Cement-glue with incorporation of cement and recycled aggregate

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Abstract

Cement-based products are a focus of high environmental impact, opposing the new environmental goals of carbon neutrality in 2050, and economic growth decoupled from resource use. Thus, efforts have been directed by the technical and scientific community to find viable alternatives that reduce the use of clinker or allow the production of more eco-efficient clinkers, as well as the reuse of construction and demolition waste. In this context, relevant research has been developed at the superior technical institute, under the EcoHydb project, which aims at the innovative production of recycled cement (RC).

This work aimed to analyze the use of CR and recycled sand (RA) in the production of a new type of cement-glue (CC) marketable, of greater eco-efficiency and sustainability, but technically and economically feasible in the market. Thus, 7 types of SCC were developed, with partial to total replacement of normal cement (CPN) by CR and normal sand (AN) by RA, which were characterized in terms of their applicability (spreading, setting time, transfer, wetting power and water requirement), mechanical strength and adhesion, under variable aging conditions (by immersion in water or heat). This work was carried out in collaboration with the company Weber.

The incorporation of CR decreases workability, with repercussions on mechanical strength and durability properties. However, it was found that for incorporation percentages of up to 50% of CR it was possible to achieve more sustainable CC with applicability and adherence conditions, before and after aging, within the standard.

1. Introduction

The cement industry is one of the largest in the world, with a wide associated value chain. The influence of this industry is felt in many sectors outside the industry, creating a variety of operations that end up having a considerable environmental impact due to the consumption of resources and emission of pollutant gases.

In 2020, 14 billion m3 of concrete were produced which led to the consumption of 4.2 billion tons of cement, 40% of which was for the residential market (association 2020) al. They are therefore materials associated with high consumption of natural resources and high release of greenhouse gases, mainly related to cement production. It is estimated that the cement industry is responsible for about 7% of total anthropogenic emissions worldwide (Kaliyavaradhan & Ling 2017; Mazurana et al. 2022), releasing approximately 800 kg of CO2 per ton of clinker produced (Kaliyavaradhan & Ling 2017). The high environmental impact of cement runs up against the European commission's stringent environmental requirements, which aim for carbon neutrality by 2050, as well as economic growth decoupled from resource use (European Deal 2019). Thus, the cement industry has defined as a priority to strongly reduce its environmental footprint. According to the National Roadmap for the decarbonization of the cement industry (Pereira et al., 2019), several strategies should be implemented in order to enable an overall reduction of CO2 emissions of 75% in 2050 compared to 1990. One of the main levers for reducing the environmental impact of cement, while carbon capture and use systems (CCS/CCU) are not economically and technically feasible, consists in reducing the clinker ratio or using alternative low-carbon cements (Carriço et al. 2020). In this context, the Department of Civil Engineering of the Institute Superior Técnico (IST) has been developing a line of research aimed at the production of recycled cement (RC), thermoactivated from waste concrete, and its incorporation into cement-based materials. Under the ongoing project, EcoHydb (funded by the foundation for science and technology, FCT), in which the present work is integrated, several studies have been conducted to explore the optimization of the production and characterization of CR, aiming its application in the development of more environmentally friendly and sustainable products.

The current state of knowledge in this area is still scarce in view of the youth of the subject. Most of the works published internationally are less than 5-10 years old, and we are still in a phase of development of VC and in the understanding of hydration phenomena and development of phases associated with it (Carriço et al. 2020). Its incorporation in cement-based materials has been essentially limited to the mechanical characterization of pastes and mortars, with few studies conducted at the level of the development of other construction products (Bogas 2021). Thus, in a new stage that follows up on the work that has been done in the EcoHydb project, we intend to explore the use of CR in more environmentally friendly products that can be marketable and increase the competitiveness of companies linked to the construction sector.

2. Experimental program

2.1 Materials

2.1.1 Cement VS Recicled Cement

The cement used in this work to produce the samples of paste and concrete to be recycled, as well as the specimens of cement-glue, was cement type I 42.5 from Secil in Outão. The recycled cement used in this work, as shown in table 2, was produced from original paste and concrete, according to the grinding, separation and thermoactivation procedures referred to in the following sections. The composition of the PO and BO is shown in Table 1, and were produced with pastes with a w/c ratio of 0.55, in order to be representative of existing current concrete.

Table 1 - Compositions

Source material	a/l	M_{cement} (Kg/m^3)	V_{water} (L/m^3)	$Vt_{hick\ aggregates}$ (L/m^3)	$V_{fine.aggregates}$ (L/m^3)
BO	0.55	360	198	406	260
PO	0.55	1032	568	-	-

Table 2 - Cement VS Recicled Cement

Parameters		Cement I 42,5 R	Parameter	Procedure	CR
Residue on the sieve 45 µm (%)		3.5	Absolute density (g/cm3)	Helium pycnometer	3.005
Specific surface (cm^2/g)		4388	$SiO_2+Al_2O_3+Fe_2O_3$ (%)	EN 196-2	19.14+5.13 +3.0
Compressive strength of the reference mortar (MPA)	2 Days	28,6			
	7 Days	40,6	CaO+MgO (%)	EN 196-2	60.79+1.77
	28 Days	52,1			
Activity index (%)		-	CaO livre (%)	EN 451-1	13.94
Expansion (mm)		0,75	Normal consistency (w/b)	EN 196-3	0.74
Ignition loss (%)		3,64	Setting time (min)	Start EN 196-3	290
$SiO_2+Al_2O_3+Fe_2O_3$ (%)		18,49 + 4.95 + 3.61		End	385
Ca0+MgO livre (%)		63,11 + 1,62			
Density (kg/m3)		0,60 + 0,80			
Setting time (min)	Start	130			
	End	210			

2.1.2 Natural and Recicled Sand

For the separation of the concrete components, the separation process developed at IST was followed, which is protected by patent(WO 2021/173022 2021). The detailed description of the followed process is presented in (Hu 2019). For the present work, only the RA resulting from this process was considered. The RA produced had a maximum size of 0.5 mm and the granulometric curve shown in Figure 1. The density of the RA was estimated according to EN 1097-6 (ref.), obtaining the value of 2500 kg/m3. For the reference adhesive cements, a fine 0-1 natural sand was used, usually used in commercial products developed by Weber. This sand with a density mass of 2600 kg/m3 had the granulometry also indicated in Figure 1.

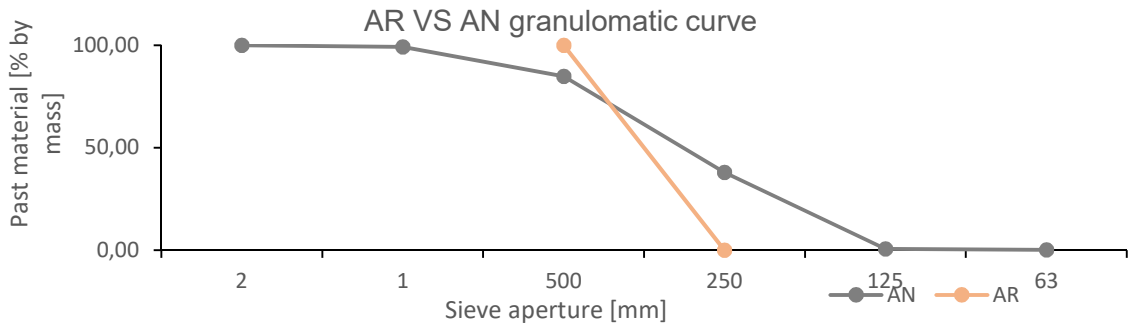

Figure 1 - AR VS AN granulometric curve

2.1.3 Additives

For the production of the adhesive cements, different additives were used in order to improve their applicability and adhesion characteristics. In this case, a water retainer (cellulose ether, EC), an adhesion promoter (RDP resin), a cohesion promoter thickener (starch ether, EA) and a setting accelerator (calcium formate, AP) were used. Their use in adhesive cements was optimized based on weber's experience in previous works, respecting the formulations currently specified for current commercial products

2.2 Adhesive cement formulation

In the present work, 7 types of adhesive cement were analyzed, as shown in Table 3, consisting of CPN and/or CR or FC, natural (AN) or recycled (RA) sand, and additives. The different types of cement-glue (CC) were defined in order to study the separate influence of CR and AN on their behavior. The consideration of FC aimed to better understand the possible contribution of CR as an active agent, promoting extra hydration products.

The reference composition was defined for the mixture composed only by CPN and NA, based on a commercial cement-glue indicated by the weber company. The remaining compositions were refined from the reference composition by trial-error tests, taking into account the different water requirements of their constituents, especially the RC and RA.

Table 3 – Compositions

Materials/Glue-cement	CPNAN	CPNAR	CRAN	CRAR	50CRAR	50FCAN	50FCAR
Cement (kg/m^3)	400	382	0	0	168	191	0
CR (kg/m^3)	0	0	352	301	168	0	167
FC (kg/m^3)	0	0	0	0	0	191	167
AN (kg/m^3)	920	0	810	0	0	880	0
AR (kg/m^3)	0	878	0	693	771	0	769
EC (kg/m^3)	3,57	3,65	3,25	3,38	3,43	3,41	3,2
RDP (kg/m^3)	33,00	31,5	29,06	24,8	27,7	31,6	27,6
EA (kg/m^3)	0,264	0,252	0,23	0,2	0,2	0,252	0,2
AP (kg/m^3)	6,604	6,301	5,81	5,0	5,5	6,312	5,5
Water (kg/m^3)	272	283	335	392	345	287	351
Relation a/l	0,68	0,74	0,95	1,3	1,03	0,75	1,05

2.3 Tests and curing process

The adhesive cement mortars were characterized in the fresh and hardened states in terms of bulk density, spreading, transfer, transmissibility, flexural strength, compressive strength and pull-off strength tests, before and after heat aging and immersion, as shown in table 4. Additionally, we also evaluated the setting time directly on the binder component of the adhesive cements.

Table 4 - Tests performed

Fresh concrete tests	Test standard
Spreading	NP EN 1015-3 (2006)
Compaction of the mixture	NP EN 196-1 (2006),
Density	NP EN 1015-6 2006)
Normal consistency and setting time	NP EN 196-3 (2005).
Transfer and wetting power	EN 12004-2 2017
Hardened concrete tests	-
Flexural strength	NP EN 1015-3 (2006).
Resistance to compression	NP EN 1015-3 (2006),
Pull off adhesion	NP EN 1348 (2000).
Pull off adhesion after heat ageing	(NP EN 1348, 2000).
Pull off adhesion after water immersion aging	(NP EN 1348, 2000).

2.3.1 Mass volume

The density in the fresh state was determined according to the standard (NP EN 1015-6, 2006), using a 1 liter cylindrical container. After the mixture was produced, the cylinder was filled and compacted in two layers. The compaction was done with 25 strokes. The density of the mixture is given by the quotient between the mass of the sample and the volume of the container.

2.3.2 Spreading

The spreading test was performed according to the standard (NP EN 1015-3, 2006). This test aims to determine the consistency of the mixture in its fresh state, measuring the deformability of the cement-glue sample when it is subjected to the repeated action of falling from a table. At the end of the test is recorded the average diameter of spread, measured in 3 different directions.

2.3.3 Normal consistency and setting time

The test for normal paste consistency and setting time was performed according to the standard (NP EN 196-3, 2005). This test is designed for binders, giving an idea of their water requirement and the speed of development of the first reactions that lead to their loss of plasticity.

2.3.4 Transfer and Wetting Power

The transfer and wetting power tests were performed according to an internal procedure defined at Weber, based on the standard (EN 12004-2, 2017) and (EN 1347, 2006). For this purpose, a square concrete slab with a side of about 30 cm and a thickness of 5 cm is used. Initially, a first layer of adhesive cement is spread, with the help of the smooth part of a toothed trowel, on the concrete slab. After this first layer, a second layer is spread using the toothed part of the trowel. This second layer is "combed", in order to facilitate subsequent adhesion conditions.

2.3.5 Bending strength

The bending strength was determined on prismatic specimens of 160x40x40 mm3, according to the standard (NP EN 1015-3 , 2006). The specimens were placed on two cylindrical supports, 100 mm apart, and then a point load was applied at mid-span of the specimen. The test is performed at a speed of 0.1 KN/s until failure, and the value of the maximum applied load is recorded. The strength is calculated according to equation 1.

$$f_t = 1{,}5 \times \frac{F_t \times l}{b \times d^2}$$

2.3.7 Compressive Strength

The compressive strength test was performed according to the standard (NP EN 1015-3, 2006), based on the two halves obtained from the bending test. Each half is placed on a 40 mm wide base in order to obtain a contact area of 40x40 mm2. This test is performed at an average speed of about 0.4 kN/s, and the maximum applied load is recorded. The compressive strength is calculated according to equation 2.

$$f_c = \frac{F_t}{A}$$

2.3.8 Pull-off adhesion test without aging

The pull-off test was performed according to the standard (NP EN 1348, 2000). Initially a thin layer of cement-glue is applied on a square concrete slab 5 cm thick, through the smooth part of a toothed trowel. Then another thicker layer is applied and "combed" with the toothed part of the trowel, presenting notches of about 6x6 mm. During the application, the trowel must be kept at an angle of approximately 60° to the slab. After 5 minutes of rest, tiles with 50 x 50 mm2 are applied on the adhesive cement, and then a force of 20 N is applied for 30 seconds. This force is applied using a 2 kg weight.

2.3.9 Pull-off adhesion test with heat aging

In the heat aging tests, the specimens are subjected to a curing scheme prior to the adhesion tests (NP EN 1348, 2000). After bonding the tiles, the specimens are initially conditioned under standard conditions, in air, for 14 days, followed by curing in a ventilated oven for 14 days at 70 °C. Then, the metallic discs are placed and the specimens are conditioned for another 24 hours under standard conditions. After this curing period, the adhesion test is performed. The tensile strength is determined by applying a force of 250 N/s at constant speed until failure.

2.3.10 Pull-off adhesion test with water immersion aging

For the water immersion aging tests, the specimens are initially air cured for 7 days, followed by 20 days immersed in water at a standard temperature, 23 ± 2 °C (NP EN 1348, 2000). After being rinsed, the metallic discs are applied and after 7 hours they are immersed again in water until the next day. Finally, the adherence test is performed. The tensile strength is performed at 28 days and is determined by applying a force of 250 N/s at constant speed until failure.

3. Results and discussion

3.1 Fresh state properties

3.1.1 Fresh density and air content

The substitution of natural sand by recycled sand led to a slight increase in the a/l ratio (3%). On the one hand, although the separation method allows a significant removal of adhered paste, the estimated content of about 5% contributes to a slight increase in the amount of mixing water. Considering 2.3% absorption in RA, this corresponds to a/l variations in the order of 0.05 (8%).

Still, the void content was similar in the CPNAN and CPNAR mixtures, leading to similar compaction efficiencies. This also suggests that the higher a/l ratio was essentially associated with the water absorbed by the aggregate. The high void content obtained in these 2 mixtures is noteworthy, much higher than that usually observed in current mortars. The replacement of CPN by CR led to a significant increase in the w/b ratio, of about 44%. However, contrary to expectations, despite the fact that the CR colored cements were produced with higher a/l ratio and that CR had a higher density than CPN, the density was 13% higher than that of the CPN colored cements. Taking into account the dosage of its constituents, this led to a void content of only 17%, almost half of that obtained with CPN.

The development of a 100% recyclable product, composed of only CR and AR, led to higher water requirements, making it difficult to achieve the target spread for reasonable a/l values. The increase in a/l with the additional introduction of RA was similar to that seen in the CPN mixtures. Again, higher density mass and lower void content were obtained than in the reference mix, confirming the low efficiency of the CE.

Finally, the partial replacement of 50% CPN by an equal content of CR led to an increase in the a/l ratio less than the intermediate of that obtained in the CPNAR and CRAR mixtures. This increase was only 17.6% compared to CPNAR. On the other hand, in contrast to what was observed in the other adhesive cements with CR, the density and the void content were of the same order of magnitude of that obtained in the reference mixture with only CPN. This indicates that the CE will have worked and been compatible with the mixed binder of CR and CPN in this mixture. In this case, the amount of adsorbed CE or the development of heat of hydration will not have been sufficient to deactivate the function of the CE,

allowing the generation and distribution of microbubbles in the cement-glue matrix. This introduction of air contributes to increased workability, bringing the a/l ratio closer to that used in reference adhesive cements with CPN. This results can be seen in table 5 below.

Table 5 - Compositions

Composition	a/l	Thickness (mm)	Mvfresh (Kg/m3)	Voids content (%)
CPNAN	0,66	141	1419	31,5
CPNAR	0,68	139	1397	31,1
CRAN	0,95	145	1598	17,2
CRAR	1,00	135	1584	15,9
50CRAR	0,80	140	1415	28,1

3.1.2 Setting Time

Considering the results in Table 6 and 7, only mixtures with up to 50%CR developed a setting time up to 24h. In general, when compared to the 1stphase, the w/b ratio and the CE content were increased, which tends to delay the setting time. Thus, the tendency was to increase the setting time in the various mixtures. Only CPNAN had its setting time reduced, since the CE content was decreased by 4%. In the opposite direction, the setting time of CPNAR increased due to the increase in w/b and CE content (3% increase).

The mixture with 100% CR had the longest delay in setting time, which was only completed after 4 days. In this case, the increase of 21% in the EC content will have contributed to a significant delay in the reactions of the CR, which is not compatible with its use at 100% in adhesive cements. Note that this delay is not characteristic of the RC, since previous studies show a higher reactivity of this cement than of CPN up to 3 days of age (Bogas et al. 2020; Real et al. 2020). It is still noteworthy the fact that the mixture with up to 50% CR and 100% RA met the end of setting at the limit. This means that mixtures with AN and up to 50% replacement of CPN by CR should meet the 24-hour limit. The replacement of 50% PCN by 50% CF in mixtures with cement or with CR increased the setting time, even when this incorporation implied a reduction in the CE content, as is the case of 50CRFCAR versus 50CRAR. This phenomenon is related to the dilution of the clinker content in the mixture, increasing the equivalent w/c of the mixture and reducing its reaction speed and development of hydrated products. Thus, although FC facilitates the applicability and wettability of the clinker-cement, it reduces its reactivity. In this case, the mixture of 50%FC and 50%CR does not allow reaching viable hardening conditions for its use in cement-glue. Curious is the fact that the CRARgel mixture showed a lower setting time than CRAR, despite having been produced with higher a/l and higher Ec content.

Table 6 - Initial and end setting time - phase 1

Test mixture	a/l	Arrest (h)	Remarks
CPNAN	0,66	12 - 15	Harden through thick and thin
CPNAR	0,68	12 - 18	Hardens through thickness
CRAR	0,95	>24	Surface hardening
50CRAR	0,80	>24	Hardens on surface

Table 7 - Start and end of setting time - phase 2

Test mixture	a/l	%EC	Arrest (h)	Remarks
CPNAN	0,68	0,27	<12	-
CPNAR	0,74	0,29	<24	-
CRAR	1,20	0,32	>24-96	96 hours arrested completed
50CRAR	1,03	0,31	24	Prey completed
50FCAN	0,75	0,27	<24	-
50CRFCAR	1.05	0,29	>24-48	48 hours lock-up completed
CRARgel	1,3	0,34	>24-48	48 hours incarceration completed

3.1.3 Wetting power and transference

Excluding CRAR at 28 days, all mixtures reached adhesion values higher than 0.5 MPa, as recommended in (EN 1348, 2007). Although adjusting a/l and the CE content improved the applicability of CRAR, it still showed an unsatisfactory transfer ability immediately after the first few minutes of application. Again, this was reflected in the higher percentage of AFT breakage observed in these mixtures. Note that the tiles were applied 5 minutes after placing the adhesive cement, a situation in which the mixture was already parched. Even so, with the new changes, only at 28 days a value below 0.5 MPa was obtained. Even after adjusting the composition and the CE content, the incorporation of RA led to a slight reduction in the average bond strength (8% at 28 days). In this case, the rupture in both cases occurred by cohesion (CFA), which indicates a lower resistant capacity of CPNAR. This is justified by the fact that the total a/l was 9% higher in CPNAR than in CPNAN. Even assuming absorption up to saturation in the RA (2.3%), the effective a/l would be 8% lower, going to about 0.67, still higher than the effective a/l of CPNAN. In any case, it is shown that the use of recycled fine sand is feasible for the production of more eco-efficient cementitious adhesives, having achieved solutions with average bond strengths higher than 1 MPa from 7 days of age. In the intermediate situation of 50% substitution of CPN by 50% CR, it was possible to achieve a balanced solution with good adherence capacity and high rate of incorporation of recycled material (100% RA and 50%CR). In this case, the adjustments imposed to the mixture, with considerable improvement in its applicability up to 20 minutes (change in the content and type of CE, more stable at high temperature), allowed the rupture to occur by CFA, for average tensions of rupture close to 1 MPA, even taking into account the 50% increment in the a/l ratio against CPNAN.

It was found that replacing 50% of CPN or CR by FC improved the mixtures' applicability. In this case, the various test specimens showed cohesion failure (CFA). On the one hand, the wetting capacity is high and on the other hand, these binder mixtures have lower hydration capacity and, as such, lower strength. Thus, the replacement of 50% CPN by 50% FC led to a dilution of the clinker content and a reduction in the equivalent w/c, obtaining a 50% reduction in bond strength at 7 days and 30% at 28 days. Still, it was possible to achieve very reasonable solutions, with stress greater than 0.8 MPa at 28 days. In the cements-glue with CR the replacement of CPN by FC, reaching a much more coefficient solution, led to little significant reductions in the average bond strength at 28 days, compared to 50CRAR. However, there were reductions of up to 44% at younger ages. This results can be seen in table 8 below.

Table 8 - Wetting power and transfer

Misture	a/l	%EC	Wetting power (%)				Transfer (%)			
			5 min	10 min	20 min	30 min	5 min	10 min	20 min	30 min
CPNAN	0.68	0.27	100	100	90	>75	70	>70	>70	>60
CPNAR	0.74	0.29	100	95	>80	>70	>50	>70	>65	20
CRAR	1.20	0.32	95	>80	50	30	<40	<10	0	0
50CRAR	1.03	0.31	100	90	50	50	90	80	50	0
50FCAN	0.75	0.27	100	100	100	>90	80	70	>75	>60
50CRFCAR	1.05	0.29	100	100	100	90	100	100	95	0
CRARgel	1.3	0.34	100	95	90	50	100	90	10	0

3.2 Hardened state properties

3.2.1 Compressive strength e flexão

Despite the difference in a/l ratio, the compressive strength of the cementitious adhesives with CPN was little affected by the replacement of AN by RA. In fact, there was a slight increase in compressive strength in the mortar with RA. Besides the variability of the test itself, this is essentially related to a possible reduction of the effective a/l of CPNAR. In fact, although the total a/l ratio was higher in CPNAR, the effective a/l is similar to lower, since part of the water absorbed in the aggregate does not participate in the hydration and increase of matrix porosity.

The lowest strengths were achieved in mixtures with 100% CR, which is associated with higher a/l.

In comparison with the reference mixtures with CPN, in fact, a greater proximity in the resistances at 7 days than at 28 days is observed. However, despite the possible lower efficiency of CR and the higher a/l ratio (43-47%), the reduction in compressive strength at 28 days was less than 29%. In the mixture with 50% CR and 50% CPN, an intermediate strength of CPNAR and CRAR was obtained.

However, given the great influence of the void content on the mechanical strength of cement base materials, for which 1% of air content can mean a 5% reduction in mechanical strength, it is not possible to conclude strictly on the relative performance of the RC compared to CPN. This results can be seen in the table 9 below.

Table 9 - Density and compressive and flexural strength of cement-glues.

Misture	a/l	Mv_{28d} (Kg/m3)	V_{V28d} (%)	Compressive Strength (MPa)			Flexural Strength (MPa)		
				7 Dias	28 Dias	CV_{28d}	7 Dias	28 Dias	CV_{28d}
CPNAN	0,66	1536	49.3	2,3	4,3	3%	1,69	1,97	5%
CPNAR	0,68	1495	49.2	3,76	5,5	3%	1,62	2,22	3%
CRAN	0,95	1684	46.6	1,92	3,3	1%	0,43	1,47	5%
CRAR	1,00	1713	46.5	3,04	3,9	5%	1,40	1,77	3%
50CRAR	0,80	1654	50.0	3,09	4,2	5%	1,43	2,12	4%

3.2.2 Pull off bond strength tests, without aging

Unlike CRAR, at 28 days, all mixtures reached adhesion values higher than 0.5 MPa, as recommended in (EN 1348, 2007). Adjusting a/l and CE content improved the applicability of CRAR, that only at 28 days shows a value below 0.5 MPa. In turn, the use of cold water and the additional correction of the w/b ratio and EC content (higher water retention) in the CRARgel mixture allowed to increase the transfer capacity, reflected in the reduction of the percentage of failure by AFT and in the increase of the average bond strength. Even after adjusting the composition and CE content, the incorporation of RA led to a slight reduction in the average bond strength (8% at 28 days). In this case, the rupture in both cases occurred by cohesion (CFA), which indicates a lower resistant capacity of CPNAR. This is justified by the fact that the total a/l was 9% higher in CPNAR than in CPNAN. Even assuming absorption up to saturation in the RA (2.3%), the effective a/l would be 8% lower, going to about 0.67, still higher than the effective a/l of CPNAN. In any case, it is shown that the use of recycled fine sand is feasible for the production of more eco-efficient cementitious adhesives, having achieved solutions with average bond strengths higher than 1 MPa from 7 days of age. The adjustments promoted in the mixture allowed to ensure a higher percentage of failure by CFA. In relation to CPNAN, there was an increase in the percentage of failure by AFT, which is justified by the slight reduction in CE content and water reduction capacity. In the intermediate situation of replacement of 50% CPN by 50% CR, it was possible to achieve a balanced solution with good adherence capacity and high rate of incorporation of recycled material (100% RA and 50%CR). In this case, the adjustments imposed to the mixture, with considerable improvement in its applicability up to 20 minutes (change in the content and type of CE, more stable at high temperature), allowed the rupture to occur by CFA, for average tensions of rupture close to 1 MPA, even taking into account the 50% increment in the a/l ratio against CPNAN. This results can be seen in table 10 and 11 below.

Table 10 - Failure mode of non-aging specimens (mean values of failure area - phase 2)

Composition	Burst Mode (7 days)	Burst Mode (14 days)	Burst Mode (28 days)
CPNAN	80 CFA 20 AFT	95 CFA 5 AFT	95 CFA 5 AFT
CPNAR	90 CFA 10 AFT	90 CFA 10 AFT	90 CFA 10 AFT
CRAR	40 AFT 60 CFA	60 CFA 40 AFT	60 CFA 40 AFT
50CRAR	CFA	CFA	CFA
50FCAN	40 AFT 60 CFA	90 CFA 10 AFT	80 CFA 20 AFT
50CRFCAR	100 CFA	100 CFA	100 CFA
CRARgel	50 CFA, 50 AFT	50 CFA, 50 AFT	50 CFA, 50 AFT

It was found that replacing 50% of CPN or CR by FC improved the mixtures' applicability. In this case, the various test specimens showed cohesion failure (CFA). Thus, the replacement of 50% CPN by 50% FC led to a dilution of the clinker content and a reduction in the equivalent w/c, obtaining a 50% reduction in bond strength at 7 days and 30% at 28 days. Still, it was possible to achieve very reasonable solutions, with stress greater than 0.8 MPa at 28 days. In the cements-glue with CR the replacement of CPN by FC, reaching a much more coefficient solution, led to little significant reductions in the average bond strength at 28 days, compared to 50CRAR. However, reductions of up to 44% were seen at younger ages.

Table 11 - Adhesion of non-aging specimens (mean bond strength values - phase 2)

Composition	μ_m (7 dias)	Cv 7 days	μ_m (14 dias)	Cv 14 days	μ_m (28 dias)	Cv 28 days
CPNAN	1,29	2%	1,25	1%	1,29	1%
CPNAR	1,03	1%	0,95	1%	1,19	2%
CRAR	0,5	5%	0,51	5%	0,4	4%
50CRAR	0,90	1%	1,04	2%	0,88	4%
50FCAN	0,645	5%	0.83	2%	0.90	2%
50CRFCAR	0,76	2%	0,58	4%	0,81	1%
CRARgel	0,52	3%	0,58	4%	0,65	2%

3.2.3 Pull-Off bonding tests, water immersion aging

All mixtures tested at this stage showed loss of adhesion capacity after immersion in water. This reduction was 26%, 20%, 5%, 28%, 43%, 79% and 80% in mixtures with CPNAN, CPNAR, CRAR, 50CRAR, 50FCAN, 50CRFCAR and CRARgel, respectively. This reduction in adhesion was generally accompanied by an increase in the percentage of AFT failure, especially in mixtures where the percentage of reduction was greater than 40%. The mixtures most affected by immersion aging were those in which 50%FC or 100% CRAR was incorporated. After immersion, the CRAR (100%CR) and 50CRFCAR mixtures showed bond strength less than 0.5 Mpa. In the CRARgel mixture, the better transfer capacity allowed for improved contact and effectiveness of the resin between the cement-glue and the tile, which was lost when the resin was attacked by saponification. It is recalled that mixtures with 100% CR have high alkalinity, due to the high free lime content in the binder.

Although 50FCAN showed an average bond strength of 0.5 MPa after immersion, the loss observed in its adhesion capacity highlights the poor contribution of its cementitious fraction to this property. The fact that the mixture with 50%CR and 50%CPN (50CRAR) showed an adequate value of bond strength, higher than 0.6 MPa, even after immersion aging, is noteworthy. The loss of adherence compared to the reference mixture CPNAN was 34%, close to the 32% obtained without immersion. This results can be seen in table 12 below.

Table 12 - Bond strength and failure mode after immersion in water (phase 2)

Compositions	μ_m (28 days)	Cv 28 days	Burst Mode (28 days)
CPNAN	0,96	1%	90 CFA 10 AFT
CPNAR	0,95	2%	90 CFA 10 AFT
CRAR	0,38	5%	95 AFT 5 CFA
50CRAR	0,63	2%	CFA
50FCAN	0,52	2%	85 CFA 15 AFT
50CRFCAR	0,17	3%	100 AFT
CRARgel	0,13	0%	50 AFT, 50 CFA

3.2.4 Pull-OFF Adhesion Tests, Heat Aging

The various tested mixtures experienced a reduction in bond strength after heat aging. These reductions were 5%, 17%, 10%, 12%, 74% and 66% for CPNAN, CPNAR, 50CRAR, 50FCAN, 50CRFCAR and CRARgel, respectively. In the CRAR blend the stress remained practically unchanged. The high sensitivity of the 50CRFCAR and CRARgel mixtures to this test is noteworthy, with significant losses of bond strength. This results can be seen in table 13 below.

Table 13 - Adhesion and failure mode after heat aging (average values of adhesion and failure area)

Compositions	μ_m (28 days)	Cv 28 days	Burst Mode (28 days)
CPNAN	1,23	1%	100 CFA
CPNAR	0,99	2%	95 CFA 5 AFT
CRAR	0,43	2%	45 CFA 55 AFT
50CRAR	0,79	1%	CFA
50FCAN	0,79	2%	70 CFA 30 AFT
50CRFCAR	0,21	3%	50 CFA, 50AFT
CRARgel	0,22	5%	50 AFT, 50 CFA

4. Conclusion

The development of 100% recycled cementitious adhesives has led to solutions with high water requirements and rapid desiccation, making them difficult to apply. The lower efficiency of the CE also contributes to this. In these mixtures with 100% RC it was necessary to significantly correct the w/b ratio and the CE content to achieve reasonable conditions of applicability, reaching solutions in which these factors were increased by up to 76% and 17%, respectively, compared to CPNAR. Even so, they always presented the lowest bond strength values, as well as the highest percentages of adhesion failure (AFT). These adhesive cements are characterized by rapid drying out, losing plasticity after a few minutes. Only acceptable wetting power and transfer conditions were guaranteed up to 10 minutes of waiting time, and the setting time was longer than 24 hours. The bond strength, even under non-aging conditions, was slightly less than 0.5 MPa. The high water requirement and loss of workability of CR makes its 100% use in adhesive cements unfeasible.

The partial incorporation of 50% CR replacing CPN combined with the use of 100% CR led to the most balanced solution in terms of technical performance and eco-efficiency. Under these conditions, the incompatibility and loss of efficiency of CE was not confirmed, allowing to achieve solutions of similar density mass to the reference mixtures with CPN. The compressive and tensile strengths were little affected by the incorporation of 50% CR, being in line with the results obtained in other previous works with cementitious base mixtures constituted by up to 40% CR.

After adjusting the a/l and the content and type of Ec, the applicability of 50CRAR was satisfactory up to 20 minutes of waiting (wetting power and transference). In turn, it was found that only up to 50% CR it is possible to guarantee setting times longer than 24 hours. Under these conditions, the average bond strength was only slightly less than 1 MPa between 7 and 28 days of age. These adhesive cements also show satisfactory behavior after immersion and heat aging, with relative losses similar to those observed in the reference mixtures with CPN.

It can be concluded that up to 50% of substitution of CR by CPN is feasible the development of commercial cements-glue with high eco-efficiency, associated with high percentages of incorporation of recycled material (50% of binder plus 100% of aggregate).

The substitution of CPN or CR by 50% FC allowed for improved applicability conditions, after proper adjustment of a/l and Ec. Its use promotes a reduction in clinker content and in the equivalent a/c ratio, reducing the reactivity of the cementitious matrix. This is reflected in the increase of the setting time and the reduction of the bond stress. The synergistic use of CF and CR allows to compensate part of the water requirement and poor applicability provided by CR, as well as the poor reactivity of CF. This led to improved solutions compared to 100% CR, with bond strengths greater than 0.8 MPa at 28 days. However, mixtures with 50% CF and 50% RC were not able to achieve viable hardening conditions, exhibiting setting times greater than 24 hours. On the other hand, the incorporation of 50% CF led to high reductions in bond strength after aging, showing its high dependence on the adhesion capacity and integrity of the resin.

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