

THE EFFECT OF MATERIAL VARIABILITY ON THE PRE-GELATION BEHAVIOUR OF PREPREG COMPOSITES

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ABSTRACT

Processes such as forming and AFP have become increasingly popular, and these manufacturing techniques require manipulation of the uncured pre-impregnated thermoset matrix composite material (prepreg). Interest in the initial state or pre-gelation behaviour of prepregs has grown due to these manufacturing trends as well as the increasing recent research connecting this regime to processing defects such as wrinkling and porosity. Historically, the pre-gelation behaviour of prepreg has been relatively unexplored due to the challenges that traditional testing methods have in handling the soft raw prepreg. It is shown here that Digital Image Correlation (DIC) can capture the prepreg behaviour through the entirety of a cure cycle on both a local and global level. This work presents the global response of the prepreg, showing the true pre-gelation response, which differs markedly from what has traditionally been presented as the expected behaviour. Additionally, the variability in local prepreg response can be linked back to local strain variations observed using DIC. With the insight into the variability of a ply and the pre-gelation strain response, the findings in this work emphasize the importance of understanding and accurately capturing the initial prepreg condition.

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1. INTRODUCTION

The composites industry has seen a shift in recent years, increasingly incorporating newer manufacturing methods such as AFP and forming. These newer methods involve handling raw material in different ways and therefore, a better understanding of the mechanisms that drive the raw material behaviour is needed. Recent research has focussed on the impact of the initial condition of the material on the development of defects such as wrinkling and porosity, which develop early during the process cycle [1–3]. However, it appears that no one has directly characterized the pre-gelation free-strain response of prepreg, and the literature typically assumes

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that the pre-gelation free strain response should follow a path similar to that depicted in Figure 1 [4].

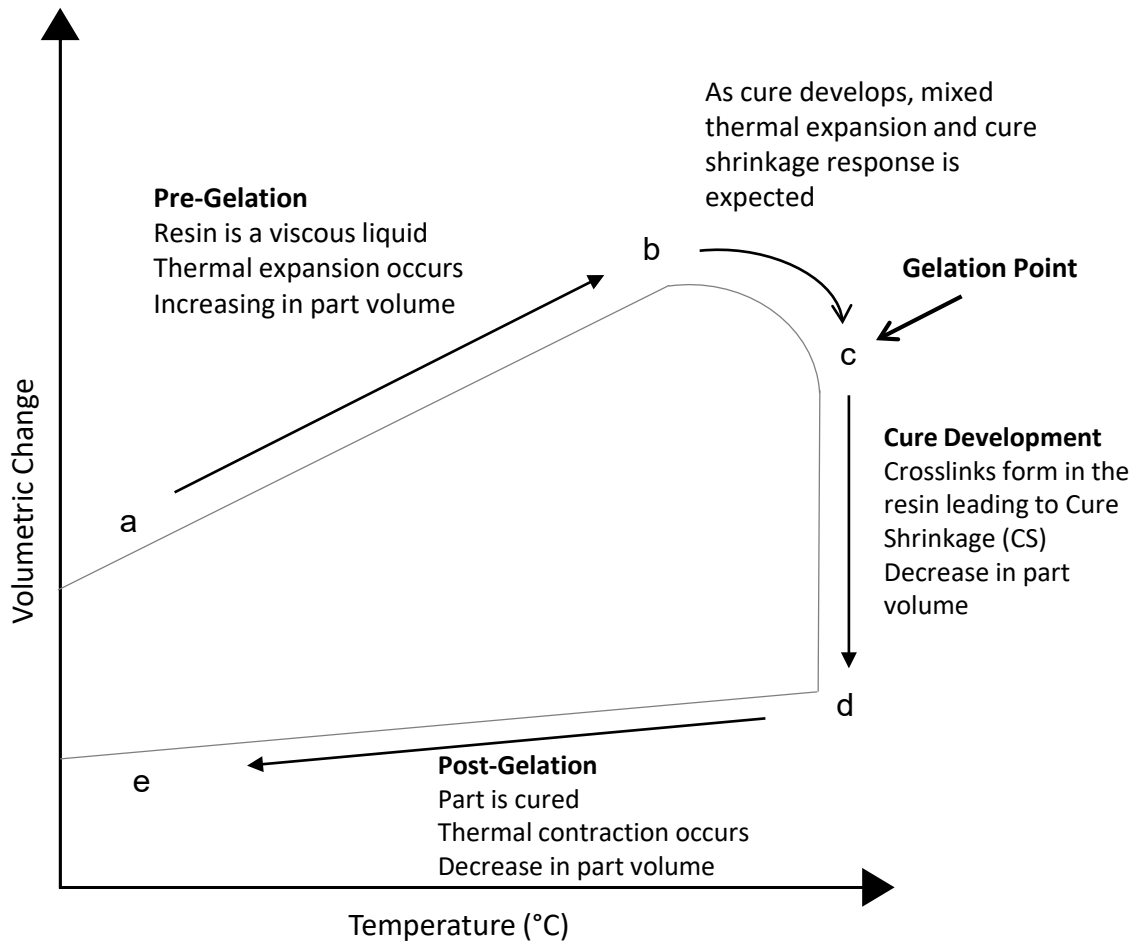


Figure 1. Schematic of the volumetric changes that a thermoset based prepreg system is traditionally expected to undergo during processing, as adapted from literature [4]. When heating initially, the resin in the ply is a liquid and is expected to expand (a-b). At some point during the heating cycle, the cure reaction will begin and will contribute to volumetric shrinkage (b-c and c-d). Finally, when the prepreg is cooled thermal contraction will shrink the prepreg (d-e)

It is typically believed that when heated, the uncured prepreg will expand through the thickness of the ply and in the direction perpendicular to the fibres. As resin cure starts, it is expected that this will lead to a corresponding shrinkage in the prepreg, with a linear relationship between strain and the degree of cure (DoC) [4]. In essence, the literature considers the resin in the pre-gelation prepreg to be acting as a very low modulus solid. This work evaluates this assumption, by directly observing the pre-gelation behaviour of prepreg using digital image correlation (DIC), a non-contact measurement technique. The results presented here are part of a more extensive study into

pre-gelation prepreg behaviour [5]. Throughout this study, the importance of the variability in the material and strain response during the early stages of processing is highlighted.

2. EXPERIMENTATION

2.1 Material

This study used an autoclave thermoset system composite material, AS4/8552 unidirectional (UD) prepreg (HexPly AS4 12K/8552; 35 %; 190 AW) [6]. Previous work has created process models for the resin cure kinetics and viscosity development [7,8]. These process models have been implemented in RAVEN simulation software [8], which has been used in this study to obtain cure and viscosity profiles, as well as the gelation point, for the different test temperature cycles.

2.2 Digital Image Correlation (DIC)

DIC systems use image analysis to observe the development of strain in a material. In the past, this technique had been used to study resin flow, crack development, and stress development in composite materials [9–15]. The use of a non-contact measurement technique in this study allowed the strain development of a prepreg sample through the complete process cycle to be captured. The DIC set-up captured the in-plane strains (ϵ_{11} and ϵ_{22}) of the UD prepreg sample.

2.2.1 DIC Equipment Set-Up

This study used a Correlated Solutions VIC-3D system. Two digital cameras were positioned approximately 100 cm above the sample position. The cameras were pointed at a 30° angle towards the sample. The cameras were connected to a computer and controlled by VIC 3D image acquisition software. LED lights placed on either side of the sample were used to illuminate the sample allowing a high contrast image to be captured.

The system was calibrated using a 9 x 4-inch calibration panel placed on the heading plate where the sample sits during testing. Several images of the calibration panels at varying positions were used to establish the height and angle of the cameras by tracking target spots on the panel.

2.2.2 Heating System

The samples were heated using a Peltier heating system controlled by an Anton Paar Rheometer. The Peltier heating system has a temperature range between -20 °C to 200 °C and was selected for its fast and accurate temperature control. Teflon tape was placed on the surface of the plate to reduce interaction between the sample and the substrate.

The temperature cycle selected for this experiment was designed to limit cure advancement during the temperature ramp segment of the cycle, isolating the thermal and chemical free strains. Figure 2 shows the cure cycle.

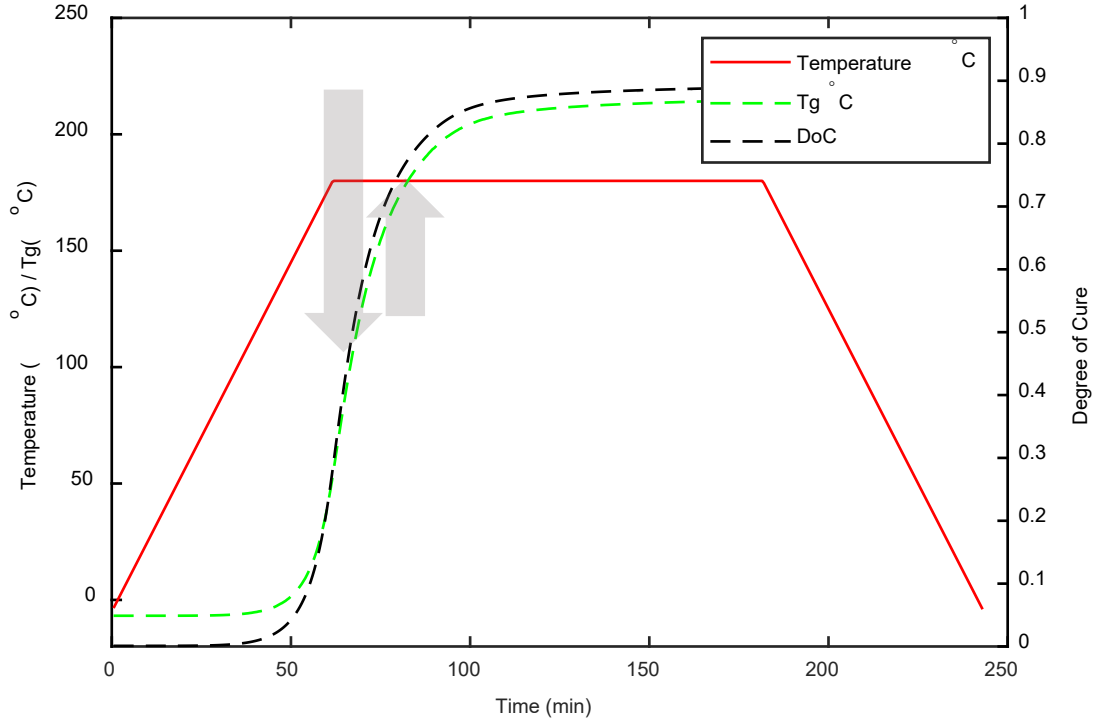


Figure 2. Temperature Cycle 1 used for DIC experiments. One hold cycle- ramp from -5 °C to 180 °C at a rate of 3 °C /min. Hold isothermally at 180 °C for 120 min. Ramp down to -5 °C at 3 °C /min.

2.2.3 Sample Preparation

AS4/8552 unidirectional prepreg was cut into squares approximately 25 mm by 25 mm. Samples of either 1 or 4 plies were made using a unidirectional lay up and de-bulked at room temperature for 10 minutes. After being laid up, samples were painted using a high-temperature Rust-Oleum white [16], and then a speckled layer of KRYLON black spray paint with a matt finish [17] was applied resulting in a random, high contrast speckle pattern.

2.2.4 Image Acquisition and Processing

After the sample was prepared and placed on the heating plate, the temperature cycle and the image acquisition were started in unison. Ever 30 seconds, an image was taken until the completion of the temperature cycle. Temperature data was recorded every second.

To process the images, the Correlated Solutions software was used. The sample was selected, and the software tracked the displacement of the speckles on the sample resulting in the local strain data. The average strain at each time step can be found for the sample during post-processing of the data. To extract the average strain data the extraction tool in the software was used. Lines running perpendicular to the fibre direction were drawn across the sample were selected using the data extraction tool. Once marked the average strain data were extracted for the selected line. The average of these results is taken, resulting in the average global strain in the transverse direction (ϵ_{22}) to the fibres.

3. RESULTS

3.1 Global Transverse Strain Behaviour

DIC results of the average transverse strain behaviour shows the pre-gelation behaviour response differs markedly from the traditionally assumed behaviour in the literature, as depicted in Figure 1. As opposed to observing a thermal expansion on the initial heating ramp, the results, as shown in Figure 3, showed only a small initial expansion at the start of the heating ramp, followed by a significant drop in strain.

Figure 3 shows the results for one DIC test performed on a 4 ply UD sample processed with the temperature cycle described in Figure 2. The results seen in the test presented here are consistent with, and representative of nine other tests with one hold temperature cycles and four tests with two hold cycles [5].

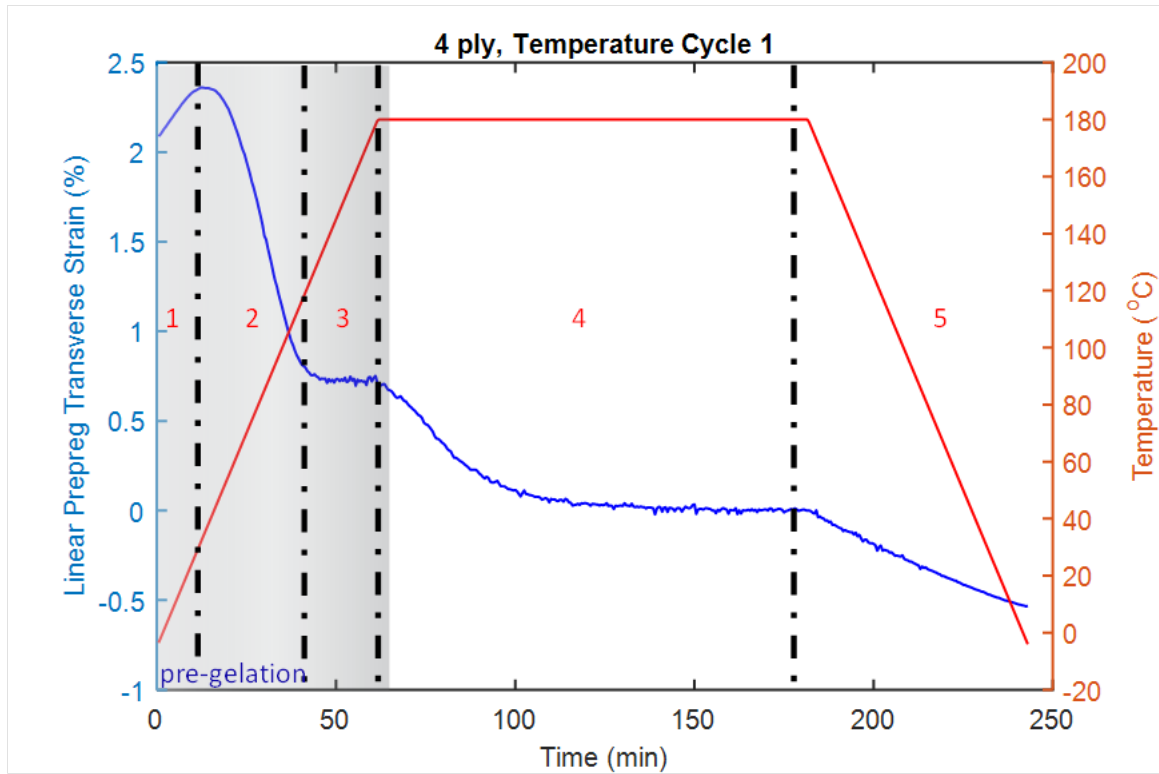


Figure 3. Measured temperature and linear prepreg transverse strain from DIC as a function of time. Zones delineate shifts in strain behaviour: Zones 1-3 identify different behaviour during the heating ramp, Zone 4 is the isothermal hold section of the test where cure advances, and Zone 5 is the cooldown ramp.

3.1.1 Observed behaviour in different zones

The average linear prepreg transverse strain response is presented in Figure 3 and Figure 4. The results have been broken down into zones based on shifts in the strain response. By examining each of the zones, a better understanding of the cause for the observed behaviour can be gained.

As the test starts, the sample is heated at a rate of 3 °C/min. Zone 1 focuses on the strain response during the start of this heating ramp, between -5 °C and approximately 30 ± 5 °C. During this time, the sample expands thermally. The observed transverse coefficient of thermal expansion (CTE) is $89 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$. For AS4/8552 prepreg or equivalent calculated linear prepreg values based on 8552 neat resin measurements, the literature reports CTE liquid values ranges from $74 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ to $373 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ [18–20]. The resin viscosity in Zone 1 is initially relatively high (above 600 KPa.s) but drops to about 9 KPa.s at 30 °C as the prepreg is heated. As the viscosity drops further during the heating ramp, the material expands less, until it begins to shrink in Zone 2. The initial CTE response of the prepreg as captured in Zone 1 is consistent with the range reported previously.

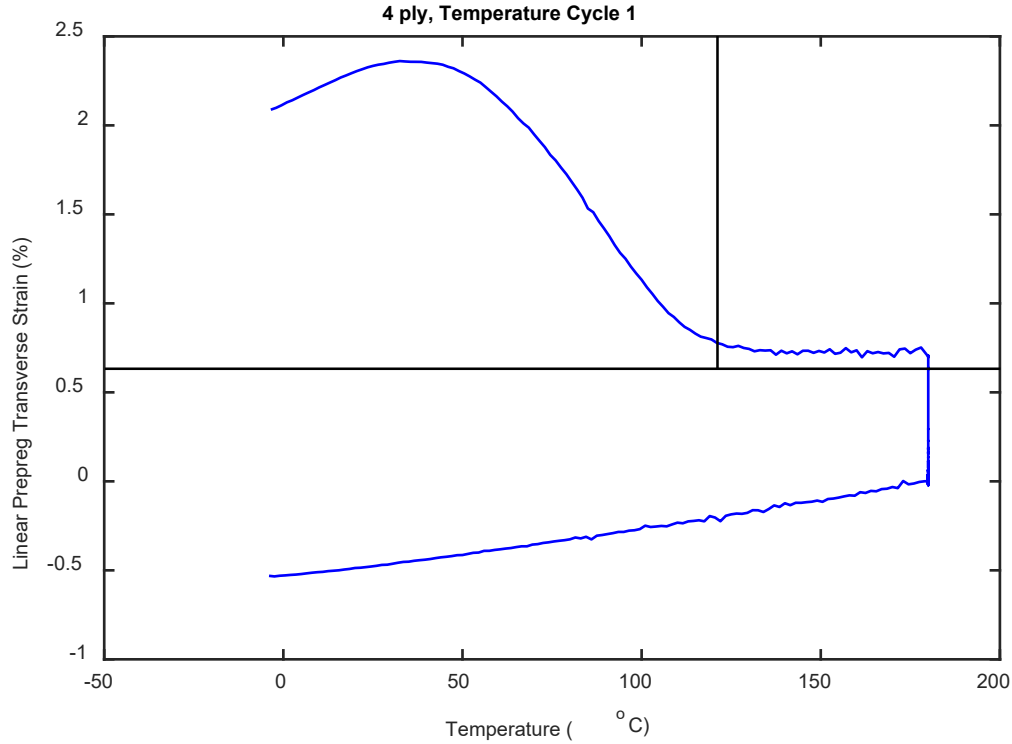


Figure 4. Measured linear prepreg transverse strain as a function of applied temperature, with Zones 1-5 from Figure 3 identified. This observed behaviour is markedly different from the traditionally expected behaviour as shown in Figure 1.

Zone 2 shows unexpected behaviour, in that the prepreg is shrinking in the transverse direction even as it is being heated. Previous work has highlighted that the prepreg is not fully saturated, and there is a variable resin distribution within the fibre bed of AS4/8552 UD prepreg. This leads to self-consolidation within the ply when heated [5,21,22]. The driving force leading to this ply saturation is the surface tension driven capillary action which occurs between the resin and the fibre bed within the core of a ply. The strain drop observed in this work can be linked back to this surface tension effect [22]. For the test presented in this paper, the strain response showed a decrease of -1.34 % between 40 °C and 105 °C. In this temperature range, the resin viscosity is low enough to flow (~3300 Pa.s down to ~20 Pa.s) and as the resin flows to fill voids within the prepreg, the ply consolidates.

In Zone 3, the material shows little change in strain. Although the sample is still being heated, cure starts in earnest (see Figure 2) and therefore in this zone, conflicting contributions of thermal expansion and cure induced shrinkage are expected. One might still expect the effect of thermal expansion to dominate initially, although at a decreasing rate, as in the trend depicted between points b and c in Figure 1. The absence of a change in strain observed in Zone 3 could be a result

of continuing resin flow at the microscopic scale, or perhaps shear flow of the resin relative to the fibre bed.

Zone 4 is during the isothermal hold and is therefore the isolated effect of cure shrinkage. Cure shrinkage of the neat resin is believed to be linear with cure [19]; however, the DIC results for the prepreg show a transition between pre- and post- gelation response. Pre-gelation, the linear coefficient of cure shrinkage (CCS), was found to be -0.37 %, and post gelation the CCS was found to be -2.18 %. The prepreg post-gelation response is consistent with the neat resin values reported in the literature [19]. The low CCS of the prepreg in the pre-gelation regime suggests that the resin cannot fully overcome the fibre-bed resistance and thus partially flows or shears relative to the fibre bed. Once the resin gels, the resin can overcome the fibre-bed resistance, and the prepreg CCS is consistent with the resin CCS.

Zone 5 represents the cool-down portion of the test. The prepreg in this zone is post-gelation, and as it cools down acts as a glassy solid. The thermal contraction yields a glassy CTE of $29.56 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ which is constant with the values of $33.3 \pm 0.07 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ reported in the literature [23].

3.2 Local Strain Variation Pre-gelation

The strain maps for each DIC image taken during the test can be used to visualize the local strain variations across a sample. 3D plots allow the visualization of the surface profile of the sample with the strain variable overlaid. Examination of these strain maps shows the non-uniformity of strain with time, particularly in the pre-gelation regime. Snapshots of the sample profile at key moments during the heating ramp are shown in Figure 5.

At the start of the heating cycle, the 3D DIC surface plot is uniform in strain (all orange), but as the sample is heated, bands of different strain magnitude appear. This variation is consistent with the change in the global strain response between Zone 1 and Zone 2. It appears that in the areas where surface-tension-driven resin flow has led to ply saturation, the strain maps show variability, with some regions showing negative strain responses double other regions. This would imply that within the ply, some regions see a larger consolidation than others. This is consistent with other studies that have shown the variability in a ply of AS4/8552 [5,24].

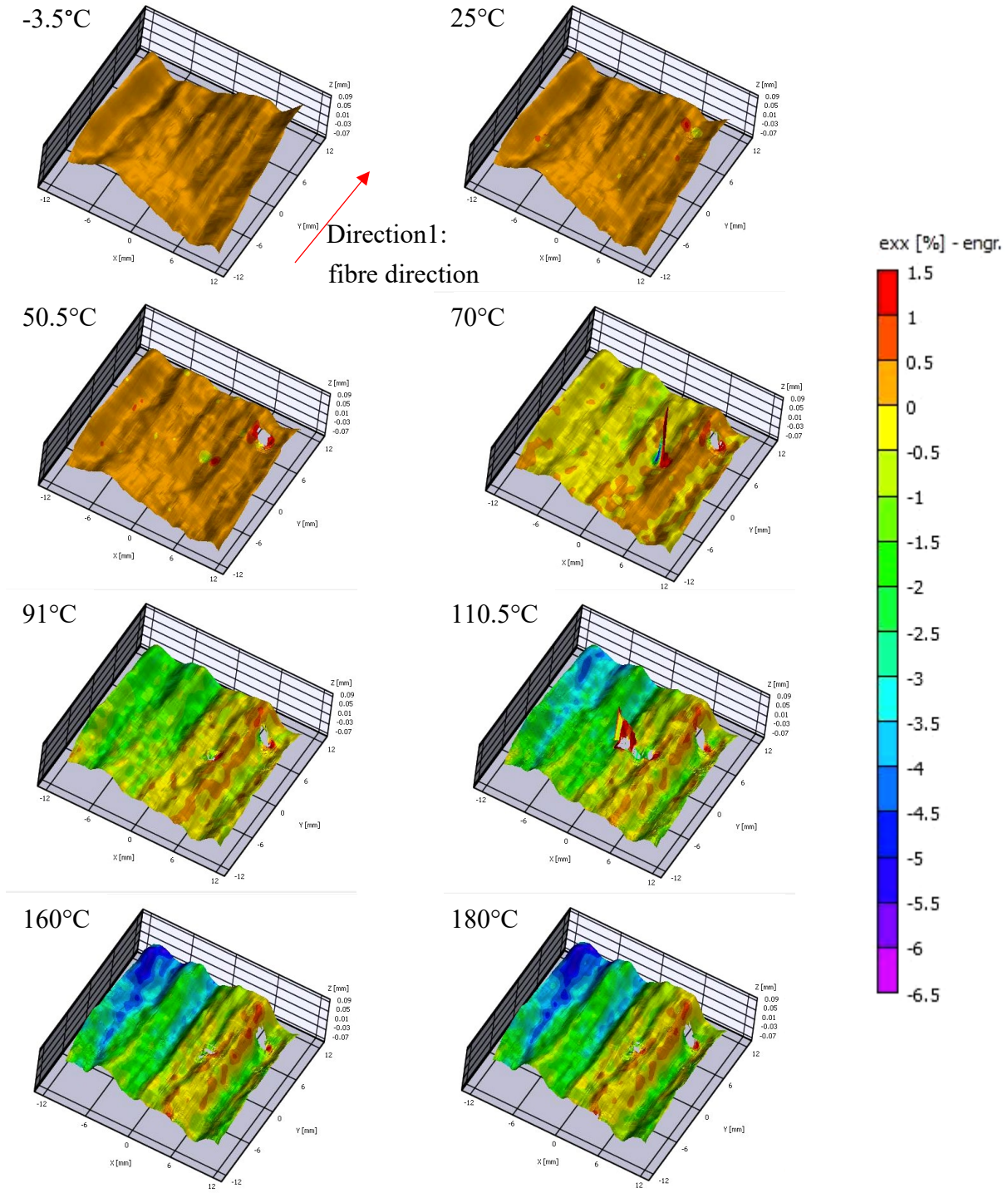


Figure 5. Strain maps for a 4-ply test of AS4/8552 processed using temperate cycle 1. Key points during the temperature ramp from -5 °C to 180 °C at a rate of 3 °C /min are shown. After 50.5 °C, the strain pattern shifts from a uniform strain (orange in colour) to a more random pattern, with some areas showing localized strain values double the average strain.

4. SUMMARY AND CONCLUSIONS

DIC image analysis results show that in the pre-gelation state, prepreg free strain behaviour is markedly different from what is typically expected, and solid-mechanics-based descriptions are not applicable. In particular, it appears that resin flow in and around the fibre-bed has a dominant impact. Only after gelation, when the modulus of resin increases, does the prepreg increasingly follow the expected behaviour. Additionally, the results introduced in this paper highlight the variability in prepreg response, particularly once the resin viscosity drops significantly during heating. The local strain patterns are consistent with the global strain trends although further study is needed to understand the details of the observed local and global response.

As manufacturing shifts towards automation using AFP and forming, understanding and control of the condition of the incoming prepreg and its subsequent handling should help the quest for defect reduction. The work presented in this paper not only gives insight into the underlying mechanisms that determine pre-gelation prepreg response but suggests a pathway to the interpretation of non-destructive imaging results gathered during automated manufacturing processes.

5. ACKNOWLEDGMENTS

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