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# ABRASION RESISTANCE OF PAINTS MADE WITH CURCUMIN PIGMENT AND PVA RESIN

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Abstract
**Polyvinyl acetate (PVA) polymer resins are commonly used in the manufacture of artisanal paints with natural inputs, giving them the classification of economic latex paint according to NBR 11702. Aiming to minimize the final cost, since PVA is the most expensive component, this study contributes to the area of building paints through the innovative idea of formulating paints with PVA resin, curcumin pigment extracted from the root of *Curcuma Longa L.*, color preservative additives and bacterides, in order to find the appropriate proportions capable of reaching the minimum limit of resistance to abrasion, so that the paint is considered to have good performance according to technical standards. Furthermore, the present study analyzed the variation in the characteristics of the paints based on the variation in the amount of PVA in the mixtures. The results indicate that the addition of PVA at extreme levels reduces the abrasion resistance and viscosity of the mixtures. Statistical analysis obtained satisfactory results when PVA made up 33% of the emulsion. With the positive results, the present intention is to begin research in the area of building paints made with curcumin pigment and PVA as a binder.**

Keywords: **Building paints; Formulation; Curcumin pigment; Polymer resin; Abrasion resistance test.**

## 1. INTRODUCTION

Artisanal paints made with natural ingredients are composed of pigments, solvents, binder, and additives. The function of the pigment is to give color to the paint mixture, the solvent keeps the pigments in a homogeneous configuration and improves the applicability of the paint mixture, evaporating after application, while the additives are used to optimize the characteristics and durability of the paints. The binder is the non-volatile part of the paint, responsible for uniting the pigment particles, forming an adhesive film when applying the paint to the surface [1, 2].

The binder is used in the manufacture of paints since cave paintings dated between the years 30.000 to 8.000 BC, in which the inhabitants of caves felt the need to use local protein inputs as a "glue" to adhere the pigments to the walls, such as eggs (albumin protein), resins and vegetable oils [3, 4, 5].

Over time, other possibilities for binders were discovered, such as natural resins extracted from acacia species (popularly known as gum arabic), gelatin (collagen extracted from the skin, tendons and bones of pigs, cows and chickens, and from the skin and scales of fish), cow's milk (casein protein) and beeswax [6, 7].

In South America and Africa, it was common to use latex (resin extracted from *Hevea euphorbiacex*) as a binder and for waterproofing surfaces, while in Mexico and Colombia it was common to use resin extracted from the "nopal" cactus (*Nopalea coccinellifera, Cactus cochenilliferus*), known as "fig tree", for the same purposes [7]. Other vegetable resins commonly used in the elaboration of natural dyes are rye flour glue with the addition of zinc sulfate ($ZnSO_4$), agave sap (*Agave tequilana*), juice of cooked banana leaves, sap of *Cactus opuntia*, sap of Euphorbia lactea, kapok oil (*Chorisia speciosa*) and cellulose glue [8].

Despite being a product of industrial origin, the use of polyvinyl acetate resin (PVA) as a binder (popularly known as "white glue") is recommended in the production of building paints due to the lack of toxicity, high solubility in water, and being classified as the best alternative to glues by the German government [9], in addition to the absence emissions of volatile organic compounds (VOCs). According to the database of the University of Delft, in the Netherlands [10], PVA white glue requires 58 MJ/kg of energy for its manufacture, equivalent to 12.9 MJ/l in recipes for paints with soil pigment 11, that is, a relatively low value compared to 65 MJ/kg of industrialized PVA paints [12]. As the objective of making natural paints is also to encourage self-production, PVA white glue is commonly found on the market, which makes it practical to use as a binder, despite being considered the most expensive component in the making of natural paints.

Currently, it is common to find recipes and experiments published by researchers of building paints area with natural ingredients in which PVA resins are used as a binder, and the colors are obtained through mineral pigments extracted from clays and lime [11, 13]. However, vegetable pigments are little explored in construction area due to their instability in the face of adverse weather conditions.

Therefore, the present study fills this gap through the development of paints formulated through the method of experimental mixture planning, in which the amounts of PVA resin in the mixtures were varied within the limits stipulated by a bibliographic review in order to evaluate its influence as a binder on the characteristics of pH, viscosity and abrasion resistance (RAU) characteristics of artisanal paints made with curcumin pigment extracted from the root of *Curcuma Longa L.*, popularly known as turmeric.

## 2. MATERIALS AND METHOD

### 2.1. Selection of components

The curcumin pigment was extracted from the turmeric root, which underwent drying and grinding processes to obtain a powder, later diluted in ethanol (ethyl alcohol 70° INPM) in the proportion of 1:10 according to the data collected based on experiences with artisans and researchers in the area of artisanal paints with natural inputs. Ethanol used for the extraction of the pigment acted as a solvent in the mixtures prepared during the formulation stage, whose choice was motivated by the satisfactory results in the extraction of the pigment according to the information collected, also due to its low toxicity, low cost and ease of access.

Cascorez polyvinyl acetate resin (PVA) was used as a binder (universal category), presenting pH 4.3 and viscosity of 7.100 cp (centipoises) at 25°C (according to the information provided by the manufacturer on the product packaging) at the moment it was added to the mixtures. Due to its high viscosity and pH around 4.5, PVA resin was selected in this stage of the study in order to achieve high levels of RAU, according to information collected in a literature review. The amount of binder was based on the consumption range of resins for latex paint from 4.3% to 13% by mass [14] and on studies on the relation between the amount of resin and the volume of pigment [15].

As a color conservative additive, *kakishibu* (green persimmon juice of the astringent type, rich in tannins) was used, prepared by the Rio Grande do Sul artisan Marion Rupp, which underwent fermentation for 24 months, with a pH of 3.4 and viscosity of 0.92 cp at 25°C at the time it was added to paint mixes. Painting chalk was used as a bactericide and fungicide due to its alkalinity, preventing the formation of stains and avoiding damage to the coatings [11, 16]. It is expected to achieve balance between the acidic pH of the additive and the alkalinity of the bactericide in mixtures planned to contain both components, in order to reach the ideal pH to result in satisfactory RAU. The amount of solids in the mixture (pigments and additives) was limited to the usual range for latex paints, in which the total solids content is 35.6% to 52% by mass, and the pigment content in the mixture is 30 % to 45.9% by mass [14].

### 2.2. Planning and experimental design of mixtures

In order to determine the correct proportion of each input, it is necessary to carry out experiments that evaluate each response variable, called the experiment's response [17]. The mixture design evaluates the variation of these responses according to the variation of each component [18], being considered an effective methodology for the elaboration of paints since it reduces the number of experiments necessary to determine the physical or rheological properties considered satisfactory for the objective to be achieved [13].

Therefore, in this article, the experimental design stage using the statistical method of modeling mixtures has as main objectives: to verify how the characteristics of the final product are affected by the variation of the proportions of the components; to define the appropriate proportions to obtain mixtures with suitable pH, viscosity and RAU values according to pre-established limits; and to analyze the results in order to obtain formulations that meet the minimum performance requirements according to the technical standard [19].

In the present study, polynomial models were used. They indicated indicated when the sum of the proportions of each component is equal to 1 and, at each change in the mixture formulation, the sum of the new proportions continues to result in 1 [14]. For this, Minitab 20.4 software was used to elaborate the experimental design in which the quadratic models were adjusted based on the main effects and double interactions. Aiming at multicollinearity in the process, it was decided to work with the pseudocomponents and analyze the significance of the terms (p-value < 0.05) as well as the coefficient of determination ($R^2$) to verify the fit of the model. The "p-value" is the probability of obtaining a test statistic equal to or more extreme than that observed in a mixture, under the null hypothesis. In the present study, the effect of each component and of the dual interactions between them was tested, considering as a null hypothesis the absence of a significant effect of each component and of the interactions with the response variable. Thus, considering 95% confidence, null hypotheses were rejected when p-value < 0.05. In this sense, it was decided to work with the pseudocomponents and analyze the significance of the terms (p-value < 0.05). The coefficient of determination ($R^2$) expresses the variability of the data explained by the model, being an adjustment measure that varies between 0 and 1 (0% and 100%), that is, the closer to 100%, the more adequate the model.

As the variations in the proportions of the components aim to provide the most adequate formulations according to the answers of the performance requirements, the present study elaborated the design of extreme vertices of degree 4, considering as variables answers the characteristics viscosity, pH and resistance to the abrasion (RAU). After inserting the minimum and maximum limits of each component in the Minitab 20.4 software, shown in Table 1, 36 mixtures were generated shown in Table 2, in which the pigment curcumin (X), the solvent ethanol (Y), the PVA resin (Z), *kakishibu* color additive (W) and paint lime bactericide (S).

**Table 1.**
**Minimum and maximum limits of each component in the mixture**

| | Proportion (%) | |
|---|---|---|
| Component | Inferior | Superior |
| X | 0.03 | 0.2 |
| Y | 0.20 | 0.37 |
| Z | 0.30 | 0.47 |
| W | 0.30 | 0.47 |
| S | 0 | 0.17 |

**Table 2.**
**Mixtures generated by Minitab 20.4 software**

| Mixture | X | Y | Z | W | S |
|---|---|---|---|---|---|
| 1 | 0.03 | 0.2 | 0.3 | 0.3 | 0.17 |
| 2 | 0.2 | 0.2 | 0.3 | 0.3 | 0 |
| 3 | 0.03 | 0.37 | 0.3 | 0.3 | 0 |
| 4 | 0.03 | 0.2 | 0.47 | 0.3 | 0 |
| 5 | 0.03 | 0.2 | 0.3 | 0.47 | 0 |
| 6 | 0.03 | 0.2 | 0.3 | 0.39 | 0.09 |
| 7 | 0.03 | 0.2 | 0.39 | 0.3 | 0.09 |
| 8 | 0.03 | 0.2 | 0.39 | 0.39 | 0 |
| 9 | 0.03 | 0.29 | 0.3 | 0.3 | 0.09 |
| 10 | 0.03 | 0.29 | 0.3 | 0.39 | 0 |
| 11 | 0.03 | 0.29 | 0.39 | 0.3 | 0 |
| 12 | 0.12 | 0.2 | 0.3 | 0.3 | 0.09 |
| 13 | 0.12 | 0.2 | 0.3 | 0.39 | 0 |
| 14 | 0.12 | 0.2 | 0.39 | 0.3 | 0 |
| 15 | 0.12 | 0.29 | 0.3 | 0.3 | 0 |
| 16 | 0.03 | 0.2 | 0.36 | 0.36 | 0.06 |
| 17 | 0.03 | 0.26 | 0.3 | 0.36 | 0.06 |
| 18 | 0.03 | 0.26 | 0.36 | 0.3 | 0.06 |
| 19 | 0.03 | 0,26 | 0.36 | 0.36 | 0 |
| 20 | 0.09 | 0.2 | 0.3 | 0.36 | 0.06 |
| 21 | 0.09 | 0.2 | 0.36 | 0.3 | 0.06 |
| 22 | 0.09 | 0.2 | 0.36 | 0.36 | 0 |
| 23 | 0.09 | 0.26 | 0.3 | 0.3 | 0.06 |
| 24 | 0.09 | 0.26 | 0.3 | 0.36 | 0 |
| 25 | 0.09 | 0.26 | 0.36 | 0.3 | 0 |
| 26 | 0.03 | 0.24 | 0.34 | 0.34 | 0.04 |
| 27 | 0.07 | 0.2 | 0.34 | 0.34 | 0.04 |
| 28 | 0.07 | 0.24 | 0.3 | 0.34 | 0.04 |
| 29 | 0.07 | 0.24 | 0.34 | 0.3 | 0.04 |
| 30 | 0.07 | 0.24 | 0.34 | 0.34 | 0 |
| 31 | 0.06 | 0.23 | 0.33 | 0.33 | 0.03 |
| 32 | 0.05 | 0.22 | 0.32 | 0.32 | 0.1 |
| 33 | 0.13 | 0.22 | 0.32 | 0.32 | 0.02 |
| 34 | 0.05 | 0.3 | 0.32 | 0.32 | 0.02 |
| 35 | 0.05 | 0.22 | 0.4 | 0.32 | 0.02 |
| 36 | 0.05 | 0.22 | 0.32 | 0.4 | 0.02 |

The production of the 36 mixtures followed the procedures listed below:

a) the amounts of pigment and solvent for dilution were weighed according to the formulations prepared by Minitab 20.4;

b) to the mixture of pigment and solvent, the weighed amount of binder was added;

c) later, the weighed amounts of additive and bactericide, when present, were added;

d) the mixture was stirred with an electric mixer at 3.000 rotations per minute for 3 minutes to become homogeneous;

e) mixtures were transferred to a container with a capacity of 500 ml to carry out the viscosity and pH measurements;

f) after measurements, applications on substrates for abrasion resistance tests were prepared.

### 2.3. Viscosity, pH and RAU

The viscosity of a paint is an important factor as it conditions its applicability to the substrate, guarantees the adhesion of the films after total drying, and determines the final performance of the paint [15]. The final consistency of each mixture was reached by varying the amount of solvent added to the mixtures, in order to reach a consistency similar to that of conventional paints and paints made with soil pigment [11, 13], therefore, the viscosity of the mixture was measured with a Ford Cup viscosimeter (QUIMIS model Q280) with orifice number 4 (diameter 4.115 mm), following the test methodology described in the technical standard [20]. Three measurements were taken in each mixture with a temperature of 25°C±1, and the result obtained through the average of the three measurements was adopted.

The pH of the mixtures was based on values considered ideal for emulsions using PVA resin, between 4.5 and 5.5 [21] and between 5.0 and 6.5 for emulsions using PVA and ethanol [22]. Studies prove that a high pH compromises the action of PVA resins [13], increasing the solubility of polymeric resins by exposing OH- groups and facilitating the interaction with water, which is desirable, as it favors contact between the polymer and the pigment, however, the vinyl acetate monomer emulsion system is sensitive to pH changes [23].

Therefore, the experimental stage of the research considered reformulating the mixtures evaluating the pH of the final interaction between all components. The pH of each mixture was measured with a Digimed model DM-23 pH meter.

The abrasion resistance (RAU) of the mixtures followed the procedures described by the technical standard24, and followed the minimum limit of 100 brushing cycles stipulated by the technical standard for economic latex classification paints [19].

## 3. RESULTS AND DISCUSSION

Figure 1 shows the 36 paint mixtures made with curcumin pigment applied to a mortar substrate of 10 cm x 10 cm, according to procedures described in NBR 15380 [25] for the RAU test. From the image it is possible to notice the difference in the shade of the yellow color as the amounts of pigment, solvent, additives vary, and according to the addition of painting lime which, due to its alkalinity, results in a change in the color of the pigment, as occurs with the bixin pigment, extracted from annatto (*Bixa orellana*) [26].

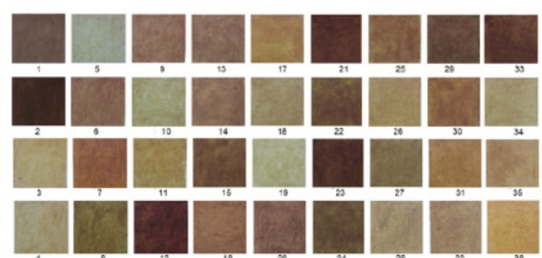

**Figure 1.**
**The squares with the images of the specimens painted with the paint mixtures are identified with the corresponding mixture number (1 to 36)**

Table 3 presents the results of the response variables analyzed at each experimental point, as well as the density (g/cm³) of each mixture and its temperature (°C) at the moment of measuring the cinematic (v) and dynamic viscosity (μ). In the table are highlighted in bold the values considered acceptable for viscosity for good application of the paint on the substrate (minimum of 70 cp and maximum of 160 cp), the pH values within the limits described according to the bibliographic review (between 4.5 and 5.5), and RAU above 100 brushing cycles considered good performance for economic category latex paints.

From the analysis of the response variables, it is possible to identify a pattern when the PVA resin is used at the minimum and maximum levels, presenting lower RAU and viscosity when the amount of PVA resin reaches the maximum in the paint emulsion, regardless of the pH value. The minimum amount of

**Table 3.**
**Density, temperature and response variables**

| Mixture | pH | Temperature (°C) | Density (g/cm³) | Viscosity $\nu$ (mm²/s) | Viscosity $\mu$ (cp) | RAU (cicles) |
|---|---|---|---|---|---|---|
| 1 | 12.2 | 24.4 | 0.82 | **162.5** | **133.2** | **137** |
| 2 | **4.6** | 25 | 0.82 | **193.7** | **158.83** | **217** |
| 3 | 4.4 | 25.4 | 0.78 | 74.42 | 58.04 | 54 |
| 4 | 4.3 | 24.1 | 0.82 | **103.8** | **85.11** | 66 |
| 5 | 4.2 | 25.3 | 0.81 | 64.71 | 52.42 | 56 |
| 6 | 12 | 25.4 | 0.88 | **90.32** | **79.48** | **220** |
| 7 | 12 | 25 | 0.88 | **96.48** | **84.9** | **128** |
| 8 | 4.2 | 24.5 | 0.77 | 77.5 | 59.67 | 59 |
| 9 | 12 | 25 | 0.92 | 79.15 | 72.82 | 123 |
| 10 | 4.4 | 24.7 | 0.76 | 63.95 | 48.6 | 48 |
| 11 | **4.5** | 25.2 | 0.78 | 77.8 | 60.69 | 60 |
| 12 | 12 | 24.5 | 0.86 | **112.84** | **97.04** | 186 |
| 13 | **4.5** | 25.5 | 0.86 | 78.96 | 67.9 | **114** |
| 14 | **4.5** | 25.3 | 0.84 | **95.32** | **80.07** | 102 |
| 15 | **4.7** | 24.5 | 0.81 | 75.19 | 60.9 | **104** |
| 16 | 11.9 | 25 | 0.81 | 86.08 | 69.73 | **120** |
| 17 | 12 | 24 | 0.81 | 80.58 | 65.27 | **113** |
| 18 | 12 | 25 | 0.82 | 76.73 | 62.92 | **124** |
| 19 | 4.3 | 24 | 0.84 | 72.11 | 60.57 | 51 |
| 20 | 11.7 | 25.2 | 0.88 | 76.92 | 67.7 | **138** |
| 21 | 11.9 | 25 | 0.85 | **99.83** | **84.85** | **163** |
| 22 | 4.4 | 24.3 | 0.85 | 77.23 | 65.64 | 77 |
| 23 | 11.7 | 24.5 | 0.84 | 82.9 | 69.63 | **121** |
| 24 | **4.6** | 24 | 0.84 | 75.69 | 63.58 | **101** |
| 25 | **4.7** | 24 | 0.83 | **89.74** | **74.49** | 69 |
| 26 | 11.8 | 24 | 0.8 | 72.8 | 58.24 | **107** |
| 27 | 11.8 | 24 | 0.83 | 86.08 | 71.45 | **118** |
| 28 | 11.7 | 25.3 | 0.81 | 77.69 | 62.93 | **145** |
| 29 | 11.7 | 25 | 0.81 | 80.77 | 65.42 | **270** |
| 30 | 4.4 | 24.5 | 0.8 | 74.53 | 59.62 | **180** |
| 31 | 11.4 | 25.5 | 0.85 | 76.27 | 64.82 | **152** |
| 32 | 12.1 | 25.5 | 0.84 | 82.23 | 69.08 | **196** |
| 33 | 8.7 | 25.9 | 0.85 | 80.89 | 68.75 | **136** |
| 34 | 10.7 | 24.3 | 0.82 | 69.8 | 57.23 | **140** |
| 35 | 11.3 | 24.3 | 0.86 | **82.04** | **70.55** | 88 |
| 36 | 11.1 | 24.5 | 0.82 | 69.61 | 57.08 | **110** |

PVA resin in the paint emulsion reached sufficient RAU when the viscosity was high, regardless of the pH value, while the minimum amount of resin resulted in insufficient RAU when the viscosity and pH values of the emulsions were lower to the stipulated limits. Furthermore, sufficient RAU was achieved when the emulsions had low viscosity, regardless of the pH value.

According to the results of the analysis of the response variables, 30% of the prepared mixtures had a viscosity between 70 and 160 cp, 19% of the mixtures had a pH between 4.5 and 5.5, and 72% of the mixtures had a RAU ≥ 100 brushing cycles.

The regression equations for the response variables viscosity, pH, and RAU are shown in Table 4, where the components are identified in the equations: pigment (X), solvent (Y), PVA resin (Z), color preservative additive (W) and bactericide (S), considering significant interactions (p-value < 0.05).

**Table 4.**
**Regression equations for the response variables viscosity, pH, and RAU**

| Response variable | Regression equation | $R^2$ |
|---|---|---|
| Viscosity | $\hat{y} = 152.57X + 57.56Y + 84.86Z + 52.58W + 130.57S - 147.2XY - 127.7XZ - 132.3XW - 181.9XS - 93.4YS - 94.1ZS$ | 93.28% |
| pH | $\hat{y} = 4.14X + 4.35Y + 4.36Z + 4.24W + 10.63S + 22.16XS + 23.06YS + 23.69ZS + 23.71WS$ | 87.34% |
| RAU | $\hat{y} = 197.6X + 47.9Y + 49.3Z + 59.3W + 141.0S + 338.0WS$ | 60.40% |

The regression analysis for the viscosity index indicates that, to obtain viscosity within the specified limits, higher levels of X and S are necessary, and lower levels of Y, Z and W. However, the X factor interacts significantly with the others components, indirectly producing a negative effect. In the same way, component S interacts indirectly with components X, Y and Z, producing a negative effect. Therefore, it is recommended to adopt intermediate and complementary levels for components X and S. It is important to highlight that components X and S are the only solid components of the mixture and, for this reason, they directly influence viscosity levels, since the increase in liquid components Y, Z and W results in a mixture with high viscosity, outside the values considered acceptable for viscosity for good application of the paint on the substrate (minimum of 70 cp and maximum of 160 cp). Therefore, to reduce or completely eliminate the S component while maintaining the levels of the X component, it would be necessary to significantly increase the levels of the Z component in order to maintain the viscosity levels within the values considered acceptable.

The regression analysis for pH indicates that, to obtain higher pH values, it is necessary to increase the levels of factor S and reduce the levels of factors X, Y, Z and W. The S component interacts significantly with the other components, producing a positive effect through this interaction. Therefore, in order to control the effects produced to bring pH values within limits, it is recommended to adopt lower levels of the S component.

The regression analysis for RAU indicates that, to obtain higher resistance values, it is necessary to increase the levels of factors X and S, and reduce the levels of factors Y, Z and W. The S component interacts significantly with W, producing a positive effect indirectly. Therefore, it is recommended to adopt higher levels of factor S to obtain high RAU results.

In order to find the mixture that best represents the joint optimization of the response variables, the present study considered developing a response optimization analysis adopting the same weight for the three variables analyzed using the Minitab 20.4 software, that adjusted the data to simultaneously fit the three response variables through the *Desirability method* [27], commonly used to determine the best process adjustment conditions, simultaneously optimizing the multiple responses.

The *Desirability method* found a model fit considered optimal (d = 97%) for each variable, in order to present the ideal composition of each component in the mixture, resulting in a product that meets the requirements performance limits stipulated for an economical latex paint. Therefore, the pH assumed a value of 5.00, the viscosity of 103.89 cp, and the abrasion resistance of 150.49 brushing cycles.

Among the 36 mixtures prepared in the experimental design, only two achieved result within the stipulated limits for the three analyzed variables, the mixtures numbers 2 and 14, which presented adequate viscosity for good applicability of the paint (in the case of paints with vegetable pigment, between 70 and 160 cp), pH between 4.5 and 5.5, and resistance to abrasion above 100 brushing cycles, thus representing the mixtures considered ideal for the elaboration of artisanal paint with *Curcuma Longa L*. pigment.

## 4. CONCLUSIONS

It is concluded that the pigment represents a relevant factor not only in fulfilling its common functions in the composition of a paint (providing color and protecting the binder), but is also capable of influencing the viscosity and the ability of the paint to resist brushing. or friction when applied to internal walls. The addition of *kakishibu* to the mixtures as a color-conserving additive increased the viscosity of the samples and reduced the pH due to its high acidity, generating satisfactory RAU results, since the high pH compromises the action of the PVA resin, according to data collected in a literature review.

Components such as solvent, binder and additive showed unsatisfactory results when used in minimum and maximum proportions, indicating the need to find intermediate proportions to achieve balance in the mixture, achieve appropriate viscosity, abrasion resistance above 100 brushing cycles, and to avoid unnecessary waste with PVA, considered the component with the highest cost in an artisanal paint.

As expected, PVA resin proved to be an effective binder in the RAU tests. However, in the samples to which the bactericide was added, the pH increased, resulting in low RAU, confirming the studies collected in a literature review that stipulated an ideal pH between 4.5 and 5.5 to obtain satisfactory RAU results. For these reasons, based on the calculations and analyzes accomplished through the *Minitab 20.4* software, a mixture that fits simultaneously within the specified limits for all variables in the matter is composed of approximately 15% pigment, 20% solvent, 33% PVA resin, 32% additive and does not contain a bactericide.

Besides its main findings, the present study intends to leverage future research in the area of architectural paints made with natural inputs and PVA resin through the initial results of the evaluation of the interaction between this and other components. It is expected to stimulate interest in the possibilities of making architectural paints with other types of chromophores and their interaction with polymeric resins, using as a basis the formulations and characteristics analyzed in the present study.

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