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REPLACEMENT OF SYNTHETIC ADDITIVES WITH LIGNIN AND TANNINS IN THE PRODUCTION OF CONCRETE ROOF TILES

André Orathes do Rego Barros¹, Raquel Marchesan², Michelle de Pereira Menezes¹, Karolayne Ferreira Saraiva¹, Rodrigo Araújo Fortes³, André Ferreira dos Santos²

¹Universidade Federal do Tocantins (UFT), Programa de Pós-Graduação em Ciências Florestais e Ambientais (PGCFA), Gurupi, Tocantins, Brasil - urso.orathes@gmail.com; michelle2015@mail.uft.edu.br ; karolayne1409@mail.uft.edu.br.
²Universidade Federal do Tocantins (UFT), Curso de Engenharia florestal, Gurupi, Tocantins, Brasil –raquelmarchesan@uft.edu.br*; andrefs@uft.edu.br.
³Instituto Federal do Tocantins (IFTO), Curso de Engenharia Civil, Palmas, Tocantins, Brasil - rodrigoaraujofortes@gmail.com

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Resumo

Lignina e tanino como substitutos ao aditivo sintético em produção de telhas de concreto. Este estudo teve como objetivo avaliar o efeito da substituição de aditivos sintéticos por aditivos naturais na produção de telhas de concreto, visando melhorar propriedades físicas e mecânicas, como absorção de água, resistência à flexão, peso seco, e conformidade com aspectos normativos. Foram formulados seis tratamentos utilizando lignina e tanino como aditivos naturais, em diferentes proporções 2,5% e 5%, além de uma formulação de referência contendo apenas aditivo sintético. Os resultados indicaram que o tratamento com lignina na concentração de 2,5% (T2) destacou-se em relação aos demais, apresentando desempenho próximo ao limite especificado pela norma NBR 13858-2 (ABNT, 2009) nos aspectos visuais, e atendendo integralmente aos critérios de peso seco, absorção de água e carga de ruptura à flexão. Em contrapartida, os tratamentos com tanino apresentaram alto índice de perdas e desempenho mecânico insuficiente. Todos os tratamentos, com exceção do T5, atenderam aos critérios de empenamento e folga entre telhas (GAP). Conclui-se que a lignina na concentração de 2,5% é uma alternativa mais indicada para substituir aditivos sintéticos em telhas de concreto.

Palavras-chaves: Aditivos naturais. Plastificantes. Aproveitamento de Resíduos.

Abstract

This study aimed to evaluate the effect of replacing synthetic additives with natural additives in the production of concrete tiles, aiming to improve physical and mechanical properties, such as water absorption, flexural strength, dry weight, and compliance with regulatory aspects. Six treatments were formulated using lignin and tannin as natural additives, in different proportions, 2.5% and 5%, in addition to a reference formulation containing only synthetic additives. The results indicated that the treatment with lignin at a concentration of 2.5% (T2) stood out in relation to the others, presenting performance close to the limit specified by standard NBR 13858-2 (ABNT, 2009) in visual aspects, and fully meeting the criteria of dry weight, water absorption and bending rupture load. On the other hand, tannin treatments showed a high rate of losses and insufficient mechanical performance. All treatments, except for T5, met the warping and gap between tiles (GAP) criteria. It is concluded that lignin at a concentration of 2.5% is a more suitable alternative to replace synthetic additives in concrete tiles.

Keywords: Natural additives. Plasticizers. Waste utilization.

INTRODUCTION

Araújo *et al.* (2022) emphasize that new technologies, including replacing synthetic components with sustainable materials, such as natural additives, have been applied to enhance the durability and strength of cementitious materials produced from Portland cement, such as concrete roofing tiles. Additives can provide several advantages, including accelerated curing and setting times of concrete, the possibility of reducing cement content, improved workability, reduced mixing water demand, and even enhanced strength and service life of both individual components and entire structures (Botelho & Marchetti, 2018; Lima *et al.* , 2018). Currently, a wide range of synthetic additives is available on the market that improve their performance when incorporated into cement. Research has demonstrated that the use of synthetic latexes, such as polyvinyl acetate (PVA), acrylic copolymers (SAE), ethylene-vinyl acetate copolymers (EVA), and styrene-butadiene copolymers (SBR)—some of the polymers employed in concrete and mortar formulations—has led to significant improvements, particularly in strength and durability (Costa *et al.*, 2015).

The woody tissue of plant species is composed of the biopolymers cellulose, hemicellulose, and lignin (macrocomponents), along with mineral compounds and extractives (low-molecular-weight compounds), which include several chemical classes such as terpenes, alkaloids, and phenolic compounds (e.g., tannins, flavonoids, lignans, and coumarins) (Sjostrom, 2013). Recently, the production and application of biopolymers have emerged as a promising alternative due to their technical and economic viability, offering significant potential for expansion. Biopolymers are polymers or copolymers derived from renewable raw materials, including those from timber and non-timber forest species. Among them, kraft lignin—a residue extracted during chemical pulp production processes (Vaz Junior *et al.*, 2020)—and tannin, obtained from the bark of genera such as *Eucalyptus* and *Acacia* for use in leather tanning, are particularly noteworthy.

Unlike many other polymers, lignin possesses a complex structure with non-repetitive monomeric units. As the second most abundant natural polymer, lignin confers rigidity, impermeability, and resistance to xylophagous organisms to plant tissues (Lapierre, 1993; Fengel & Wegener, 1984). It is a three-dimensional polymer and constitutes a principal structural component of the plant cell wall. Lignins are widely distributed among both angiosperms and gymnosperms. While gymnosperm lignins predominantly exhibit guaiacyl and syringyl units, angiosperms present all three types of lignin units: syringyl, hydroxyphenyl, and guaiacyl (Malavasi *et al.*, 2016).

Structural modifications occur during wood pulping and lignin precipitation processes; consequently, no isolation method yields lignin in its native state as found in plants. Such structural changes directly affect the lignin's potential applications. Using kraft lignin as an additive is highly promising, given that it is a macromolecule inherently present in wood's chemical constitution and contributes to particle adhesion.

Doherty *et al.* (2011) argue that developing value-added products from lignin will greatly enhance the economics of biomass-based product manufacturing. At the same time, it will improve the sustainability of natural resource utilization and enhance environmental quality by reducing greenhouse gas emissions and toxic discharges.

Tannins have been used since antiquity for leather tanning and have long been employed in the wine industry and in the production of wood adhesives and preservatives. In some countries, such as Australia and South Africa, tannins have been commercially utilized for some time (Pizzi & Mittal, 2017). According to Zhou and Du (2019), tannins are extracted from sustainable natural materials and are widely used for the preparation of adhesives due to their phenolic structure. Tannins can be categorized into hydrolyzable and condensed types, with the latter being the principal focus of wood adhesive research and accounting for 90% of global tannin production. Nevertheless, as with lignin, no extensive studies still investigated their use as natural additives in concrete and mortar formulations for civil construction.

Using natural additives in concrete has become increasingly recognized as a promising alternative in the construction sector, addressing the growing demand for more sustainable and efficient practices. According to Borges, Motta, and Pinto (2019), natural additives offer advantages such as low cost, biodegradability, and recyclability, in addition to promoting economic benefits within the agricultural sector. Given this context, research into natural additives may contribute to developing a new type of concrete roofing tile exhibiting properties comparable to or superior to those produced with synthetic adhesives, while simultaneously helping to mitigate environmental impacts associated with the toxicity and degradation of discarded tiles. The general objective of this study was to evaluate the effect of replacing synthetic additives with natural additives (lignin extracted from *Eucalyptus* and tannin extracted from *Acacia*) on the performance of concrete roofing tiles, aiming to improve their physical and mechanical properties.

MATERIALS AND METHODS

Tile Production

The tiles were produced and tested at the TETO tile industry in the municipality of Gurupi, state of Tocantins, Brazil. The materials used in tile production included: Portland cement type II (CP II-E), washed sand as fine aggregate, potable water, a synthetic plasticizing additive (lignosulfonate), and the natural additives powdered tannin (Acacia spp.) and kraft lignin (Eucalyptus spp.). TETO itself supplied the cement, sand, and synthetic additive. At the same time, a company in Montenegro, Rio Grande do Sul, donated the tannin, and the lignin was donated by a pulp and paper company in Limeira, São Paulo.

The tiles were produced following the recommendations of standard NBR 13858-2 (ABNT, 2009), using a mortar mix proportion, referred to as "trace," of 1:3:0.45 for cement, fine aggregate (washed sand), and potable water, respectively. The synthetic additive is used in the industry's standard mix at a proportion of 2.5% of the cement weight. In this study, the synthetic additive was partially or completely replaced by the natural additives lignin and tannin in each treatment.

Six treatments and one control were established, each using a mix consisting of 50 kg of cement, 150 kg of fine aggregate, and 22.5 liters of water. The mix resulted in 75 concrete tiles per treatment, for a total of 525 tiles. The dosage of 2.5% additive was maintained in treatments T1, T2, T4, T5, and the control, while treatments T3 and T6 received a double dosage (5%) to test tile efficiency.

The dimensions of the tiles were also set according to the standard and standardized by the company, with 50 cm in length, 33 cm in total width, 30 cm of effective width, 50 mm profile depth, approximately 5 kg of weight, and a permissible weight of 40 kg/m².

The tile production process begins with mixing the mortar in an industrial mixer, with all components added to achieve the most uniform incorporation possible: cement first, then washed sand, water, and finally the additive. The resulting mortar is deposited onto a conveyor belt that carries it to a dosing hopper, gradually feeding it into the extruder. Simultaneously, aluminum molds move along a dedicated conveyor, passing through the extruder, where a force of 180 KgF is applied to shape the mortar.

The tiles emerge from the extruder as a continuous bar and are cut by a knife to the mold size, every 50 cm. The molds are manually removed and placed on shelves in a mobile rack, which is then transferred to a drying kiln for 24 hours at an average temperature of 60°C and relative humidity around 50%.

After this period, the tiles are removed from the kiln and shelves, demolded, and placed in curing bays for final curing.

Tests for Determining Tile Properties

The tests to determine the concrete tiles' visual, physical, and mechanical properties were based on standard NBR 13858-2 (ABNT, 2009), which defines the requirements and test methods.

At seven days of curing, 12 tiles were randomly selected from each treatment: six for absorption and dry weight tests, and six for the flexural rupture load test. For the 28-day curing period, 18 tiles were selected from each treatment, distributed as follows: six for flexural rupture load testing, six for absorption and dry weight, and six for warping and GAP tests (the clearance between the lower and upper faces of the highest wave crests of two overlapped tiles).

In total, 210 tiles were tested, covering the different treatments and curing periods. Additionally, all produced tiles were visually inspected to check conformity with the established standards.

Visual Aspects

Standard NBR 13858-2 (ABNT, 2009) states in item 4 that concrete tiles must not present defects that compromise their quality, such as cracks, bubbles, faults, disaggregation, or breakage. Furthermore, item 5.5 specifies that up to 2% breakage during tile transport does not invalidate batch acceptance. Thus, tile loss due to defects was monitored by counting the number of tiles lost during demolding and handling at the seven- and 28-day periods.

Dry Weight

The dry weight of the tiles was determined according to NBR 13858-2 (ABNT, 2009). Initially, the tiles were submerged in potable water for 24 hours. They were then placed in an oven at 105 ± 5°C until reaching a constant weight and subsequently weighed on a digital scale to obtain the dry weight in kilograms. The maximum allowed value is 5.0 kg per tile or 40 kg/m².

Warping

Warping was assessed in tiles after 28 days of curing. Each tile was placed on a perfectly flat horizontal surface with the underside facing downward. The greatest deviation from the plane was measured using a feeler gauge. According to NBR 13858-2 (ABNT, 2009), this deviation must not exceed 1.5 mm.

Tile GAP

The GAP determination test was conducted on tiles with 28 days of curing by measuring the clearance between the upper and lower faces of the highest wave crests of two overlapped tiles. This clearance must be less than or equal to 6 mm. However, creating an air layer that prevents water absorption by capillarity is necessary, thereby allowing ventilation between tiles, as per NBR 13858-2 (ABNT, 2009).

Water Absorption

The water absorption test was performed in accordance with NBR 13858-2 (ABNT, 2009). The tiles were immersed in a reservoir with potable water for 24 hours. After immersion, surface water was carefully removed, and the tiles were weighed on a digital scale (10 kg capacity) to record their wet mass. Subsequently, the tiles were placed in an oven at 105 ± 5°C until reaching constant weight and were weighed again to determine the dry mass. Water absorption was calculated based on the wet-to-dry mass ratio, adhering to the standard's maximum limit of 10%.

Flexural Rupture Load

A hydraulic press with a 10-ton capacity and a 1000 KgF load cell was used to determine the flexural rupture load. The test followed the procedures outlined in NBR 13858-2 (ABNT, 2009). First, the tiles were immersed in water for 24 hours. Subsequently, each tile was placed individually on two fixed support bars of the press, with the specimen centered transversely beneath an articulated steel bar. The load was then applied progressively, without shocks, and the rupture load values were recorded.

The standard classifies tile strength based on the profile depth. For this study, the tiles produced had a standard depth of 50 mm, classifying them as Class "A," which requires a minimum rupture load of 240 KgF.

Data Analysis

The experiment followed a completely randomized design (CRD) with a 6 × 2 factorial scheme (five additive treatments and one control, across two curing periods), excluding treatment T5, which was disqualified due to total tile loss. Initially, the Shapiro-Wilk normality test was applied, and upon confirming data normality, an analysis of variance (ANOVA) was conducted to determine significant differences among treatments. Tukey's test was applied for mean comparisons at a 5% probability level when significant differences were observed, using Excel and Sisvar software.

RESULTS

Visual Aspects

For the parameter concerning the visual aspects of the tiles, the most recurring defects were breakage and disintegration during demolding and handling, particularly in treatments T4, T5, and T6 (Figure 1). Table 1 presents the percentage of tiles lost due to these defects.

Figure 1. Adhesion to form (A), disaggregation (B), and breakage of the tiles (C).
Figura 1. Adesão à forma (A), desagregação (B) e quebra dos ladrilhos (C).

Table 1. Defects occasioned by the unmolding and handling of tiles.
Tabela 1. Defeitos ocasionados pela desmoldagem e manuseio de ladrilhos.

Treatments	N° of tiles produced	Adhesion to mold	Disintegration	Breakage	N° of tiles lost	Losses (%)
T1 - Lignin/Synthetic additive	75	0	0	25	25	33,33
T2 - Lignin 2.5%	75	0	0	8	8	10,66
T3 - Lignin 5%	75	0	0	14	14	18,66
T4 - Tannin/Synthetic additive	75	15	26	12	53	71,00
T5 - Tannin 2.5%	75	20	40	15	75	100,00
T6 - Tannin 5%	75	14	13	9	39	52,00
Control	75	0	0	6	6	5,30

According to NBR 13858-2 (ABNT, 2009), a breakage rate of up to 10% during tile transportation does not disqualify the batch's quality. In this regard, only the control sample met the standard (5.30%).

Among the natural additives, treatment T2, with a loss rate of 10.66%, was closest to the acceptable threshold and can be considered satisfactory, followed by treatment T3, with 18.66%. Treatments T1, T4, T5, and T6 exhibited significantly higher loss rates, with treatment T5 standing out at 100% loss. Thus, five out of the six treatments were disqualified concerning the visual aspects requirement.

Dry Weight

Table 2 shows the means determined by the Tukey test for the tiles' dry weight (kg) according to the curing period and the additives used. There was no significant difference between the means regarding the interaction of factors or the curing period. However, a significant difference at the 5% probability level was observed for the treatments involving additives. Treatment T4 exhibited the lowest dry weight (4.60 kg), while treatment T3 and the control sample showed the highest averages (5.03 kg).

Table 2. Average values, Tukey test, and Weight Variation Coefficient (Kg) of the tiles.
Tabela 2. Valores médios, teste de Tukey e Coeficiente de Variação de Peso (Kg) dos ladrilhos.

Curing period (days)	Dry weight (Kg)	Pr > Fc
7	4,86 a (3,27)	ns
28	4,88 a (4,46)	
Additives	**Dry weight (Kg)**	**Pr > Fc**
T1 - Lignin 1.25% + Synthetic additive 1.25%	4,84 b (1,42)	*
T2 - Lignin 2.5%	4,87 b (0,76)	
T3 - Lignin 5%	5,03 a (2,43)	
T4 - Tannin 1.25% + Synthetic additive 1.25%	4,60 c (1,43)	
T6 - Tannin 5%	4,86 b (1,11)	
Control - Synthetic additive 2.5%	5,03 a (1,16)	
Interaction	**Dry weight (Kg)**	**Pr > Fc**
Curing period × Additives	-	ns

Note: ^{ns}: not significant at 5% probability; * = significant at 5% probability. Means followed by the same lowercase letter in the column do not differ statistically according to the Tukey test. Values in parentheses correspond to standard deviation and coefficient of variation (%), respectively.

Warping

Regarding warping, all treatments, including the control sample, complied with the requirements established by NBR 13858-2 (ABNT, 2009), with the distance from the farthest point of the tile to the reference plane not exceeding 1.5 mm.

Tile Gap

The average tile gap values (Table 3) were also within the limits of NBR 13858-2 (ABNT, 2009), which specifies a maximum allowable gap of 6 mm. All treatments and the control sample exhibited gap values ranging from 3 mm to 4.2 mm, except for T5, which presented a 100% loss rate.

Table 3. GAP between concrete tiles.
Tabela 3. GAP entre telhas de concreto.

Treatments	Average GAP* (mm)	Standard Deviation	Coefficient of Variation (%)
T1 - Lignin/Synthetic additive	4,20	0,00	0,00
T2 - Lignin 2.5%	3,22	0,29	8,87
T3 - Lignin 5%	3,00	0,00	0,00
T4 - Tannin/Synthetic additive	3,00	0,00	0,00
T6 - Tannin 5%	3,00	0,00	0,00
Control	3,00	0,00	0,00

Note: * GAP refers to the clearance between the lower and upper faces of the crests of the highest waves of two overlapped tiles.

Water Absorption

Table 4 presents the mean water absorption values of the tiles per treatment and for the control sample. An interaction between treatments and curing periods was observed, where only treatment T4 failed to meet the NBR 13858-2 (ABNT, 2009) specification of a maximum 10% water absorption for both curing periods, with averages of 11.17% at 7 days and 10.64% at 28 days.

Table 4. Average values, Tukey test, interaction between factors, and coefficient of variation of Absorption (%) of the tiles.

Tabela 4. Valores médios, teste de Tukey, interação entre fatores e coeficiente de variação de absorção (%) dos ladrilhos.

Absorption (%)		
Treatments	Curing period (days)	
	7	28
T1 - Lignin/Synthetic additive	6,78 cA (4,75)	6,51 bA (5,85)
T2 - Lignin 2.5%	5,47 dA (21,76)	4,77 cB (2,87)
T3 - Lignin 5%	6,45 cB (6,30)	7,75 bA (6,61)
T4 - Tannin/Synthetic additive	11,17 aA (6,19)	10,64 aA (7,92)
T6 - Tannin 5%	8,01 bB (14,82)	9,46 aA (0,25)
Control	5,67 d A (9,27)	3,77 cB (15,32)

Note: ^{ns}: not significant at 5% probability; * = significant at 5% probability. Means followed by the same lowercase letter in the column and the same uppercase letter in the row do not differ statistically according to the Tukey test. Values in parentheses correspond to standard deviation and coefficient of variation (%).

The control sample exhibited the lowest absorption averages for both curing periods (5.67% and 3.77%, respectively), followed by treatment T2, with 5.47% at 7 days and 4.77% at 28 days. Among the treatments with natural additives, the treatment with 2.5% lignin again stood out as the most efficient.

Flexural Strength (Breaking Load)

Table 5 shows the behavior of tile resistance to flexural breaking load for the treatments with natural additives and the control sample.

Table 5. Average values, Tukey test, interaction between factors, and coefficient of variation of the flexural failure load (KgF) applied to the tiles.

Tabela 5. Valores médios, teste de Tukey, interação entre fatores e coeficiente de variação da carga de ruptura por flexão (KgF) aplicada às telhas.

Flexural failure load (Kgf)		
Treatments	Curing period (days)	
	7	28
T1 - Lignin/Synthetic additive	227,13 abB (3,56)	274,60 bA (9,03)
T2 - Lignin 2.5%	241,06 aB (1,90)	280,28 bA (2,62)
T3 - Lignin 5%	200,06 bB (12,43)	225,56 cA (14,63)

Flexural failure load (Kgf)		
Treatments	**Curing period (days)**	
	7	**28**
T4 - Tannin/Synthetic additive	28,05 cA (17,59)	31,77 dA (12,84)
T6 - Tannin 5%	25,70 cB (20,4)	52,56 dA (19,33)
Control	247,33 aB (6,86)	313,63 aA (6,87)

Note: ^{ns}: not significant at 5% probability; * = significant at 5% probability. Means followed by the same lowercase letter in the column and the same uppercase letter in the row do not differ statistically according to the Tukey test. Values in parentheses correspond to standard deviation and coefficient of variation (%).

For this parameter, an interaction between treatment and curing period was also observed, with tile resistance increasing over time but decreasing with higher dosages of natural additives, especially for treatments T3, T4, and T6. The control sample exhibited the highest resistance averages at 7 and 28 days, followed by treatment T2 (2.5% lignin). Both met the NBR 13858-2 (ABNT, 2009) requirements already at 7 days of curing.

The treatments most negatively affected by adding natural additives were those with tannin in their composition (T4 and T6). Breaking load values were significantly below the recommended standard for the produced tile profile (50 mm; 240 KgF), ranging between 31.77 and 52.56 KgF at 28 days of curing.

For the treatments incorporating lignin (T1, T2, and T3), only T3 failed to meet the standard at 7 and 28 days, with mean values of 200.06 KgF and 225.56 KgF, respectively. Treatment T1 met the standard at 28 days of curing (274.6 KgF), and treatment T2 showed the best performance at both 7 and 28 days, with resistance values above the recommended level: 241.06 KgF and 280.28 KgF, respectively.

Table 6 presents the analysis of variance for the additive treatments and curing periods, the interaction between the two factors, and the coefficient of variation for the parameters dry weight (kg), absorption (%), and flexural breaking load ($F_{\text{máx,}}$ KgF).

Table 6. Analysis of variance coefficient of variation of statistical factorial analysis.
Tabela 6. Análise do coeficiente de variância da análise fatorial estatística.

ANOVA	**Pr>Fc**		
FV	**Dry weight (Kg)**	**Absorption (%)**	**$F_{\text{máx}}$ (Kgf)**
Additives	0,0000 *	0,000 *	0,0000*
Curing period	0,3389 ^{ns}	0,5277 ^{ns}	0,000*
Additives × Curing period	0,5636 ^{ns}	0,000 *	0,0038*
CV (%)	1,51	9,24	9,39

Note: ^{ns}: not significant at 5% probability; *: significant at 5% probability; CV (%): Coefficient of variation.

Table 7 shows the correlation between dry weight, flexural strength, and absorption during the 7- and 28-day curing periods.

Table 7. Pearson correlation between the parameters analyzed for the tiles after 7 and 28 days of curing.
Tabela 7. Correlação de Pearson entre os parâmetros analisados para os ladrilhos após 7 e 28 dias de cura.

7 days of curing			
Parameters	**Dry weight (Kg)**	**Flexural failure load (KgF)**	**Absorption (%)**
Dry weight (Kg)	1		
Flexural failure load (Kgf)	0,5332	1	
Absorption (%)	-0,7049	- 0,7731	1

28 days of curing			
Parameters	**Dry weight (Kg)**	**Flexural failure load (KgF)**	**Absorption (%)**
Dry weight (Kg)	1		
Flexural failure load (KgF)	0,7126	1	
Absorption (%)	-0,6653	-0,9159	1

At 7 days of curing, the correlation between flexural breaking load and dry weight of the concrete tiles was moderate and positive, indicating that heavier tiles generally exhibited higher flexural strength. Conversely, the correlation between absorption and dry weight was strong and negative, demonstrating a pronounced inverse relationship: the lower the tile's dry weight, the higher its water absorption rate.

Moreover, the correlation between absorption and flexural breaking load was strong and negative at 7 days of curing, revealing a significant inverse relationship: the lower the water absorption, the greater the flexural strength.

At 28 days of curing, the correlation between flexural breaking load and dry weight remained strong and positive, maintaining the trend observed at 7 days: heavier tiles presented greater flexural strength.

Similarly, the correlation between absorption and dry weight remained strong and negative at 28 days, continuing the inverse relationship: lower dry weight was associated with higher water absorption.

The correlation between absorption and flexural breaking load also remained strong and negative at 28 days, confirming that lower water absorption corresponds to higher resistance to flexural breaking load.

DISCUSSION

The results presented in Table 1 demonstrate that using tannin as a natural additive for the production of concrete roof tiles proved inefficient, with treatment T5 resulting in total loss among the treatments evaluated. One explanation for the negative behavior of tannin in concrete tiles is its low moisture resistance, caused by the early formation of methylene bridges between long and rigid flavonoid polymers. These bridges immobilize the bond between cement and tannin (steric hindrances), rendering it ineffective, as the distances involved in the chain and the viscosity prevent the formation of additional methylene bridges, leading to few cross-links and, consequently, weak bonds (Fechtal & Riedl, 1993).

Another hypothesis proposed by Tostes *et al.* (2004), in a study involving tannin addition to wood panels, is its high potential for water absorption. The authors observed that the greater the amount of tannin added to the panels, the higher the water absorption rate, weakening particle bonding and thus reducing mechanical strength. Since cement depends on water to react and form strong bonds necessary for concrete formation, it can be inferred that tannin excessively consumes the water in the mix used for tile production, thereby affecting the water-to-cement ratio and impairing the essential reactions required for the curing process of the tiles.

Powdered Kraft lignin proved effective in various aspects; however, regarding the parameter of visual aspects, the treatments did not fully meet the standard’s requirements. Nevertheless, treatment T2 presented an average close to the acceptable limit, suggesting opportunities for further studies with lower dosages than 2.5%.

The results obtained for the dry weight of the tiles (Table 2) were within the recommendations of NBR 13858-2 (ABNT, 2009), which stipulates a maximum dry weight per tile of 5.0 kg or 40 kg/m². It is extremely important that these values do not exceed the allowed limit, particularly per square meter, as this could lead to additional construction costs due to the need to reinforce the roof structure and, most importantly, could compromise roof safety. In a study conducted by Damasceno *et al.* (2015) on concrete tiles manufactured with different insulating materials, the authors also found values in compliance with the standard, ranging between 4.5 and 5.0 kg.

All treatments were satisfactory regarding warping (values below 1.5 mm) and tile gap (maximum gap of 4.2 mm for treatment T1), all remaining within the recommended limits established by NBR 13858-2 (ABNT, 2009).

Table 4 reveals a trend of reduced water absorption rates with longer curing periods (from seven to 28 days). However, with increased dosages of natural additives in the tile mix (from 2.5% to 5.0%), the opposite occurred for both lignin and tannin, meaning that absorption rates increased at 28 days of curing. This indicates that the optimal dosage for controlling water absorption in the tiles is 2.5% of the natural additive.

In a study conducted by Dantas *et al.* (2020) on the performance of mortars with partial substitution of cement and additives by lignin, the authors observed average absorption rates close to 5% when using 2.5% natural lignin at 30 days of curing, results similar to those achieved by treatment T2.

These findings are significant because water absorption in tiles is related to the material's porosity, which can directly influence the quality of the roofing by causing water infiltration (Silveira *et al.* , 2018). This indicates that lignin did not affect the tiles' porosity at any curing period.

The flexural strength property of the tiles must meet the standard value established by NBR 13858-2 (ABNT, 2009), as the tiles must support handling, transportation, and roof assembly (be self-supporting), as well as maintenance loads and loads related to environmental conditions (e.g., water weight, falling branches), ensuring roof safety. Alongside water absorption, flexural strength is considered the most important property to be evaluated.

CONCLUSION

- The research demonstrated that replacing synthetic additives with lignin and tannin yielded distinct results in the performance of concrete roof tiles. Lignin, particularly at a 2.5% dosage (T2), showed promise as a natural additive, meeting the primary normative requirements related to flexural strength and water absorption at seven and 28 days of curing, while exhibiting a positive evolution of these properties over time.
- Despite a slight inadequacy regarding visual aspects, treatment T2 with lignin was considered satisfactory. On the other hand, treatments involving tannin did not meet the normative parameters regarding flexural strength and visual aspects, even in the best case observed for treatment T5.
- Thus, it can be concluded that lignin extracted from *Eucalyptus* shows potential as a sustainable substitute for synthetic additives in concrete tile production. In contrast, tannin extracted from *Acacia* did not perform satisfactorily under the conditions tested.

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