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Characterization of Water Absorption, Thickness Swelling and Diffusion Coefficient of Natural Fiber/Epoxy Bio-Composites for Applications in Aquatic Environment

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

The aim of this work was to study the suitability of wood flour, fluted pumpkin stem and rice husk flour/epoxy bio-composites for application in aquatic environment. The water absorption, thickness swelling and diffusion coefficient of wood flour, fluted pumpkin stem flour, rice husk flour/epoxy bio-

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composites were investigated. Composites were prepared by the hand layup method at varying weight percentages of filler reinforcement (10, 20 & 30%). The increase in the addition of the fillers enhanced the water absorption, thickness swelling and diffusion coefficient of the composites. However, after 14 days of immersion, all the composites attained an equilibrium state. It was observed that with 30wt% filler loading, the fluted pumpkin stem flour epoxy composites showed the highest water absorption of 40.5%, diffusion coefficient of 10.80x10⁻⁸m²/s, followed by the wood flour epoxy composite with water absorption of 35.3%, diffusion coefficient of 8.17x10⁻⁸m²/s, while rice husk flour epoxy composites had the lowest water absorption of 16.5%, diffusion coefficient of 6.12x10⁻⁸m²/s. The same trend was observed for the composite showed 1.62%, followed by fluted pumpkin stem flour composite 1.40% and rice husk flour composite 0.35% with 30wt% filler loading. A similar trend was observed for the composite 0.35% with 30wt% filler loading. A similar trend were observed for the composites with 20 and 10 wt% filler loading. A similar trend were observed for the composites with 20 and 10wt% filler loading. This study can be useful to comprehend the effect of water absorption on the performance of natural fiber composites and help in creating designer compositions for use in building, construction and automotive.

Keywords: Natural fiber composite; water absorption; thickness swelling; diffusion coefficient; epoxy resin.

1. INTRODUCTION

"The attraction of using natural fibers as a replacement for manmade fibers in composites as reinforcements has surged extensively in modern times. The importance of utilizing natural fibers has risen owing to their outstanding features including lightweight, non-abrasive, combustible, nontoxic and biodegradable" [1,2]. composite "The conventional applies reinforcements such as glass fiber, the carbon fiber and numerous other reinforcing elements. Natural fibers such as flax, bamboo, hemp, jute and so forth have been employed recently as reinforcements in the composites. A natural fiberreinforced composite exerts the merits such as low density, low cost, easily recyclable and biodegradable in nature, so therefore it is referred to as a potential substitute for manmade fibers. Utilization of natural fibers elicits a understanding comprehensive of surface adhesive cohesion and its chemical composition" [3-5]. "Natural fibers are a blend of primary components such as cellulose, hemicelluloses, lignin, wax and pectin. To produce improved adhesion between reinforcements and matrix, fiber surface modifications has to be performed by chemical modifications such as NaOH treatments. Many researchers have studied on the fiber modifications of the fibers surfaces to achieve enhanced mechanical properties" [6-11].

"The hydrophilic nature of the natural fibers plays a vital function in the ageing of bio-composites by immersion in water or in a wet atmosphere. Moisture absorption has become a barrier in the development of the use of the natural fibers in composite materials due to degradation and decrease in the properties of composites in wet conditions. Bio-composites used for outdoor applications need to be investigated for water absorption, thickness swelling and diffusion coefficient in a wet environment. Moisture absorption is one of the main parameters to assess the quality of the bio-composite materials. Water absorption is considered a shortcoming considering its migration into the material and this can lead to perturbing of the matrix-fiber interface as well as affects the stability and reduces the overall strength of bio-composites" [12]. Munoz et al. [13] analyzed "the water absorption and change on flax fiber reinforced bio-epoxy composites for mechanical properties focusing on sustainable green composites and eco-friendly preparation using RTM (resin transfer molding) process. Diffusion coefficient, water uptake values, flexural properties were studied showing diffusion coefficient in the order of 10⁻⁶ mm²/sec". Gomes et al. [14] investigated "the absorption capability for water in sisal fiber reinforced polymer matrix composites at temperatures of 25, 50 and 70°C with 44.6% sisal fibers and 55.4% polyester matrix in proportion manufactured using hand layup technique and water absorption were analyzed. Results revealed favorable water absorption, moisture content gradients were higher in the surface planes, while diffusion coefficient was dependent on the moisture content, geometrical shape and temperature. It is thus imperative that this problem is addressed so that natural fiber may be considered as a viable reinforcement in composite materials". The mechanisms of water transport in hybrid composite was investigated by Melo et al. [15] who reported that the water absorption of all formulations approach the non-Fickian diffusion model.

The rice husk was observed to be better in terms. of its water absorption properties when it was compared with other types of fillers. Muthuraj et al. [16] reported that "composites having rice husk showed lower water absorption compared to other types of fillers, such as wheat husk, wood fibers, and textile waste. This observation was ascribed to the higher hydrophobicity of rice husk compared to other fillers". Yusuf et al. [17] compared "composites containing rice husk with composites having bamboo stem fiber. They reported that composites with rice husk were better in terms of their lower water absorption and swelling thickness as a result of the reduced attraction of rice husk to water". Shevkh et al. [18] compared "rice husk and bagasse ash in an HDPE composite. The rice husk/HDPE composite was seen to have lower water absorption and thickness swelling properties. This was ascribed to the lower accessible -OH group on the surface of rice husk compared to bagasse fibers". Mohamed et al. [19] compared "the water absorption properties of various contents of hybrid kenaf/rice husk in а polypropylene composite". Similarly, other researchers found that a higher rice husk content exhibited lower water absorption properties. The reason being that kenaf has larger voids with more hydroxyl groups that can interact with water.

The aim of this study is to explore the suitability of wood flour, fluted pumpkin and rice husk flour epoxy composites for building, construction and automotive applications. Therefore, the water absorption, thickness swelling and diffusion coefficient of fluted pumpkin stem flour, rice husk flour and wood flour epoxy bio-composites were investigated.

2. MATERIALS AND METHODS

2.1 Materials

In the present work, three different natural fibers/agro-waste materials were used, namely, wood flour, fluted pumpkin stem flour and rice husk flour (density 0.22-1.14 g/cm³) were sourced locally and processed. Epoxy resin (Ampreg 21) with a density of 1.2 g/m³ and the epoxy curing agent/hardener (Ampreg 21) with a density of 1.02 g/cm³, brown color and hardening time of 24-36 at 23°C were purchased from AMT Composites Company Johannesburg, RSA.

2.2 Mold Preparation

The square plate mold with dimensions: $480 \times 480 \times 3$ mm was specifically fabricated for the development of the samples was assembled with base and top plates, for the easy removal of the samples without sticking. The mold cavity was thoroughly cleaned in order to avoid particles interaction with the epoxy resin and were coated the inner layers of the mold with polyvinyl alcohol (PVA), for easy removal of the composite samples.

2.3 Preparation of the Composites Test Samples

Natural-fiber-reinforced epoxy resin was fabricated using hand-lav-up technique to ascertain the water absorption and thickness swelling tendencies of the composites. Newly manufactured ampreg-21 epoxy resin, which belongs to the larger family of epoxides was used as the matrix material and wood flour, fluted pumpkin and rice husk flour agro-waste materials used as reinforcing fillers. The ampreg 21 epoxy resins and the Ampreg 21 hardener were mixed in a ratio of 100:33 by weight as prescribed in the manufacturer data sheet and all the composites were fabricated at room temperature. This experimental work fabricated three composite sheets at different percentage weights of polymer matrix and reinforcing fillers: 90/10, 80/20 and 70/30 percentage weight loadings of epoxy resin and the different natural fiber agro-waste fillers respectively.

2.4 Water Absorption and Thickness Swelling Test

Rectangular composite panel samples of dimension 22.4mm x 66.2mm were used for water absorption and thickness swelling tests as per ASTM D-570-98 [20]. Thirty samples were dried with three representing each composite sheet. All the samples were heated in an oven at 160°C for about 30 min in order to dry any initial moisture that may be present in the samples and later cooled at room temperature condition for 4hrs. The initial weights and corresponding thicknesses of these samples were measured thereafter, to the nearest 10-3 g and 10-3 mm. respectively. All the samples were later immersed in distilled water and after every 24 h, the composite samples were periodically taken from the water bath and the sample surface was wiped using tissue paper. After that, the composite samples were weighed to determine the mass of the water they absorbed. Composite samples were weighed regularly until the water absorption percentage attained a saturation level. In the present investigation, all the composites attained saturation at 10 days of immersion. After the 14th day there was no increase in the water absorption percentage, hence, for analysis, water absorption percentage for 14 days was reported (24, 48, 72, 96, 120, 144, 168, 192, 216, 240, 264, 288, 312 and 336 h) at room temperature (22±3°C). All the samples were measured with the digital balance and vernier caliper and the results were analyzed. The moisture absorption and the thickness swelling were then estimated using the relations.

 $W(\%) = W_t - W_0 / W_0 \times 100\%$ (1)

 $S(\%) = T_t - T_0 / T_0 \times 100\%$ (2)

Where W_o is the oven-dried mass of the specimen before it was immersed in distilled water, while W_f is the final mass of the specimen after its immersion in water. Three samples were analyzed for each panel and were reported. Where T_o is the initial thickness (mm) of the composite specimen after it was oven-dried and T_f is the final thickness of the specimen after it was immersed in distilled water.

2.5 Determination of Diffusion Coefficient

The diffusion coefficient of the different natural fiber composites were calculated using the Fickian equation,

$$\partial C/\partial t = \chi \partial^2 C/\partial x^2 \tag{3}$$

In which the accumulation, $\partial C/\partial t$ [cm⁻³s⁻¹], is proportional to the diffusivity χ [cm²/s] and the 2nd derivative of the concentration, $\partial^2 C/\partial x^2$ [cm⁻⁵].

3. RESULTS AND DISCUSSION

3.1 Water Absorption and Thickness Swelling Behavior

"Lignocellulosic fibers exhibit weak resistance to water absorption as a result of this natural fiber reinforced composites have negative impact on dimensional stability properties when subjected to water in the environment. To comprehend the durability of composites based on the field of application, it's vital to investigate the water absorption and thickness behavior of natural fiber composites. In general, water molecules enter natural fiber composites by three varying mechanisms: diffusion of water content within the micro-gaps between the polymer chains; capillary transport into the micro-gaps and flaws in the interfaces between the fibers and the matrix" [21-23]. The moisture is basically absorbed by the interface between the fiber and the matrix, as well as by the fiber through hydrogen bonding.

The percentage water absorption and thickness swelling of the composites were plotted against the number of days of immersion as shown in Figs. 1-6. The results were expected because the higher the natural fiber content in the composites, the higher the water absorption rate as well as the higher the thickness swelling. The percentage of water absorption and thickness swelling increased with number of days of immersion in water and attained equilibrium after 14 days.

From Figs.1-6, it is observed that the composites with 30wt% filler loading showed the maximum water absorption and thickness swelling, followed by the composites with 20wt%. However, the degree of water absorption and thickness swelling of the composites varied according to the type of natural fiber utilized. This is to say that the wood flour, fluted pumpkin stem flour and rice husk flour/epoxy composites, showed differences in the amount of water absorption and thickness swelling properties at the 10, 20 and 30 wt% filler reinforcement. This could be attributed to the differences in the chemical composition of the natural fibers (i.e., to differences in the cellulose, hemicelluloses and lignin contents of the different types of natural fibers used [24]. "Higher reinforcement increased water absorption and thickness swelling as a result of increased contact between water molecules and fiber. Moreover, the percentage of water absorption and thickness swellina increased linearly from the onset of the immersion period, showing the steady absorption of water by the natural fiber composites. Similar results were found in napier grass fiber/polyester composites" [25].

For all the composites exposed to water, the amount of water penetration increased linearly and attained a saturation state after 14 days of immersion. From the water absorption and thickness swelling results, it is evident that the lowest and highest water absorption and thickness swelling was observed at 10wt% and 30wt% of fiber volume of the composites respectively. 10wt% fiber volume of composites showed reduced water absorption and thickness swelling, and this is due to the amount of matrix is higher at the lower fiber weight percentage. The hydrophobic nature of the matrix restricts the interaction of the water molecules with the fiber content. Aji et al. [26] reported that after 14 days of immersion, kenaf/pineapple leaf fiber had minimum water absorption of 4% obtained by varying the kenaf and pineapple leaf fiber reinforcement by an equal amount.

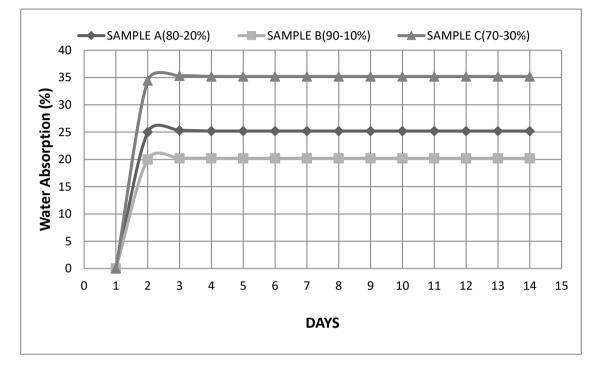


Fig. 1. Water absorption of wood flour/epoxy composites

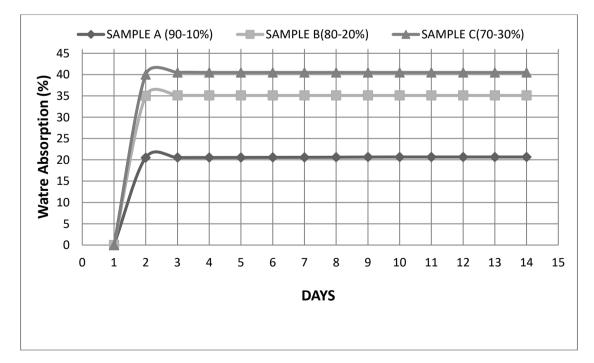
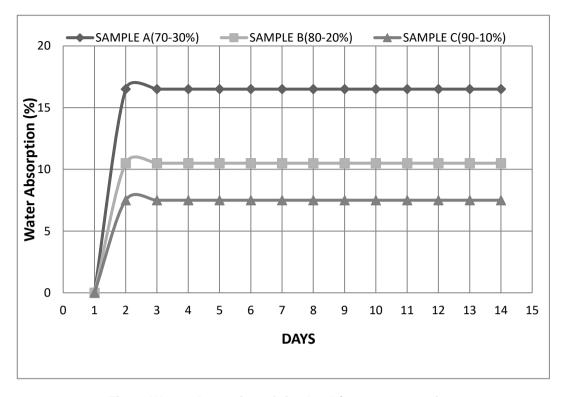


Fig. 2. Water absorption of fluted pumpkin stalk flour/epoxy composites



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Fig. 3. Water absorption of rice husk/epoxy composites

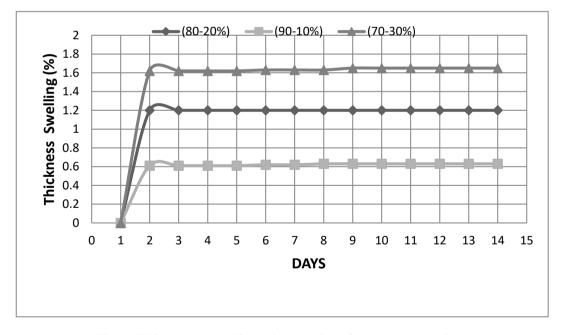


Fig. 4. Thickness swelling of wood flour/epoxy composites

3.2 Diffusion Coefficient

Water molecules penetrate into natural fiber composites by the diffusion mechanism, and the diffusion coefficient can be computed using the experimental data. It has been reported that the diffusion coefficient can be used to determine the permeability of water molecules. For example, it has been reported that sisal fiber composites attained equilibrium quickly when the diffusion coefficient was at its maximum. Table 1 gives the values of the diffusion coefficients "D" for the three types of natural fiber composites. It was observed that the diffusion coefficient increased with the increase of the wood flour, fluted pumpkin and rice husk flour fiber content from 10wt to 30wt% respectively. It can be seen that the 10wt% fiber composites show the lowest diffusion coefficient values for the wood flour, fluted pumpkin and rice husk flour/epoxy composites. The 30wt% fiber composites gave the highest diffusion coefficient followed by 20wt% fiber composites. Similar findings have been reported by other researchers [27] where it was found that higher fiber weight percentages resulted in higher diffusion coefficient, water absorption and swelling.

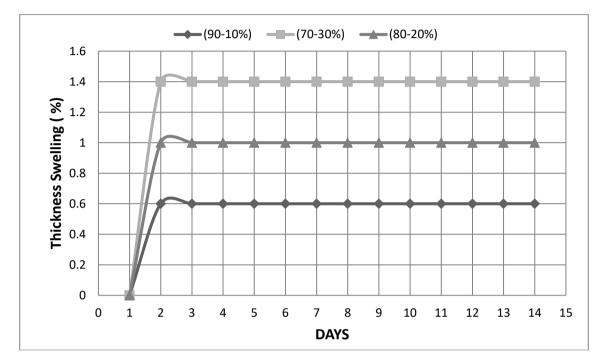


Fig. 5. Thickness swelling of fluted pumpkin stalk/epoxy composites

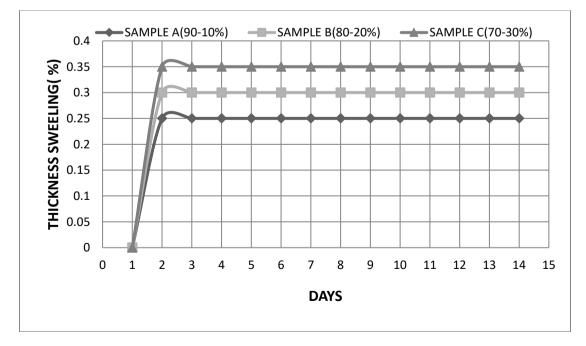


Fig. 6. Thickness swelling of rice husk/epoxy composites

Composite Samples	Concentration (c) =(Wi -Wo)	Thickness (x) = (T1-T2)	Duration (s)	Diffusion coefficient (m ² /s)
Sample A (Ri	ce husk flour/Epox	y Composites)		
90:10	0.11	0.03	604800	1.11x10 ⁻⁸
80:20	0.09	0.04	604800	3.96x10 ⁻⁸
70:30	0.13	0.03	604800	6.12x10 ⁻⁸
Sample B (W	ood flour/Epoxy Co	omposites)		
90:10	0.09	0.12	604800	2.66x10 ⁻⁸
80:20	0.23	0.01	604800	5.18x10 ⁻⁸
70:30	0.1	0.02	604800	8.17.10x10 ⁻⁸
Sample C (FI	uted pumpkin stem	/Epoxy Composite	s)	
90:10	0.36	0.01	604800	3.63x10 ⁻⁸
80:20	1.03	0.03	604800	6.443x10 ⁻⁸
70:30	1.9	0.05	604800	10.80x10 ⁻⁸

Table 1. Diffusion coefficient of natural fiber/epoxy composites

It was observed that with 30wt% filler loading, the fluted pumpkin stem flour epoxy composites showed the highest water absorption of 40.5%, diffusion coefficient of 10.80x10⁻⁸m²/s, followed by the wood flour epoxy composite with water absorption of 35.3%, diffusion coefficient of 8.17x10⁻⁸m²/s, while rice husk flour epoxy composites had the lowest water absorption of 16.5%, diffusion coefficient of 6.12x10⁻⁸m²/s. The same trend was observed for the composites with 20 and 10 wt% filler loading. In which the composites with 20wt% filler loading showed improved water absorption and diffusion coefficient values compared to the composites with 10wt% filler loading. In terms of thickness swelling, the wood flour composite showed the highest value of 1.62%, followed by fluted pumpkin stem flour composite 1.4% and rice husk flour composite 0.35% with 30wt% filler loading. A similar trend was observed for the composites with 20 and 10wt% filler loading. Where by the composites with 20wt% filler loading showed higher thickness swelling values compared to the composites with 10wt% filler loading respectively.

4. CONCLUSIONS

The wood flour/fluted pumpkin stem flour/rice husk flour/epoxy bio-composites were prepared by varying filler concentrations using the hand lay-up technique. The water absorption, thickness swelling and diffusion coefficient properties of the natural fiber/agro-waste composites was investigated for their suitability of application in water-rich environment. The absorption, thickness swelling water and diffusion coefficient of the composites increased with increase in filler concentration which

depends upon the type of natural fiber/agrowaste material utilized. The composites with 30wt% filler concentrations showed the maximum water absorption, thickness swelling and diffusion coefficient values, followed by the composite with 20wt% filler loading, while the composite with 10wt% filler loading showed the lowest values.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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