In-Situ Measurement of Resin Shrinkage in Epoxy Composite

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ABSTRACT

Resin shrinkage due to the chemical changes and thermal gradients plays an important role in the residual stress evolution during composite manufacturing. In this paper, the resin shrinkage is measured in the neat resin and composite with the help of digital image correlation (DIC) in-situ during the curing cycle. The neat resin is cured inside a specially designed autoclave with viewports while the images are captured at regular intervals of time with the help of DIC to measure the displacements and strains during the curing cycle. An average compressive strain of 0.00896 is observed during the curing process. We also observed the effect of fibers on the shrinkage of the resin sample using carbon fiber, fiberglass, and crepe bandage as fillers. We noticed that as the stiffness of filler increased the strain in the matrix decreased.

1. INTRODUCTION

This study aims for the real-time measurement of chemical and thermal shrinkage in a commercial epoxy resin during the cure cycle in an autoclave with the help of digital image correlation. The total reduction in the volume of a composite or polymer due to curing is called polymerization shrinkage, and it has two main components, namely chemical shrinkage, and a thermal component. The conversion of the monomer molecules into a polymer network is accompanied by a closer packing of the molecules, which leads to bulk contraction [4-6]. The chemical shrinkage is a direct consequence of this crosslinking of the thermosetting polymer, and its characterization is thus of crucial interest for modeling the residual stresses [1, 2] and shape evolution of composite part during curing [3]. The shrinkage caused due to the heating and cooling cycles in the curing process called thermal shrinkage. There are number of reasons responsible for the residual stresses and distortions in the composite like the differential thermal expansion of matrix and fiber, cure shrinkage, and tool-part interaction. Residual stress formed in the composite while it is being cured leads to the distortion in the composite like warpage, delamination, and cracks. These distortions reduce the dimensional tolerance of the as-manufactured composite components, which in-turn results in prestressing during assembly, in-service defects, and increased cost.

The cure cycle temperature profile consists of three stages, which involve heating, isothermal stage, and a cool down. In the heating stage and a part of the isothermal stage, the resin is in the liquid form and thermally expands [7]. There is chemical shrinkage due to crosslinking during gelation and post gelation in the isothermal stage [34]. Finally, in the third stage, the solid composite is cooled to room temperature experiencing thermal contraction. Residual stress and the

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resulting deformation is often attributed to the cooling stage. The effects of thermal stress lead to distortion in flat plates when prepreg lay-ups are not balanced and symmetric [8, 9]. Spring back and spring on effects due to the thermal expansion mismatch in the tools are also due to these thermal stresses [10]. Given the complexity of the stress and strain evolution during curing cycle, it is important to understand the behavior of resin while it is being cured which helps to know the residual stress evolution and the factors affecting distortions and dimension instability in composites.

Many methods are available in the literature for finding the dimensional changes in thermoset resins using various dilatometric techniques. For example, volume dilatometric methods such as capillary-type dilatometry [11-12] measure the cure shrinkage by determining the linear change in the height of the column of liquid (water or mercury) connected to a reservoir of the thermoset test material. Plunger type dilatometry [13-15] is similar to a syringe wherein the pressure changes during curing lead to changes in plunger position. Gravimetric methods [16-17] are based on monitoring the change in buoyancy of the resin to measure chemical shrinkage when kept at an isothermal cure.

Other non-volume dilatometric methods include approaches like dynamic mechanical analyzer [18-19]. In a thermal, mechanical analyzer [20] a small resin sample is sandwiched between two silicon slides and the dimensional change is monitored during an isothermal cure. Rheometer [21] or Pycnometer [22-23] is used to measure the volume of the sample during curing to estimate shrinkage. Ultrasonic waves [24] have been used to measure the evolution of the thickness of resin while it is curing. Fibre Bragg gratings [25-28] have also been used to measure cure shrinkage.

Apart from these experimental approaches, computational models at various scales have been used to understand this phenomenon. Molecular dynamics (MD) simulations [29], which consider the atomic details have been very successful in exploring the crosslinking behavior of polymers in a cure cycle, providing significant information difficult to obtain through experiments. Coarser thermomechanical analyses using finite element analysis have been extensively used to predict the end of cure curvature of the composite structures and for designing tool surface compensation [10].

A recent study by Kravchenko et al. [30] used a digital image correlation (DIC) method to measure polymer chemical shrinkage and coefficient of thermal expansion for the resin sample. Recent advancements in DIC methods allow for measuring strain fields during mechanical and thermal testing [31-33]. The speckle pattern was bonded on to the resin sample and cure induced strains were measured using DIC. The primary advantage of using DIC is the ability to measure the strain in-situ in the manufacturing environment. The objective of this paper is to use a similar approach to measure the strain both in a neat resin as well as in a composite. Further, we extend this approach to pressurized autoclaves, which are often used in the industry for composite manufacturing. The experimental details are provided in section 2 followed by results and discussion in section 3.

2. EXPERIMENTATION

2.1 Materials

The resin studied for this experiment is 105 epoxy resin/206 hardener produced by West system. This resin forms a high-strength and moisture resistant solid with excellent bonding and barrier coating properties. 3K, 2*2 twill weave carbon fiber and 6 oz fiberglass fabric produced by Fiberglast and a commonly available cotton crepe bandage are used as fibers with varying stiffness to observe the filler behavior on the resin shrinkage.

2.2 Experimental Setup and Procedure

An autoclave (See Figure 1(a)) with a high temperature and pressure capability is used to cure the thermosetting resin sample. The autoclave is equipped with specially designed borosilicate viewports (highlighted in Figure 1) which enable studies on in-situ characterization while the composite specimen is cured. The cure cycle recipe is implemented using the autoclave's programmable logic controller (PLC).

Digital image correlation (DIC) is used to measure the resin shrinkage during cure and the associated strains. The DIC system comprises two 6-megapixel high-resolution cameras and is capable of measuring both in-plane and out-of-plane displacements and strains. A thin film of the speckle pattern is applied on the resin sample, and the movement of these speckle pattern is captured by the DIC due to cure induced strain in the resin sample. The DIC is setup with the autoclave as shown in figure 2. The advantage of using the DIC method is that it allows for in-situ measuring of the stress-free strain due to the crosslinking and thermal deformation of the resin [30]. The displacements and strains in the resin sample are calculated using the post-processing tool Vic-3D.

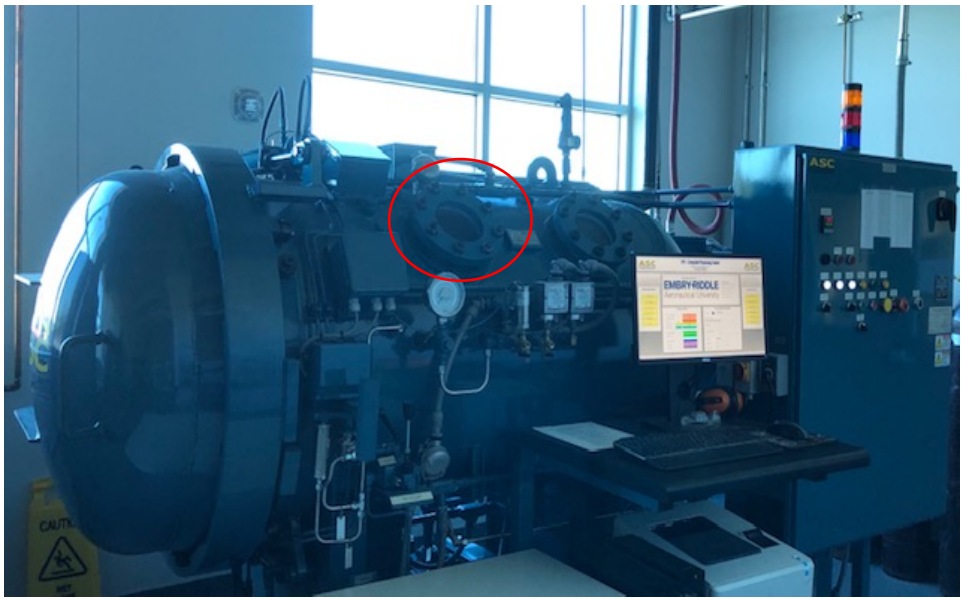


Figure 1. ASC autoclave with borosilicate glass viewports.

A flat aluminium plate is sanded to remove any remnants and impurities from the plate, and a thin layer of releasing agent from Fiberglast is applied on to the plate. A 7*7 cm sealant tape is used to create a container for epoxy on the aluminum plate. The 105 epoxy and the 206 hardener with the volume ratio of 5:1 (20 g of epoxy with 4 g of hardener) are thoroughly mixed and poured on to the aluminum plate dammed by the sealant tape. If a speckle layer is sprayed on top of the resin sample, it will partially dissolve in the resin, making it difficult to capture the strains. Therefore, a speckle layer is sprayed on a thin layer of peel ply and bounded on to the resin sample. Any movement in the speckle pattern during the cure cycle is captured by the DIC.

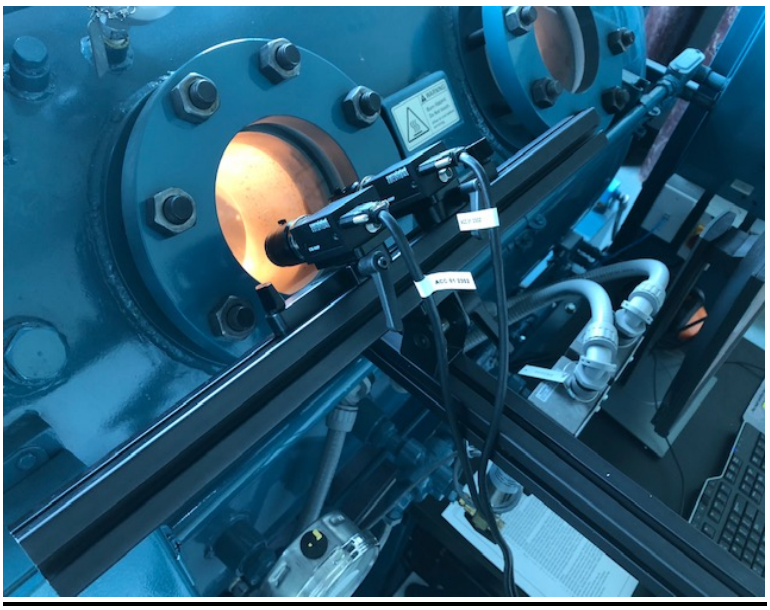
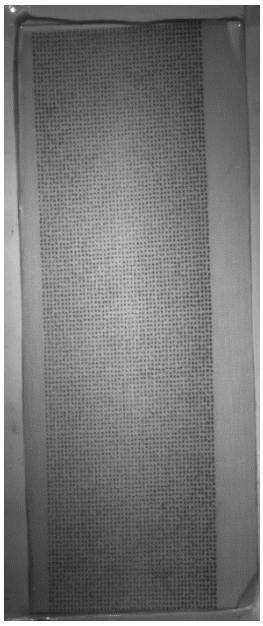
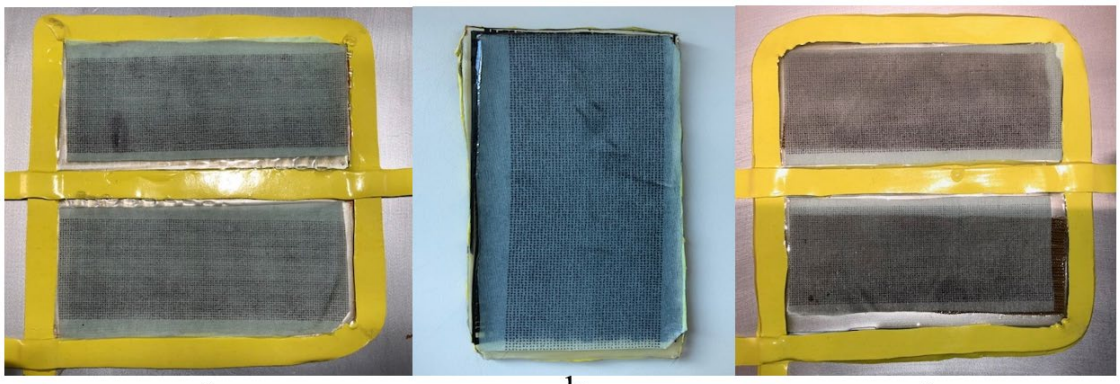
(a)  (b) 

Figure 2. (a)The digital image correlation (DIC) setup near the view-port of the autoclave. (b) The speckle pattern bonded on top of the resin sample before curing.

Figure 2(b) shows the speckle pattern bonded on top of the resin sample. The stiffness of the peel ply is assumed to be small compared to that of the resin sample at gel state due to a significant difference in size between the resin sample with the thickness of 0.1 inch, and that of peel ply is 0.0045 inch. The experimental setup in figure 2(a) shows the DIC assembled near the autoclave viewport. The aluminum plate containing a resin sample is placed inside the autoclave. During our initial experimentation process, we observed that the airflow inside the autoclave causes additional movement in the resin sample. To minimize this movement, we place two aluminum plates across the resin sample shielding it from the direct airflow. Finally, the thermal cure profile is applied to the resin sample including an isothermal stage of 120 ^{0}C for two hours.



a b c

Figure 3(a) shows the Glass fiber composite specimen. A similar procedure was followed for carbon fiber, and crepe bandage, Figure 3 (b) and (c) show the samples with carbon fiber and crepe bandage respectively.

The DIC cameras capture timed images of the sample every 30 seconds throughout the cure cycle. The strains and displacements are then analyzed using the post-processing tool Vic-3D. Through this procedure, we can relate the strains to cure state in all the three stages of the cure cycle. Additional experiments were carried out to observe the effect of including glass fiber, carbon fiber and crepe bandage as fillers in the epoxy resin. First, the resin is poured on to the aluminum plate inside the sealant tape, and then the 7*7 cm glass fiber fabric is placed on top of the resin, and finally, the peel ply containing speckle pattern is placed on top of it.

3. RESULTS AND DISCUSSION

3.1 Neat Epoxy

The average strain in the specimen is calculated using the post-processing tool Vic-3D as shown in the contour plot in figure 4. These in-plane strains correspond to the post cure stage. The average strain evolution in the sample during the cure cycle is plotted in figure 5. The overlapping cure cycle profile helps understand this evolution. From the graph, it is observed that during the heating cycle the strain value is -0.00698, at an isothermal stage the strain value is -0.0091, and finally at the cooling stage the strain value is -0.01082.

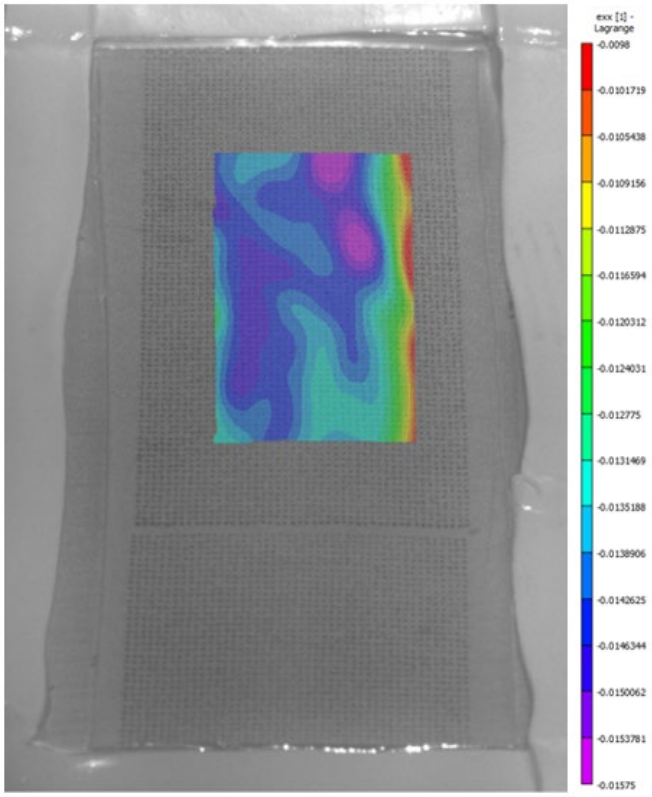


Figure 4. Average strain calculated in X-direction using post-processing tool Vic-3D.

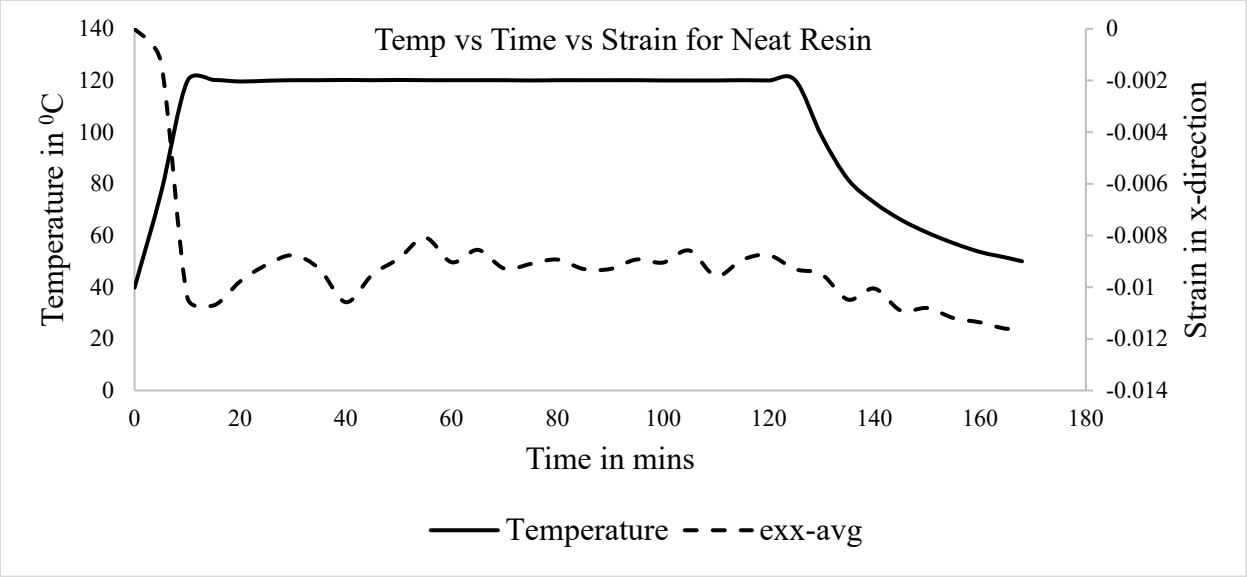


Figure 5. Strains plotted against curing time and temperature axis for neat resin.

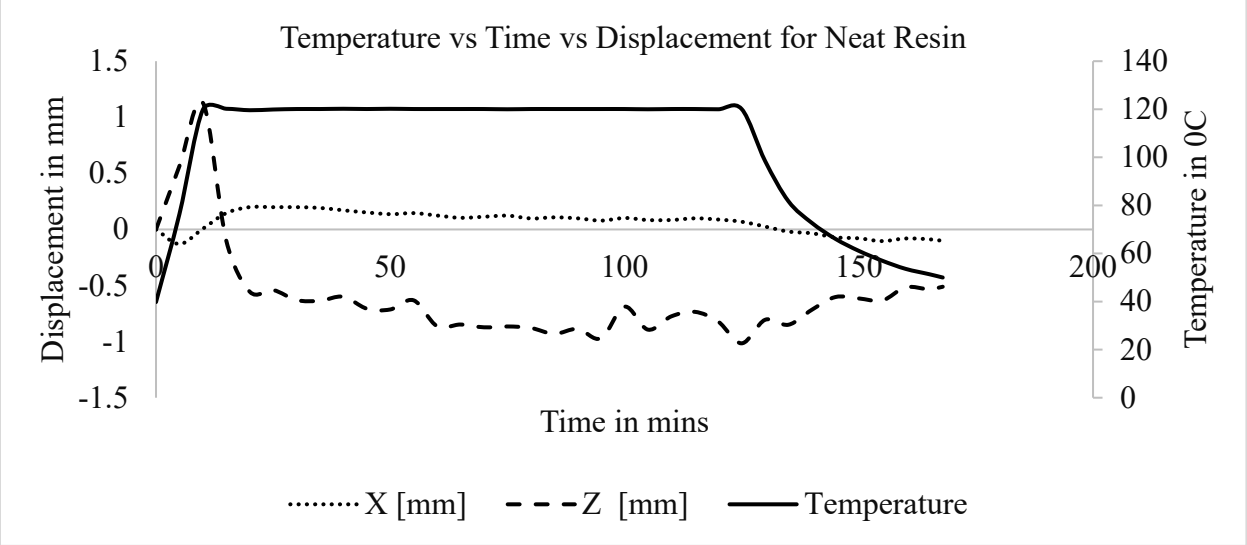


Figure 6. The average displacement in X and Z direction is plotted against temperature and cure time scale for neat resin.

We can observe from the figure 6 that the X-displacement initially oscillates due to competing for thermal and chemical interactions, however subsequently in the heating cycle, it monotonically decreases leading to compressive strain in the resin sample. Similarly, in the Z direction, the displacement increases in the resin sample during the heating cycle due to thermal expansion and decreases drastically during the isothermal stage due to polymerization reaction contracting the resin sample and finally, a small increase in the displacement in the cooling stage.

3.2 Effect of Fillers

The displacement values in the X-direction and Z-direction are calculated using the post-processing tool Vic-3D for all the fillers. These values are compared with the neat resin in figure 7 and 8 respectively. We can observe from the figure that the in-plane displacement of fiberglass is different from other fillers. It may be noted that fiber glass fabric has a fiber thickness of 0.009 inches compared to the much lower fiber diameter in carbon and cotton fibers. Figure 9 shows the variation of the average in-plane strain (ε_{xx}) during the cure cycle for neat resin and the composites with different fillers.

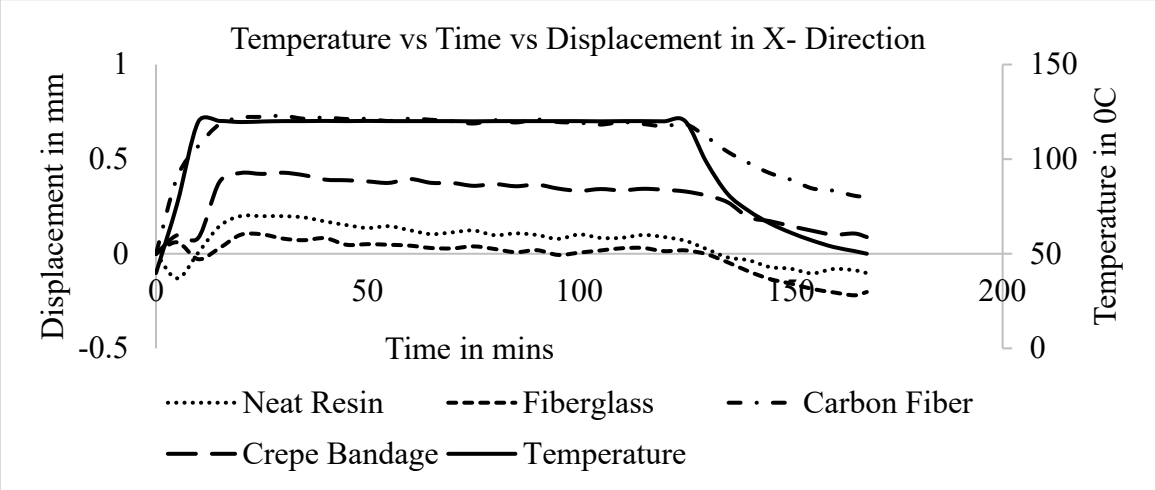


Figure 7. Displacement in X-direction in fillers and neat resin.

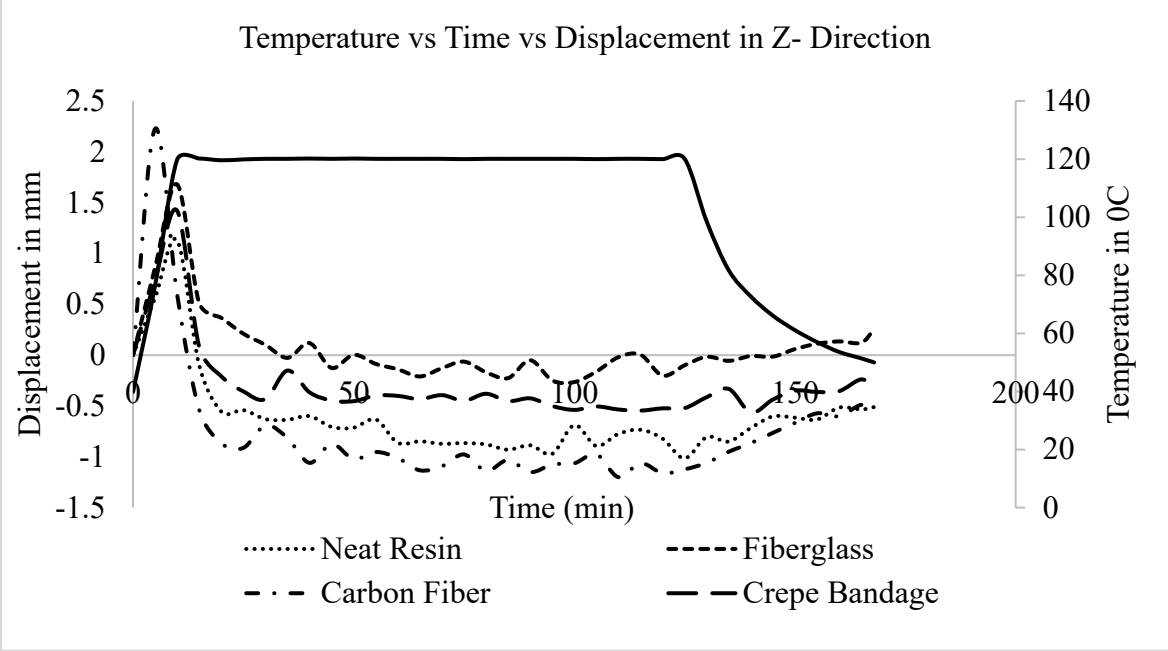


Figure 8. Displacement in Z-direction in fillers and neat resin.

From figure 9, we can interpret that the neat resin undergoes larger strain when compared to that of the composite specimen. The filler modulus has a significant impact on the observed strain as shown in the Table 1. The lowest stiffness cotton fibers do not alter the strain profile of the neat resin significantly. Glass and carbon fibers have higher stiffness and therefore reduce the observed strains. We can observe from the Table 1 that the strains in the neat resin and filler are different in all the three stages of the cure cycle. The stiffness of the fillers, affects their response to the thermal stresses. Therefore, the presence of stiffer fillers reduces the strain in the composite.

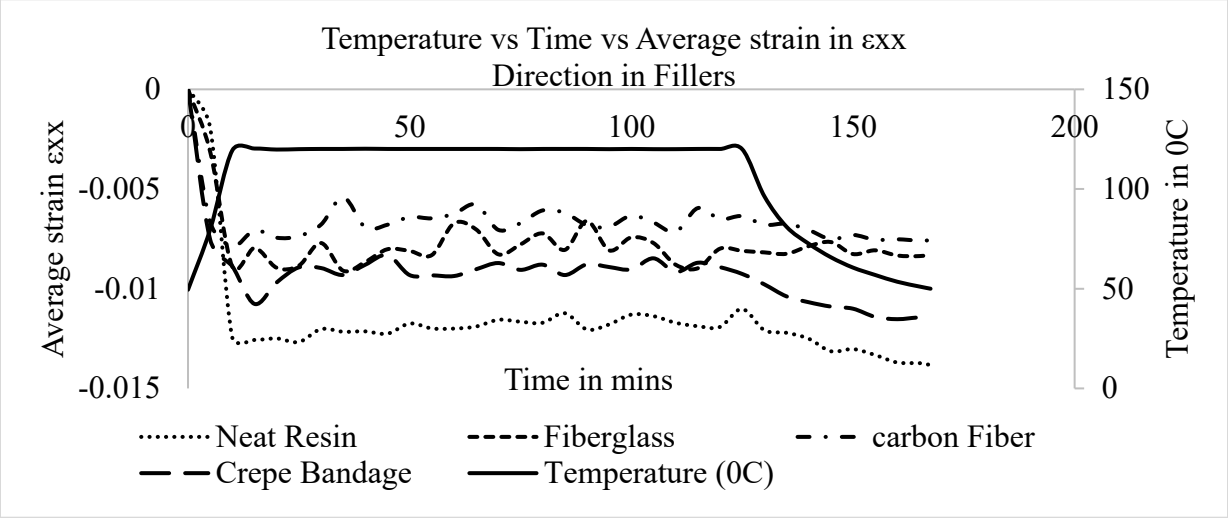


Figure 9. Average strain in the ε_{xx} direction in fillers and neat resin

Stiffness ↓

Particulars	Heating stage	Isothermal stage	Cooling stage
Plane Resin	-0.00698	-0.00910	-0.01082
Crepe Bandage	-0.00444	-0.00907	-0.01097
Glass fiber	-0.00578	-0.00798	-0.00814
Carbon Fiber	-0.00607	-0.00648	-0.00729

Table 1. Strain in the X-direction for neat resin, glass fiber, carbon fiber and crepe bandage in all the three stages.

4. CONCLUSION

In summary, we demonstrate the use of DIC to measure the shrinkage in polymeric resins during the curing process within a specially designed autoclave. Through this procedure, we can obtain strain evolution during the various stages of the cure cycle, in both neat resin and composite. We compare the strain results of the neat resin sample with those of composite using carbon fiber, glass fiber and, crepe bandage as fillers. We find that stiffer fillers lead to lower overall strain. The experimental approach developed in this paper can be used to understand the shrinkage and the resulting residual stresses during composite processing.

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