

COMPARISON OF DENSITY MEASUREMENT METHODS FOR UNIDIRECTIONAL FLAX-EPOXY POLYMER COMPOSITES

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Abstract. *Density is a decisive factor in determining one of the most important advantages of flax fiber-reinforced polymer (FFRP) composites, i.e. their relatively low weight leading to high specific properties. As a fundamental physical property of composites, density enters in many engineering design and quality control calculations and its value is a determining factor for several applications. Especially, we need precise material properties including density so as to develop efficient numerical models to these materials. In this work, three density measurement methods were evaluated and compared: Helium-gas pycnometry and Archimedes with two different immersing liquids, water and ethanol. The results show that Helium-gas pycnometry and Archimedes with ethanol gave similar and repeatable results, whereas using water resulted in much lower values. The density values were all in the range of reported values. However, Helium-gas pycnometry or Archimedes using ethanol are recommended for more precision in measuring the density of FFRP composites.*

1 INTRODUCTION

Towards increasing demand for replacing conventional synthetic fibers with natural fibers as reinforcement in fiber-reinforced polymer (FRP) composites, flax fibers have found their place in the industry to replace man-made glass fibers. Flax is one of the oldest plant-based fibers and together with hemp, jute, ramie and kenaf fibers are classified as bast fibers which are nowadays extensively used in the composite field [1, 2]. Flax fibers are extracted from the stems of the flax plant and are composed of about 64% of cellulose. They normally come in

bundles and at the mesoscopic level, a bundle contains 10 to 40 elementary fibers linked together by pectin. The elementary fibers, which have a 5-7-side polyhedron shape, have an extremely complex microstructure composed of concentric primary and secondary walls forming an open channel in the middle called lumen that contributes to water absorption [2, 3].

Flax fibers are used in different forms as reinforcement in polymer composites, for instance, monofilament fibers, mats, rovings, yarns and fabrics. Nowadays, various fabrication techniques are used to manufacture polymer composites, e.g. resin transfer molding (RTM), vacuum infusion, hand lay-up, film stacking, compression molding, injection molding and pultrusion [2, 3]. In some studies, a variety of surface treatments have been applied to flax fibers to improve fiber-matrix adhesion, however, it is preferred to eliminate any extra procedure from the manufacturing process and use the fibers without any modifications [3-7]. J. Zhu [3] reported the resin transfer molding as a preferred processing method to produce high performance flax fiber-reinforced epoxy composites.

Flax has been used to reinforce many polymer matrices, including thermoplastics, thermosets and biopolymers, and each type has its advantages and disadvantages. Thermoset polymers are superior in terms of mechanical properties, chemical resistance, thermal stability and overall durability. As a thermoset polymer matrix, epoxy offers high mechanical performance and durability. It has attracted many researches and is the most commonly used matrix to produce flax fiber-reinforced polymer composites [2, 4, 8-10]. For that reason, epoxy has been chosen for matrix in the flax composite of this study.

Flax/epoxy composites have many environmental advantages, economic benefits, high-performance and low density, factors of the highest importance for many industries [2, 3, 11-13]. These composites, having specific properties comparable to those of glass fiber-reinforced polymer (GFRP) composites, are potential alternatives for GFRPs in many structural and non-structural applications. They have found their ever-widening applications in many industries, such as automotive, construction, packaging, sporting and leisure [2, 3, 14, 15]. Consequently, these composites need to be known very well in terms of physical and mechanical properties.

As a fundamental physical property of composites, density enters in many engineering design and quality control calculations and its value is a determining factor as to their application. Density determines one of the most important advantages of flax fiber-reinforced polymer (FFRP) composites, i.e. their relatively low weight leading to high specific properties. So we need precise material properties including density so as to develop efficient numerical models to these materials.

Although there are some works regarding the measurement of fiber density [16] and some others dedicated to the density measurement of flax fibers [17], the sensitivity of FFRP composites to different measurement methods has not been studied yet. The literature data are scattered, ranging from 1.117 to 1.32 g/cm³ [2, 18] and considering the fact that the density value depends on factors like the fiber volume fraction, fabrication process, measurement method and the condition of fibers used, it is often not possible to use values published in other studies. For the Finite Element Modeling (FEM) of the machining process of the FFRP composites used in this study, and in order to have a precise density value for them, this study has been conducted. Three methods to determine the density of unidirectional flax composites are evaluated and compared: Helium-gas pycnometry and Archimedes using two different immersing liquids, water and ethanol. The values will be compared and the possible sources of variations will be discussed.

2 MATERIALS AND TEST SPECIMENS

Unidirectional flax fibers in the form of tapes with a surface density of 200 g/m², commercially named FlaxTape™ (LINEO - France), were used as reinforcement and Marine 820 epoxy resin and Marine 824 hardener (ADTECH® Plastic Systems) as the polymer matrix to produce flax fiber-reinforced composite laminates. The flax/epoxy laminates were molded using the RTM process, as reported to be the preferred processing method to produce high-performance flax fiber-reinforced epoxy composites [3]. All laminates were molded in 300 mm × 300 mm dimensions, with [0]_n and [0/90]_n stacking sequences for the preparation of the test specimens. The thicknesses of the laminates were precisely calculated and controlled to result in a fiber volume fraction of 41% for all composites. The laminates were post-cured at 70°C and prepared for the cutting of specimens.

The test specimens were cut to fit in the considered location of test equipment and 5 specimens were prepared from identical laminates to check the repeatability of the tests. The samples were all stored in room conditions for 24 hours before testing to stabilize the temperature and humidity levels.

3 EXPERIMENTAL PROCEDURES

Three conventional methods, which are frequently used by researchers to determine the density of composites, were applied to measure the density of the FFRP composites: Helium-gas pycnometry, Archimedes (water used as immersing liquid) and Archimedes (ethanol used as immersing liquid).

3.1 Helium-gas pycnometry

Helium-gas pycnometry is a very quick, clean and convenient density measurement method, based on which some commercially available equipment and the ASTM-D4892 [19] standard test method are developed. In this method, based on the volume of displaced gas and Boyle's law, the solid phase volume of a material sample of known mass is measured and accordingly the density is calculated. For this purpose, two chambers are used and the solid volume of the specimen is calculated based on equation 1;

$$V_s = V_c - \frac{V_x}{\frac{P_1}{P_2} - 1} \quad (1)$$

where the subscript V_s refers to the sample volume, V_c to the sample chamber volume, V_x to the expansion chamber volume, P_1 and P_2 to the pressures after fill and after expansion respectively.

The disadvantage of this method is that from a certain level of pressure, Helium can enter into the open cavities so as to measure the density of the constituent materials of the composite rather than the composite density in its specific form and configuration. This issue will be evaluated by comparing the results with those of other methods.

An ACCUPYC® II gas displacement pycnometer device (a product of Micromeritics Instrument Corporation) was used in this work. The specimens and equipment are shown in figure 1. The test was automatically repeated by the equipment 10 times for each sample, the average value was taken as the density of composite and the standard deviation was calculated.

3.2 Archimedes method

This is a buoyancy method working based on the Archimedes' principle. It is simple to implement and has been traditionally used for measuring the density of materials. The ASTM D792 standard test method [20] has been developed based on this principle and is recommended for measuring the density of plastic materials. It has been used for measuring the density of composite in some works [21, 22]. A Sartorius YDK03 density measurement kit was used in this study, as shown in Figure 2. The density was calculated according to equation 2;

$$\rho = \frac{W(a) \cdot [\rho(fl) - \rho(a)]}{W(a) - W(fl)} + \rho(a) \quad (2)$$

where ρ refers to the density, W refers to Weight, (fl) refers to immersion fluid and (a) to air. In order to monitor the absorption of liquid by the composite samples, the surface of the coupon was dried immediately after testing and the coupon reweighed.

3.2.1 Water used as immersing liquid

Water is the most commonly used liquid with this method. Due to the hydrophilic nature of natural fibers, immersing their composites in water may cause absorption of water by the fibers, thus affecting the value of measured density. In some studies water has been used to measure the density of natural fiber composites [21, 22]. In this method, the weight of the samples in the water started to increase without stabilizing. This is an evidence of absorbing water (rather than the removal of bubbles that stops in a short period of time) which was validated after testing by reweighing the samples. To avoid this problem, before immersing the samples, they were rapidly wetted to remove the air bubbles surrounding the sample and they were weighted in the water in a short time after immersion to avoid absorption. In this study, the validity of using water as immersing liquid for measuring FFRP composites will be investigated by comparing the three measuring methods.



Figure 1: ACCUPYC® II Helium Gas Pycnometer

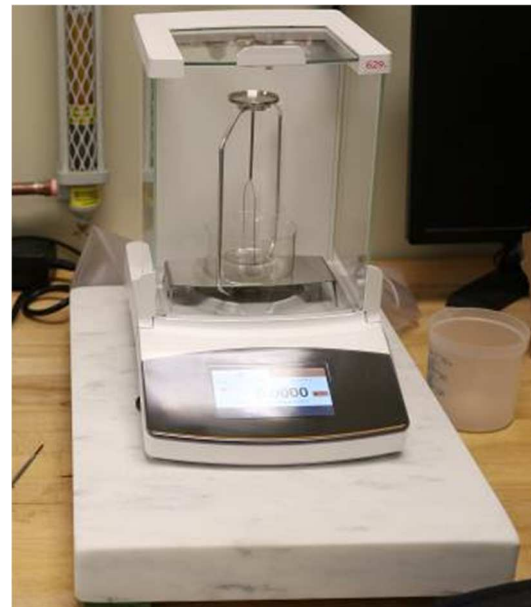


Figure 2: Sartorius YDK03 density measurement kit

3.2.1 Ethanol used as immersing liquid

As an alternative to water, ethanol was used. It can be easily cleaned off, it doesn't affect the samples and it can be safely used in room conditions. It has been used by some researchers to measure the density of natural fibers [23, 24]. The weight of samples in the ethanol was very well stabilized after a reasonable immersion time.

9 RESULTS AND DISCUSSION

For each method at least 5 tests were performed and the mean value of the measured densities are summarized and reported in table 1. As can be seen, the gas pycnometry method resulted in nearly the same value for different samples.

Using Archimedes' method with water, in order to avoid the absorption of water by the samples, they were weighted in a short time after immersion. This resulted in lower specimen weight in the liquid (which may be due to remaining air bubbles) and consequently, in a lower density value (following Equation 2). Considering this issue and the fact that the samples need to be immersed in the fluid for a certain period of time to remove the air bubbles, it is concluded that measuring the density of FFRP composite by this method doesn't result in precise values.

Archimedes' method with ethanol allowed an immersion times long enough to remove the air bubbles and reach a stable weight. After testing, no weight change was observed, meaning that almost no ethanol was absorbed. Secondly, the very small coefficient of variation in Table 1 (for repeated samples) verify that a single physical property is measured. Finally, because the resulting value is very well correlated with the result of gas pycnometry method, this means both methods can be recommended to evaluate the density of natural fiber composites.

Table 1: Measured density values and its variations

Method	Gas Pycnometry (with Helium)	Archimedes (with water)	Archimedes (with ethanol)
Density (g/cm ³)	1.2813	1.2594	1.2804
Standard deviation	0.0005	0.0065	0.0009
Coefficient of variation [†] (%)	0.04	0.52	0.07

[†] Standard deviation/Mean value *100

11 CONCLUSION

The density of FFRP composites was measured by three different methods, the results were compared to each other and discussed. According to the findings of this study, Archimedes' method using water as immersion fluid is not recommended for measuring the density of FFRP composites. Instead, Archimedes' using ethanol and the gas pycnometry methods are recommended as the results obtained from these methods were nearly the same.

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