Volume XXI 2018
ISSUE no.1
MBNA Publishing House Constanta 2018

Scientific Bulletin of Naval Academy

SBNA PAPER • OPEN ACCESS

Water absorption behaviour of reinforced epoxy laminates

To cite this article: Cristian Munteniţă, Vasile Bria, Adrian Cîrciumaru and Iulia Graur, Scientific Bulletin of Naval Academy, Vol. XXI 2018, pg. 418-423.

ISSN: 2392-8956; ISSN-L: 1454-864X

doi: 10.21279/1454-864X-18-I1-063

Water absorption behaviour of reinforced epoxy laminates

Cristian Munteniţă, Vasile Bria, Adrian Cîrciumaru, Iulia Graur

Abstract. In this work, the studied materials are formed with a graduated matrix and reinforced with different types of fabrics. These composites are similar regarding the reinforcement, but the difference between them consists of the epoxy matrix types. Any application of a polymer composite in an outdoor environment invariably involves water absorption and variable thermal conditions. The degradation of composite materials was observed after each cycle, water absorption and frozen-unfrozen, from a qualitative (visual) point of view, until the materials reached a saturation degree (the mass of specimens remaining constant after a certain number of cycles). This aspect had led to the conclusion that materials can no longer undergo quantitative changes. The aim of this work is to examine the behavior of materials subjected to environments that simulate atmospheric conditions (water absorption and frozen-unfrozen). The purpose of these materials was to be used in order to form some spare parts of vehicles such as, in case of an impact, to ensure pedestrians protection since they could have controllable strength. This aspect pays a very important role both in economically and environmental terms.

Keywords: water *absorption, frozen-unfrozen, epoxy laminates.*

1. Introduction

Some of the most common fibres being used as reinforcement in composite materials include flax, hemp, jute, carbon, glass and aramid [1]. The major drawback in using natural fibers in composites is that the fibers are hydrophilic meaning that they attract water, whilst the matrix materials are usually hydrophobic, meaning that they repel it [2]. Accordingly, there must be good interphase properties between the fibres and the matrix. The incorporation of fibres often resulted in a large variability of composite properties which are influenced by natural conditions such as temperature, humidity, moisture, climate, and manufacturing process [3]. One of the main applications of epoxy thermosets is their use as matrices in polymer composites. However, their relatively low toughness and their high capacity of absorbing water are two undesirable properties of these polymers which have limited their proliferation into some critical areas [4].

Epoxy polymers are characteristically hydrophilic, which means that they have strong affinity towards water. This nature of epoxy resins makes them susceptible to high moisture absorption; in general, depending on the nature of the epoxy resin, the equilibrium moisture uptake can be in the range of 1–7%. Water absorption into a polymer matrix leads to change in both chemical and physical characteristics and affects the mechanical properties through different mechanisms such as hydrolysis, and swelling [5].

Therefore, it is very important to know what effects reinforcing fibres will have on the water absorption behaviour and variable temperature conditions resistance [6]. The degree of environmental degradation that occurs in a fiber reinforced polymer composite structure is linked directly with the amount of moisture that is absorbed. But the moisture absorption kinetics of epoxy resins differs widely and also changes with physical ageing [7]. In general, when ingress of water in epoxy systems takes place, two possibilities are assumed for then: (i) the water is either strongly interacting with specific (polar) groups of the epoxy matrix or (ii) clustered together in free volume micro voids as ‘‘free water’’ [8, 9]. It is believed that in the water environment, water molecules will enter rapidly the interphase of the composites between the fibre and the resin because of the capillarity [10, 11, 12, 13]. The polymer matrix and the fibre/matrix interphase can be degraded by a hydrolysis reaction of unsaturated groups within the resin [14]. Debonding may occur at fibre/matrix interphase [15]. The hydro elasticity of a polymer or of a composite is defined as its reversible dimensional response to liquid or vapour penetration [16, 17]. The weight gain was calculated according to the following equation [10, 18]:

$$M_t(\%) = \left(\frac{W_t - W_0}{W_0}\right) \times 100 \tag{1}$$

Where W_t is the weight of the sample at time *t* and W_0 is the initial weight of the sample.

The present study focuses the temperature water absorption effect: frozen-unfrozen, because composites are susceptible to heat and moisture when operating in changing environmental conditions [7]. Composites with common structural polymer matrices often absorb moisture and this has profound effects in their mechanical, thermal, dielectric and barrier property performance. It is, therefore, not surprising that experimental and theoretical work on moisture absorption in composites was and still is an active research topic across many disciplines [19, 20, 21]. A concern with using fibre composites is the long-time exposure to high humidity environment and frozen-unfrozen atmospheric conditions.

2. Materials

The polymers matrix consisted of bisphenol A diglycidyl ether-based epoxy resin (HT2 Resin, Epiphen DE 4020, HT and C) were used. Epoxy resin is an important matrix used for fibre reinforced polymer. Due to its features, epoxy resin has been used in manufacturing applications such as adhesives, coatings, electronic and aerospace structures [22].

Fibre-reinforced composites materials consist of fibres of high strength and modulus embedded in or bonded to a matrix with distinct interfaces (boundaries) between them. In this form, both fibre and matrix retain their physical and chemical identities, yet they produce a combination of properties that cannot be achieved with either of the constituents acting alone [23]. The composites were formed by wet lay-up method with each layer of reinforcement being imbued with pre-polymer mixture of the epoxy system and being placed into a glass mould with dimensions of 280x210x4mm.

These composites consists of 15 layers of reinforcement made of five different types of fabrics as follows: the first three layers - C240 fabric at 0^0 orientation only with epoxy resin; the following three layers - C/A68 fabric at 45^0 orientation, epoxy resin with 5% (wr) 1-Methyl,2-Pyrrolidinone; the next three layers - C160 fabric at 0^0 orientation, epoxy resin with 10% (wr) 1-Methyl,2-Pyrrolidinone; before the last ones layers – A61 fabric at 45^0 orientation, epoxy resin with 15% (wr) 1-Methyl,2-Pyrrolidinone and the last three layers – CT60 fabricat 0^0 orientation, epoxy resin with 20% (wr) 1-Methyl,2-Pyrrolidinone. So, as matrix were chosen four epoxy systems and the hybrid epoxy laminates had been denoted: L- epoxy laminates with L epoxy resin; C- epoxy laminates with C epoxy resin; E- epoxy laminates with Epiphen epoxy resin and HT- epoxy laminates with HT epoxy resin.

The purpose of thermal treatment was to eliminate any suspicion that leads to the change in the specimen weight, which will be referred to below, after extracting from the oven, weight that will be taken into consideration as the initial weight. The surface of the material is glossy, the only portion with a high degree of porosity being the material edges. It is worth noting that the experiment was not one compared to a standard material, because the studied materials are formed with a graduated matrix and reinforced with different types of fabrics.

3. Water absorption tests

Water absorption tests were conducted in accordance with ISO R62 [18]. Three specimens were used for the water absorption tests. These numbers of specimens are in accordance with the relevant standards [1]. All the specimens were dried in an oven at 60^{o}C temperature for as long the time as the weight of each specimen became stabilized (120 min). The next step consisted in weighing the test specimens to determine the initial weight. This value plays an important role throughout the experiment as all the weights increases or decreases of the specimens were reported at the initial weight. The experiment consisted of consecutive cycles (absorption - immersion in distilled water at 23^{o}C and freezing - placing the specimens in a freezing space at -7^{o}C). The weight of each specimen was measured constantly after each cycle. A cycle consisted of absorption - freezing at different time intervals: 24 hours, 48 hours, 168 hours and 336 hours. These cycles were repeated until no mass variations were observed, about 10 reps / weights (equilibrium moisture absorption of samples, was assumed to be reached when the weight gain of samples was less than 0.01%).

The variation of water absorption degree observed during the experiment was an expected one and the phenomenon is explained graphically in Figure 1. Initially, the absorption was different depending on the material and the fabric type. In the freezing phase no variations in samples weight or surface destruction were observed following a visual analysis. This can be explained by the quality of the interfacial fabric - epoxy resin, resulting in a very compact material from a structural point of view.

4. Results and discussion

The following figures shows the water absorption at 23^{o}C and freezing at -7^{o}C, consecutive cycles (absorption - immersion in distilled water, freezing - placing the specimens in a freezing space). Fig.1 shows a cycle consisted of absorption - freezing for 24 hours. The trends observed in this figure are similar for all composite materials. The rate of mass increase due to water absorption and then, when freezing, the mass remains almost the same as in the absorption.

Fig.2 shows a cycle consisted of absorption - freezing for 48 hours. This figure shows the water absorption for over 48 hours, consecutive cycles (absorption - immersion in distilled water - placing the specimens in a freezing space). Water absorption increases for L, E and HT epoxy systems. For C epoxy system the mass after water absorption and frozen cycles decreases. A comparison of Figs. 1 and 2 reveals that the amount of water absorbed is higher for 48 hours, concerning the L epoxy system. It can be seen that for 48 hours, the tested composite materials, absorb significantly more water than those tested for 24 hours. Water can cause matrix swelling, physical damage of the interphase and hydrolysis of the material.

Fig.3 shows graphical values of the specimen masses after the 10 cycles at 168 hours, from which we can see a stabilization of the values of the masses compared to the initial mass. As in Fig. 3, in Fig. 4 even if the time of a cycle was doubled, 336 hours, it can be seen that the sample mass is constant. From this point of view, we believe that the last test cycle confirms the observed saturation degree in the previous cycle (138 hours).

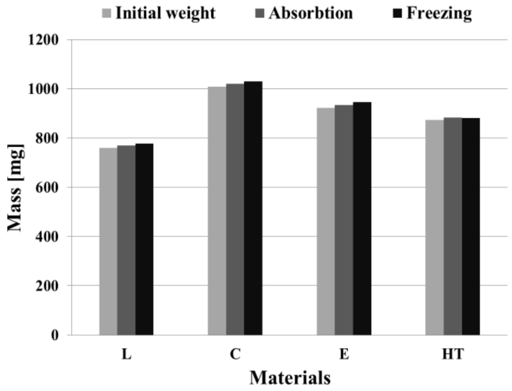

Figure 1. Water absorbtion at 23^{o}C and freezing at -7^{o}C, consecutive cycles (absorption - freezing) for 24 hours testing period

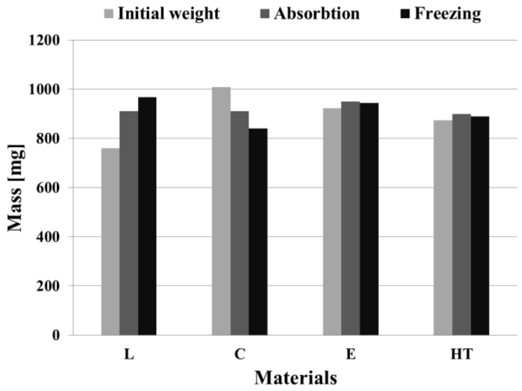

Figure 2. Water absorbtion at 23^{o}C and freezing at -7^{o}C, consecutive cycles (absorption - freezing) for 48 hours testing period

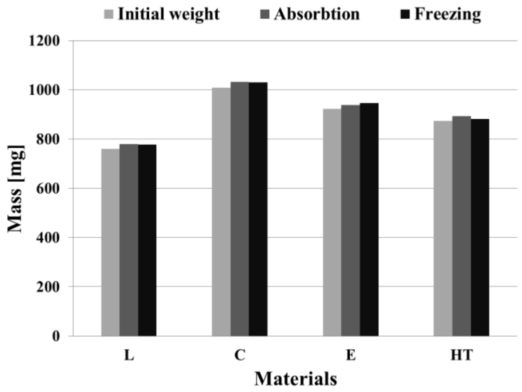

Figure 3. Water absorbtion at 23^{o}C and freezing at -7^{o}C, consecutive cycles (absorption - freezing) for 168 hours testing period

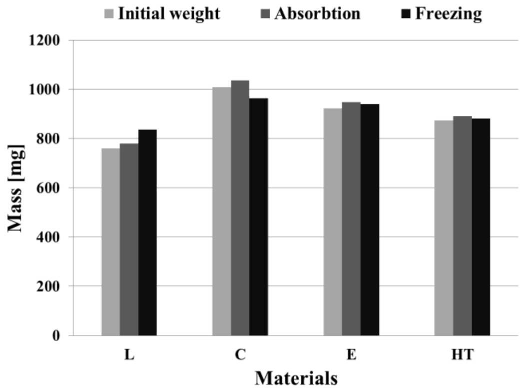

Figure 4. Water absorbtion at 23^{o}C and freezing at -7^{o}C, consecutive cycles (absorption - freezing) for 336 hours testing period

5. Conclusions

The experimental results obtained from the complete frost-absorption cycle are presented and commented in detail. Water absorption properties of laminates made of carbon, aramid and glass and four different resins namely L, HT, E and C has been made.

The four materials show different behaviours for the first two cycles (24 hours and 48 hours) and stability in the third and fourth cycles (168 hours and 336 hours). We cannot say that the structure of the material influenced in a specific way their behaviour after various cycles. The effect of moisture (water absorption and frozen-unfrozen) on the properties of polymer composites is an important issue and further studies are necessary.

References

[1] S. RASSMANN, R.G. REID, R. PASKARAMOORTHY, Effects of processing conditions on the mechanical and water absorption properties of resin transfer moulded kenaf fibre reinforced polyester composite laminates, Composites: Part A 41 (2010) 1612–1619.

[2] BLEDZKI AK, GASSAN J., Composites reinforced with cellulose based fibres, Prog Polym Sci 1999; 24:221–74.

[3] ADRIANA TICOALU, THIRU ARAVINTHAN, FRANCISCO CARDONA, A review on the characteristics of gomuti fibre and its composites with thermoset resins, Journal of Reinforced Plastics and Composites 32(2) 124–136.

[4] J MIJOVIĆ, LIN KF, The effect of hygrothermal fatigue on physical/mechanical properties and morphology of neat epoxy resin and graphite/epoxy composite J. Appl. Polym. Sci. 30 (1985) 2527.

[5] SALAH U. HAMIM, RAMAN P. SINGH, Effect of Hygrothermal Aging on the Mechanical Properties of Fluorinated and Nonfluorinated Clay-Epoxy Nanocomposites, Int Sch Res Notices, 2014.

[6] D. OLMOS, J. GONZA´LEZ-BENITO, Visualization of the morphology at the interphase of glass fibre reinforced epoxy-thermoplastic polymer composites, European Polymer Journal 43 (2007) 1487–1500.

[7] B.C. RAY, Temperature effect during humid ageing on interfaces of glass and carbon fibers reinforced epoxy composites, Journal of Colloid and Interface Science 298 (2006) 111–117.

[8] MUSTO P, RAGOSTA G, MASCIA L. Vibrational spectroscopy evidence for the dual nature of water sorbed into epoxy resins. Chem Mater 2000;12(5):1331–41.

[9] D. OLMOS, R. LOPEZ-MORON, J. GONZALEZ-BENITO, The nature of the glass fibre surface and its effect in the water absorption of glass fibre/epoxy composites. The use of fluorescence to obtain information at the interface, Composites Science and Technology 66 (2006) 2758–2768.

[10] J. COMIN, Polymer Permeability, Chapman and Hall, London, 1994.

[11] MAROM, G. A. D., L. J. BROUTMAN, Moisture penetration into composites under external stress, Polymer Composites 2.3 (1981): 132-136

[12] B.C. Ray, Effects of crosshead velocity and sub-zero temperature on mechanical behaviour of hygrothermally conditioned glass fibre reinforced epoxy composites, Materials Science and Engineering A, Vol. 379, Iss 1-2, 2004.

[13] ZHANG JIHUA, ZHAN MAOSHENG. Visual experiments for water absorbing process of fibre-reinforced composites. J Compos Mater 2004;38: 779–90.

[14] KOOTSOOKS A, MOURITZ AP. Seawater durability of glass- and carbonpolymer composites. Compos Sci Technol 2004;64:1503–11.

[15] KRYSTYNA IMIELINSKA, LAURENT GUILLAUMAT. The effect of water immersion ageing on low-velocity impact behaviour of woven aramid-glass fibre/epoxy composites. Compos Sci Technol 2004;64: 2271–8.

[16] CHENGQIANG REN, TIEHU LI, FAJU SONG, XIAOYAN SUN, QILANG LIN, Influence of additives on the permeability of impregnating coal-tar pitch, Materials Letters 60 (2006) 1570–1574.

[17] CHRISTOS J. TSENOGLOU, SYLVIA PAVLIDOU, CONSTANTINE D. PAPASPYRIDES, Evaluation of interfacial relaxation due to water absorption in fiber–polymer composites, Composites Science and Technology 66 (2006) 2855–2864.

[18] D 570 – 98, Standard Test Method for Water Absorption of Plastics

[19] KUMOSA L, BENEDIKT B, ARMENTROUT D, KUMOSA M, Moisture absorption properties of unidirectional glass/polymer composites used in composite (non-ceramic) insulators. Composites A 2004;35:1049–63.

[20] WEITSMAN YJ, YA-JUN GUO Y-J. A correlation between fluid-induced damage and anomalous fluid sorption in polymeric composites. Compos Sci Technol 2002;62:889–908.

[21] WOO M, PIGGOTT MR. Water absorption of resins and composites: IV. Water transport in fiber reinforced plastics. J Compos Technol Res 1988;10(1):20–4.

[22] H. ALAMRI, I.M. LOW, Effect of water absorption on the mechanical properties of nano-filler reinforced epoxy nanocomposites, Materials and Design 42 (2012) 214–222.

[23] P.K. MALLICK, Fiber-Reinforced Composites: Materials, Manufacturing, and Design, Third Edition, CRC Press, 2007.

Acknowledgments

The authors would like to acknowledge the financial contribution of the Project 12 P01 024 21 (C11) /31.08.2012 (code SMIS 50414).