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The use of red mud and kaolin waste in the production of a new building material: Pozzolanic pigment for colored concrete and mortar

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ABSTRACT

The state of Para is one of the greatest producers of mineral substances in Brazil. Kaolin for paper coating industry and bauxite for alumina and aluminium production are amongst the most important commodities. The latter is responsible for the generation of red mud, a well-known residue from Bayer process. The kaolin processing plants are also responsible for the generation of large amounts of wastes, in this case, very fine-grained kaolinite (white mud). The research aimed to find a final destination for such residues by developing new building materials. The proposed material is a pozzolanic pigment produced through calcination and grinding of mixtures of red mud and kaolin waste. The pozzolanic pigments provided increased mechanical strength and color stability of the colored mortars relative to the inert pigment. The pozzolanic characteristics of the pigments reduced the leaching of the mortars. The new material has proven to be promising in its application as an innovative construction material, with the possibility of using these residues in a market that is little explored in Brazilian civil construction: colored mortars and concrete.

Keywords: Red mud; Kaolin waste; Colored concrete; Pozzolanic pigment.

1. INTRODUCTION

The state of Pará is one of the most privileged regions of the planet in terms of exploitation of mineral resources. Mining projects in Pará are characterized by the quality of the product, the marketed values, and the size of the production volume, which also makes them responsible for generating significant amounts of waste or byproducts that are released and deposited in the environment. Among the most diverse types of residues generated by intensive mineral activity are the production volume of kaolin waste (KW) generated by the extraction and processing of kaolin, and the red mud (RM) from the Bayer process for extracting alumina from bauxite. It is estimated that the deposited amounts of KW and RM since the implantation of kaolin mills and Alumina do Norte do Brasil SA (ALUNORTE) are 15 and 60 million tons, respectively. The first with approximately 450 thousand tons are deposited annually in sedimentation ponds [1–4] and the second with approximately 4.7 Mt of this residue are generated annually [5].

The KW has excellent technical characteristics, which has been demonstrated by the various researches, indicating the application potential not only in the field of building, but also in the refractory and advanced ceramic industries [6–12]. It consists essentially of extremely fine kaolinite, presents excellent uniformity and easy handling. All these requirements are excellent for the production of a highly reactive pozzolan from the calcination and grinding of pure kaolinite clays with very low inert minerals called metakaolin or metakaolinite.

RM itself would not be a toxic waste if it were not for its causticity, so much so that the United States Environmental Protection Agency does not classify it as a hazardous waste [13]. However, other researchers consider it to be toxic precisely because of the high alkalinity and ion exchange capacity, which constitute a high risk to neighboring populations [14, 15]. The chemical and mineralogical characteristics of RM impose difficulties to its use due to the variety of minerals present. In general, RM consists of a complex assembly of minerals ranging from those not dissolved in the process such as aluminum oxides and hydroxides (gibbsite, boehmite and diaspore), the iron oxides and hydroxides (hematite e goethite), rutile, anatase, calcite, dolomite, kaolinite, besides the neo-formed ones as sodalite and cancrenite and others that are in the form of traces as the oxides of V, Ga, P, Mn, Mg, Zn, Th, Cr and Nb.

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In addition to the trend in the world aluminum industry for the densification of waste in order to reduce the generated volume of RM, many attempts have been made to take advantage of it instead of simply depositing it because of the high costs of RM disposal in the residue disposal area (DSR). However, the vast majority of the studies did not find a satisfactory application from the economic point of view [16]. Among the various applicability of RM, it is important to highlight the direction of the efforts for the use in Construction as a raw material for the manufacture of building materials. The feasibility of such a solution would be of threefold benefit, since the large-scale consumption of building materials could substantially eliminate or mitigate the problem of waste disposal; would add economic value to RM and would be easier to deploy on a universal scale.

The initiatives for the application of this residue in construction materials, we can highlight: glass ceramic tiles [17], paving blocks [18] and cement production [19–21]. In addition to the use in concrete, where durability studies have already been developed [22]. In Brazil, research has observed the influence of mud on cement setting time [23], pozzolanicity index [11, 24–26], evaluated the pozzolanic property of dry and calcined Brazilian red mud at different temperatures.

Inorganic dyes can also be produced from bauxite residue after calcination at high temperatures, the materials obtained present new crystalline structures and when added to ceramic glazes as additives produce glazes with stable and high intensity colors [27]. The use of bauxite residue as a pigment is acceptable if it brings performance advantages in addition to environmental and economic advantages. According to CARNEIRO *et al*. [28], the addition of 3% by weight of this calcined material has a better performance than in natura dry, and is a more stable coloring agent than commercial hematite.

PERA *et al*. [29], claim that calcined RM could be used to produce colored concrete as a low-cost pigment compared to conventional pigments. In addition to the economic aspect, it presents some other technical advantages such as the elimination or reduction of efflorescence, common pathology in concrete structures and that is extremely harmful to concrete and colored mortars because it is responsible for the appearance of efflorescence on the surface, substantially damaging the color of the material surface [30]. Another positive aspect would be the possibility of being used in larger percentages, without loss of resistance, which commonly occurs with conventional inorganic pigments, which are inert.

A disadvantage of the use of RM as a building material would be free or exchangeable sodium present in sodalite, which could render its use in Portland cement based products unreliable due to the activation of alkali-aggregate reactions and the crystallization of salts on the surface (efflorescence). Due to this possibility, the present paper starts from the hypothesis that the combined calcination of RM with KW, in addition to increasing the pozzolanic activity, the exchangeable sodium present in the sodalite can be stabilized by solidification (S / S) in the formation of the structure of silicates and calcium aluminosilicates from the cement hydration and the pozzolanic reactions between metakaolinite and Portland cement.

Another advantageous aspect of the combined use of KW and RM are the distances between the residue disposal areas. Both residues are generated in the municipality of Barcarena, within a radius of 5 km between the DSR of ALUNORTE and the sedimentation ponds of Imerys. The aim of this article was to evaluate the technical feasibility of the pigments from the RM and KW mixtures by investigating their effect on the properties of colored mortars such as: compressive strength, color stability. In addition to contributing to an environmental assessment of RM with the performance of leaching tests on the pigments and mortars that incorporate these pigments.

2. MATERIALS AND METHODS

2.1. Raws materials and pigments

The raw materials used for the study were KW from Imerys and RM from ALUNORTE. Pigments were prepared from three different proportions of RM and KW, and individually calcined RM (CRM).

The materials RM and KW were delivered with different moisture content, Kw (18–20%) and RM (34%) were previously dried and ground to break them into smaller fragments and then co-calcined and pulverized. The raw materials were dried in an electric oven (model Q-317B, brand Quimis) at a temperature of 105 ºC until they reached a constant mass. Grinding was carried out for one hour in a ball mill (model CT 242, brand SERVITECH). The mill container with a capacity of 10 liters was 2/3 full, 1/3 occupied by the load of balls with high alumina content and the other 1/3 by the sample to be ground. The remaining part was intended for the space required for the mobility of the materials. The material loading corresponding to one third was about 1.5 kg. Alumina balls of 10 mm and 30 mm with masses of six kilograms and three kilograms, respectively, were used.

Table 1: Pozzolanic pigments investigated in the experimental program.

POZOLANIC PIGMENT	% RM	% KW
RK19	10	90
RK55	50	50
RK91	90	10
CRM	100	0

After comminution, the three different pigments were co-calcined at 800°C, as was the CRM. The calcination procedure consisted of calcining the samples in stainless steel crucibles in a muffle at a temperature of 800°C. The residence time of each sample in the muffle was 90 minutes. After this time, the samples were removed from the muffle and cooled in air, undergoing thermal shock. The muffle used is of the brand Quimis, model Q-318D, electrically heated, maximum temperature of 1200ºC with digital controller. The pigments were then ground for three hours using the same equipment and grinding procedure as described above.

The commercial pigment adopted as reference was Bayerferrox 120® (BF). Table 1 shows the composition of the pozzolan pigments investigated in the test program.

The mineralogy of the raw materials and the pigments was evaluated by X-ray diffractometry (XRD) by the powder method (model Empyrean, PANalytical), with $\theta - \theta$ goniometer, sealed copper (Cu) ceramic X-ray tube, 2,200 W long fine focus and Ni kβ filter, X'Celerator model liner detector (PSD), with active length of 2,122º 2θ and 128 channels. The instrumental conditions used will be: voltage of 40 kV and current of 35 mA; 0.04º rad soller slits (our incidences and diffracts); scanning range from 9 to 2θ; 0.01º 2θ step size with 20 s time/step in continuous scan mode; 1/4th rad divergent slit and 1/2nd rad anti-scatter; irradiated sample size of 15 mm; Offer anti-slit scattering, 50 mm diffract and circular motion sample with motion frequency of 1/s. The total collection time was 10 min. The chemical characteristics of the raw materials were carried out using energy dispersive X-ray fluorescence spectrometry (EDS, Model 700 HS, Shimadzu). The particles size distribution of the RM and KW obtained by laser grain size measurements (CILAS Model 715 E 701). The specific surface obtained of the raw materials was measured using the BET method (Micromeritics ASAP 2020). The densities of RM and KW were determined according to the requirement of Brazilian standard NBR 16.605.

2.2. Mortars mix proportions

The mortars were produced with structural white Portland cement from the Mexican company CEMEX, equivalent in Brazil to CPBE 32. The fine aggregate was a silica natural sand (density 2.65 kg/dm^3, absorption 0.8% and fineness modulus 2.4). The superplasticizer additive used was Glenium 51®, polycarboxylate ether based, with a mean density of 1.05 g/cm^3.

Mortars were produced according the binder to sand ratio of 1:3.5 (in mass) and a water/binder ratio kept constant of 0.55. Six mortar were molded, one relative to Portland Cement (controle mix) and five with 10% substitutions of the cement by the pozzolanic and commercial pigments (CRM mix, RK19 mix, RK55 mix, RK91 mix and BF mix). In mortars with 10% of pigments the superplasticizer additive (polycarboxylate ether based) was used. A seventh mortar with 10% cement replacement per calcined KW was cast exclusively for the leaching test on mortars (CKW mix).

2.3. Methods

The compressive strength was carried out in mortars in accordance with the procedures described in Brazilian standard NBR 7215. All the mortars samples were cast into 50 x 100 mm cylinders for the determination of compressive strength at 1, 7 and 28 days. For each age, three samples were molded, totalizing 12 specimens for each type mortar. After casting, all the samples were placed in the laboratory environment at 23.0 ± 2.0 ºC for 24 hours. After that, the molds were removed and the mortar specimens were stored in satured lime water as specified in NBR 7215 until the time of testing.

The environmental assessment of dry RM and pozzolanic pigments was carried out through leaching test described in Brazilian standard NBR 10 005. The environmental assessment of mortars produced with pozzolanic pigmentos was carried out according to the Dutch standard [31]. The test consisted in immersing cylindrical specimens of mortar of dimensions 50 × 100 mm at the age of 28 days in acrylic tanks and covered by lixiviant (liquid/solid = 5). Acidified demineralize water (pH close to 4) was used at each renew lixiviant. After 64 days, the leachate extract obtained was filtered through a 45 μm membrane and identified by inductively coupled plasma – optical emission spectrometry (ICP – OES) (Varian, model VISTA MPX CCD simultaneous). These analyzes were restricted only to control mortars and with a 10% substitution of CRM, RK91, RK55 and RK19.

The color provided by pigments to Portland cement mortars and their stability to the natural and controlled environments were evaluated by colorimetry. The color was determined using the CIELAB color space [32, 33], from the initial and final L*, a*, and b* values were obtained using a Konica Minolta spectrophotometer, model CR-400. To assess the color stability, the total color-difference parameter (ΔE) was calculated using successive measurements performed at different times. This work used the results from the CIEDE1976 (ΔE76) [33, 34]. The color-difference (ΔE76) between the a and b points in an object is the Euclidian distance between the color stimulus in both points, and approximately represents the color-difference perceived by the color stimulus in the CIELAB color space. The Equation 1 calculates this vector magnitude.

$$\Delta E^*_{76} = ([\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2} \quad (1)$$

Where: $\Delta L^* = L^*b - L^*a$; $\Delta a^* = a^*b - a^*a$; $\Delta b^* = b^*b - b^*a$.

In both laboratory and field analyzes, chromatic measurements were performed for the ages of 1 and 238 days. In these tests, sixprismatic specimens of 100 × 100 × 30 mm dimensions were molded for each type of mortar, three for control environment and three for exposure to natural environmental in the Amazon region. In the laboratory, the specimens were under controlled conditions of temperature and humidity, in the case 65 ± 5% and 23 ± 2 ºC respectively. In the field, the specimens were submitted to conditions of the equatorial rainy climate of Belém, remaining in this natural environment for approximately eight months, covering the dry and rainy seasons. For each specimen, six color measurements were performed on the surface of 100 × 100 m^2. The value of the chromatic variation ΔE76 per mortar specimen was obtained from the mean of these six measurements. To ensure that the measurements always occurred at the same points of the specimens, a mask with horizontal and vertical quadrants (2 × 2 cm^2) was made.

3. RESULTS AND DISCUSSION

3.1. Characterization of raw materials and pigments

The physical and chemical characteristics of RM and KW are shown in Table 2. The Figures 1–2 show the x-ray diffractograms of raw materials and pigments. RM is composed of hematite and goethite, besides gibbsite, anatase and sodalite. Fe present is in the form of hematite (Fe_2O_3) and goethite (FeOOH), both responsible for the intense red color of the RM. Al_2O_3 present in the RM is in the form of aluminum hydroxide of the gibbsite type ($Al(OH)_3$) as well as in the crystalline structure of sodalite ($Na_{7,6}(Al_6Si_6O_{24}).(CO_3)_{0,93}.(H_2O)_{2,93}$), newly formed mineral during the bauxite digestion process. SiO_2 and Na_2O present in RM are also part of the structure of

Table 2: Physical and chemical characteristics of RM and KW.

PARAMETERS	RM	KW
SiO_2	18.48	45.27
Al_2O_3	23.26	39.24
Fe_2O_3	31.89	0.57
TiO_2	6.91	0.45
Na_2O	11.58	0.21
K_2O	0.11	<0.05
CaO	1.55	<0.05
MgO	0.10	<0.05
MnO	0.19	<0.05
P_2O_5	0.09	0.08
LOI	5.85	14.12
Density (kg/m^3)	3.05	2.55
BET specific surface (m^2/g)	15.0	18.0
$D_{50\%}$ (µm)	3.0–4.0	3.0–6.0

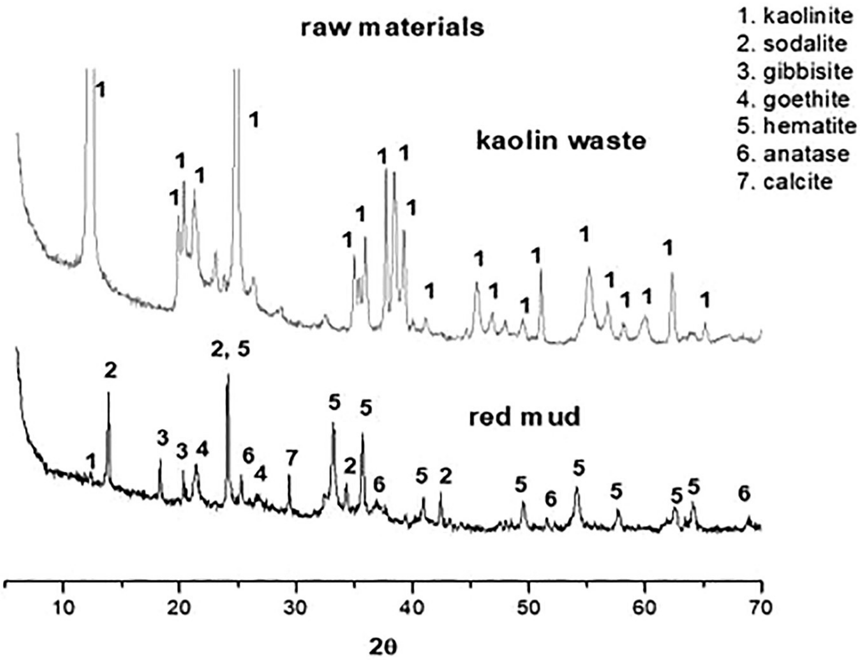

Figure 1: X-ray diffractogram of raw materials.

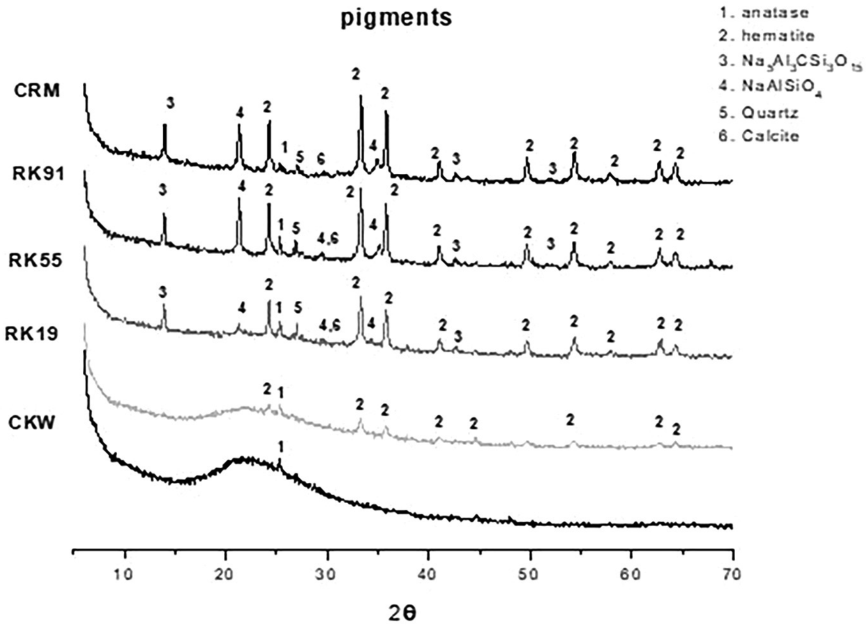

Figure 2: X-ray diffractogram of pigments.

sodalite. TiO_2 is in the form of anatase and CaO is part of the structure of calcite. The high content of Fe explains the high density of 3.00 kg/dm^3 of RM. Another relevant aspect is the high concentration of sodium, either free or weakly bound to the sodalite structure, which can cause problems related to surface efflorescence if the RM is incorporated in the production process of building materials.

The KW consists essentially of a well crystallized kaolinite with a low degree of defects, with sharp high intensity peaks at angles 2θ from 12,46^o and 25,05^o and triplets with well individualized reflections. When it is calcined, the crystalline structure is disordered, being identified in the x-ray diffraction as an amorphous band between the angles 2θ 10º and 30º (Figure 2).

In the RM calcined at 800 ºC, the gibbsite disappeared; however, the presence of boehmite or γ-alumina which are the minerals formed from the dehydration of gibbsite at temperatures above 300 ºC were not identified. Possibly because of the low amount and the high background in the X-ray diffractogram of RM. These hydroxides and aluminum oxides are the pozzolanically active phases in RM. The goethite became hematite, intensifying the red color of the RM. The sodalite decomposed and from this process alkaline oxides of the denser structure of the type $NaAlSiO_4$ and $Na_5Al_3CSi_3O_{15}$ were formed. Anatase and quartz traces were also detected.

The pozzolanic pigments from the mixtures of RM and KW (RK91, RK55, RK19) are composed of the same minerals of the CRM (hematite, $NaAlSiO_4$, $Na_5Al_3CSi_3O_{15}$, quartz and anatase), as well as metakaolinite from KW incorporated into RM. The latter is the phase responsible for the pozzolanic activity of these pigments. The difference in the mineralogical composition of the pigments results from the higher or lower concentration of hematite or metakaolinite, depending on the type of pigment. The commercial pigment BF, as expected, is composed only of hematite, well crystallized, with high intensity, inherent in the controlled production process of this material (not shown).

3.2. Compressive strength

Figure 3 shows the development of compressive strength of reference mortar and with 10% of pigments. Figure 4 shows the relative compressive strengths compared to PC mortars for the ages of 1, 7 and 28 days. The mortars RK55 and RK19, with higher kaolinite incorporations in the pigments, 50 and 90%, respectively, presented the highest strengths for all ages observed. The increases vary between 11 and 86% and are attributed to the high reactivity of metakaolinite.The metakaolinite present in the pigments RK19 and RK55 reacts with the calcium hydroxide from the hydration of the cement to form new products such as stratlingite (or hydrated gehlenite), of lower density than the C-S-H formed in the ordinary Portland cement paste (reference), providing in this way greater filling of the voids in the cement matrix, which results in greater mechanical strength and pore refinement [35–37].

The mortars with calcined RM and predominance of RM in the composition (RK91) also presented higher compressive strengths than the reference mortar and BF. However, the increases were smaller than those obtained with RK19 and RK55 mortars, between 1 and 17%. The increased resistance presented by these mortars is due to the pozzolanic and filler effects of RM. In the case of the pozzolanic effect, although it is lower than that of kaolinite, it is attributed to the hydroxides and aluminum oxides present in gibbsite. The filler effect is caused by the very fine RM particles which accelerate the hydration reactions of the cement, providing a substantial surface increase for the nucleation of new hydration products. However, this effect occurs more pronounced in the first 24 hours, whereas for later ages the pozzolanic effect is more preponderant. For this reason, the increase in compressive strength of CRM and RK91 mortars at the ages of 7 and 28 days is not as pronounced as for RK19 and RK55 mortars because of the low pozzolanic activity of RM.

In relation to the commercial pigment BF, the incorporation of 10% on the Portland cement caused a loss of mechanical resistance in relation to the reference mortar. This is attributed to the fact that it is an inert mineral admixture, which does not react with Portland cement.

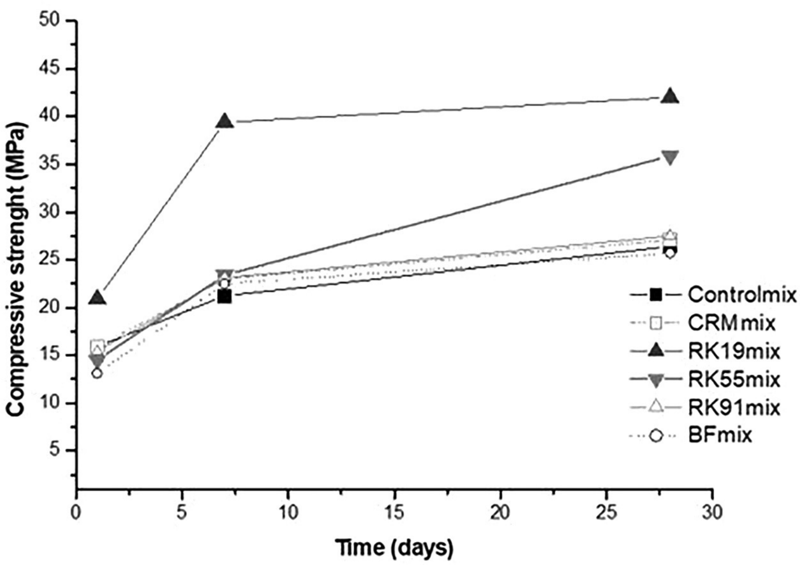

Figure 3: Compressive strength of mortars at 1, 7 and 28 days.

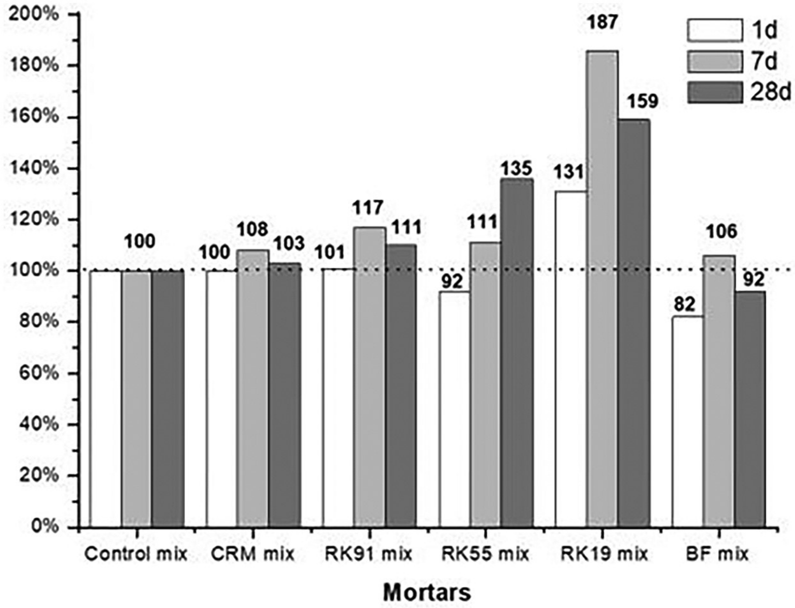

Figure 4: Compressive strength of mortars normalized to the strength of control mix.

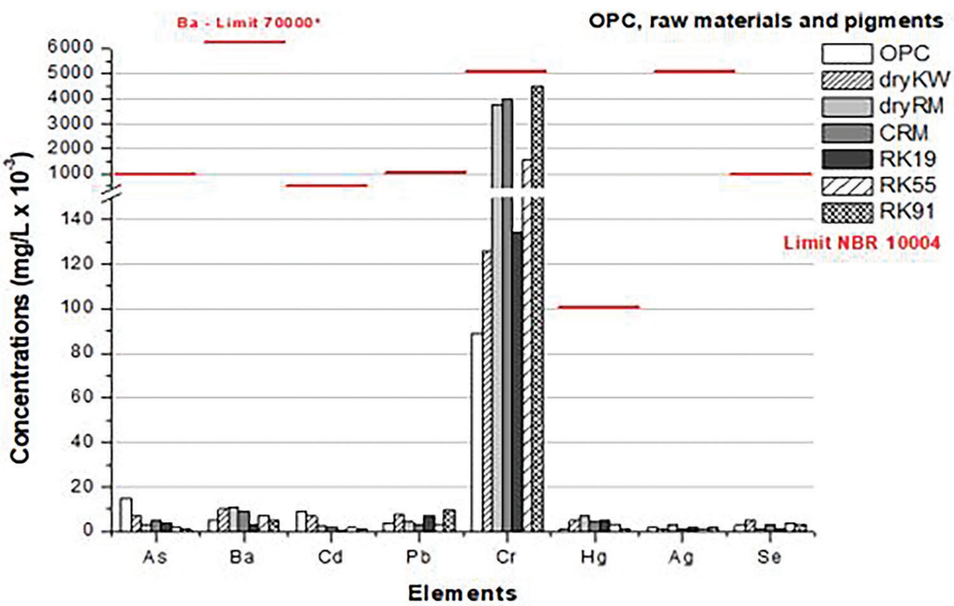

Figure 5: Concentrations of elements leached from raw materials, cement and pigments.

All pigments from the RM and KW mixtures provided significant increases in compressive strength for all ages evaluated, which means that they can replace the cement in percentages even greater than 10% without loss of strength mechanical and even increase the color tonality of mortars and concrete products. The difference between these pigments and BF is that they are pozzolanic, in other words, capable of reacting with Portland cement.

3.3. Environment assessment

The concentration of heavy metals leached from dry RM, pozzolanic pigments and white Portland cement are shown in Figure 5. The results show that dry and calcined MR as well as pozzolanic pigments are classified as non-hazardous waste (Class II) because the contents do not exceed the limits established in Annex F of the Brazilian standard NBR 10004, with only Cr approaching the limit of 5 mg/L.

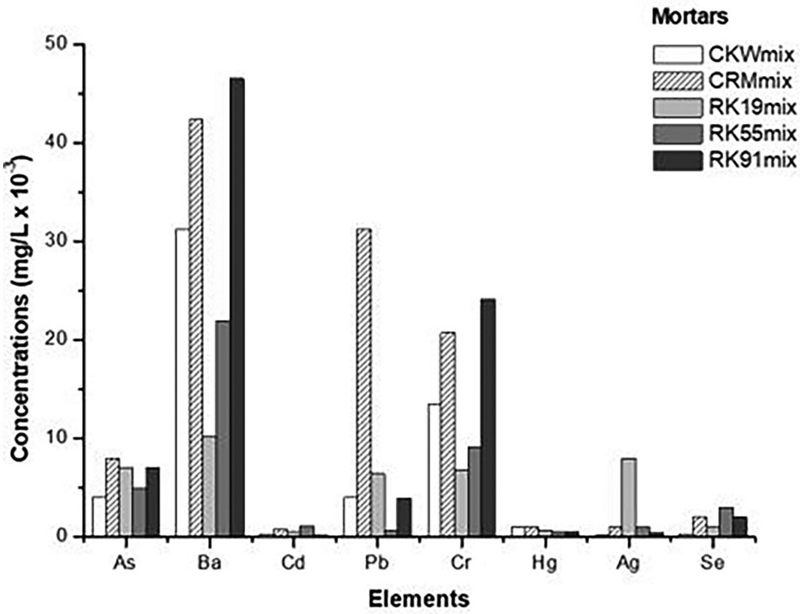

Figure 6: Concentrations of elements leached from mortars.

Table 3: Concentrations of sodium leached from the pigments obtained in the test as well as the theoretical values.

PIGMENTS	Na_{TESTE} (mg / L)	$Na_{THEORETICAL}$ (mg / L)	REDUCTION (%)
Dry RM	152	–	–
Dry KW	8.9	–	–
CRM	133	–	13.0
RK19	20.73	23.21	10.6
RK55	48.69	80.45	39.5
RK91	104.58	137.69	24.0

The leached elements of the mortars are shown in Figure 6. All mortars produced, with or without pigments, had concentrations below the limits defined in annex F of the Brazilian standard NBR 10004. These results are a consequence of the previous ones, taking into account that neither the pigments nor Portland cement had concentrations above the established maximum limits.

The concentrations of sodium leached from the pigments obtained in the test as well as the theoretical values calculated from the weighted average of the sodium contents of each of the dry raw materials (RM and KW) are shown in Table 3. The sodium leached content of RM is 152 mg / L. However, when calcined alone there was a 13% reduction of sodium in the leached extract. Calcination was responsible for the conversion of sodalite to alkali oxides of more compact structures such as $NaAlSiO_4$ and $Na_5Al_3CSi_3O_{15}$, which probably reduced the sodium ion exchange capacity. The pozzolanic pigments also showed reduction of the leached sodium determined in the test compared to that calculated from the percentages of dry raw materials. The reduction range varied between 10 and 40%, indicating that during the firing the incorporation of kaolinite increased the conversion of sodalite to alkaline oxides of dense structure.

The concentrations of sodium leachate from the mortars determined in the test and the theoretical values calculated from the weighted average of the sodium contents of the calcined RM and KW calcined mortar are shown in Table 4. The concentrations of sodium in the leached extract of the mortars with the pozzolanic pigments were lower than those calculated from the individual concentrations of each of the calcined RM and KW mortars. The reductions in sodium concentration varied between 22 and 58%, being the highest for mortars with the highest percentages of metakaolinite (RK55 and RK19). The results show that sodium was stabilized by solidification (S / S) from the formation of the structure of silicates and calcium aluminosilicates from the pozzolanic reactions between metakaolinite and Portland cement. The incorporation of KW to RM not only brought benefits to the mechanical properties but also allowed the encapsulation of the free sodium present, reducing the efflorescence phenomenon.

Table 4: Concentrations of sodium leachate from the mortars determined in the test and the theoretical values.

MORTARS	Na_{TESTE} (mg / L)	$Na_{THEORETICAL}$ (mg / L)	REDUCTION (%)
CRM mix	127.6	–	–
KW mix	5.5	–	–
RK19 mix	7.6	17,8	57.2
RK55 mix	47,7	66.8	28.4
RK91 mix	89.9	115.8	22.3

3.4. Color stability in mortars

Table 5 shows the color parameters values periodically measured on 100 cm^2 of each sample before (sub-index 1 = age 1) and after (sub-index 2 = age 2) 8 months exposure to an natural (N) or control (C) environment. The ΔE76 average values calculated by using those parameters are also shown.

With regard to the color stability, reported that if ΔE76 values are > 1.5, color differences in mortar surfaces can be perceived at naked eye. In line with this statement, the ΔE76 values of all mortars studied were higher than this limit value, which means that in the natural environment of the Amazon region it is very difficult for the colors of the colored mortars to remain stable over time. However, the results showed that the less noticeable changes in mortar color (ΔE76 between 3.50 and 5.7) occurred in those that incorporated pozzolan pigments with a higher percentage of metakaolinite (RK55 and RK19). As seen in item 3.3, the presence of metakaolinite reduced the efflorescence by fixing the sodium in the structure of the products of the pozzolanic reactions, causing not only the increase of the mechanical resistance but also the greater color stability to mortars. In mortars with pigments consisting essentially of calcined RM (CRM and RK91), the perception of the color change was very pronounced, with values of ΔE76 higher than 9 units. However, the worst result was obtained for the mortar with the incorporation of commercial pigment (BF), with ΔE76 equal to 13.6. In the environment under controlled conditions of humidity and temperature and without exposure to solar radiation, the color stability is maintained regardless of the type of pigment used.

Table 5: Color stability parameters.

ENVIRONMENT	MORTAR	i	li	ai	bi	ci	hi	ΔE76
N	BF mix	1	45.6	24.4	13.5	27.9	28.3	13.6
		2	50.3	15.1	7,7	16.9	26.9	
N	CRM mix	1	57.7	21.1	19.4	28.7	42.6	12.1
		2	57.5	12.8	10.6	16.6	39.6	
N	RK19 mix	1	74.8	7.6	14.2	16.1	62.0	5.7
		2	71.8	5.2	9.9	11.2	62.4	
N	RK 55 mix	1	63.1	14.4	16.1	21.5	48.1	3.5
		2	61.8	12.3	13.5	18.3	47.5	
N	RK 91 mix	1	58.6	18.4	17.8	25.6	44.0	9.4
		2	59.0	12.2	10.7	16.2	41.2	
C	BF mix	1	45.3	24.3	13.2	27.6	28.5	0.7
		2	45.2	23.7	12.8	26.9	28.4	
C	CRM mix	1	59.0	20.7	19.2	28.3	42.9	2.1
		2	57.9	19.3	18.1	26.4	43.1	
C	RK19 mix	1	73.7	7.6	13.5	15.5	60.6	1.5
		2	74.9	7.2	12.7	14.7	60.4	
C	RK 55 mix	1	58.0	19.1	18.2	24.4	43.7	1.4
		2	58.1	18.1	17.3	25.0	43.8	
C	RK 91 mix	1	61.9	13.8	15.8	21.0	48.9	1.3
		2	63.1	13.8	15.9	21.1	49.0	

Visual assessment of ΔE76: 0.5–1.5: slight; 1.5–3.0: obvious; 3–6: very obvious; 6–12: large.

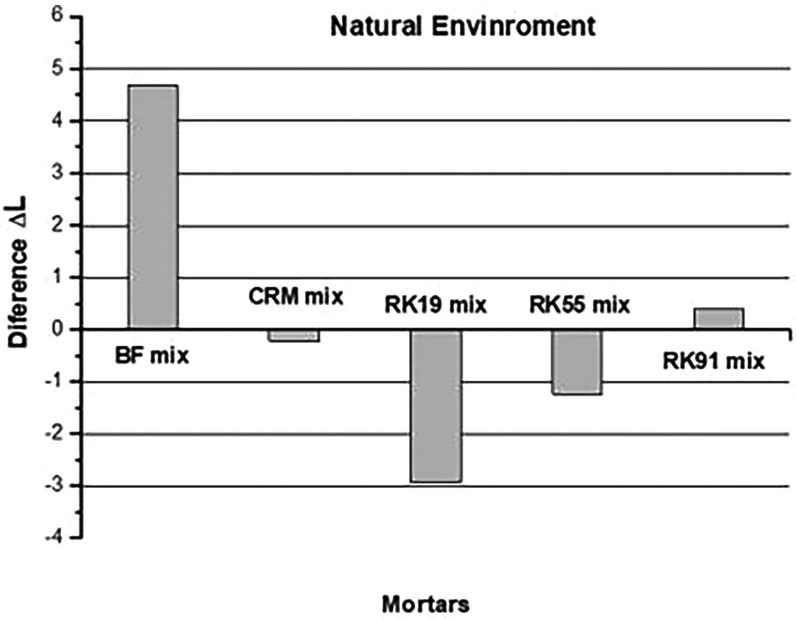

Figure 7: ΔL in mortars.

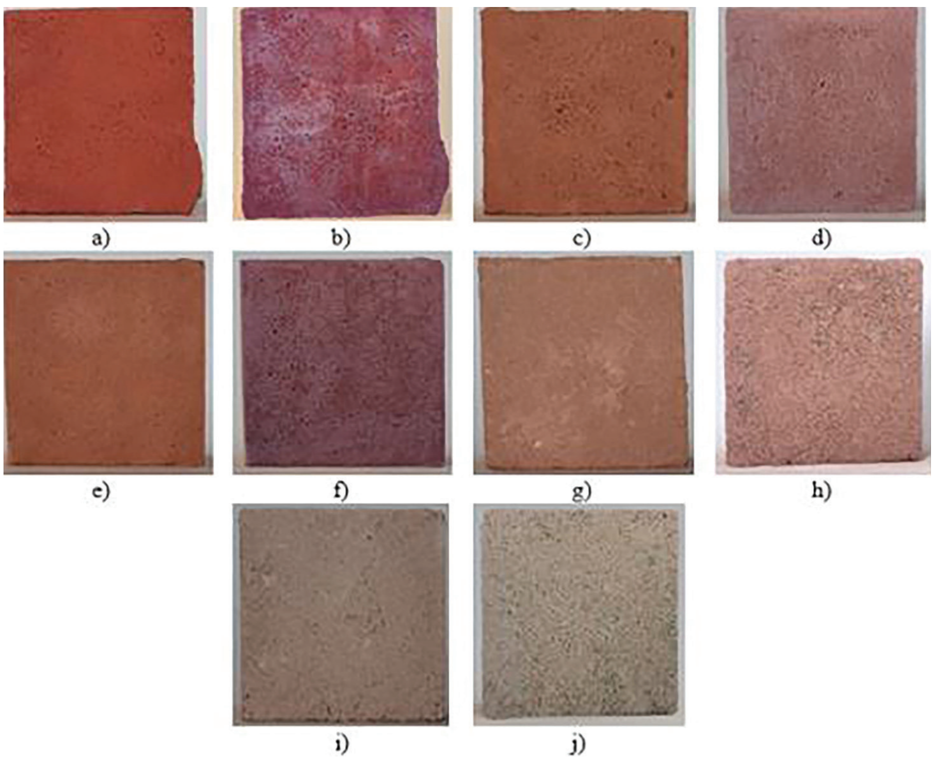

Figure 8: Color changes of BF mortar and with pozzolanic pigments during 8 months of exposure to the natural environment: a) and b) BF mix at 1 and 238 days; c) and d) CRM mix at 1 and 238 days; e) and f) RK91 mix at 1 and 238 days; g) and h) RK55 mix at 1 and 238 days; i) and j) RK19 at 1 and 238 days.

Regarding the luminosity, in the natural environment the ΔL values for the mortars with the pozzolanic pigments ranged from –0.22 to –2.9 units whereas for the mortar BF it was of + 4.68 (Figure 7). This means that for the former, the surfaces have become darker because of the deposition of dirt by the wind, a fact that is inevitable. However, in the mortar with the commercial pigment BF the surface became lighter due to the efflorescence (Figure 8). The pigment BF is inert, therefore, it does not react with salts originating from the hydration of the Portland cement that ascends to the surface, whereas the pozzolanic pigments, preferentially those with higher concentrations of metakaolinite (RK55 and RK19), mitigate the efflorescence due to the pozzolanic reactions. Under controlled laboratory conditions, there were practically no changes in the luminosity of the surfaces of all the mortars studied.

With regard to Δa, parameter that expresses the red color, regardless of the environment, the pigment BF provided a higher value of a* for the mortar, which means a greater red color intensity compared to the pozzolan pigments. This is due to the higher concentration of hematite in the commercial pigment composition. However, pozzolan pigments compensate for the loss in hue with a greater replacement of Portland cement since they allow this because of the higher mechanical strengths. In the natural environment the mortars with pozzolanic pigments presented negative variations between 2 and 8 whereas the one with pigment BF the negative variation was superior to 9 units. This means that the originally red color was substantially altered due to efflorescence and deposition of particulates. The results again demonstrate that pozzolan pigments provide greater color stability than the commercial BF pigment.

4. CONCLUSIONS

The incorporation of pozzolanic pigments from RM and KW mixtures provided substantial increases in compressive strength in relation to commercial BF pigments, reaching gains of 80% at seven days and 67% at 28 days, which is extremely advantageous for the case of colored concrete floors.

The pozzolanic pigments reduced the concentrations of sodium leachate in the mortars between 22% and 57% due to the presence of metakaolinite, which, through pozzolanic reactions, promotes the stabilization of the chemical element by solidification.

As a result of this mitigation of sodium leaching, the pozzolanic pigments also provided greater color stability to the mortars compared to the commercial pigment BF, which had the worst result, with ΔE76 equal to 13.6, while the pozzolan pigments had the highest percentage of metakaolinite (RK55 and RK19), with better performance, showed less noticeable changes in the color of the mortar (ΔE76 between 3.50 and 5.7). Not yet meeting threshold values of naked eye perception of ΔE76 > 1.5 in the Amazon environment.

The pozzolanic pigments showed superior behavior to the commercial pigment in terms of mechanical resistance, reduction of sodium leaching and greater color stability due to the reduction of efflorescence. Only the color intensity was lower, however, this aspect can be mitigated with greater incorporations of pozzolanic pigment without prejudice to the mechanical properties.

In summary, the results demonstrate the promising character of the RM-KW mixtures for the production of pozzolanic pigments as well as mineral admixture for pozzolanic cements of excellent durability and mechanical behavior.

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