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On the methodological measurements of viscosity of unidirectional Flax/PP composites: Towards a benchmark

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Abstract Understanding the viscosity behaviour of the pre-impregnated thermoplastic composites is crucial for optimizing manufacturing processes and ensuring the end product's performance. Reviewing prior research on this topic reveals that viscosity measurements have encountered numerous difficulties, resulting in unclear interpretations and complexities in data processing. The variations in viscosity can be influenced by factors such as cavity thickness, temperature, and compression rate. The objective of this work was to enhance the reliability of results, taking a step toward initiating a benchmark for viscosity measurements of pre-impregnated thermoplastic. The study focuses on transverse squeeze flow experiments of UD flax/polypropylene (PP) for viscosity measurements.

Introduction

Different flow and deformation mechanisms are involved during the processing and thermoforming of composite materials, as discussed for various composites and processing techniques in the literature [1]. During the consolidation of pre-impregnated thermoplastic tapes with continuous fibres, a major physical phenomenon is the squeeze flow of the thermoplastic polymer composite [2]. By utilizing a lubrication assumption, an incompressible fluid, and considering a simple geometry, an analytical solution for the governing equations can be derived. This model establishes a correlation between the transient change of the thermoplastic polymer (TP) composite thickness and several factors such as process conditions (pressure, temperature, and time), component geometry, and TP viscosity. A precise understanding of the TP viscosity correlation with temperature and shear rate is crucial for extracting meaningful predictions from the squeeze flow model [3].

Previous studies have investigated the rheological characteristics of TP composites, revealing that the viscosity of TP composites is one to several orders of magnitude higher than the neat TP. For instance, for polypropylene composites, the viscosity falls within the range of 10^4- 10^6 for temperatures between 175-195 °C. Some research only considered the neat resin viscosity dominating the flow during laminate compaction, assuming that no fibres act at the tape surface [4–6]. Some authors used effective tape viscosity [7–11]. Others rely on literature data [9,11,12] instead of directly characterizing the effective viscosity of the tape under investigation.

It was shown before that tapes from different suppliers may exhibit distinct rheological behaviour, even when composed of similar fibres and polymer types [7,13]. This can be due to the presence of a resin-rich layer, different fibres distribution, and molecular weight of the polymer.

Nevertheless, the viscosity results were not consistently reproducible in consecutive replicate tests. Several studies have experimentally demonstrated a power-law model for viscosity variation with shear rate, describing the non-Newtonian behaviour of the thermoplastic at elevated temperatures. The significant role of TP viscosity in squeeze flow modelling and compression moulding has led researchers to spend several years developing squeeze flow approaches for viscosity measurement [14]. The procedure involves compressing thermoplastic composite specimens at elevated temperatures under isothermal conditions while concurrently recording displacement and applied pressure. Subsequently, the squeeze flow model is applied to compute the viscosity parameters.

Since there is no standard method of measuring the effective viscosity of UD composites, researchers and different laboratories worldwide were required to develop their in-house squeeze flow test instruments. To take a step towards standardisation, the involvement of a large part of the research community is necessary to create a viscosity database and organize a large-scale benchmark. However, even with these efforts, based on our lessons from three consecutive worldwide permeability benchmarks [15–17], certain issues need attention regarding understanding the fundamentals of variability and uncertainty in composite viscosity measurements. This approach aims to propose a unified procedure and implement more controlled experimental conditions to elucidate the factors contributing to variability, enabling a better comparison of experimental results. To achieve this goal, the ongoing collaborative work between LIST and KU Leuven has conducted several experiments to investigate the test conditions for measuring the viscosity of thermoplastic composites. The study outlines how various processing parameters, such as specimen thickness, thickness reduction rate, and temperature, can impact the reproducibility of the viscosity measurements.

While the complexities of viscosity measurement have been extensively studied in the context of continuous synthetic fibre-reinforced thermoplastic composites, the analogous investigation for natural fibres appears to be comparatively limited. The existing studies are limited to short flax fibres with low fibre volume fractions where standard rheometers are applicable [18–20].
To this end, using a squeeze flow test apparatus and a previously derived Newtonian squeeze flow model [21], this paper investigates the viscosity of the UD flax/PP composites.

Material and methods

The material under examination is a unidirectional prepreg tape composed of flax fibres and semi-crystalline PP, EcoRein/UD50 (BPREG Composites and Textiles A.Ş., Bursa, Turkey). The tape specifications are listed in Table 1.

Table 1 - The technical specifications of the flax/PP unidirectional tape.

Supplier	Fibre (wt %)	Polymer type	Thickness (mm)	Width (cm)	Areal density (g/m^2)
BPREG	50	Semi-crystalline PP	0.3	60	300

The samples were prepared first by cutting layers and drying them at 80°C for at least 12 hours to diminish the moisture content. The blank was prepared to the desired thickness and consolidated utilizing a hot press at 190 °C under a pressure of 5 bar for 15 minutes of dwell time inside a mould with a spring mechanism to control the final thickness. The laminate was then cooled under pressure before demoulding. The laminate production steps with its different thermal cycle steps are shown in Fig. 1.

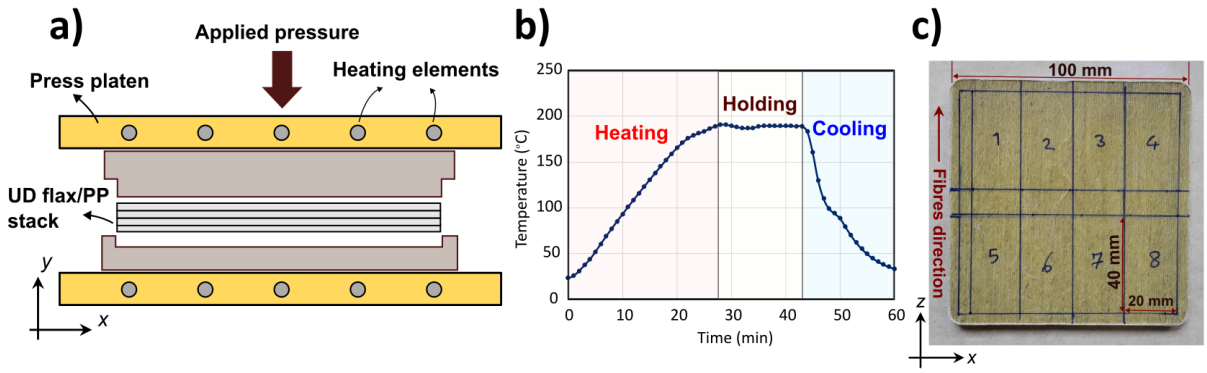

Figure 1 - Specimen preparation: (a) hot press moulding of the UD flax/PP prepregs, (b) recorded thermal cycle of the production process, c) a produced plate prepared for cutting the test specimens.

Samples of L (length) 40 × W (width) 20 mm (with fibres oriented in the length direction) were cut from the plates using a band saw cutting machine and then dried in a vacuum oven for 15 hours at 80 °C before testing. The compression test setup used is very similar to the one used for the compressibility of fabrics [22]. It consists of two circular steel plates with a diameter of 80 mm, aligned parallel to each other and fixated in a universal testing machine (Instron 5985, Instron, USA) equipped with a 30 kN load cell. The setup inside a ventilated hot-air oven is shown in Fig. 2 and b. The oven was first heated to the desired temperature (T) and then opened to put the specimen inside. Samples were allowed to equilibrate with the oven temperature, checking with thermocouples embedded on the bottom plate and the specimen (Fig. 2a and b). Fig. 2c shows the force and temperature cycle after putting the specimen inside when the specimen temperature starts to rise toward the desired temperature. Two temperatures of 180 and 190 °C were used to study the temperature dependency of the measurements. Kapton films were placed between the sample and the steel plates to avoid sticking. When the temperature reached, a force ramp was applied to achieve a constant force of 100 N. Of course, it takes some time for the machine to reach the constant force value, and this transient region can affect the results (see Fig. 2c). The test starts at t_1 when the temperature on the specimen reaches the desired value (T) and continues until t_3, as plotted in Fig. 2c. t_2 minus t_1 is the ramp time that is required for the machine to build up the force to the required constant value. This time can be defined based on the compaction rate in the first stage of the machine profile. The model derived by Lin and Advani [21] is used to measure the composite viscosity (μ_{mf}):

$$\mu_{mf} = \frac{5}{8}\frac{Ft}{L_i W_i^3}\frac{(h_i^2 h_f^5)}{(h_i^5 - h_f^5)}$$

where L_i and W_i are the initial length and initial half-width of the laminate, respectively. h_i represents the initial thickness and h_f is the final thickness of the laminate due to a constant force of F during the evaluation time of t. The model is derived from the continuity and motion equations for an incompressible fluid (negligible void content). Moreover, a lubrication assumption is made that is valid for a specimen width much higher than its thickness. A no-slip boundary condition is assumed at the top and bottom tool-material interface. This assumption is validated after the test by the intact initial surfaces of the compacted specimen. After the test and removing the Kapton film, there were no signs of material flow (slipping) on the initial surface area of the material [13].

Ideally, the compaction displacement is measured using Digital Image Correlation, but this was not possible for the thin samples inside the oven. Thus, the crosshead displacement was used but corrected for system compliances based on Instron instructions. A total of eight batches were prepared, as listed in Table 2. These batches involved two distinct thicknesses of 5.3 and 8.2 mm, temperatures set at 180 and 190 ºC, and three different initial compaction rates of 0.5, 5, and 20 mm/min until a constant force of 100 N was achieved. The thicknesses are referred to as t5 (h_i = 5.3 mm) and t8 (h_i = 8.2 mm) in the batch names for simplicity.

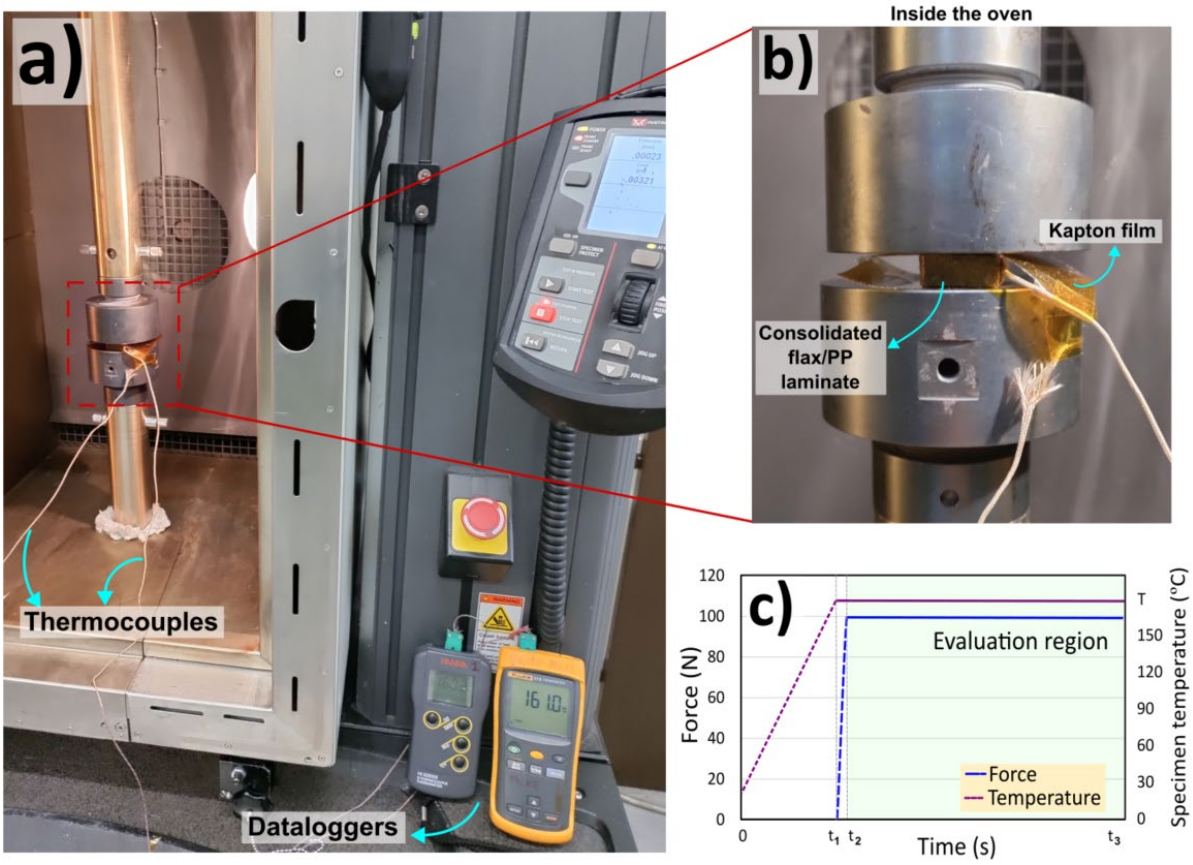

Figure 2 - Experimental setup for the squeeze flow test. (a) compression setup inside the oven, (b) a magnified view of the specimen mounted for the constant volume test (larger compression plates), (c) temperature and force cycle after mounting the specimen inside (t = 0) until the end of the test (t_3).

Table 2 – Different testing conditions and coding for each test batch.

Batch	Thickness (mm)	Test temperature (ºC)	Initial rate (mm/min)	Constant force (N)
t5_T180_r05	5	180	0.5	100
t5_T180_r5			5	
t5_T180_r20			20	
t5_T190_r05		190	0.5	
t8_T180_r05	8	180	0.5	
t8_T180_r5			5	
t8_T180_r20			20	
t8_T190_r05		190	0.5	

Results and discussion

Reduction in thickness over time is depicted in Fig. 3, along with the corresponding force changes. In both cases, it is evident that the adjusted thickness following compliance calibration is just slightly higher than the thickness reduction determined by the original displacement of the machine. This confirms the negligible compliance of the setup, allowing the direct utilisation of the original displacement of the machine for data reduction. Nevertheless, it is crucial to validate this step to guarantee accurate thickness reduction measurement, independent of machine compliance, as emphasized in [8].

It is worth mentioning that for all tested samples, significant dimensional change was observed in the transverse direction, confirming squeeze flow as the primary deformation mechanism, as there was no increase in the specimen length due to fibres inextensibility and much higher extensional viscosity in the longitudinal direction [23–25]. Additionally, as expected and shown in Fig. 3, a reduced initial rate requires a significantly longer time to reach the steady force region (100 N) essential for viscosity measurements. Fig. 4 presents the relationship between force increase and thickness reduction for both investigated thicknesses. As shown in Figs. 4a and b, a lower initial rate leads to a higher thickness reduction before achieving the constant force necessary to enter the evaluation region. This is more severe for the thicker specimens (t8).

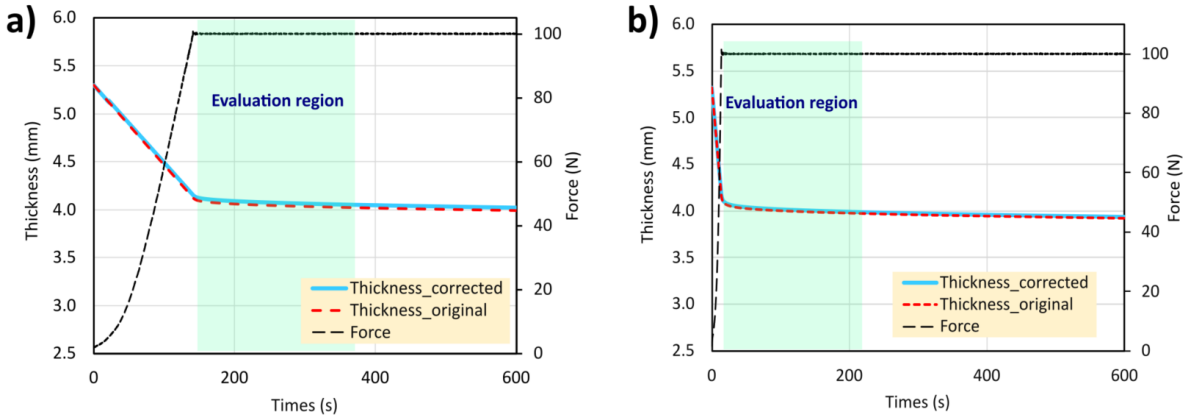

Figure 3 – Thickness reduction curves before and after compliance calibration for specimens with the thickness of 5 mm and from batch (a) t5_T180_r05, (b) t5_T180_r5.

The obtained viscosity values are reported in Table 3. As shown, all the values are in the range of 10 MPa·s and higher. This is at least an order of magnitude higher than the values reported for short flax/PP composite up to 30 wt% of flax fibres which were in the range of 10^4-10^6 Pa·s for [18,19]. Looking at the effect of the testing parameters, as expected, a higher temperature of 190 ºC resulted in a lower viscosity value for both thicknesses at a constant initial compaction rate of 0.5 mm/min. However, increasing the initial rate at a constant thickness and temperature resulted in a lower viscosity. This can be explained by Fig. 4, which shows a higher deformation of the r05 specimens before reaching the constant load region. Increased compaction and reduced thickness would result in more interaction and fibre entanglement (locking) that can act as a barrier impeding material flow, ultimately contributing to elevated viscosity.

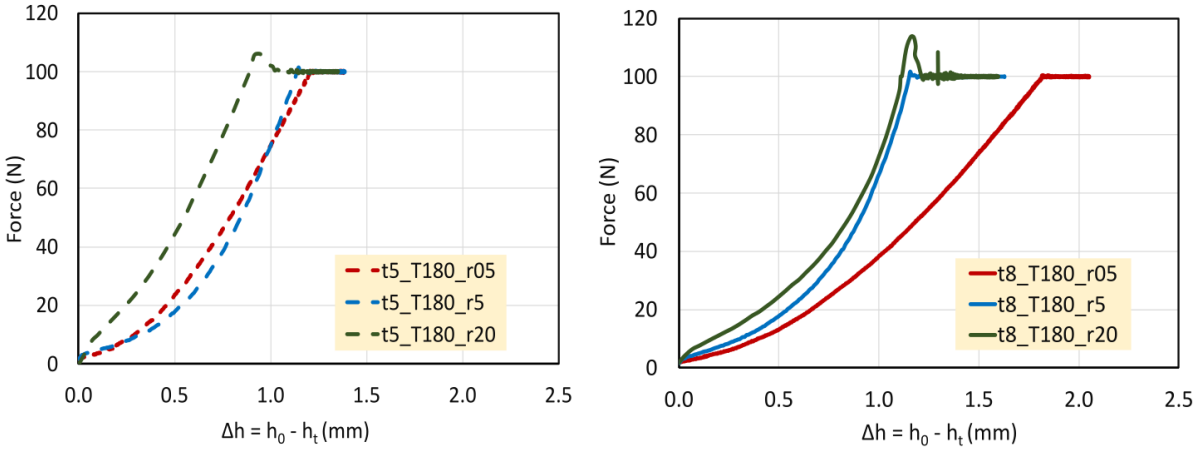

Figure 4 – Thickness reduction for specimens from different batches tested at 180 ºC. (a) thickness of 5 mm, (b) thickness of 8 mm.

This could be more severe in the case of natural fibres as the fibres are not perfectly straight, and they pose variations in the fibres diameter. It is worth noting that the highest standard deviation is obtained for both batches with the lowest compaction rate of 0.5 mm/min (r05) at 180 ºC, while the coefficient of variation (CoV) is higher for t5_T180_r05. A higher temperature of 190 ºC (T190) shows a lower standard deviation and CoV than those tested at 180 ºC (T180). However, more experiments are still needed for some batches, but the standard deviation is lowered by increasing the initial compaction rate for the cases tested at 180 ºC.

It should be noted that the material is treated as a Newtonian fluid in the current work, which serves as one of the first works on the viscosity measurement of UD flax/PP laminates. However, a shear-thinning behaviour is expected by increasing the shear rate for most thermoplastic tapes, which is not considered here. More complex models can be studied using the models based on the Carreau fluid [26] or power law behaviour [27].

Table 3 – Measured effective viscosity of UD flax/PP batches with the standard deviation and coefficient of variation.

Batch	Number of tested specimens	Min. viscosity ($\times 10^6$ Pa·s)	Max. viscosity ($\times 10^6$ Pa·s)	Avg. viscosity ($\times 10^6$ Pa·s)	Standard deviation ($\times 10^6$ Pa·s)	CoV (%)
t5_T180_r05	5	18.5	34.9	28.2	7.5	26.8
t5_T180_r5	2	10.4	11.3	10.8	0.6	5.8
t5_T180_r20	2	6.0	10.5	8.2	3.1	38.1
t5_T190_r05	4	18.8	27.9	22.3	4.4	4.4
t8_T180_r05	4	60.5	75.5	66.6	6.5	9.8
t8_T180_r5	3	37.3	43.7	41.0	3.3	8.1
t8_T180_r20	3	34.0	37.3	35.7	1.7	4.6
t8_T190_r05	4	53.9	60.2	56	2.9	5.1

Summary

This paper reported the viscosity data for UD flax/PP composites tested at different conditions, which may be a base for the formulation of a benchmarking exercise. The primary objective was to identify critical aspects of the test method necessary for obtaining reliable results and to propose a recommended test procedure for viscosity measurements in reinforced thermoplastic composites. Transverse squeeze flow experiments were conducted to achieve this goal. Although several models are available for this measurement based on the assumption of fluid behaviour, a simple

Newtonian model is considered here to initiate the exercise. The experiments involved compressing a consolidated laminate of UD flax/PP between two parallel plates. The results revealed that the viscosity data depends on the thickness, exhibiting a power law pattern as a function of initial thickness and thickness reduction rate. Altering the sample thickness from t5 mm to t8 mm and conducting the experiment at a comparable temperature and initial rate did not yield consistent results. In these cases, the viscosity can be up to three times higher than that measured in the t5 samples.

Furthermore, at a constant temperature, a higher scatter was observed at the given target thickness for lower initial thickness reduction rates. The measured viscosity shows lower values by increasing the compaction rate, as less time is available for more compaction and fibre locking before entering the evaluation region. The results show that employing a higher initial compaction rate before reaching the constant force regime could be beneficial to avoid possible fibre locking that would cause a considerably higher and erroneous viscosity measurement.

As a recommendation, it can be suggested that the viscosity measurement should be conducted across various laminate thicknesses from thin (1-2 mm) to thick laminate (10-12 mm) while employing a high compaction rate. This approach aims to minimise uncertainty associated with the measured viscosity and improve the reliability of the results. Moreover, thermal expansion at elevated temperatures may be considered as it might alter the initial dimensions of the specimens.

In addition to the mentioned influencing parameters, a comprehensive benchmark exercise could also investigate other parameters such as void content, time effect in the evaluation region, holding time inside the oven, and other models available in the literature.

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