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COMPATIBILITY BETWEEN NATURAL AGRO-INDUSTRIAL BY-PRODUCTS AND SYNTHETIC MATERIALS, A BASIC ELEMENT IN OBTAINING BIOCOMPOSITE MATERIALS WITH POTENTIAL FOR USE IN CONSTRUCTION

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Abstract

Agriculture is one of the main sources generating significant quantities of natural agro-industrial by-products, such as stalks, fibrous residues, husks and cobs, etc. Based on the principles of the circular economy, such natural materials can have numerous possibilities for superior valorization, often as biocomposite materials, in various fields, not least in construction. Starting from the recognized potential for valorization of natural agro-industrial by-products in this sector, the paper presents aspects regarding the importance of compatibility and interaction between two natural materials, namely rice husks and industrial hemp fibers, and a synthetic resin as their binder. The implications of the compatibility and interactions between such types of materials and how this can influence the evolution over time of some characteristics of the biocomposite material expected to be used in the construction field as coatings are also presented.

Key words: *adhesion, coatings, hemp, pre-reaction time, rice husks.*

INTRODUCTION

It is currently known that organic raw materials can be used in a sustainable way, with increased efficiency and lower processing costs. The reducing waste, reusing and maximizing the value of resources can generate valuable products, promoting sustainable development and maximizing ecological and socio-economic benefits (Coelho Vianna et al., 2024).

The agricultural sector is one of the main sectors generating impressive amounts of waste (Ufitikirezi et al., 2024; Capanoglu et al., 2022; Pradhan et al., 2024), which has led to concern among public opinion and the scientific community.

Data published by the Food and Agriculture Organization (FAO) shows that agriculture produces over 140 billion tons of biomass each year, with over 2 tons per day in some rural areas (Ufitikirezi et al., 2024). It is also recognized that, by valorizing agricultural waste, farmers and agricultural industries can contribute through sustainable practices to resource efficiency and the development of the circular economy (Ufitikirezi et al., 2024).

Agricultural wastes, such as straw, stalks, fibrous residues, husks and cobs, constitute a significant part of the waste that can be recovered through various techniques, being able to be transformed into fuels and value-added materials (Ufitikirezi et al., 2024; Capanoglu et al., 2022; Wazed Ali et al., 2022). Through such an approach, the circular economy aims to minimize the quantities of waste generated, by optimizing their superior use, through viable processes, in order to increase their value and establish circular practices in the agri-food sector.

At the same time, it promotes the conservation and economic value of products, trying to extend the maximum lifespan of their components and obtain the highest possible added value (Coelho Vianna et al., 2024).

In the spirit of the principles of the circular economy, within the experimental research presented in this paper, the aim is to obtain innovative construction materials in the form of covering products. In order to achieve higher added value, will be integrated two natural local agro-industrial by-products, namely rice husks - plant material resulting from the food industry, and industrial hemp - in the form of yarn, resulting from the use of seeds in the food sector, in the textile industry, etc.

Rice, one of the staple foods for almost half of the world's population, is cultivated on all continents, in over 100 countries (Pradhan et al., 2024), resulting in one of the abundant natural resources, namely rice husks.

However, this natural by-product is usually abandoned in a post-harvest area, resulting in an average of 2 million tons accumulated per year (Raki-in et al., 2021), a quantity that, if stored uncontrolled, seriously affects the environment (Raki-in et al., 2021; Milawarni et al., 2023; Pradhan et al., 2024).

The construction industry can promote sustainable development and minimize this type of natural agro-industrial by-product considered as waste, by incorporating rice husks into various construction products.

For example, embedded into bricks and blocks as a lightweight aggregate, rice husks lead to improved thermal properties and reduced overall weight (Selvaranjan et al., 2021).

Also, using this natural material as a filler or binder in the production of insulation materials, such as panels, boards or filler insulation, improves the thermal and acoustic insulation properties of the resulting products (Muthuraj et al., 2019; Sembiring et al., 2016; Aravind et al., 2020).

In other cases, combining rice husks with other lignocellulosic materials and binders has resulted in particleboards and fiberboards, which can be used in various construction applications, such as flooring, wall panels and furniture components (Manatura et al., 2023). Moreover, the integrating of these husks into a matrix of natural or synthetic polymers, as a reinforcing material, results in biocomposites that can be used in a number of other construction applications, such as decking, cladding and interior finishes (Faruk et al., 2012; Wang et al., 2020).

As for hemp, especially in the form of hemp pulp, it is currently being exploited through integration into numerous types of building materials, with the recognized role of improving both the thermal insulation properties and acoustic performance of the resulting building product, as well as the humidity and durability of the environment with which it comes into contact.

Among the numerous hemp-containing materials, for example, a few products recognized as being used/usable in construction are mentioned, namely: Hempcrete, a lightweight material obtained by mixing hemp powder with a thermally insulating lime-based binder, the resulting mixture being able to be used as a non-load-bearing filling material in walls, floors and roofs, with very good thermal and acoustic insulation and the ability to regulate humidity levels (Arrigoni et al., 2017; Demir et al., 2020; Piot et al., 2017; Yadav et. al, 2022); insulation materials in the form of panels, sticks or filling insulation (Abdellatef et al., 2020), with good thermal and acoustic insulation properties; boards usable as floors, wall panels and even furniture components, made by mixing hemp pulp with other lignocellulosic materials and binders; floors, cladding elements and interior finishes, practically biocomposite materials resulting from combining hemp pulp with a matrix of synthetic or natural polymers (Cigasova et al., 2015; Dhakal et al., 2015; Requile et al., 2019); hemp concrete which, although it does not have high strength or portability characteristics, has low thermal conductivity, high water vapor permeability, fire resistance, biodegradation, good acoustic properties and therefore can be used for walls, roofs and floors, for insulating walls on their outer side (Gołębiewski, 2017).

All these examples regarding the creation of biocomposite products usable/used in the construction field and the study of their characteristics clearly demonstrate, at an international level, the interest of researchers in obtaining, studying and promoting for use new construction materials that contain natural agro-industrial by-products, with properties comparable to those of traditional materials, made on a synthetic basis.

MATERIALS AND METHODS

In this work, rice husks (Figure 1) and hemp threads (Figure 2) were used, natural agro-industrial by-products provided by local producers.

Figure 1. Rice husks

Figure 2. Hemp fiber

The experimental research aimed at the superior valorization of these natural materials by obtaining innovative materials with ecological characteristics/coating products, with potential for use in the construction field.

Two series of innovative products were obtained and studied, namely: Series 1 - products with added rice husk content and Series 2 - products with mixed content of rice husk and hemp fibers.

In Series 1, two innovative basic products were first obtained, by embedding rice husks in an acrylic binder.

From a compositional point of view, these biocomposite materials were designed in such a way as to mainly study the influence of the constituent elements - the synthetic binder and the vegetable component. Two variants of acrylic resins were used as binder, denoted by R1 - acrylic base for interior/exterior coatings, respectively R2 - silicone acrylic base for interior/exterior coatings, the mass ratio of mixing rice husks: binder being 1:10, resulting in the first stage the products SP1RI, SP1R2.

In addition to the influence that compositional factors have on the main characteristics of the products, it was also aimed to study the influence that the method of preparing the plant material before application would have in this regard. For this purpose, a pre-reaction time was introduced, during which the binder and natural components were maintained in contact, before the application of the biocomposite.

The notion of pre-reaction time, specific to two-component anti-corrosion epoxy protections, is the time in which, before being put into operation, the two components are mixed, homogenized and left to interact, the lack or non-compliance with this time affecting the characteristics and performance of the anti-corrosion protection. Starting from this principle, in the case of coatings made from innovative products that integrated the rice husk, a pre-reaction time of 30 minutes was established for the rice husk-binder mixture. Thus, starting from the initial products, SP1R1 and SP1R2, without pre-reaction time, the corresponding products, SP1R1/30, SP1R2/30, were made, for which the pre-reaction time of 30 minutes was considered.

In order to determine the influence that the nature of the embedded agro-industrial by-products has on the characteristics of the resulting bilayer coatings, additional biocomposites with mixed plant content were made and tested.

Series 2 of innovative materials was made respecting, in principle, the same approach as in the case of the products in the Series 1.

This time, four basic innovative products with mixed plant content were initially obtained from rice husks and hemp threads of an average length of approx. 1 cm, embedded in the same types of binders, namely resin R1 and R2, respectively. The length at which the hemp threads were cut, and the content of this natural component were conditioned by obtaining a homogeneous biocomposite material, with good workability when applied to a concrete substrate. The mixing ratios of the hemp:rice husks:binder components were 1:2.5:40, resulting in the products marked SP1R1T and SP1R2T, and 1:3:40, respectively, for the products marked SP2R1T and SP2R2T.

As in the case of the innovative products in Series 1, for the products in Series 2, the influence of the 30-minute pre-reaction time between the synthetic binder and the mixture of the two natural agro-industrial by-products was studied. This resulted in four other new recipes of biocomposite materials, denoted as follows: SP1R1T/30, SP1R2T/30, SP2R1T/30 and SP2R2T/30.

It is worth emphasizing that, from the beginning, for each product obtained in the two series, there was compatibility between the synthetic and vegetable components, resulting in homogeneous materials with good workability. As a result, each of the twelve biocomposites was applied to concrete support surfaces, in two layers, with an interval of 24 hours between them, the coatings being applied and subsequently maintained under standard laboratory atmospheric conditions, namely at a temperature of 23 ± 2°C and a relative air humidity of 50 ± 5%.

At this stage of the experimental research, the innovative products obtained were characterized by determining the average total thickness, and adhesion to concrete, a first evaluation of the resulting coatings, from the point of view of the potential for use in construction. Considering the organic, acrylic nature of the R1 and R2 binders used, as well as the average values of the thicknesses of the resulting bilayer coatings, ranging between 3.26 and 6.43 mm, without taking into account at this stage the influence of the pre-reaction time on the total thickness of the coating, the determinations were carried out referring to the condition specified in the technical specification SR EN 15824 - "Specifications for external renders and internal plasters based on organic binders", and the test conditions were those provided for in SR EN 1542 - "Products and systems for the protection and repair of concrete structures. Test methods. Measurement of bond strength by pull-off".

Referring to these documentations, the resulting coatings being atypical in terms of component materials, it was considered appropriate to monitor the evolution of the adhesion values to concrete for 56 days, establishing the measurements to be carried out at 7, 28 and 56 days after application. Since for each series of products obtained, the appearance of the bilayer coatings is very similar, regardless of the resin used, the absence or presence of pre-reaction time, in the following, exemplary images of the samples made will be presented.

Thus, for the products in the Series 1, the appearance of the SP1R2/30 coating is presented as an example, with the addition of rice husks and pre-reaction time, as well as the way the coating breaks when performing adhesion by pulling, after 7 days from the application (Figure 3), respectively after 28 days from the application (Figure 4).

For the coatings in Series 1, without analyzing for the moment the influence of the pre-reaction time, the average thickness values varied between 3.26 mm and 6.43 mm, and the average adhesion values to the concrete substrate, over the 56 days, between 0.22 MPa and 0.36 MPa (Figure 5).

Figure 3. Appearance of the SP1R2/30 coating containing rice husks and the way it breaks when pulled off, for determining adhesion at 7 days

Figure 4. Appearance of the SP1R2/30 coating containing rice husks and the way it breaks when pulled off, for determining adhesion at 28 days

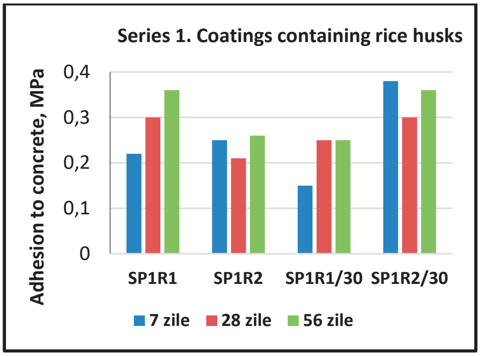

Figure 5. Variation of the adhesion to concrete for the two-layer coatings of Series 1

Given the diversity of materials used to obtain products from the Series 2 and, as a result, the possibility of particular manifestations of these coatings over time, adhesion to the substrate was monitored at shorter time intervals compared to previous cases, namely at 7, 28, 42 and 56 days after the application of the two-layer coatings on concrete substrate surfaces. Also in Series 2, regardless of the resin used, the absence or presence of pre-reaction time, the coatings obtained had a very similar external appearance. As a result, the appearance of the SP1R1T coating, having the hemp: rice husks: binder ratio of 1: 2.5: 40 (Figure 6), respectively of the SP2R1T/30 coating, characterized by the hemp: rice husks: binder ratio of 1:2.5:40 (Figure 7), are presented below as examples, as well as the way the coatings break when performing adhesion by pulling, 28 days after application.

Figure 6. Appearance of the SP1R1T coating, containing rice husks and hemp fibers, and the way it breaks when pulled off, for determining adhesion at 28 days

Figure 7. Appearance of the SP2R1T/30 coating, containing rice husks and hemp fibers, and the way it breaks when pulled off, for determining adhesion at 28 days

For the coatings in Series 2, without analyzing at this time the influence of the pre-reaction time, the average values of the thickness of the bilayer coatings containing rice husks and hemp fibers varied between 3.81 mm and 5.09 mm, and the average values of the adhesions to the concrete support, during the 56 days, between 0.35 MPa and 0.39 MPa (Figure 8).

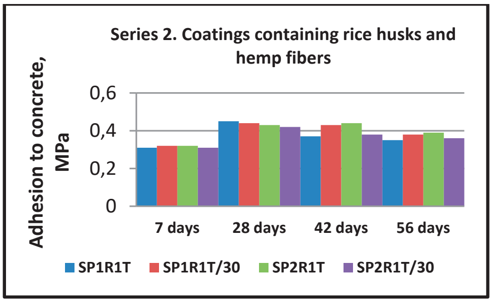

Figure 8. Variation of the adhesion to concrete for the two-layer coatings of Series 2

RESULTS AND DISCUSSIONS

In the experimental research presented in this paper, was mainly studied the possibility of obtaining biocomposite materials, by integrating, in acrylic binders, rice husks or/and mixtures in variable proportions of rice husks and hemp fibers, from the perspective of material compatibility.
At the same time, the evaluation of the potential for use in construction of the obtained materials, in the form of plaster-type coatings, was pursued. The extent to which the adhesion of the resulting coatings to concrete was influenced by

the type and content of the two natural materials, by their application conditions, meaning by the pre-reaction time between the main components, was also studied.

A first response considered favorable under the mentioned aspects was the existence of compatibility between the specified natural and synthetic materials, resulting in biocomposites containing rice husks (SP1R1, SP1R2, SP1R1/30 and SP1R2/30), respectively in those with mixed rice husks and hemp threads (SP1R1T, SP2R1T, SP1R1T/30 and SP2R1T/30). The establishment of the usability of the eight innovative product variants was achieved by analyzing and correlating the experimental results obtained after determining the average total thickness and the adhesion to concrete of each bilayer coating.

Thus, a further positive outcome was the formation of two-layer, rustic-style plaster coatings after applying both categories of biocomposites onto concrete surfaces. The coatings containing rice husks achieved average total thicknesses ranging from 3.26 mm to 6.43 mm, while those with mixed plant content exhibited thicknesses between 3.81 mm and 5.09 mm.

The third favorable aspect was that regarding the potential for use in construction of this category of innovative plaster-type coatings, given the good adhesion to the concrete support surface. This statement is supported by the fact that, at the end of the monitoring period of the coatings, after 56 days from application, the condition specified in the technical specification SR EN 15824 was met, according to which the adhesion of a plaster with an organic binder must be greater than or at least equal to 0.3 MPa.

In this context, from the point of view of adhesion to the support, we consider that six of the eight variants of the innovative coatings obtained have a potential for use in construction, however, additional research is necessary to achieve an experimentally substantiated, detailed and multidisciplinary characterization of the respective coatings. This potential for use as a multidisciplinary coating is suggested by the specific characteristics that each of the two integrated natural agro-industrial by-products has, implicitly by the properties that they are likely to confer on the products, respectively the plasters in which they are included.

All these favorable and encouraging aspects being specified, it is interesting and useful to highlight a series of specific aspects observed regarding the effects of integrating hemp fibers into the composition of such innovative materials.

Reference is made to a series of findings, resulting from the comparison of biocomposites/coatings containing only rice husks vs. those with mixed content, of rice husks and hemp threads.

The first finding is the existence of a good interaction between plant materials and the synthetic binder, with the obtaining of biocomposites with good workability. This allowed their application in two layers on concrete support surfaces. The resulting average adhesion values, at 28 days after application, generally had an upward trend in all eight coatings, those with mixed plant content having higher adhesion than those of coatings containing only rice husks. All the favorable aspects were highlighted during the determination of the adhesion by pulling, in most cases, through an adhesive rupture mode, between the coating and the concrete, which indicated a cohesive character of the coating, with a potential for continued increase in adhesion to the substrate over time.

However, by the end of the monitoring of the adhesion to the substrate for the two series of coatings, 56 days after their application, a slow but clearly different evolution of this characteristic was observed. Namely, if in the case of the coatings in Series 1, containing exclusively rice husks, the adhesions had an upward trend until the end of the analyzed period, in the case of the coatings in Series 2, in which hemp threads were also introduced, the trend was slightly downward, for all four coatings.

Even in this context, the final values of the average adhesions to concrete were higher for coatings with mixed vegetal content, values that even though in a decreasing trend, were above the limit value of 0.3 MPa.

We consider that such an evolution of the adhesion values was generated by the synergistic effect of two aspects: firstly, by the compositional difference, by the addition of hemp threads, and secondly, by the introduction of the pre-reaction time. Comparing the experimental results obtained on the coatings from the two series, it is found that the effects of the second factor were highlighted both by the increase in the average thickness values of the coatings, and by the average values of the adhesions to the support, during the 56 days. We consider it also useful to point out that the cumulative effect of the two mentioned factors was more pronounced in the case of coatings containing hemp, which underlines the importance of this type of agro-industrial by-product in ensuring compatibility and maintaining good interaction between the natural and synthetic components of this type of innovative coating.

CONCLUSIONS

The paper presents experimental research that had as its main purpose the study of the possibility of obtaining biocomposite materials, by integrating, in a synthetic binder, two local natural agro-industrial by-products, namely rice husks or/and hemp fibres, from the perspective of their compatibility and interaction, as well as the possibility of using the obtained materials in construction.

Eight innovative products were obtained and studied: four products with rice husk content, and four products with mixed content of rice husk and hemp fibers.

In addition to the influence that compositional factors have on the main characteristics of the coatings, it was also aimed to study the influence that the method of preparing the plant material before application would have in this regard. For this purpose, a pre-reaction time of 30 minutes was introduced, during which the binder and natural components were maintained in contact, before application of the resulting biocomposites.

From the point of view of adhesion to the concrete support, we consider that six of the eight variants of the innovative coatings obtained have a potential for use in construction. However, additional research is necessary to achieve an experimentally detailed and multidisciplinary characterization of the respective coatings. After applying each biocomposite–on concrete surfaces, two-layer plaster-type coatings with a rustic appearance were obtained.

At the end of monitoring the adhesion of the coatings to the concrete substrate, it was observed that in the case of those containing exclusively rice husks, the adhesions had an upward trend, and in the case of those in which hemp fibers were also introduced, the trend was slightly downward. Even so, the values fell within the adhesion limits provided by the specifications for classic organic plasters for concrete. Referring to the compatibility and interaction between the synthetic binder and the two natural materials, mainly the hemp fibers, that this research pointed out, we consider that additional research are necessary to establish the long-term evolution of these aspects.

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