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STUDY OF ADHESION BEHAVIOR OF THE SISAL FIBER WITH SURFACE TREATMENT IN PORTLAND CEMENT BASED MATRIX

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Abstract. *Choosing the use of vegetable fibers as reinforcement has been considered beneficial because these require less energy for extraction and may improve the mechanical properties of the composite. However, modifying the characteristics of natural fibers by applying some type of treatment may provide an increase in the anchoring ability of the fiber to matrix. Thus, this study aims to apply a treatment using expanded polystyrene and rice husk silica in natural sisal fibers, in order to evaluate its influence on the fiber-matrix adhesion capacity through pullout tests using fiber embedment length of 50 mm, for the control age of 7 and 28 days. The absorption capacity and surface microstructure of the sisal fiber were also evaluated. Based on the obtained results, the water absorption index of treated sisal fibers was considerably reduced and the optical microstructure images showed possible fiber sealing through the proposed treatment, besides agglutination points along the fiber, which may have been a possible contributing factor to lower matrix-fiber adhesion. It was verified, through the statistical analysis of the pullout load values, that the proposed treatment did not promote increase of the adhesion capacity of the fiber with the cementitious matrix.*

Keywords: *sisal fiber, surface treatment, pullout test, cementitious composites.*

1. INTRODUCTION

The use of vegetable fibers as reinforcement in composite materials have been considered an effective alternative when referring to lightweight composites, since their mechanical properties approximate or even surpass those associated with synthetic fibers (Defoirdt et al., 2010). In cement composites, from the inclusion of natural fibers tries to improve characteristics of the mechanical properties, besides adding to it a better cost-benefit and environmental preservation (Ferreira et al., 2015).

The composites reinforced with sisal fibers have been widely used due to their high impact strength, tensile and flexural (Li et al., 2000). These properties may be correlated to the fact that sisal fiber presents one of the highest values of modulus of elasticity and mechanical strength among natural fibers (Bledzki and Gassan, 1999). Despite its excellent tensile strength, sisal fibers in their natural state provide low chemical adhesion and high water absorption, promoting a variation in their morphology, which directly interferes with fiber-matrix adhesion capacity, occurring a great loss adhesion with the matrix (Li et al., 2008).

The adhesion behavior is an essential feature of fibers reinforced composites, as it significantly influences their performance when exposed to mechanical efforts (Ferreira, 2012). In connection therewith, Ferreira et al. (2015) presented a comparison of pullout load for sisal fiber with 25 mm embedment length for different surface treatments. In Figure 1(a) it is observed that all the treatments resulted in an improvement in the adherence between the sisal fiber and Portland cement matrix, being these related to the increase of stiffness, adhesion and mechanism of friction, mainly for polymer and hybrid treatments. The formation of a styrene butadiene copolymer film on the surface of the fiber was observed through scanning electron microscopy (SEM), besides anchoring points of the polymer, promoting a bridge between fiber and matrix, strengthening interfacial bonding, as may be seen in Fig. 1(b).

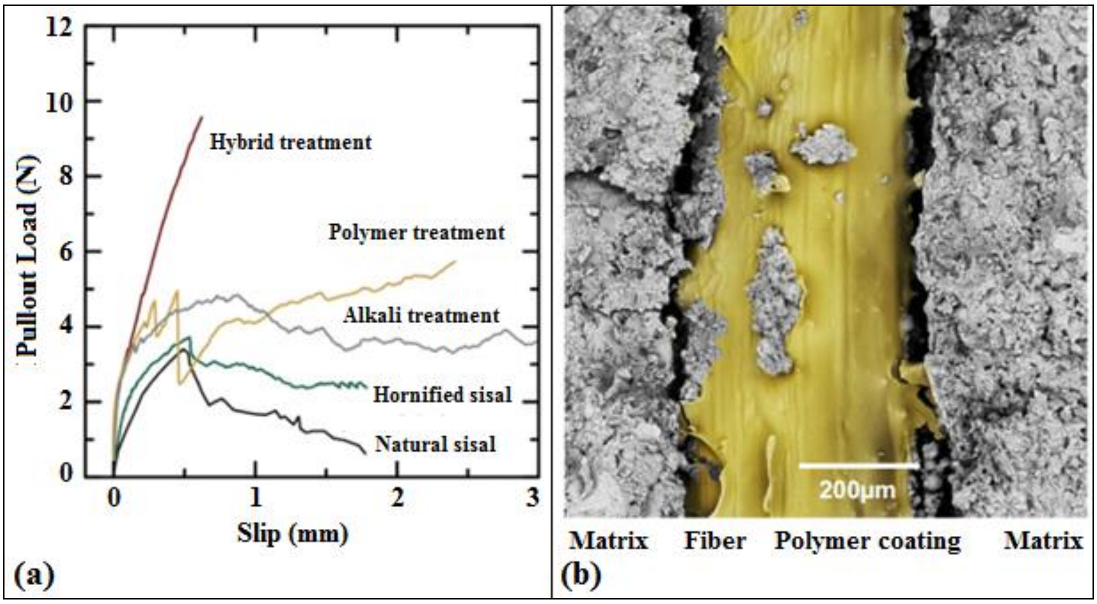

Figure 1. (a) pullout behavior slip curves (L = 25 mm) for different types of fiber surface treatments and (b) interface matrix-polymer-sisal fiber. Adapted from Ferreira et al. (2015).

Fidelis et al. (2018) verified through pullout tests an increase in fiber-matrix interfacial bonding to polymer-coated jute fibers. Similarly, Zukowski et al. (2018), found higher values of adhesion force using composites with alkali-treated fibers, where the pulled out fiber was shown with a connected segment of the matrix yet.

However, the complex microstructure and significant heterogeneity of natural fibers are preponderant factors that tend to influence their interaction with cement matrices, whose identification requires greater advances in the current state of knowledge (Ferreira et al., 2018).

Rice husk silica (RHS) is a mineral addition that interacts chemically in the cement environment, as it provides a bond with the calcium hydroxide present in the Portland cement during its hydration. RHS particles may promote roughness on the surface of the fibers; possibly becoming a physical agent that will collaborates with greater anchorage between the fiber and the matrix. In this context, with the objective of evaluating the influence of the surface treatment on the maximum fiber-matrix adhesion load in pullout tests, the current study used expanded polystyrene (EPS) based polymer treatment, dissolved in toluene, with the addition of RHS to the natural sisal fibers, using a free matrix of calcium hydroxide. Besides this, for a better understanding of the mechanical adherence behavior, the water absorption capacity index (AI) and surf ace microstructure of the sisal fiber were evaluated by optical microscopy.

2. METHODOLY

The treatment used in the sisal fibers consisted in impregnating EPS thermoplastic polymer with addition of RHS to the fibers. The solubilization of the EPS in the solvent was performed by shaking in ultrasonic bath equipment, enabling the synthesis of both materials in order to promote a bond between the fiber and the silica particles through the adhesion bridge formed by the use of the polymer. For this methodology, the initial amounts of solvent and polymer were based on those performed by Roca Bruno (1996).

Thus, tests were realized by fixing the amount of solvent and varying in mass the amounts of EPS and RHS, verifying the solution viscosity and the solution's adherence capacity through immersing the sisal fibers to promote the treatment. Thereby, based on the fractions analyzed, in order to allow the association between EPS, RHS and sisal fiber, the amounts indicated in Tab. 1 were obtained.

Table 1. Proportion of materials to promote surface treatment in the sisal fibers.

Expanded polystyrene	17 g
Toluene P.A. ($C_6H_5CH_3$)	100 ml
Rice husk silica	2.5 g

In order to verify the modification in the sisal fiber structure the water absorption capacity of the fibers was evaluated according to the methodology proposed by Toledo Filho (1997) to obtain the water absorption index for the treated and untreated sisal fibers. For both fiber classifications, two samples with various fibers in filaments were separated, which had the same initial dry weight.

As stated by Toledo Filho (1997), the result for the AI was obtained through Eq. (1), in which "P_{dry}" represents the weight of the dried fiber in kiln and "P_{wet}" the wet weight, after saturation, thus verifying how much the treatment used influenced the water absorption capacity of the sisal fibers.

$$AI = \frac{P_{wet} - P_{dry}}{P_{wet}} \quad (1)$$

In order to relate the AI results to what occurred on the fiber surface through the proposed treatment, optical microscopy analysis was performed for untreated and treated fiber using optical metallographic microscope, model Kontrol - IM713.

The matrix used to produce the composites is an adaptation of the matrix used by Ferreira (2012), for which the proportion of materials utilized by the author was maintained. The composition of the calcium hydroxide free cement matrices is shown in Tab. 2 and these were prepared according to NBR 7215 (ABNT, 1996).

Table 2. Consumption of the materials used in the proportion of the cement matrix.

Matrix (kg/m³)								
Material	Cement	Sand	Metakaolin	Fly ash	Water	W/C* ratio	SP**	Spread (mm)
Free CH Matrix	362	542	289	434	434	0,40	25	≥450

** W/C: water/cement materials*
*** SP: superplastificizer (by solids of Blend [Cement + Fly ash + Metakaolin])*

Adapted from Ferreira (2012).

The specimens for the pullout test were produced using MDF plates, which have 32 mm circular recess to allow the fitting of the PVC molds. The plates contain central bore to ensure complete alignment of the fibers within the composite (Silva et al., 2011), as shown in Fig. 2.

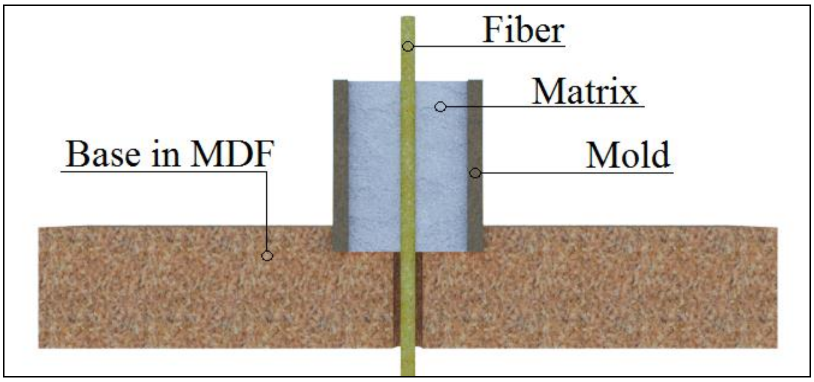

Figure 2. Schematic drawing showing fiber alignment.

The fibers were then introduced and the PVC molds were embedded in the MDF plate. The molding of the specimens with mortar consisted of filling the molds manually with the aid of a bag of confectioners (Ferreira, 2012), a procedure similar to what may be verified in Fig. 3.

After filling the PVC molds, the MDF plate was superimposed on the molds, as shown in Fig. 4. Afterwards, the fiber was aligned in order to provide greater rigidity to the set and to allow better uniformity of the fiber in the matrix medium.

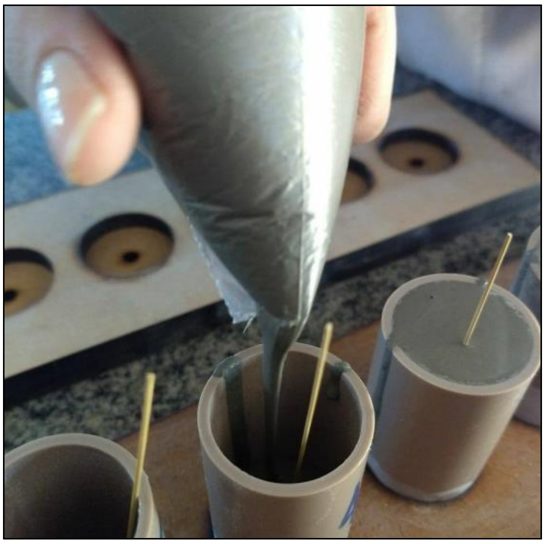

Figure 3. Procedure for molding test specimens for pullout test.

Figure 4. Top fitting of MDF plates in the PVC molds.

The pullout tests was performed on a Shimadzu AGS-X with 500N load cell and displacement speed of 0.1 mm/min. For the pullout test were produced 40 specimens with 50 mm embedment length of sisal fiber inside of specimen. These were tested at the control ages of 7 and 28 days being equally divided into: sisal fiber reinforced composites without surface treatment (UNTREATED), reference specimens; and composites reinforced with sisal fiber subjected to polymeric treatment with RHS (TREATED). Figure 5 shows the specimens used to pullout tests and the tests configuration, where it is possible to visualize aligned positioning of the fiber with respect to the center of the PVC mold.

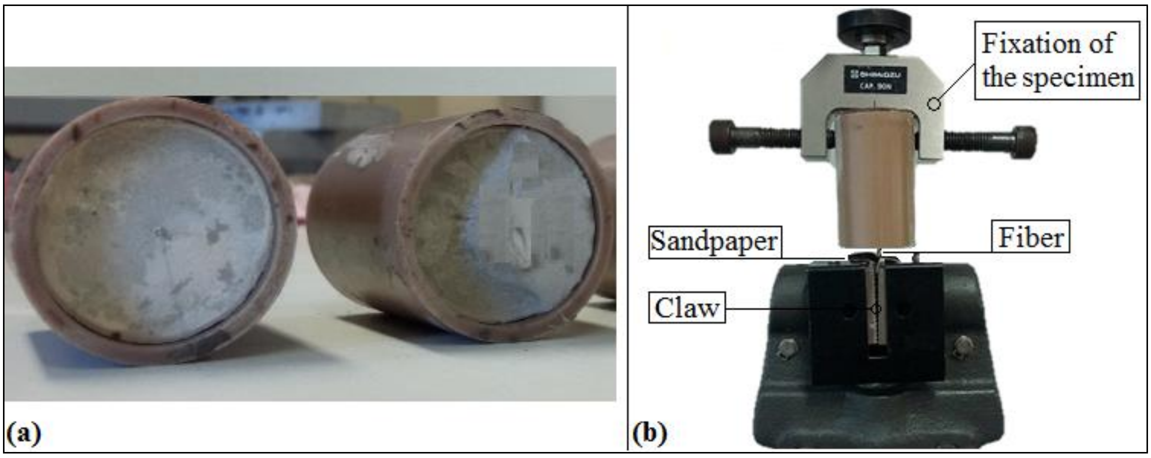

Figure 5. (a) Specimens with 50 mm embedment length and (b) configuration of the pullout test.

3. RESULTS AND DISCUSSION

For the evaluation of the water absorption index of the fibers, for which an AI of 84.67% was obtained for the natural fibers of sisal and 15.18% for the fibers subjected to the polymeric treatment based on EPS and RHS, showing that after the treatment application there was a decrease of about 70% in AI.

Based on the literature, for other types of treatment in natural fibers, this behavior may be explained due to the stiffening polymeric structure of the fibrocells due to the treatment process used, allowing a greater packing of the internal fiber structure (Ferreira, 2016).

As expanded polystyrene consist an apolar compound of a hydrophobic nature, ie, this polymer is characterized by having little or no interaction with water (Shi, et al., 2016). Consequently, hydrophobic materials tend to form a film on the surface in contact, which may have prevented paste (cement and water) from penetrating into the pores of the fiber and, for this reason, decreases the adhesion.

Figures 6 and 7 show the images obtained by optical microscopy for untreated and treated sisal fiber. Analyzing the images obtained from the surface of the sisal fibers with optical microscope, it may be noted that the treatment used created a possibly discontinuous layer in the longitudinal wrapping of the fiber, promoting agglutination points of the EPS polymer solution with RHS. Evaporation of the solvent, causing a high increase in the viscosity of the mixture when exposed to the ambience, may be indicated as one of the factors for this to happen.

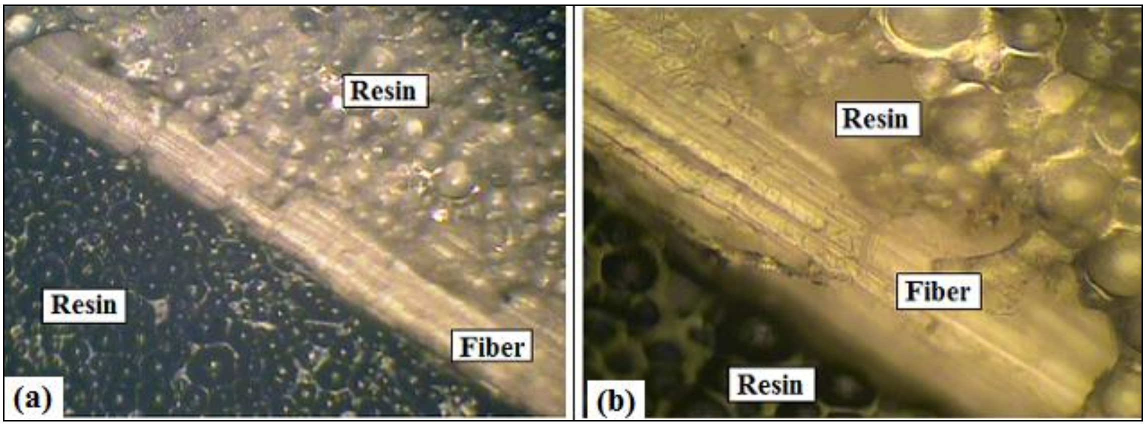

Figure 6. Optical microscopy images of the untreated sisal fiber surface: (a) 100x magnification and (b) 200x magnification.

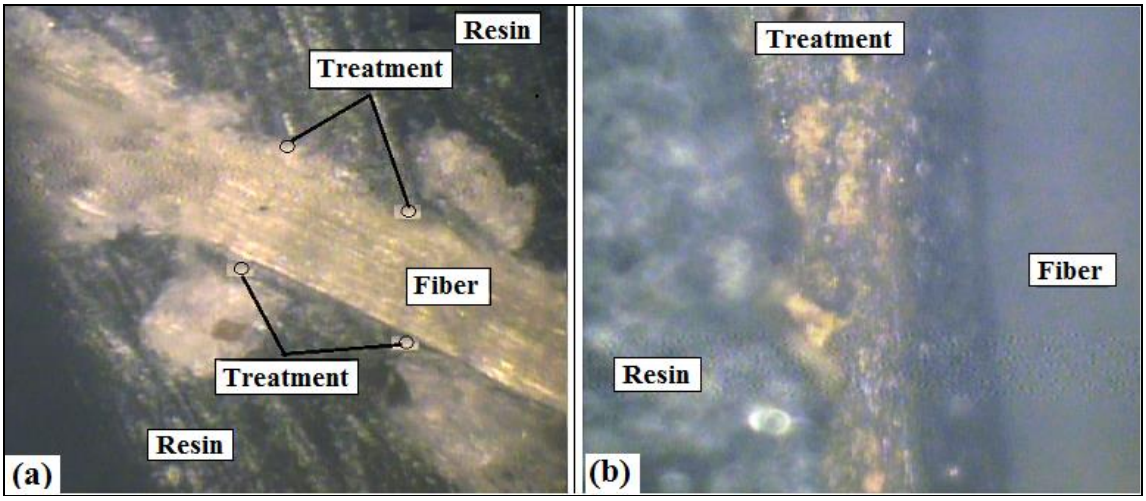

Figure 7. Optical microscopy images of the treated sisal fiber surface: (a) 100x magnification and (b) 200x magnification.

Another relevant aspect was that the treatment that consisted in the dissolution of the polymer and the silica in solvent, the non-uniformity of the treatment might have been caused by the heterogeneous absorption of this by the pores of the sisal fiber, causing excess points. Furthermore, it is possible that there was no interaction of the natural fiber with the solvent used for the solubilization of the mixture, or up to even with the polymer, since this would act as the binding element among polymer-silica-fiber.

The results for the cementitious composites reinforced with sisal fibers, untreated and treated, such as the value average, minimum and maximum to the adhesion load and displacement, standard deviation and coefficient of variation for the specimens were verified at both control ages of 7 and 28 days, as shown in Tab. 3 and 4.

Table 3. Results for composites reinforced with untreated and treated sisal fibers at the control age of 7 days.

Control Age	UNTREATED		TREATED	
7 days	Displacement (mm)	Pullout load (N)	Displacement (mm)	Pullout load (N)
Average	1.53	6.40	1.25	5.24
Minimum	1.04	2.11	0.33	1.58
Maximum	2.89	10.16	2.69	14.63
Standard Deviation	0.53	2.64	0.73	3.91
Coefficient of Variation	34.84%	41.17%	58.61%	74.56%

Table 4. Results for composites reinforced with untreated and treated sisal fibers at the control age of 28 days.

Control Age	UNTREATED		TREATED	
28 days	Displacement (mm)	Pullout load (N)	Displacement (mm)	Pullout load (N)
Average	1.33	7.73	0.96	5.73
Minimum	0.45	2.18	0.18	2.08
Maximum	2.28	15.12	1.83	13.32
Standard Deviation	0.69	4.51	0.62	3.39
Coefficient of Variation	52.10%	58.41%	64.81%	59.11%

Regarding the results obtained for the specimens using untreated and treated fibers with the same embedment length, it may be observed that the use of the treatment seems to impair the adhesion performance of the fibers to the matrix. The average pullout loads supported by reinforced composites using natural sisal fibers was 18.09% and 25.85% higher at 7 and 28 days, respectively, compared to that obtained for those using treated fibers. The deviations obtained also indicate that the treatment resulted in larger variability in the results for pullout load and displacement, which may be justified by the non-uniformity of the process used to treat the fibers.

Thus, in order to confirm whether the evaluated treatment actually decreased the fiber-matrix bonding interaction, ANOVA was performed for the data obtained for the maximum pullout forces of the fiber inserted into the cementitious matrix. From the statistical analysis, with 95% reliability by Fisher's method, it was determined that there was no significant difference between the pullout forces supported by the natural or treated sisal fiber reinforced composites, as well as between the control ages analyzed, ie, there was no change in the mechanical behavior of adhesion of the composites.

Therefore, it may be proven that the treatment with EPS and RHS polymer did not cause changes in the fiber to the point of improving its adhesion interaction with the Portland cement matrix, but maintained the same pullout load levels supported using natural fibers of sisal.

Moreover, since, according to Shi et al. (2016), the type of polymer used does not interact with water and acts to repel its molecules, the humidification process may not have been able to reopen the capillary voids of sisal fibers. This process occurs when the fiber interacts with the paste (cement and water), thus impairing the stiffness by slipping, due to the lower adhesion between the fiber and the cementitious matrix.

Besides, it is possible sealing of the fiber by the treatment, creating a very thick and irregular layer of EPS combined with RHS, as it was realized a manual process, subject to variability in the constancy of the treatment for each fiber. Such an occurrence may have promoted a sliding layer, giving to origin points susceptible to decrease of fiber adherence to the matrix. Thus, the dimensional variation provided by the coating created for the treated sisal fiber caused the effort transmission capacity to be reduced, weakening the interfacial bond.

Based on the coefficients of variation shown in tables 3 and 4, it was noted that the levels reached by the pullout load are variable, which may be explained not only by the non-uniformity of the treatment, but also by the fiber diameter irregularity. In accordance with Silva (2009), the sisal fibers has a hierarchical structure with variable morphology, having irregular cross-section. Therefore, in order to obtain accurate results on pullout stress and deformation, the irregular areas of the fibers should be considered and the breaking stress calculated.

According to Naik et al. (2019), the characterization of fiber-matrix interfacial bonds using conventional fiber pullout tests may not be suitable for cementitious composites reinforced with natural fibers. Therefore, the authors propose a modified pullout test method for the indirect characterization of the natural fiber and cement matrix interfacial properties, which may be still a factor that favor the results found in this study.

For a better understanding of the interaction between natural fibers and cementitious matrices, which is a primordial characteristic in the control of the mechanical behavior of natural fiber reinforced composites (N-FRCC), Ferreira et al.

(2015) characterized geometrically and mechanically three types of natural fibers, sisal, curaua and jute. These results emphasize that the geometric and mechanical properties that control the behavior of these fibers are in some way comparable to other types of "industrial" fibers already used as reinforcement in FRCCs. The authors performed an inverse identification procedure, based on a simplified bilinear bond slip law; assuming invariant along the embedded length of the fiber, the procedure demonstrated its ability to identify the interaction between fiber and matrix binding.

4. CONCLUSIONS

This study aimed to analyze the influence of polymeric treatment with EPS and RHs in sisal fibers to examine their adhesion behavior when these were used to reinforce cement Portland matrices bases. Thus, to verify the ability of the bond of adhesion at the fiber-matrix interface, the composites with sisal fibers in the natural state and treated sisal fibers were compared. The water absorption capacity of the fibers was evaluated and fiber surface modifications were verified by optical microscopy, before and after the treatment, in order to understand the behavior of the composites subjected to the pullout tests.

Concerning to water absorption capacity, it was observed that the treated fibers obtained a lower absorption index than for those in the natural state. As found in the literature, this fact may have been caused due to the decrease in the fiber capillary void index, which were possibly closed progressively from the application of the treatment. This fact may also have occurred because of the hydrophobic nature of the polymer, since this material type has no interaction with water and acts to repel its molecules.

According to the data obtained, it may be concluded in view of the adhesion capacity of the fiber with the cementitious matrix that the treatment used did not significantly alter the mechanical properties of adhesion of the sisal fibers, maintaining pullout load levels equivalent to natural fibers. Thus, this treatment was possibly not sufficient to promote an increase of adhesion at the fiber-matrix interface, creating a distinct interface of fiber-polymer-matrix interaction. This may have occurred because the treatment did not change the modification of the morphology of the sisal fiber, which would cause the reformation in the walls of its fibrocells, allowing a lower variability in the load supported by the fibers during to the pullout tests.

The manual process of performing the treatment may have caused dispersion in the results, since there is not total control of the uniformity of the treatment along the fibers, as evidenced by the analysis of optical microscopy images. Thus, because the diameter checks were not performed to obtain the pullout stress, it was not possible to evaluate how inconstancy in the fiber diameters after the treatment may have influenced in variation of the results and, therefore, made it difficult to obtain a dimensional standard of sampling.

Therefore, the proposed treatment to standardize fiber structure, in order to improve its adhesion capacity with cimentitious matrix, was not effective. However, from the ineffectiveness of the superficial treatment presented, it is necessary to analyze alternatives of chemical and/or physical treatments to modify the mechanical properties of the fibers or to explore the combination of the polymeric treatment with the addition of rice husk silica to another type treatment already studied.

5. ACKNOWLEDGEMENTS

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7. RESPONSIBILITY NOTICE

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