehdikhani, L. Gorbatikh, I. Verpoest et al., "Voids in fiber-reinforced polymer composites: A review on their formation, characteristics, and effects on mechanical performance," *Journal of Composite Materials*, vol. 53, no. 12, pp. 1579–1669, 2019.

[3] S. F. M. de Almeida and Z. d. S. N. Neto, "Effect of void content on the strength of composite laminates," *Composite Structures*, vol. 28, no. 2, pp. 139–148, 1994.

[4] P.-O. Hagstrand, F. Bonjour, and J.-A. Månson, "The influence of void content on the structural flexural performance of unidirectional glass fibre reinforced polypropylene composites," *Composites Part A: Applied Science and Manufacturing*, vol. 36, no. 5, pp. 705–714, 2005.

[5] B. Neitzel and F. Puch, "Application of capacitive sensors and controlled injection pressure to minimize void formation in resin transfer molding," *Polymer Composites*, vol. 44, no. 3, pp. 1658–1671, 2023.

[6] C. H. Park, A. Lebel, A. Saouab et al., "Modeling and simulation of voids and saturation in liquid composite molding processes," *Composites Part A: Applied Science and Manufacturing*, vol. 42, no. 6, pp. 658–668, 2011.

[7] E. Ruiz, V. Achim, S. Soukane et al., "Optimization of injection flow rate to minimize micro/macro-voids formation in resin transfer molded composites," *Composites Science and Technology*, vol. 66, 3-4, pp. 475–486, 2006.

[8] H. Teixidó, J. Staal, B. Caglar et al., "Capillary Effects in Fiber Reinforced Polymer Composite Processing: A Review," *Frontiers in Materials*, vol. 9, 2022.

[9] B. Neitzel and F. Puch, "Optical Detection of Void Formation Mechanisms during Impregnation of Composites by UV-Reactive Resin Systems," *Journal of Composites Science*, vol. 6, no. 11, p. 351, 2022.

[10] S. Gueroult, A. Lebel-Lavacry, C. H. Park et al., "Analytical modeling and in situ measurement of void formation in liquid composite molding processes," *Advanced Composite Materials*, vol. 23, no. 1, pp. 31–42, 2014.

[11] B. Neitzel, *Formation of Voids due to Transitions in Permeability and Cavity Diameter during Resin Injection Processes*, Aachen, 05.09.2022.

[12] N. Patel, V. Rohatgi, and L. J. Lee, "Micro scale flow behavior and void formation mechanism during impregnation through a unidirectional stitched fiberglass mat," *Polymer Engineering and Science*, vol. 35, no. 10, pp. 837–851, 1995.

[13] Y.-T. Chen, H. T. Davis, and C. W. Macosko, "Wetting of fiber mats for composites manufacturing: I. Visualization experiments," *AIChE Journal*, vol. 41, no. 10, pp. 2261–2273, 1995.

[14] C. Lystrup, A. George, B. Zobell et al., "Optical measurement of voids in situ during infusion of carbon reinforcements," *Journal of Composite Materials*, vol. 55, no. 6, pp. 775–786, 2021.