

Influence of *Arapaima gigas* fish scales as reinforcement in DGEBA/TETA epoxy composite for flooring applications: mechanical and thermal behavior

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Abstract

Given the ever-growing environmental concerns, repurposing natural materials is a viable approach to developing novel green materials. Thus, the use of fish scales as reinforcement in composites has great potential. This work aimed to evaluate the influence of using arapaima fish scales as reinforcement in DGEBA/TETA epoxy composites on the mechanical and thermal properties. Composite plates using 30 vol% of arapaima fish scales were analyzed through compression tests, DMA, TGA, and DSC. The compression tests of 30 vol% arapaima/epoxy composites showed a significant increase both in Young's modulus (from 710.88±98 at 0% to 1406.20±241 MPa at 30 vol% of scales) and compressive strength (from 20.80±7.14 at 0% to 68.4±11.69 MPa at 30 vol% of scales). The DMA and TGA results showed an improvement in the thermal properties of the composites as compared to the neat epoxy resin, while the DSC results demonstrated that the use of the fish scales as reinforcement did not impair the thermal stability of the resin. These results support the potential of arapaima fish scales as reinforcement in polymeric composites for flooring applications that require high compressive strength and thermal stability.

Keywords: Arapaima fish scales; Epoxy; Green composites; Flooring; Thermal behavior; Mechanical properties.

1 Introduction

Since its development, epoxy resin has been applied in a broad range of fields, such as electronics, automobiles, aeronautics, chemistry, civil engineering, and others [1]. It is used in many applications due to its low cure shrinkage, great insulation capacity, and chemical-resistant properties. Nonetheless, the epoxy resin presents negative aspects as well, such as low impact resistance, high flammability, and poor weather resistance, as well as environmental concerns which reduce the number of possible applications [2]. In view of that, many different materials have been suggested as fillers/reinforcements in composite systems.

One of those systems, natural fibers reinforced polymer composites have been the subject of study in many papers over the past decade [3-9]. The concern for environmental preservation and the interest in the development of sustainable materials have fueled those and still drive current works in the hope of a greener future. These fibers present appealing characteristics such as good mechanical resistance, low cost/weight ratio, and biodegradability [9].

Additionally, nature has engineered other fibrous structures with interesting properties, such as the fish scales, which can be considered as a novel reinforcement in composites. The use of these materials is a two-fold solution since they are usually discarded, and their associated cost is lower than the epoxy.

Through millions of years of evolutionary processes, nature generates materials with extremely resistant structures, which in turn provide them protection against predators [10-12]. Pirarucu (from Tupi language: red fish, 'pirá' fish, 'urucum' red) or arapaima (i.e., *Arapaima gigas*), a well-known fish in Brazil, is one of the world's largest freshwater fish and has scales that are about 1mm thick and can reach up to 10 cm in length [12]. These scales have an internal structure divided into layers that present a mineralization gradient, i.e., the mineral phase (hydroxyapatite nanocrystals) is mostly present in the external, exposed surface and less pronounced in the innermost layers. These layers are formed by parallel-oriented type-I collagen fibers, approximately 50 μm thick, which are reinforced with hydroxyapatite nanocrystals.

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These lamellae are stacked with orientations that vary by a characteristic angle, between 45-90°, forming a Boulingand-type structure [13-16]. This characteristic structure renders the arapaima scales as one of the toughest biological materials.

Considering their application as reinforcement in epoxy composites, the scales' thermal behavior is a critical aspect to be evaluated. Uskokovic et al. [17] studied the thermal stability of collagen-based biocomposites with hydroxyapatite precipitates, observing the phase transformations present in the material with increasing temperature, mainly in the native structure of collagen fibers. Torres et al. [18] studied the effect of the water content in the scales of *Arapaima gigas* on the thermal transitions of their components, observing that collagen's denaturation temperature tends to increase as the water content of the scales increases [18].

Despite the extensive literature regarding the structure and properties of different fish scales, only a few papers have investigated the use of these naturally engineered intricate materials as reinforcement in composites. Many of these studies have focused on the thermal, mechanical, and tribological properties of fish scales reinforced composites (FSRC) [19-23]. The use of rohu scales (*Labeo rohita*) as reinforcement in epoxy composites showed promising results since they resulted in better resistance to erosion wear when compared to neat epoxy resin [19]. In one of the few studies exploring arapaima scales as reinforcement, Bezerra et al. [24] analyzed the structure and some mechanical properties of the scales and their composites with epoxy resin, reporting an increase in the flexural modulus of the epoxy matrix with the addition of 30 vol% of arapaima scales [24].

The current literature suggests that fish scales may be a novel alternative for the development of sustainable flooring materials since the fish scales can decrease flammability and increase the mechanical properties of the epoxy matrix. In this context, the objective of the present study is to assess the mechanical behavior of epoxy composites reinforced with arapaima scales through compressive tests and to analyze the thermal stability of the arapaima fish scales and respective epoxy composites through dynamic-mechanical analysis (DMA), thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). These results provide a preliminary understanding of arapaima scales reinforced composites as novel, more sustainable materials for the flooring industry.

2 Materials and methods

2.1 Materials

Arapaima fish scales were obtained from the Municipal Market in Manaus, Brazil. The scales were thoroughly washed in water and left to dry at ambient temperature. Subsequently, the plain scales were subjected to a flattening process to facilitate their use as reinforcement of the composite material,

which consisted in rehydration for 3 days and drying under compression at 80 °C for 3h, as proposed by Bezerra et al. [24]. No posterior treatment was applied to the scales. The epoxy resin selected for this study was the DGEBA/TETA, with a proportion of 13 parts of hardener for 100 parts of resin, supplied by Epoxyfiber, Rio de Janeiro, Brazil. Considering that previous papers report that composites with 30 vol% of natural fibers tend to show the best overall properties [4,6-8], the composite plates were prepared with this content of arapaima scales (EP+30%AS) via a compression molding process, in a process described elsewhere [24].

2.2 Methods

2.2.1 DSC and TGA/DTG

Differential scanning calorimetry (DSC) was carried out for both the arapaima scales, the epoxy resin, and the composite with 30 vol% of scales in a Hitachi, model DSC 7020. The test samples, of approximately 5mg, were enclosed in aluminum pans and the temperatures ranged from 18 to 200 °C with a heating rate of 10 °C/min, under a nitrogen flow of 60 mL/min.

Samples were extracted and weighed (10 mg) from the EP+30%AS composite. The thermogravimetric analysis and its derivative (TGA/DTG) were performed in a TA Instruments Systems, Q500. The analysis was carried out with a heating rate of 10 °C/min and a temperature range of 25–800 °C.

2.2.2 Dynamic mechanical analysis (DMA)

Following ASTM D4065-12 [25], dynamic mechanical analysis was carried out in order to assess the storage module (E'), loss module (E''), and tangent δ ($\tan \delta$) related to the viscoelastic behavior of the EP+30%AS composite. In addition to that, the glass transition temperature (T_g) of these composites was determined through the method described in ASTM D7028-07 [26]. Rectangular specimens of EP+30%AS composite were prepared, with dimensions of 50 mm x 13 mm x 5 mm. The test was carried out on a TA Instruments model Q/800, in three-point flexion mode at 1 Hz frequency and heating rate of 3°C/min. The tests were carried out in a nitrogen atmosphere with a temperature range of -50 to 150 °C.

2.2.3 Compression tests

Complying with ASTM D6641-16 standard [27], compression tests were performed on ten specimens prepared from the plain epoxy and epoxy composites reinforced with 30 vol% of arapaima scales. First, the composite plates were cut into smaller pieces with dimensions of 10 mm x 10 mm x 20 mm, then the compression tests were performed on an EMIC DL10000 equipment, using a crosshead speed of 5 mm/min.

3 Results and discussion

3.1 DSC and TGA/DTG

Figure 1 shows the DSC and DDSC (i.e., derivative) curves of the arapaima scales for the first heating run. During the first heating run, an endothermic peak was observed at 104.0 °C. This peak may be due to residual moisture loss in the scales and to denaturation of the collagen protein in the scales' structure. According to Torres et al. [18], this process can be divided into two steps: first, the separation of collagen triple helices into individual ones, and second, the unfolding of these helices into random coils. The authors reported that an increase in the moisture content of the scales results in higher temperatures at which denaturation occurs. The calculated enthalpy of denaturation here (i.e., area below the peak), of 265 J/g, was similar to the value found by Torres et al. [18], of 220 J/g, for the scales with 0% water content. However, the temperature of the denaturation peak (104 °C) was higher than the value described (73.3 ± 4.6 °C) for the 0% water content scales. This finding may imply better thermal stability of the scales acquired in Manaus, Brazil. No significant transitions were observed for the second and third heating runs of the scales.

Figure 2 presents the DSC and DDSC curves obtained for the first (Figure 2a) and second (Figure 2b) heating runs of the neat epoxy resin. The first heating run revealed an endothermic peak at 59.10°C, associated with moisture loss in the epoxy. The second heating run, shown in Figure 2b, depicts a transition at 119.0 °C related to the glass transition temperature (T_g) of the epoxy resin. The cooling and consecutive heating runs confirmed the glass transition peak at 119.0 °C.

The DSC and DDSC curves for the first (Figure 3a) and second (Figure 3b) heating runs of the composite with 30 vol% scales are shown in Figure 3.

In the curve from the first heating run, presented in Figure 3a, a significant endothermic peak was observed at 96.6 °C, which was associated with the release of moisture from the composite along with a less pronounced endothermic peak at approximately 72.5 °C, attributed to the denaturation process of the collagen in the scales. After the first heating run, when the moisture is eliminated and the thermal history erased, it is possible to visualize the glass transition temperature (T_g) at 103.6 °C, shown in Figure 3b. One can observe a decrease in the glass transition temperature with the addition of the arapaima scales to the epoxy resin. This occurrence was expected and is reported in the literature for composites reinforced with fish scales and other natural materials [9,20,28,29]. It is attributed to the interference of the reinforcement with the three-dimensional structure of the polymer macromolecules. The scales function as a plasticizer, facilitating the movement of the matrix chains and decreasing its glass transition temperature.

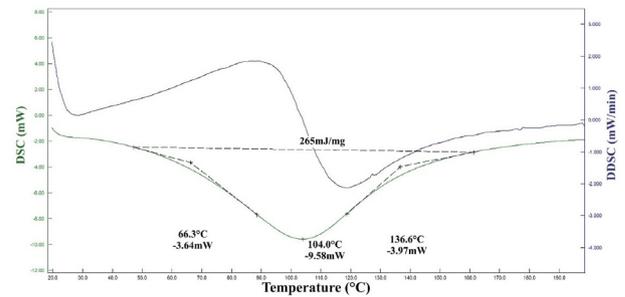


Figure 1. DSC curves of the arapaima scales.

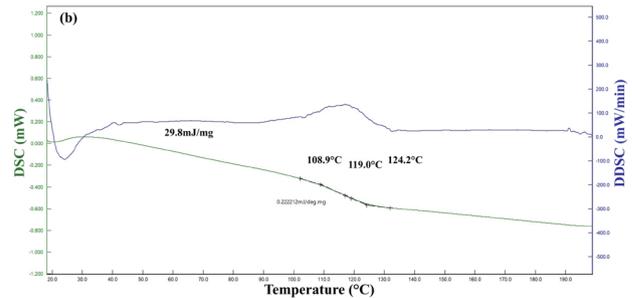
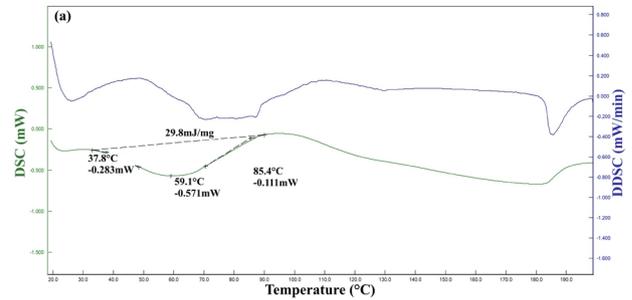


Figure 2. DSC curves of the neat epoxy resin: (a) first and (b) second heating runs.

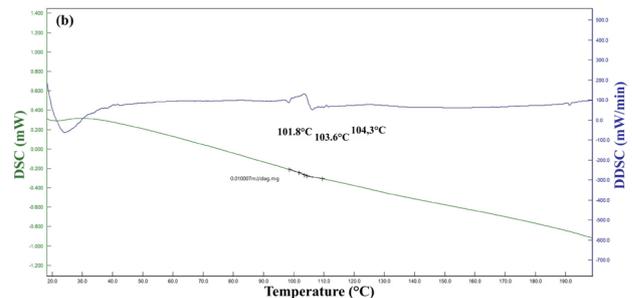
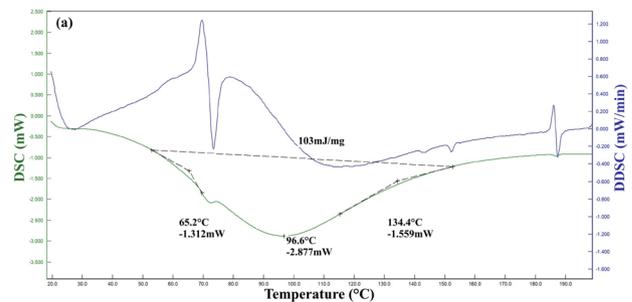


Figure 3. DSC curves of the 30 vol% arapaima scales reinforced composite: (a) first and (b) second heating runs.

Figure 4 shows the thermogravimetric analysis curves and their first-order derivative for the samples of the composite with 30 vol% arapaima scales. A small loss of mass (3.8%) is seen at temperatures near 100.0 °C extending up to 200.0 °C, which is associated with the moisture released by the epoxy matrix [28-31]. A greater mass loss is seen starting at 330.8 °C (T_{onset}), related to the degradation of the resin and the organic components of the scales. Upon further inspection of the DTG curve, it is possible to detect the highest rate of degradation of the material at the temperature of 372.0 °C (T_{max}). Additionally, a total loss of mass of 85.86% was observed.

The 30 vol% arapaima scales reinforced composites showed thermal degradation behavior similar to the reported in previous works for natural materials reinforced composites but presenting higher degradation onset (T_{onset}) and maximum degradation rate (T_{max}) temperatures [20,28-31]. A comparison of the temperatures (T_{onset} and T_{max}) and the residue at 600.0 °C from the present work and the literature values are presented in Table 1.

3.2 DMA

Dynamic mechanical analysis (DMA) allows the evaluation of the stability of the composite with increasing temperature, its glass transition temperature, and its viscoelastic nature when stimulated by dynamic loading. These analyzes were performed on epoxy resin composites reinforced with 30 vol% of arapaima scales. The storage modulus (E'), loss modulus (E''), and $\tan \delta$ curves were obtained from the results and are shown in Figure 5.

Considering the $\tan \delta$ curve from the DMA analysis, one can determine the T_g of the composite with 30 vol% of arapaima scales as equal to 100.6 °C. This value is slightly higher than that found by other authors for the neat epoxy resin but lower than the one obtained via DSC analysis. This value and others are shown in Table 2. The decrease in the glass transition temperature may indicate that the incorporation of arapaima scales improved the mobility of the epoxy resin chains. Consequently, the thermal behavior of the 30 vol% scales reinforced shows a slight difference in comparison to the neat resin.

The storage modulus reached around 6500 MPa at a temperature of 25 °C and the loss modulus around 550 MPa at its maximum point, for the composites with 30 vol% of arapaima scales.

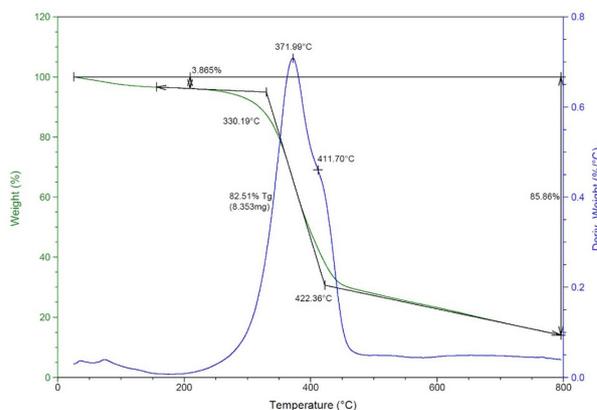


Figure 4. TGA and DTG curves for the composite reinforced with 30 vol% of arapaima scales.

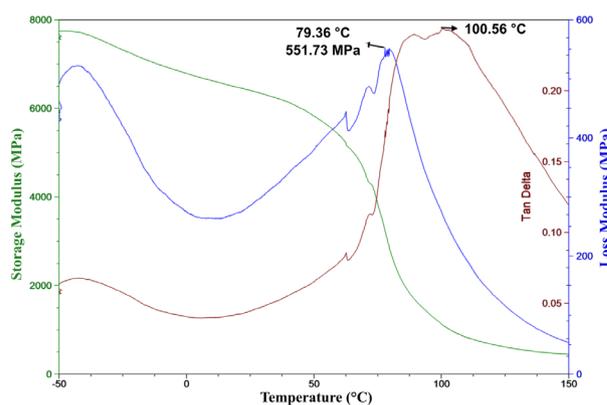


Figure 5. DMA curves for the storage modulus (E'), loss modulus (E'') and $\tan \delta$ for the composites reinforced with 30 vol% of arapaima scales.

This represents an average increase of 73 and 120%, in relation to the values reported for neat epoxy resin by Luz et al. [7], which represents a significant gain in viscoelastic properties. Furthermore, in Figure 5 it is possible to observe a sudden drop in the storage modulus at temperatures in the range of 60-85 °C. This drop is due to relaxations in the polymer matrix associated with the glass transition temperature (T_g) from the crystalline to the amorphous state. The relaxation peak (α), represented by the maximum point of the loss modulus curve, occurred at temperatures close to 80 °C.

Table 1. Comparison of the thermogravimetric parameters from the 30 vol% scales reinforced composites and other epoxy composites in the literature

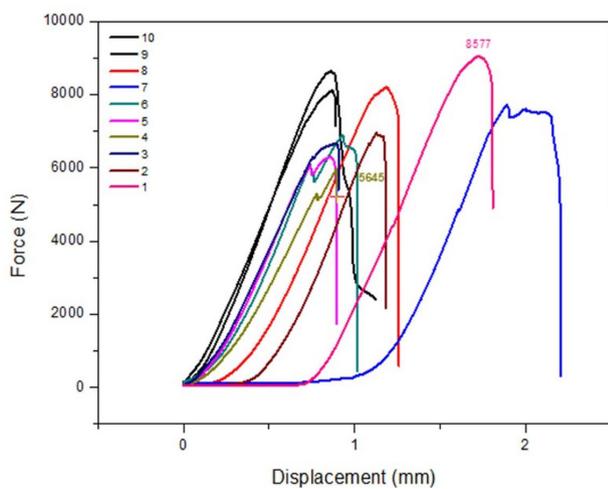
| Composite | T_{onset} (°C) | T_{max} (°C) | Residue at 600 °C (wt%) | Reference |
|-------------------------------|------------------|----------------|-------------------------|--------------|
| Epoxy+30 vol% Arapaima-scales | 330.8 | 372.0 | 14.1 | Present work |
| Epoxy+30 vol% Sedge-fiber | 292.0 | 334.2 | 22.6 | [28] |
| Epoxy+30 vol% Caranan-fiber | 298.1 | 334.2 | 25.0 | [29] |
| Epoxy+30 vol% Carnauba-fiber | 325.9 | 368.5 | 18.1 | [30] |
| Epoxy+30 vol% Hemp-fabric | 301.9 | 351.5 | 21.1 | [31] |
| Epoxy+20 vol% Fish-scales | 295.0 | 363.0 | 4.9 | [20] |
| Neat Epoxy resin | 343.0 ± 1.6 | 385.9 ± 4.5 | 18.4 ± 2.2 | [28,30,31] |

Table 2. T_g , E' at 25°, E'' and α values obtained for the epoxy composite reinforced with 30 vol% of arapaima scales compared to other epoxy composites reinforced with natural fibers

| Composite | T_g (°C) | Storage modulus (MPa) at 25° | Maximum loss modulus (MPa) | Relaxation peak α (°C) | Reference |
|-------------------------------|------------|------------------------------|----------------------------|-------------------------------|--------------|
| Epoxy+30 vol% Arapaima-scales | 100.6 | 6500 | 550 | 80 | Present work |
| Epoxy+30% Coir-fibers | 73.2 | 3800 | 430 | 75 | [7] |
| Epoxy+30% Figue-fabric | 89.4 | 1300 | 180 | 85 | [8] |
| Epoxy+30% Caranan-fibers | 113.1 | 1378 | 140 | 101 | [29] |

Table 3. Mechanical properties of neat epoxy resin and composite reinforced with 30 vol% of arapaima scales

| Material | Young's Modulus (MPa) | Compressive strength (MPa) | Deformation (%) |
|------------------------------|-----------------------|----------------------------|-----------------|
| Epoxy+30vol% Arapaima scales | 1406 ± 241 | 68.40 ± 11.69 | 5.61 ± 1.90 |
| Neat epoxy | 711 ± 98 | 20.80 ± 7.14 | 2.89 ± 1.19 |

**Figure 6.** Force vs displacement curves from the compression tests of epoxy composites with 30 vol% of arapaima scales.

This value is close to those obtained in other composites reinforced with natural fibers, as shown in Table 2.

3.3 Compression tests

Uniaxial compression tests were performed to evaluate the mechanical behavior of the composites under compressive loads. The force versus displacement curves for the different specimens are shown in Figure 6. Upon inspection of the curves, it is possible to verify the typical behavior under compressive loads for the EP+30%AS composites: first, a slightly nonlinear rise in force, followed by a linear region and catastrophic failure after reaching the maximum force.

Figure 6 presents the values for the upper and lower maximum forces of the different specimens, namely 8.6 and 5.6 kN. The displacement at break for these two specimens were 1.8 and 0.9 mm, respectively.

After the maximum force, the specimens suffered a catastrophic failure. The primary failure mechanism was a brittle failure due to delamination at the interface between the

scales and epoxy phases. The secondary failure mechanism was the buckling of the inner resin/scale layers with some delamination under compressive force, due to the initiation and propagation of cracks at the interfaces of the scales with the epoxy matrix, resulting in crack propagation in the matrix. The mechanical properties of composites are markedly improved with the addition of a reinforcement, considering that the reinforcement phase has higher strength and stiffness values than the matrix phase. In view of this, the reinforcing effect of the arapaima scales on the epoxy matrix should be observed through the increase in compressive strength properties, as shown in Table 3.

Table 3 presents a comparison of the results from the compression tests of the EP+30%AS composite and the neat epoxy. One can clearly notice a significant increase in Young's modulus and compressive strength of the epoxy resin after the incorporation of arapaima scales. These results agree with what has been reported for the arapaima scales reinforced composites. Bezerra et al. [24] reported that by using the whole scales, the epoxy resin can become stiffer under flexural loads mainly due to the intricate, gradually mineralized Bouligand structure of the scales.

Equivalent results were obtained when smaller and thinner fish scales were applied as reinforcement [19-23]. This consistency indicates that the fish scales are effectively enhancing the epoxy resin properties.

4 Conclusions

The DSC analysis showed that the incorporation of the arapaima scales in the epoxy resin did not significantly affect the glass transition temperature (T_g), which was in agreement with the results obtained through the DMA method. The TGA results showed a slight decrease in the temperatures of the degradation onset (T_{onset}) and maximum degradation rate (T_{max}) with the addition of arapaima scales to the epoxy resin. However, these values were higher than those for other natural materials reinforced composites, which may indicate an advantage to the use of the arapaima scales.

The compression tests showed an improvement in the mechanical properties with the incorporation of 30 vol% of arapaima scales, in which the epoxy's Young's modulus was doubled and its compressive strength increased by 30%. A decrease in flammability and an increase in weather resistance have also been reported in the literature with the incorporation of fish scales in the epoxy. The results presented in this work along with the ones reported in the

literature indicate that the application of the arapaima scales as reinforcement in epoxy composites is a viable alternative for greener materials in the flooring industry.

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