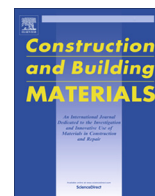




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# Construction and Building Materials

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## Laboratory-scale study and semi-industrial validation of viability of inorganic CDW fine fractions as SCMs in blended cements



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### HIGHLIGHTS

- Cementitious potential as SCMs with optimal substitutions of 5–7% of clinker.
- Filler effect and pozzolanicity induced by the recycled SCMs.
- Neoformation of hydrated phases and C-S-H gel.
- Accelerated hydration and setting time and maintain of the mechanical strength.
- Pilot validation with the manufacture of 184 t of blended cement.

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### ABSTRACT

The construction industry and more particularly cement manufacture industry are European Green Deal strategic priorities for the circularity of Europe's construction and demolition waste (CDW) stream with a view to reducing CO<sub>2</sub> emissions. The industry is engaged in a number of strategies to that end, one of which is to manufacture new low-carbon, lower clinker/cement ratio cements by replacing portland clinker with inorganic fractions of CDW featuring hydraulic or pozzolanic properties. Against the backdrop of that global challenge, the present study explores the cementitious potential of the limestone and siliceous concrete fines and shatterproof building glass found in CDW as supplementary cementitious materials (SCMs) in new blended cements. The research was conducted in two stages: generation of new laboratory-scale knowledge; and industrial validation of the viability of using the highest volume waste streams. The laboratory-scale findings revealed that the presence of the filler effect and pozzolanicity in micronised inorganic fractions of concrete and building glass waste induces the neoformation of hydrated phases and C-S-H gel. Those two developments improve the short- and long-term physical and mechanical properties of the new blended cements at optimal replacement ratios of 5–7%. The order of material effectiveness in shortening setting times, increasing the heat of hydration and maintaining mechanical strength was observed to be as follows: limestone concrete > siliceous concrete > glass waste. Laboratory analysis was followed by a pilot study consisting in the manufacture of 184 t of blended cement in which 5% of the clinker was replaced by recycled concrete. Higher product performance than the commercial reference cement confirmed the industrial, technical, economic and environmental viability of the new product, estimated to hold potential for CO<sub>2</sub> emissions abatement on the order of 41 kg CO<sub>2</sub> eq./t of cement, which could translate into 80 Mt CO<sub>2</sub> eq./year worldwide.

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### 1. Introduction

As one of the key elements in the construction value chain, cement manufacture will play a significant role in attaining climate

neutrality by 2050 [1–3]. Four strategies are being implemented by the European cement industry to neutralise CO<sub>2</sub> emissions: process electrification; CO<sub>2</sub> capture, use and purification; reduction in energy and raw material consumption; and design of new low-carbon cements to lower the clinker/cement ratio from the present 0.8 to 0.6 by 2050. Research around the fourth focuses on the pursuit of new mineral additions with hydraulic or pozzolanic proper-

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ties [4]. Those strategies are geared to transitioning to a clean and circular economy, lowering pollution and emissions and protecting biodiversity. The EU's Action Plan also specifies the investments and tools that will be needed to move the process forward.

The search for new mineral additions focuses on ubiquitous, high-volume waste or by-product streams such as CDW. That waste comprises around 52% concrete (Rc), 27% ceramic elements (Rb), 15% natural aggregate (Ru), 1% asphalt (Ra) and a further 1% glass (Rg) [5]. Europe alone generates around 374 Mt/year of CDW [6], only 50% of which is recycled, most in low added value applications such as granular material for road fills and sub-bases [7]. There is, then, a good deal of room for improvement in the recovery and use of the mineral resources in CDW for high added value use. Advanced technologies have been developed in recent years to identify, separate and recover mineral fractions [5,8–11] with latent hydraulic or pozzolanic properties such as concrete, ceramic [12,13] materials, glass and mineral wool for high added value applications including new SCMs in binary, ternary or quaternary cement manufacture [4,14,15].

The reuse of recycled concrete has traditionally been constrained to its application as coarse aggregate in new concretes [16], at a value of up to €10/t. As the fines fraction (<5 mm) is harder for this valorization in concrete due to its intrinsic properties [17,18], however, alternative applications must be sought. That fraction is now being repurposed as an SCM in cements at an added value of close to €30/t and as a soil stabilizer in the road construction sector and other municipal applications [19–22]. It holds particular interest for its high concentration of cement paste and cementitious potential [4,14].

The concretes used in Europe can be divided into those bearing siliceous (41%) and those with limestone (46%) aggregates [23]. The chemical and mineralogical differences between those two types of recycled concrete necessitate separate exploration of their behaviour as SCMs in cements. A number of solutions is presently being studied to induce both hydraulicity and pozzolanicity in recycled concrete mineral fractions for use as SCMs in cement, including micronisation and joint milling with clinker at cement plants [24,25], along with advanced heating-air classification systems (HAS) to produce ultrafine high cement paste particles from the fines fraction of recycled concrete. Although the benefits of HAS technology products have been recently addressed by the present authors [4], less is known about the effect of micronised concrete fractions on cement matrices.

Another inorganic fraction in CDW with pozzolanic potential is shatterproof glass waste (WG). As no clear option for valorising WG is presently in place, new applications must be found for that waste stream. Research on valorising glass-based MSW (bottles and vehicle scrap) as SCMs in cements on the grounds of its pozzolanicity [26,27] to replace up to 30% clinker with waste glass powder has revealed that an initial decline in early age properties relative to traditional cement is reversed after 90 days. Nonetheless, the use of ground shatterproof building glass as an SCM has not been explored in any depth.

The present study focuses on the use of the two basic types of inorganic fractions in recycled CDW about which least is known for application as SCMs in new blended cement manufacture, namely the fine fractions of recycled limestone (RCL) and siliceous (RCS) concrete, and shatterproof building glass waste (WG). As the scant research conducted on those three streams to date has been limited to laboratory analysis, this study addresses issue from the following innovative perspectives.

- Study of three types of inorganic fractions from CDW crushing and recycling: recycled limestone (RCL) and siliceous (RCS) concrete fines (<5 mm) and shatterproof building glass (WG), analysed for cementitious potential of the recycled concrete

particles not immediately after crushing as in other studies, but after exposure to the elements during storage in valorisation plant yards.

- Collection of scientific information on the hydration products to enhance the understanding of the cementitious processes involved in CDW mineral fraction air-jet milling/micronisation.
- Comparison of the cementitious activity of the recycled fractions of siliceous and limestone concrete and glass, identifying the benefits of each and the optimal replacement ratios for use as SCMs in new blended cements.
- Implementation of an industrial pilot study to assess the consistency between the laboratory- and plant-scale findings and establish the industrial viability of the use of such recycled fractions as SCMs, comparing the two sets of results to analyse the effect of milling on the efficiency of the cementitious activity of the micronised inorganic fractions.

The ultimate aim is to lower the environmental impact of cement manufacture without affecting the product's economic and technical viability by reducing the use of raw materials and CO<sub>2</sub> emissions. The benefits of this process have been proven by authors [4,28] adopting similar approaches although with other types of technologies. In such studies CO<sub>2</sub> emissions abatement was estimated to be of the same order of magnitude as the cement replacement. Consequently, the use of SCMs at a 5% replacement ratio would lower CO<sub>2</sub> emissions by around 41 kg CO<sub>2</sub> eq./t of cement, which on the worldwide scale would signify a yearly abatement of 80 Mt of CO<sub>2</sub> eq.

## 2. Experimental

### 2.1. Materials

Seven samples of inorganic CDW fractions were used in this study (Table 1). A total of 100 kg of each of three recycled three limestone and three siliceous concrete fines (<5 mm), consisting respectively in concretes initially manufactured with natural limestone or natural siliceous aggregates, were selected and sampled according to the standard CEN/TR 15310-1 [29] in order to maximize the homogeneity. The six waste management plants supplying the materials were located in different areas of Spain, three in the Basque Country where limestone quarries prevail and three in the Region of Madrid, where siliceous aggregate quarries predominate. In addition, 90 kg of shatterproof glass were recovered from the selective demolition of a residential building in the Basque Country.

All the fractions of the starting recycled concrete consisted in fines with a particle size of < 5 mm obtained by crushing and sieving the initial concrete waste that had been stored in the open air in management plant yards. The fines fraction was chosen for this study for two reasons: first, for its suitability as a mineral addition in cement given its cement paste content; and second, to enhance

**Table 1**  
Origin, labelling and initial particle size distribution of the recycled CDW studied.

Origin	Mgmt plant	Label	Starting fraction
Basque Country Recycled limestone concrete aggregate	Gutram	RCLG	<5 mm
	Lasuen	RCLL	
	Volbas	RCLV	
Region of Madrid Recycled siliceous concrete aggregate	Tecrec	RCST	<5 mm
	Casar	RCSC	
	Salmedina	RCSS	
Basque Country Shatterproof glass waste	Demolition	WG	0–40 mm

the circularity of this fraction which compromises the durability of new cement eco-matrices due to its high water absorption, impurities and the presence of undesirable compounds present in the CDW itself (sulfates and chlorides) or the result of environmental pollution (clay minerals) during storage at the management plant.

The European standard EN 197-1: 2011 compliant Spanish commercial OPC used, CEM I 52.5 R, was supplied by Cementos Lemona, S.A., a facility operating out of Bilbao, Basque Country, Spain. That same cement plant manufactured the cements for the industrial pilot from: a) 92.8% portland clinker with a Rietveld-determined mineralogy consisting in 63.02% C<sub>3</sub>S, 13.67% C<sub>2</sub>S, 6.33% C<sub>3</sub>A, 10.59% C<sub>4</sub>AF and 6.09% minority phases; b) 2.6% of 98% pure quarried natural gypsum; and c) 4.6% of recycled limestone fines (RCLG) as a supplementary cementitious material, as described in the following section.

## 2.2. Laboratory-scale preparation of materials

The recycled concrete fines (<5 mm) were dried in the laboratory at 105 °C for 24 h to eliminate moisture and subsequently ground in a ball grinder to a particle size of < 63 µm. The glass waste was initially ground to < 5 mm before grinding. The ground samples are depicted in Fig. 1.

The blended cements were prepared by replacing 5%, 7% or 10% of the OPC with each micronised inorganic addition, blending the two dry materials in a high speed power mixer. The ratios used were adopted further to recommendations in Moreno-Juez et al. [4,26,27].

The blended cement mortars were batched and cured as laid down in European standard EN 196-1:2018 [30], at water/binder = 1/2 and sand/binder = 3/1. The 4 × 4 × 16 cm<sup>3</sup> prismatic specimens prepared were demoulded after 24 h and cured in water for 2 d, 28 d or 90 d. Mortar labels and batching are given in Table 2.

## 2.3. Plant-scale preparation of materials

After laboratory testing, the use of inorganic fractions of limestone concrete from recycled CDW as a mineral addition was validated at an industrial facility (Cementos Lemona, S.A.). Given the complexity of the test and the lack of prior references, the industrial trial was conducted on only one of the laboratory-tested samples. RCLG, recycled limestone concrete from the Gutram waste management plant, was selected for the trial on the grounds of local availability of the resource (limestone aggregate in the Basque Country), volume (concrete accounts for 50% of CDW, com-

pared to 2% glass) and mechanical performance as reported in the literature and observed in the present study. Further details on this selection are given in the section on pilot test results.

At the plant, the < 5 mm recycled concrete was mixed directly with the clinker during the grinding to ensure a uniform blend. A commercial OPC was manufactured as a reference for the industrial trial. The resulting cements were labelled OPC-Pilot and 5%RCLG-Pilot, batched as listed in Table 3.

The natural gypsum used was sourced from a quarry located in the Spanish province of Burgos. Clinker chemical and mineral composition are given in Table 4.

Industrial production proceeded as follows:

- crushing, sieving and recovery of 25 t of < 5 mm recycled concrete aggregate (RCLG) from a local waste management plant transported to the Cementos Lemona facility
- manufacture of 184 t of 5%RCLG-Pilot blended cement with 169.8 t of clinker, 8.5 t of recovered mineral RCLG, 4.8 t of commercial gypsum and 1.3 t of ferrous sulfate for 3 h and 45 min, for an output of 49 t/h, and milling the commercial OPC-Pilot cement under identical conditions.

## 2.4. Experimental methodology

The experimental programme was divided into three stages (Fig. 2): characterisation of the starting mineral additions prepared from recycled inorganic concrete fines; laboratory testing; and industrial-scale manufacture and validation.

## 2.5. Instrumental techniques and test methodology

### 2.5.1. Instrumental techniques

The starting materials were prepared as pressed pellet for chemical analysis on a Philips PW-1404 XRF spectrometer fitted with an Sc-Mo X-ray tube.

Particle size distribution was dry-determined with a Malvern Mastersizers 3000 laser diffraction analyser featuring two light sources, He-Ne (red) and LED (blue), and a measuring range of 0.01 µm to 3500 µm.

Mineralogical analysis of both the starting materials and phase variations during the pozzolanic reaction was conducted by powder X-ray diffraction (XRD) on a PAN Analytical X'Pert Pro Xray 40 mA/45 kV diffractometer fitted with a copper anode, scanning at an angular range of 5–60° (2θ). Rietveld refinement, calculated with Match v.3 and Fullprof software, was deployed to quantify the mineralogical phases, identified against the Crystallography Open Database (COD) collection of crystal structure patterns. Rutile was used as an internal reference at a concentration of around 5% in all cases to be able to determine the amount of amorphous phase.

Mineralogical phase morphology was studied under an Inspect FEI Company electron microscope with a W source DX4i EDX analyser and Si/Li detector. The semi-quantitative chemical analysis values shown are the mean of 10 analyses per spot.

**2.5.1.1. Test methods.** Recycled concrete fines pozzolanicity was measured using a variation on the standardised method for determining pozzolanicity in pozzolanic cements EN 196-5:2011 [31] consisting in soaking the samples in a saturated lime solution (17.683 mm/L) at 40 °C through the test time. The lime fixed by the experimental material is the difference between the lime in the reference solution and the amount present in the problem solution upon finalisation. The mineralogical phases were monitored during the pozzolanic reaction on the residue filtered out of the solution after rinsing with ethanol to detain the reaction and drying in an electric oven at 60 °C for 24 h.

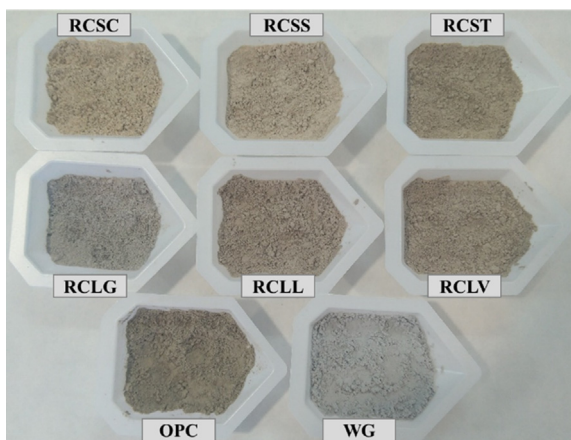


Fig. 1. Ground recycled concrete fines and glass waste.

**Table 2**  
Laboratory mortar batching.

Mortar	OPC (g)	Sand (g)	Water (g)	SCM (g)	W/B
OPC	450.0	1350	225	0.0	0.5
5% RCLG/RCST/WG	427.5	1350	225	22.5	0.5
7% RCLG/RCST/WG	418.5	1350	225	31.5	0.5
10% RCLG/RCST/WG	405.0	1350	225	45.0	0.5

**Table 3**  
Industrial plant cement batching.

Material	OPC-pilot	5%RCLG-pilot
Clinker (%)	96.7	92.1
Gypsum (%)*	2.6	2.6
Ferrous sulfate (%)**	0.7	0.7
RCLG	0.0	4.6

\* Setting regulator; \*\* Cr(VI)-reducing agent

**Table 4**  
Chemical and mineralogical composition of clinker used for the industrial pilot test.

%	Clinker	%	Clinker
SiO <sub>2</sub>	21.23	C <sub>3</sub> S	63.02
Al <sub>2</sub> O <sub>3</sub>	4.61	C <sub>2</sub> S	13.67
CaO	65.9	C <sub>3</sub> A	6.33
Fe <sub>2</sub> O <sub>3</sub>	3.48	C <sub>4</sub> AF	10.59
MgO	1.4	C <sub>3</sub> S + C <sub>2</sub> S	76.69
SO <sub>3</sub>	2.38		
Na <sub>2</sub> O	0.22		
K <sub>2</sub> O	0.69		
P <sub>2</sub> O <sub>5</sub>	0.15		
Cl <sup>-</sup>	0.03		

The heat of hydration released (heat flow and total heat) by the blended cements in the first 48 h at a constant T = 25 °C was studied with a TA Instruments Q2000 calorimeter, proceeding as described in Spanish and European standard EN 196-6:2011 [32].

Cement paste initial and final setting times were determined with an Ibertest AutoVicat analyser to the methodology laid down in Spanish and European standard EN 196-3:2017 [33]. Cement paste fresh state consistency was assessed on a spread table as recommended in standard EN 1015-3:2000 [34].

The 2 d, 28 d and 90 d mechanical strength of 4 × 4 × 16 cm<sup>3</sup> mortar specimens was found on an Autotest 200/10-SW test frame

featuring special gear to determine bending and compressive strength as per standard EN 196-1:2018 [30].

### 3. Results and discussion

#### 3.1. Recycled inorganic CDW characterisation

##### 3.1.1. Laser diffraction-determined particle size

Fig. 3 graphs the particle size distribution curves for the micronised mineral fractions later used as additions, as well as for the starting OPC cement.

The cumulative particle size curves revealed some similarity among the materials studied, albeit with differences in the 0.9 μm to 63 μm range. Generally speaking, particle size in the recycled concrete fines was smaller than, and in the glass waste (WG) very similar to, distribution in the reference cement. The limestone recycled concrete fines (RCL) were slightly finer than the siliceous mineral additions (RCS), primarily because limestone is a softer material. These minor differences in the starting materials are clearly mirrored in the D10 (mesh diameter through which 10% of the sample passes), D50 (50% passes) and D90 (90% passes) values (Table 5).

##### 3.1.2. XRF-determined chemical composition

The chemical compositions of the OPC and the mineral additions analysed as determined with XRF is given in Table 6. Qualitatively speaking, all the samples drawn from recycled CDW exhibited chemical compositions similar to that of the commercial OPC, with SiO<sub>2</sub> and CaO as the major components. Quantitatively, however, perceptible differences were found depending on the nature of the starting waste (limestone or siliceous concrete, shatterproof glass) in terms of both majority oxides and loss on ignition (LOI). In keeping with the literature [26,27], the glass waste used here comprised silica and sodium essentially, with 70.30% SiO<sub>2</sub>

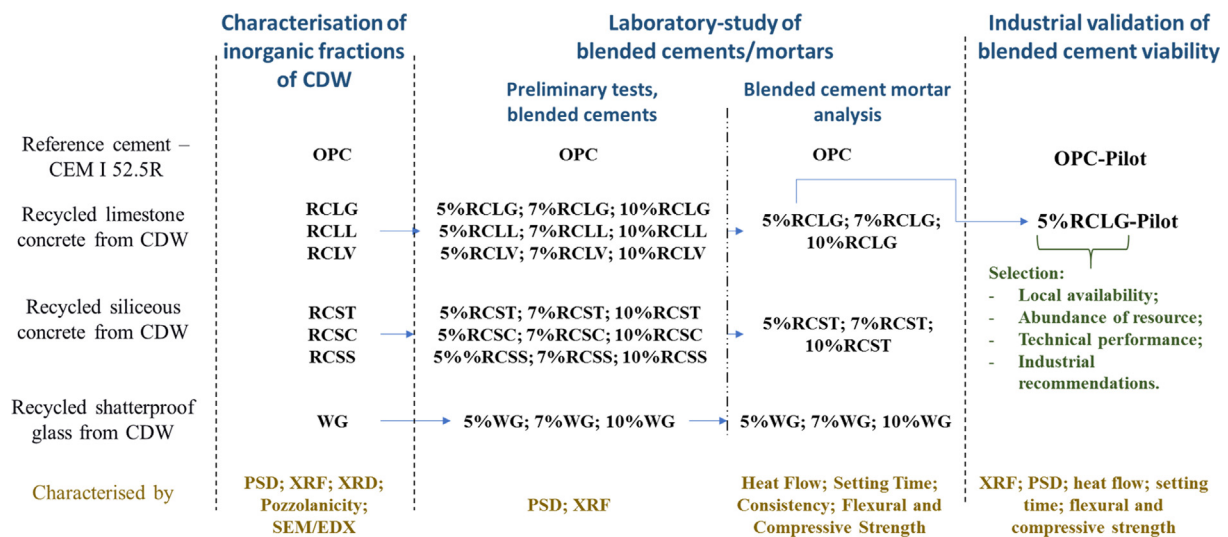


Fig. 2. Experimental programme flow chart.

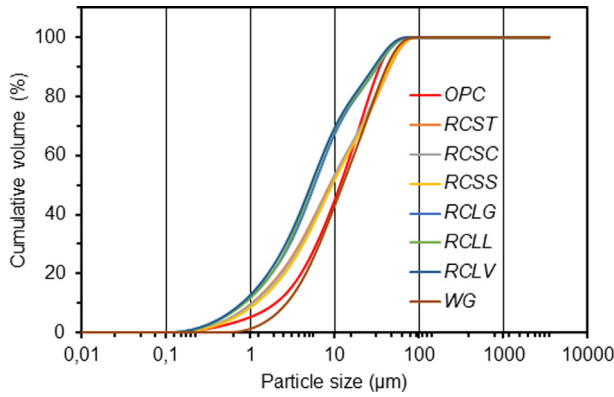


Fig. 3. Particle size distribution of the micronised samples.

and 13.3% Na<sub>2</sub>O. That high sodium content might have adverse effects on hydration and pozzolanic reaction rates, as well as on blended cement performance [35,36].

The acidic oxides (SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>) together accounted for 59.4–69.7% of the additions derived from siliceous concrete, 13.5–32.15% of the limestone concrete fines and 72.29% of the glass waste. Based on those values, the requirements set out in U.S. standards [37] for the use of fly ash and natural and calcined pozzolans in concretes for qualification as pozzolan classes N and F (≥70%) would be met only by GW, although nearly by RCSS (69.7%). All the recycled CDW fines exhibited higher LoI than the OPC due to the calcite in their composition, particularly in the limestone concrete materials, where it comprised 25% to 34% of the total. The

Table 5  
D10, D50 and D90 values (in µm) for starting samples.

	OPC	RCLG	RCLL	RCLV	RCST	RCSC	RCSS	WG
D10 (µm)	1.99	0.82	0.86	0.79	1.04	1.08	1.17	2.70
D50 (µm)	11.60	5.77	5.38	5.22	8.42	8.90	9.37	12.40
D90 (µm)	34.20	31.90	34.40	29.20	44.90	43.20	44.0	41.40

Table 6  
XRF-determined chemical composition in reference cement and waste materials.

%	OPC	RCLG	RCLL	RCLV	RCST	RCSC	RCSS	WG
SiO <sub>2</sub>	11.22	9.34	23.27	12.10	49.97	49.22	58.00	70.30
Al <sub>2</sub> O <sub>3</sub>	2.89	2.88	6.58	3.78	8.98	8.01	9.56	1.07
CaO	69.81	50.33	38.66	45.93	18.65	21.38	14.48	9.64
Fe <sub>2</sub> O <sub>3</sub>	3.70	1.20	2.30	2.49	2.30	2.19	2.12	0.92
MgO	0.93	1.12	0.78	0.92	1.37	1.58	1.11	3.64
SO <sub>3</sub>	3.36	0.85	0.59	0.67	2.53	0.88	0.72	0.24
Na <sub>2</sub> O	0.33	0.18	0.41	0.25	0.80	0.63	0.90	13.26
K <sub>2</sub> O	0.76	0.47	1.07	0.72	3.35	2.61	3.83	0.26
TiO <sub>2</sub>	0.20	0.14	0.39	0.42	0.28	0.30	0.30	0.07
P <sub>2</sub> O <sub>5</sub>	0.14	0.03	0.08	0.09	0.11	0.12	0.10	-
Cl <sup>-</sup>	0.05	0.03	0.02	0.04	0.01	0.03	0.02	0.03
LOI	3.22	33.20	25.70	32.40	11.5	12.9	8.69	0.43

Table 7  
Mineralogical composition of starting waste (%).

%	Mica	Kaolinite	Quartz	Feldspar	Calcite	AM
RCLG	10	-	10	11	52	17
RCLL	12	10	14	13	40	11
RCLV	7	Traces	12	10	62	9
RCSC	6	-	49	6	28	11
RCSS	4	-	58	10	16	12
RCST	4	-	48	8	24	16
WG	-	-	-	-	-	100

presence of limestone would be expected to have a direct effect on blended cement paste hydration kinetics.

### 3.1.3. Rietveld quantitative analysis

From a qualitative perspective (Table 7), the recycled concrete fines had similar mineralogies, consisting in mica, quartz, feldspars and calcite, with kaolinite detected in RCLL (10%) and RCLV (traces) only. In contrast, wide quantitative differences were found depending on the nature of the aggregate used to manufacture the original concrete. In the recycled limestone concrete (RCL), calcite accounted for 50% to 62% of the total and mica, quartz and feldspars for 7–14%. The siliceous recycled concrete (RCS) exhibited 48–58% quartz and 4–28% mica, feldspar and calcite. All the samples contained 9–17% amorphous matter (AM). The presence of mineralogical phases such as mica, feldspar and kaolinite was the outcome of both the impurities found in the natural siliceous and limestone aggregate used in the primary concretes and possible clay contamination during storage in the waste management plant yards [38]. No indication of crystalline mineralogical phases was observed in the glass waste.

## 3.2. Pozzolanicity of the starting waste

### 3.2.1. Pozzolanicity

The percentage of lime fixed by the recycled materials after soaking for 28 d in the CDW/Ca(OH)<sub>2</sub> system is graphed in Fig. 4. Further to the data, all the samples reacted with the lime in the medium, although reaction rates differed. Three types of behaviour were observed. The recycled limestone concrete samples fixed 22–34%; the siliceous concretes 51–58%; and the glass waste 95% of

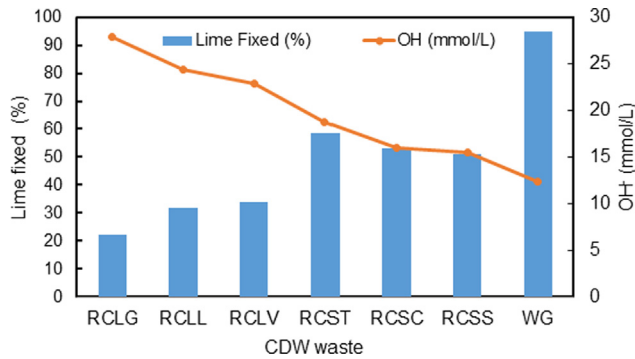


Fig. 4. Lime fixed and OH<sup>-</sup> contents in 28 d test for pozzolanicity.

the total lime initially present. The differences among the three groups would be primarily related to the variation in the sum of their acidic content described in the preceding item. The fixed lime values observed here were comparable to those for other traditional and non-traditional pozzolans: glass exhibited pozzolanicity similar to silica fume, rice husk and grass [39,40]; siliceous concrete waste to fly ash and natural pozzolans [41]; and limestone concrete waste to siliceous-manganese slag [42]. The concentration of hydroxides (OH<sup>-</sup>, mmol/L) shows a correlation with the lime fixed by the recycled materials, showing, in general terms, that the concentration of hydroxide in the medium decreases with the increase of the pozzolanic activity. The Fig. 4 clearly shows the 3 types of recycled materials with different concentrations of hydroxide: the group of limestone concrete waste (RCL) with an OH<sup>-</sup> concentration between 27.9 and 22.9 mmol/L, followed by the siliceous concrete waste (RCS) with 18.8–15.5 mmol/L and finally by the glass waste (WG with 12.39 mmol/L).

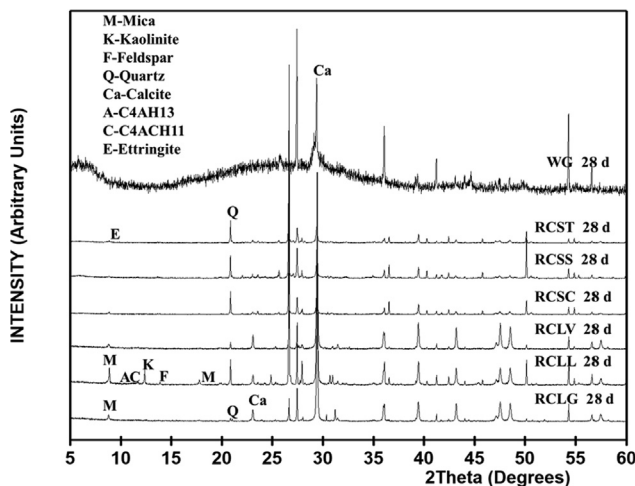


Fig. 5. Mineralogical phases in the waste after 28 d test for pozzolanicity.

Table 8  
Mineralogical phases in the waste after 28 d test for pozzolanicity.

%	Mica	Kaolinite	Quartz	Feldspar	Calcite	C <sub>4</sub> AH <sub>13</sub>	C <sub>4</sub> ACH <sub>11</sub>	Ett	AM
RCLG	10	–	9	9	51	–	–	–	21
RCLL	8	8	14	8	48	traces	traces	–	14
RCLV	5	–	10	4	64	–	–	–	17
RCSC	traces	–	49	6	28	–	–	–	17
RCSS	traces	–	58	4	16	–	traces	–	22
RCST	traces	–	48	8	24	–	–	traces	20
WG	–	–	–	–	33	–	–	–	67

Rietveld quantitative analysis findings for the mineralogical phases after 28 d are presented in Fig. 5 and listed in Table 8. Very small amounts of phases forming during the pozzolanic reaction, including tetracalcium aluminate hydrate (C<sub>4</sub>AH<sub>13</sub>), carboaluminate hydrate (C<sub>4</sub>ACH<sub>11</sub>) and ettringite (Ett), were detected in addition to the crystalline phases present in the recycled mineral fractions of the CDW (mica, kaolinite, quartz, feldspars and calcite).

The amount of amorphous material tended to rise in all cases as a result of hydrated phase formation, although the quantities were too small to be detected as crystalline phases in X-ray diffraction. The glass/lime system exhibited a particularly high calcite content, 33%, associated with the solubility of the Na<sub>2</sub>O in the glass, which when dissolving in the saturated lime solution induced portlandite precipitation [43] and subsequent carbonation during storage and testing.

### 3.2.2. SEM/EDX morphological analysis

SEM/EDX morphological studies of the solid pozzolanic reaction products corroborated the XRD findings, clearly identifying C-S-H gels in all the samples and neo-formed C<sub>4</sub>ACH<sub>11</sub>, and C<sub>4</sub>AH<sub>13</sub> and ettringite in all but the glass waste. As an example of this assessment can be observed in the Fig. 6 and Table 9 for the RCLG sample.

Further to reports by Frías et al. [38], formation of these new hydrated phases during the pozzolanic reaction in the pure pozzolan/lime system, not identified in the starting materials, would be traceable to the presence from the outset of clusters with open morphologies and fairly non-uniform, porous surfaces that enhanced their activity with lime. Such clusters, for which no mineralogy could be clearly determined, would be the outcome of early hydrated phase (C-S-H, calcium aluminates, ettringite and similar) decomposition when exposed to atmospheric conditions (CO<sub>2</sub>, rain) during storage in management plant yards (particle size < 5 mm).

### 3.3. Characterisation of laboratory-scale blended cements

In light of the findings attesting to the scant impact of origin (management plant) on the lime fixation values within each family of laboratory-prepared blended cements (Fig. 4), just one member of each group, deemed as representative, was characterised: RCLG, RCST and glass, all at replacement ratios of 5%, 7% and 10%.

#### 3.3.1. Particle size distribution of starting blended cements

Laser diffraction particle size analysis revealed nearly identical volume density in all the blended cements selected at replacement ratios of 5–10% (Fig. 7).

That similarity in particle size distribution was further supported by the characteristic D10, D50 and D90 values (Table 10). All the cements analysed exhibited practically the same fineness, which was also very similar to the values observed for the reference OPC. Those findings would rule out the possible role of fineness in any possible differences in the physical–mechanical properties of the new blended cement matrices.

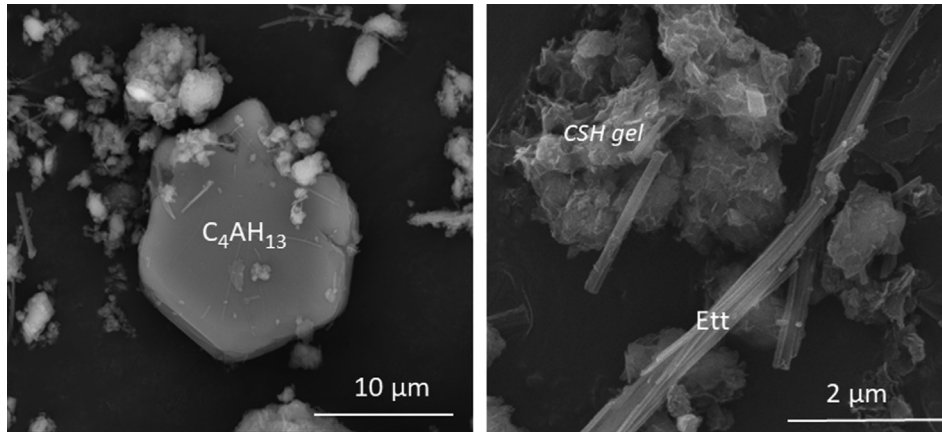


Fig. 6. Details of hydrated phases in 28 d test for pozzolanicity (RCLG sample).

Table 9 SEM/EDX microanalysis of hydrated phases (RCLG sample).

Oxide (%)	C-S-H gel	Ettringite	C <sub>4</sub> AH <sub>11</sub>	C <sub>4</sub> AH <sub>13</sub>
Al <sub>2</sub> O <sub>3</sub>	8.9 ± 1.6	16.3 ± 1.9	29.6 ± 2.4	28.6 ± 1.5
SiO <sub>2</sub>	33.5 ± 2.7	6.7 ± 1.5	11.7 ± 1.6	1.2 ± 0.4
SO <sub>3</sub>	2.2 ± 1.3	32.6 ± 3.3	5.9 ± 0.9	6.5 ± 2.1
MgO	1.4 ± 0.9	–	0.7 ± 0.2	–
CaO	52.4 ± 4.3	44.4 ± 2.5	52.2 ± 3.7	63.7 ± 4.0
Fe <sub>2</sub> O <sub>3</sub>	1.6 ± 1.0	–	–	–

Table 10 D10, D50 and D90 values for blended cements.

	Blended RCL-OPC	Blended RCS-OPC	Blended WG-OPC	OPC
D10	1.82–1.59	1.82–1.76	2.03–1.77	1.99
D50	10.9–10.1	11.8–10.8	11.3–10.3	11.6
D90	33.7–33.2	34.2–34.9	34.1–34.9	34.2

3.3.2. XRF-determined chemical composition of blended cements

Analysis of the blended cements prepared with recycled concrete fines revealed that their chemical composition, as in the reference OPC, was silico-calcareous. Nonetheless, due to the nature of the starting CDW, quantitative variations were observed in the main chemical oxides, CaO and SiO<sub>2</sub>. Content of the latter rose and of the former declined with rising replacement ratio. The SO<sub>3</sub> and Cl contents, limited in European standard EN 197-1 [44] presently in effect, are listed in Table 11, along with the minority oxides, which may have a direct effect on blended cement rheology.

The above data confirmed that all the blended cements analysed lay within the maximum sulfate and chloride contents allowed for type II ordinary cements (6–20%) by the existing standards [44], whilst the materials with 5% replacement would qualify as type I cement. The inclusion of this type of mineral additions at rates of 5–10% barely altered the sulfate and chloride content in the starting OPC (3.36% SO<sub>3</sub> and 0.05% Cl).

Another oxide that, although not limited in standards for ordinary commercial cements must be monitored, is Na<sub>2</sub>O equivalent (Na<sub>2</sub>O eq.). Whereas cements RCST and RCLG exhibited values very

Table 11 SO<sub>3</sub>, Cl, Na<sub>2</sub>O eq. and minority phase content in blended cements.

Oxide	5–10% RCST	5–10% RCSG	5–10% WG	Ceiling value
SO <sub>3</sub> (%)	3.31–3.27	3.23–3.00	3.38–3.37	≤3.5–4.0
Cl (%)	≤0.05	≤0.05	≤0.05	≤0.1
Na <sub>2</sub> O eq.(%)	0.91–1.07	0.81–0.79	1.46–1.98	–
Minority oxides				
V <sub>2</sub> O <sub>3</sub> (%)	≤0.02	≤0.02	≤0.03	–
Cr <sub>2</sub> O <sub>3</sub> (%)	≤0.04	≤0.02	≤0.04	–
ZrO (%)	0.02	0.02	0.02	–
Rb <sub>2</sub> O (%)	≤0.01	< D.L.	≤0.02	–
SrO (%)	≤0.12	≤0.12	≤0.12	–
NiO (%)	< Detection limit		≤0.01	–

similar to those observed for the reference OPC (Na<sub>2</sub>O eq. = 0.83), the inclusion of glass waste in OPC at 5% to 10% induced sodium oxide rises of 76% to 139% relative to the commercial product. Studying potentially reactive aggregate, some authors have reported the adverse effect of the presence of high alkali contents on both cement particle hydration rate [35,45] and the formation of expansive gels, possibly as a result of the aggregate-alkali reac-

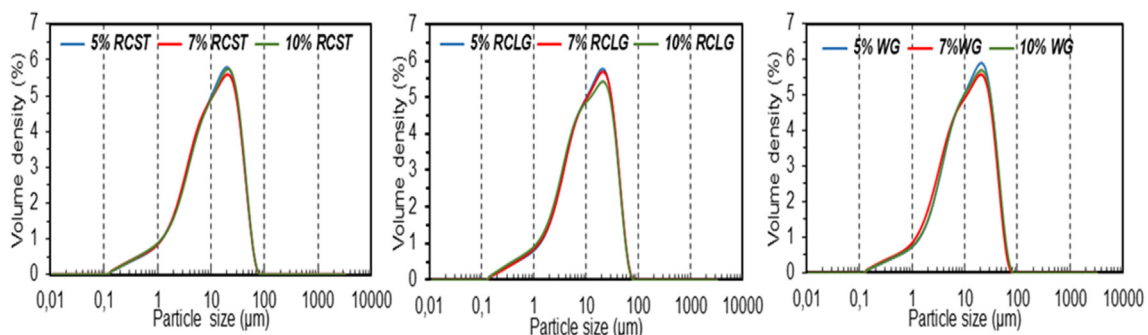


Fig. 7. Particle size distribution in blended cements.

tion, to the detriment of the technical properties of construction members [46,47]. Idir et al. [48] observed that reaction to take place with particle sizes > 150 μm, a value much higher than the < 63 μm used here.

In another vein, the minority V, Cr, Zr and Rb oxides in all the cements analysed were found at concentrations of under 300 ppm to 400 ppm, with only Sr oxide reaching 1500 ppm. The Ni oxide content, in turn, was beneath the XRF detection limit. At such small quantities, none of those elements would have an adverse impact on cement particle hydration rate [49].

### 3.4. Physical-mechanical behavior of blended cement matrices

#### 3.4.1. Heat of hydration

Depending on the nature of the active additions in cement, total heat of hydration released and its variation over time are known to differ from the values observed in reference mortars [50]. That may have a beneficial or adverse (microcracking, shrinkage) effect on the performance of cement matrices. The effect of mineral additions sourced from CDW on blended cement hydration kinetics can be studied in terms of heat of hydration, for factors such as pozzolanicity, nature and fineness of the addition play a significant role in that physical property.

Fig. 8 compares the total heat released during blended cement hydration at 6 h, 24 h and 48 h to the values for the reference OPC. In the first 6 h, more heat was clearly released by the blended cements than by the reference, with significant differences between the former depending on the nature of the recycled concrete. The 28% rise detected in mortars prepared with RCLG at 5–7% replacement declined substantially to ~10% when RCST was added and to ~5% when WG was the addition. No clear pattern in heat of hydration was observed in any of the mortars with rising replacement ratio. In contrast, in the 24 h samples heat of hydration clearly tended to be slightly higher in the recycled limestone RCLG concrete than in OPC, and lower than the reference in the other two types of additions (RCST and WG).

The behaviour observed for these concrete fines would be primarily related to the nature and percentage added [51–54], inasmuch as any effect of fineness was ruled out [55,56], given the inter-sample similarity of the values for that parameter (Table 10). Further to the literature, the mechanisms primarily involved in hydration and specifically in heat of hydration are related to the filler and chemical effects of the mineral addition. The former is particle size-associated, for the generally smaller particles in additions disperse the cement grains and accelerate hydration [57,58] by acting as nucleation sites and seeds for hydrated phase formation [59]. The chemical effect may consist in the generation of secondary hydrated phases, the outcome of the pozzolanic reaction between the addition and portlandite [60,61], or calcium carboaluminate hydrate formation as the product of the reaction between the calcite present in these recycled concrete fines and the calcium aluminate phases in the cement [62]. Along these lines, authors such as Berodier [63] and Wang [64] reported that as active, low pozzolanicity additions had no effect on early age hydration kinetics, the filler effect prevailed.

In light of the present findings and bearing in mind the aforementioned factors affecting heat of hydration, the much higher value for that parameter in the RCLG mortars than in the reference, RCST and glass samples was attributable essentially to its high limestone content. That component would react rapidly with the calcium aluminate phases in the cement to form carboaluminate hydrates [65], producing an effect that would prevail over the pozzolanic (low RCLG activity) and filler (very similar fineness in the different wastes) effects. In earlier studies, however, other authors [63,66] concluded that limestone additions favoured C-S-H nucleation due to minor surface dissolution that would raise dissolved calcium concentration, thereby enhancing C-S-H formation. Those observations are consistent with research conducted with SEM/EDX and NMR by Frías et al. [38], who showed that during the pozzolanic reaction in the pure limestone waste/lime system, limestone favoured a rise in the Q<sup>2</sup> signals in C-S-H gel as well as C<sub>4</sub>AH<sub>13</sub> formation, developments not observed for siliceous waste. The low heat of hydration observed for the GW-blended mortar was unexpected, however, given the wholly amorphous nature of glass and its 70% (theoretically reactive) silica content (Tables 6 and 7), which would initially determine behaviour similar to that of silica fume. Fig. 8 shows that the glass waste exhibited inert behaviour in the first 48 h of hydration. That apparently abnormal development would be related to findings reported by Jawed and Skalny [35], according to which alkaline ions in the cement lower Ca<sup>2+</sup> ion solubility, inhibiting or retarding hydrated phase formation (and in this study, the pozzolanic reaction).

#### 3.4.2. Setting time

Hydration kinetics were supplemented with the setting time results. Fig. 9 compares the setting times for the 10 cements analysed.

The inclusion of recycled mineral additions barely altered the initial setting time recorded for the reference (technique error = 5 min) in any of the cements analysed, although in eight of the nine blended cements studied the cement hydrated slightly earlier, in a clear correlation with the heat released. The exception being the paste prepared with 10% glass waste, where hydration was slightly retarded. Initial setting time was not impacted by the nature of the recycled mineral (limestone, siliceous, glass), possibly because of the small replacement ratios used, nor the presence of minority elements due to the low concentrations involved (Table 11). Further to the values observed, all the cements were standard-compliant (>45 min) for type 52.5 cements. Final setting time, not specified in the existing standards, also exhibited values close to the OPC times although varying slightly with the

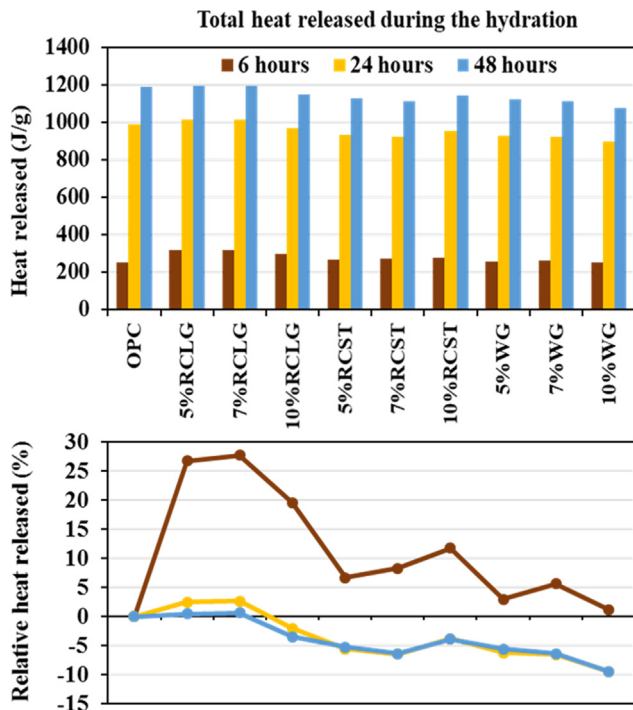


Fig. 8. Total heat released by blended mortars and OPC reference.



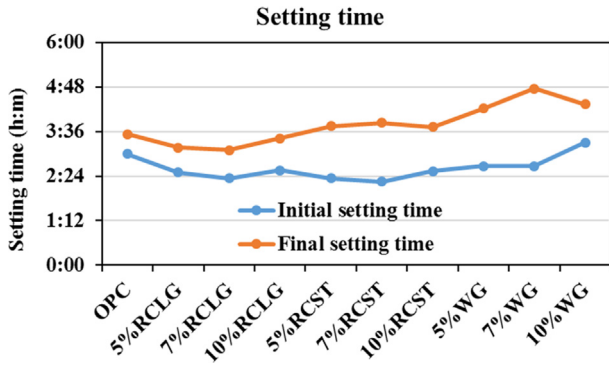


Fig. 9. Initial and final setting times for blended cement pastes.

nature of the mineral addition, with setting concluding first in RCLG followed by OPC, RCST and WG in that order.

If setting time is defined as the difference between the initial and final times, the recycled limestone concrete (RCLG) set slightly more quickly than the OPC, whereas the recycled siliceous concrete (RCST) and glass waste set more slowly than the reference. That waste composition-based behaviour might be put to beneficial use depending on the application [67,68].

3.4.3. Consistency

Only minimal differences were observed in consistency between the cement pastes bearing recycled CDW-based additions and the reference OPC, all prepared with a w/b ratio of 0.5 (Fig. 10). Nonetheless, the materials behaved somewhat differently depending on the nature of the mineral addition used. In most cases, the blended mortars had a slightly smaller slump than the reference. The effect of including recycled limestone and siliceous concrete fines was more perceptible at higher replacement ratios, whereas in the samples bearing glass waste, although the slump rose with the ratio, it was larger than in the reference for the cement paste bearing 10% replacement only.

These findings for mortars were in line with the water specified for normal consistency to conduct the standardised initial setting time test (Table 12). The inclusion of up to 10% RCLG and RCST did not raise the water demand above the value needed for the reference material. Adding glass at a ratio of 10% reduced demand slightly (by around 3%), as attested to by the consistency of the mortars prepared with that percentage of addition, which was greater than in the reference OPC prepared with the same w/b ratio. Those findings would be related to the higher density and lower porosity in glass than in concrete waste, which would translate into lower sorptivity [69].

3.4.4. Compressive and flexural strength

The compressive and flexural strength of 2 d, 28 d and 90 d mortars prepared with blended cements (determined on standard-

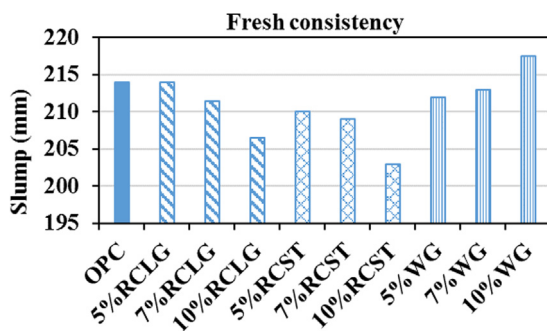


Fig. 10. Setting time in OPC and in mortars prepared with blended cements.

Table 12 Water demand to ensure normal consistency in blended cement pastes.

	OPC	RCLG			RCST			WG		
Waste (%)	0	5	7	10	5	7	10	5	7	10
H <sub>2</sub> O (g)	150	150	152	151	152	152	151	151	148	146

ised 4 × 4 × 16 cm<sup>3</sup> specimens) are compared to the mechanical performance of the reference mortar in Fig. 11.

Both compressive and flexural strength appeared to decline slightly relative to the reference in nearly all cases. Where the additions had a recycled limestone or siliceous concrete base and replacement ranged from 5% to 7%, strength was very similar to the reference mortar, with declines of around just 5%. Raising the replacement ratio to 10% induced declines by around the same amount. That pattern is a clear indication that both compressive and flexural strength loss were related to the replacement ratio and tolerable insofar as they were not higher than that percentage [4,70–73]. The strength findings corroborated the results described in preceding sections, according to which the presence of recycled limestone or siliceous concrete fines at low replacement (<10%) induce no significant changes relative to the reference pastes and mortars.

The mechanical behaviour of mortars prepared with glass waste differed from the performance observed for RCLG and RCST. In the 2 d and 28 d glass blended mortars, compressive and flexural strength declines were similar to one another across the range of replacement ratios (5–10%) and curing times. These relative compressive and flexural strength losses were greater at higher replacement ratios, with compressive strength tumbling by 24% and flexural strength by 22% in the 28 d mortars. In the 90 d mortars mechanical strength patterns changed significantly, practically offsetting the initial loss for a net of just 2–5% in both types of strength, except for the mortar with 10% replacement, where compressive strength slid by 12% relative to the reference.

That 28 d turning point in mechanical strength in mortars additioned with glass waste differed from the pattern expected on the grounds of the high pozzolanicity and amorphous nature of the addition (Fig. 4). As noted earlier, the presence of a high alkali content in the glass would have an adverse effect on the rate of both the hydration and pozzolanic reactions and a similarly detrimental impact on the performance of glass blended cement matrices [35,36,45–47]. Analysing a different type of ground glass waste, other researchers [15,26,27] detected similar declines in 28 d mechanical strength but higher 90 d performance than in the reference. A possible explanation for such behaviour might lie in the presence of a high alkali content (Na<sub>2</sub>O eq.) in the porous blended mortar solution. Under such conditions early age (<28 d) pozzolanicity would be neutralised and portland cement hydrated to more porous, less dense alkali-containing phases. The result would be a decline in mechanical strength of a greater magnitude than the replacement ratio (Fig. 11) [74–76]. At later ages, however, the drop in alkali concentration would raise the solubility of the Ca<sup>+</sup> ions in the solution, inducing a pozzolanic reaction that would contribute directly to recovering the loss of mechanical strength. That rationale is in line with earlier research in which mechanical strength was observed to rise to levels higher than the reference mortar in 90 d or older glass blended mortars. Such rises were also dependent upon glass fineness and replacement ratio [48].

Despite the loss in compressive strength observed, nearly all the blended mortars containing 5–10% recycled CDW limestone or siliceous concrete or glass additions would meet the minimum mechanical strength requirements laid down in European standards for both 2 d (>30 MPa) and 28 d (52.5 MPa) mortars and

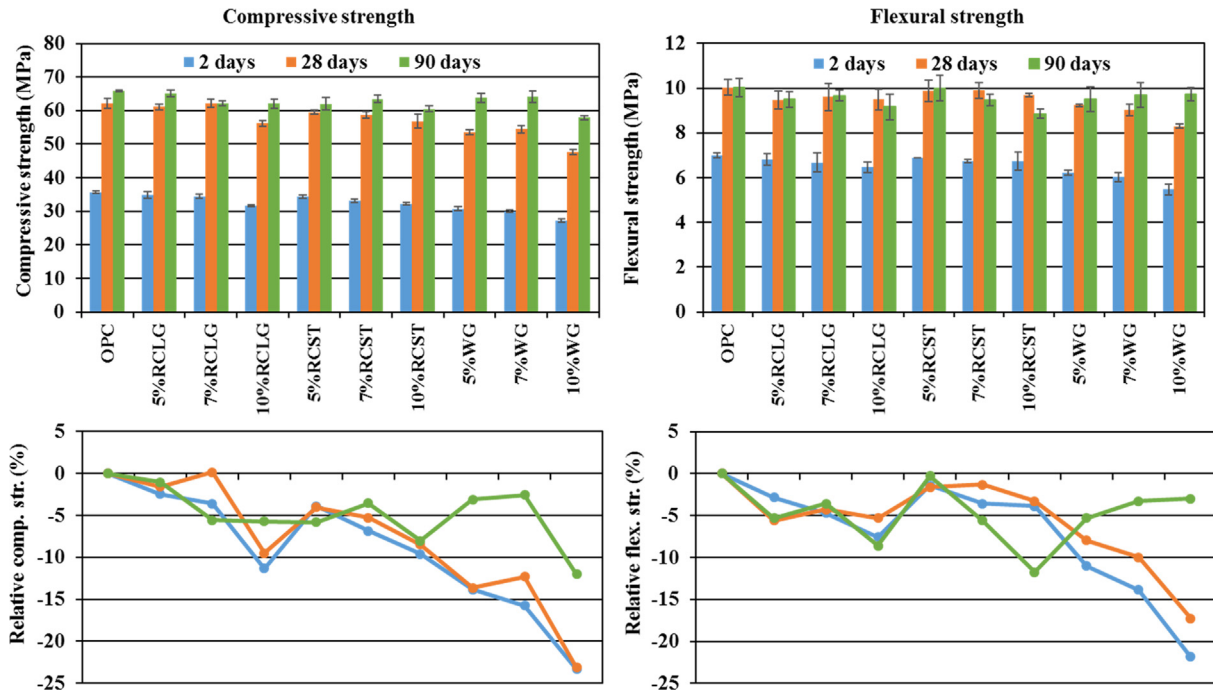


Fig. 11. Compressive and flexural strength of blended cement and reference OPC mortars.



Fig. 12. Limestone concrete fines (RCLG) from the Gutram valorisation plant used in the pilot test.

would be eligible for the same strength class as the reference OPC (CEM I 52.5 R). Both the 2 d and 28 d mortars prepared with 10% glass waste, however, would drop from class 52.5 R ( $\geq 20$  MPa) to class 42.5 R.

### 3.5. Full-scale validation

The laboratory-scale study was supplemented with a plant-scale pilot test to validate the use of CDW-based mineral additions as partial portland clinker replacements. The trial consisted in manufacturing blended cement bearing 5% RCLG (5%RCLG-Pilot) and ordinary portland cement (OPC-Pilot) as a reference all on the same day using the same raw materials and facility (Cementos Leona) (Fig. 12). The criteria informing the choice of the mineral addition used for validation and its batching are listed below.

- Local availability of CDW: the cement plant is located in the Basque Country where the natural aggregate used is limestone. The CDW valorisation plant supplying the recycled limestone concrete (RCLG) is sited at <30 km from the cement plant.
- Abundance of limestone concrete fines: as more concrete fines than waste glass were available, the cement plant opted for the former CDW stream for reasons of industrial practicality.

- Amount used: both the existing literature [4] and the present laboratory-scale findings recommended replacing 5–7% of the clinker with recycled mineral waste. The cement plant, in turn, recommended a ratio of not over 5% for the addition to qualify as a minority component further to the existing standard on cement specifications [44].

The chemical, physical and mechanical data for the two industrial cements are given in Tables 13 and 14.

Table 13  
Chemical composition of OPC-Pilot and 5%RCLG-Pilot.

%	OPC-Pilot	5%RCLG-Pilot
SiO <sub>2</sub>	20.31	20.11
Al <sub>2</sub> O <sub>3</sub>	4.4	4.31
CaO	64.24	63.64
Fe <sub>2</sub> O <sub>3</sub>	3.45	3.48
MgO	1.4	1.38
SO <sub>3</sub>	3.34	3.65
Na <sub>2</sub> O	0.26	0.26
K <sub>2</sub> O	0.67	0.69
Cl <sup>-</sup>	0.05	0.06

**Table 14**  
Physical and mechanical properties of the industrial cements manufactured.

	Particle size distribution (µm)			Setting time (h:m)		Total heat released during the hydration (J)			Compressive strength (MPa)		Flexural strength (MPa)	
	D10	D50	D90	Start	End	6 h	24 h	48 h	2 d	28 d	2 d	28 d
	OPC-Pilot	1.3	10.9	32.6	3:00	3:32	237	933	1137	37.5 ± 0.91	63.22 ± 1.46	6.9 ± 0.26
5%HcG-Pilot	1.3	11.1	32.6	2:50	4:02	267	1057	1265	37.14 ± 0.30	63.47 ± 1.66	7.03 ± 0.4	10.13 ± 0.45
Difference (%)	0.0	1.8	0.0	-6.0	14.0	12.7	13.3	11.3	-1.0	0.4	4.8	1.0

Characterisation of the blended cement manufactured at the industrial facility revealed the following.

- The chemical composition of the blended cement bearing 5% RCLG was similar to the composition of the commercial OPC-Pilot manufactured immediately after in the same facility and under identical conditions.
- The particle size distribution was essentially identical in the blended and commercial cements.
- The blended cement released slightly more heat of hydration than the reference cement, denoting a rise in early-age hydration kinetics with the RCLG addition.
- Blended cement mechanical strength was comparable to reference cement performance and met the requirements for classification as 52.5 R further to the European standards presently in place.

The analysis of the essential industrial-scale parameters showed the two materials to be similar and therefore comparable to the laboratory-scale findings despite the minor differences in the blended cements in terms of material mixing, milling and batching. That confirmed the reliability of the laboratory-scale tests as a method for designing blended cements and their translation to orders of magnitude suitable for industrial-scale production.

The industrial pilot also proved the technical viability of using recycled concrete fines as a partial clinker replacement. Similar studies using specific technologies for the production of cement additions from recycled concrete have proven to be highly viable from the economic, energy and environmental perspectives [4,28]. Here, as the addition was ground with the clinker, no additional technologies were required. The conclusion that may be drawn from the foregoing is that, provided the recycled mineral additions are supplied by valorisation facilities near the cement plant, their use holds vast economic, energy and environmental promise, for it would reduce raw material mining and consumption and significantly lower CO<sub>2</sub> emissions. Further to the aforementioned studies, potential CO<sub>2</sub> emissions abatement may be estimated to be very similar to the clinker replacement ratio. On those grounds, replacing 5% clinker would lower emissions by 41 kg CO<sub>2</sub> eq./t cement, for a worldwide savings of over 80 Mt CO<sub>2</sub> eq./year.

### 3.6. Conclusions

The conclusions that may be drawn from the present study of recycled concrete fines (<5 mm) stored in management plant yards and shatterproof glass waste as possible portland clinker replacements are set out below.

- The three recycled CDW analysed, limestone and siliceous concrete and shatterproof glass, can be valorised as SCMs, thereby constituting an eco-efficient as well as socially and economically viable alternative to stockpiling. This kind of recycled CDW are not included in the standard EN 197-1:2018 and

has only been tested in this way for the scientific and technical purposes of this study. The mineralogy of the three types of waste consists primarily in quartz and limestone (depending on the aggregate used to manufacture the original concrete), along with feldspars in the concrete fines and a wholly amorphous material in the glass.

- None of the phases characteristic of hydrated cement was identified in the starting recycled concrete. SEM/EDX analysis identified clusters of structurally undefined materials, however, that might constitute a mix of such hydrated phases, altered or decomposed as a result of weathering. Such porous, scantily compacted clusters with reactive surfaces afford this waste lime-fixation capacities, whose chemical composition-governed efficacy is, in descending order: glass waste > recycled siliceous concrete fines (RCS) > recycled limestone concrete fines. Such pozzolanicity is corroborated by the neo-formation of hydrated phases such as C-S-H gel, C<sub>4</sub>AH<sub>11</sub>, C<sub>4</sub>AH<sub>13</sub> and ettringite in the siliceous and limestone solid concrete waste and C-S-H gel only in the glass waste after soaking in a lime solution for 28 d.
- Heat of hydration monitoring showed the reaction kinetics in the 24 h and 48 h mortars prepared with 5% to 10% of recycled waste to be very similar to the behaviour observed in the reference mortar. Nonetheless, more heat was released in the 6 h experimental materials than in the control, in the following descending order: recycled limestone concrete > recycled siliceous concrete > glass > reference.
- At laboratory scale, the mechanical strength of the mortars prepared with recycled limestone or siliceous concrete is 5% to 10% lower than in the reference, depending on the replacement ratio, although that difference narrows to 2–3% in the 90 d mortars. The 28 d difference is wider in the case of glass waste, at 14–24%, but narrows to 2–12% in the 90 d mortars. These relative declines in strength are of scant significance, however, for in all cases the blended cement mortar are compliant with the short (2 d) and medium (28 d)-term standard requirements for strength class CEM I 52.5, the same class as the starting commercial cement. Further to those findings, replacement ratios of 5–7% are recommended to ensure highest performance in materials bearing recycled limestone or siliceous concrete or glass waste.
- Full-scale manufacture at an industrial plant of a blended cement containing 5% recycled limestone concrete fines (RCLG) confirmed the viability of using cement additions deriving from that type of waste. The end product exhibited characteristics similar to those of commercial portland cement manufactured with the same technological resources. The benefits would consist in lower raw material and energy consumption and CO<sub>2</sub> emissions abatement at a rate of 41 kg CO<sub>2</sub> eq./t of cement which could translate into a reduction of 80 Mt CO<sub>2</sub> eq./year worldwide. Such benefits are in line with circular economy strategies and the Sustainable Development Goals.

The overall conclusion is that blended cements manufactured with 5–7% inorganic CDW would comply with standard chemical

(sulfate and chloride ceilings), physical (initial setting) and mechanical (compressive strength) requirements, with performance comparable to that of commercial OPC. This study suggests new lines of research geared to expanding the use of shatterproof glass as an SCM by reducing its particle size to < 45 µm, optimising the replacement ratio and further analysing the long-term (>90 d) performance and durability of the resulting blended cements. In this sense, and due to the importance of durability aspects, additional tests are being carried out to evaluate the durability of the blended cements studied in this work.

### CRedit authorship contribution statement

**J. Moreno-Juez:** Conceptualization, Methodology, Formal analysis, Investigation, Resources, Data curation, Writing - original draft, Writing - review & editing, Visualization, Project administration. **Iñigo J. Vegas:** Conceptualization, Methodology, Writing - original draft, Writing - review & editing, Supervision, Funding acquisition. **M. Frías Rojas:** Conceptualization, Methodology, Formal analysis, Investigation, Resources, Writing - original draft, Writing - review & editing, Visualization, Project administration, Supervision, Funding acquisition. **R. Vigil Villa:** Formal analysis, Investigation, Resources, Writing - original draft, Writing - review & editing. **E. Guede-Vázquez:** Resources, Writing - review & editing, Supervision, Funding acquisition.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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The authors assume full responsibility for the views expressed in the paper and hold the European Commission harmless for any use that might be made of the information it contains.

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