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Moreno-Juez, J.; Vegas, Inigo J.; Gebremariam, Abraham T.; García-Cortes, V.; Di Maio, F.

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# Treatment of end-of-life concrete in an innovative heating-air classification system for circular cement-based products

J. Moreno-Juez <sup>a, \*</sup>, Iñigo J. Vegas <sup>a, b</sup>, Abraham T. Gebremariam <sup>c</sup>, V. García-Cortés <sup>a</sup>, F. Di Maio <sup>c</sup>

<sup>a</sup> Tecnalia, Basque Research and Technology Alliance (BRTA), Astondo Bidea, Edificio 700, Parque Tecnológico de Bizkaia, 48160, Derio, Spain

<sup>c</sup> Delft University of Technology, Faculty of Civil Engineering, Resource and Recycling, the Netherlands

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#### ABSTRACT

A stronger commitment towards Green Building and circular economy, in response to environmental concerns and economic trends, is evident in modern industrial cement and concrete production processes. The critical demand for an overall reduction in the environmental impact of the construction sector can be met through the consumption of high-grade supplementary raw materials. Advanced solutions are under development in current research activities that will be capable of up-cycling larger quantities of valuable raw materials from the fine fractions of End-of-Life (EoL) concrete waste. New technology, in particular the Heating-Air classification System (HAS), simultaneously applies a combination of heating and separation processes within a fluidized bed-like chamber under controlled temperatures (±600 °C) and treatment times (25-40 s). In that process, moisture and contaminants are removed from the EoL fine concrete aggregates (0-4 mm), yielding improved fine fractions, and ultrafine recycled concrete particles (<0.125 mm), consisting mainly of hydrated cement, thereby adding value to finer EoL concrete fractions. In this study, two types of ultrafine recycled concrete (either siliceous or limestone EoL concrete waste) are treated in a pilot HAS technology for their conversion into Supplementary Cementitious Material (SCM). The physico-chemical effect of the ultrafine recycled concrete particles and their potential use as SCM in new cement-based products is assessed by employing substitutions of up to 10% of the conventional binder. The environmental viability of their use as SCM is then evaluated in a Life Cycle Assessment (LCA). The results demonstrated accelerated hydration kinetics of the mortars that incorporated these SCMs at early ages and higher mechanical strengths at all curing ages. Optimal substitutions were established at 5%. The results suggested that the overall environmental impact could be reduced by up to 5% when employing the ultrafine recycled concrete particles as SCM in circular cement-based products, reducing greenhouse gas emissions by as much as 41 kg CO2 eq./ton of cement (i.e. 80 million tons CO2 eq./year). Finally, the environmental impacts were reduced even further by running the HAS on biofuel rather than fossil fuel.

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#### 1. Introduction

In response to the drive towards efficient building practice using circular cement-based products, efforts have been directed towards the recycling of End-of-Life (EoL) concrete for subsequent reuse in various low-to-intermediate grade construction applications. However, the recycling of concrete waste following the paradigms

E-mail address: jaime.moreno@tecnalia.com (J. Moreno-Juez).

of the circular economy (from End-of-Life concrete to new concrete) is still at an early stage. Although the report from the World Business Council for Sustainable Development (WBCSD) (World Bussiness Council for Sustainable Development, 2012) noted advances within concrete recycling across Europe, most construction and demolition waste has either been down-cycled into sub-base aggregates or dumped at landfill sites (Vegas et al., 2008, 2011; Poon et al., 2006; Nataatmadja and Tan, 2001). Despite the large number of studies on recycling concrete aggregates, only a few describe fine recycled aggregates (<4 mm) and even fewer report the use of ultrafine fractions (<0.125 mm).

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Corresponding author.

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<sup>&</sup>lt;sup>b</sup> External Associated Member of the CECEAR Research Group, Universidad Autónoma de Madrid, 28049 Madrid, Spain

Seeking maximum yields (in terms of both amount and quality) from recycled aggregates, several techniques have been studied over the past few years for the selective recovery of different size fractions. Coarse recycled aggregates (≥4 mm) and related technological developments are the most frequent areas of study, as it is easier to sort the aggregate fractions and clean them after crushing. One example is the Pre-Weakening Treatment Station (PWTS) (Bru et al., 2017) where high voltage (tens of kV) electrical discharges of short duration are passed between electrodes and counterelectrodes. The PWTS is used for the selective recovery of the natural aggregates contained in the concrete waste. One of the most tested technologies for the recovery of coarse aggregates is Advanced Dry Recovery (ADR) that uses kinetic energy to break the covalent water bonds between fine particles formed by moisture (De Vries et al., 2009). The energy consumed by ADR technology is sustainable and affordable when compared with conventional crush and screen-based technologies. Concrete made of coarse recycled concrete aggregates ( $\geq 4$  mm) have compressive strengths that are comparable with natural aggregates and, in some cases, even better performance has been reported (Malešev et al., 2010) (Lotfi et al., 2014). The performance of coarse recycled concrete aggregates is already well established in new concrete formulations (Malešev et al., 2010; Razaqpur et al., 2010; Brito and Soares, 2017; Arroyo et al., 2019), although knowledge of the mechanical performance of the fine (<4 mm) and the ultrafine fractions (<0.125 mm) is still limited.

Major constraints on the use of finer EoL concrete fractions are their high levels of moisture and impurities that make them unsuitable for use in new circular cement-based products (Fan et al., 2016; Vieira et al., 2016; Braga et al., 2010; Fern, 2016). The same constraints are an obstacle to recycling 100% of EoL concrete (Khoshkenari et al., 2014) (Zhao et al., 2015). The greater circularity of the fine recycled concrete aggregates (the <4 mm fraction constitutes a large fraction, anywhere between 30 and 40% by weight of the whole mass of crushed concrete (Etxeberria and Vegas, 2015)) can help to guarantee both the supply of cost-effective mineral resources for the production of new circular cement-based products and the reduction of certain environmental impacts that are mainly associated with the logistics of raw mineral materials that enter the building supply chain. Further treatment is then necessary for complete separation of the hydrated cement paste from the sandy fraction and removal of both moisture and impurities by thermal treatment, which is the principal method for recycling the fine fraction (Shui et al., 2008; Florea et al., 2013, 2014; Gastaldi et al., 2015). Microwave heating (Lippiatt and Bourgeois, 2012) is a strong candidate for the selective liberation of multiphase materials such as concrete. It exploits their thermal, dielectric, and mechanical properties, to generate stress gradients that can lead to grain boundary fracture, and embrittlement, however that technology is still at an early stage. Another emergent technology is the innovative Heating-Air classification System (HAS), in which the moisture and the contaminants of the EoL fine-concrete aggregates (0-4 mm) are removed, by a simultaneous combination of heating (600 °C) and separation processes within a fluidized bed-like chamber that yields upgraded fine aggregates (0.125-4 mm) and ultrafine cementitious particles (<0.125 mm), mainly composed of hydrated cements, thereby adding value to the finest fractions of the EoL concrete waste stream.

A key proof of concept (TRL3), at the lab scale, of this innovative technology has previously been reported in (Lotfi and Rem, 2016, 2018). The experimental results showed that the combination of

heat and air classification in the HAS resulted in proper separation of fine (0.125–4 mm) and ultrafine (0–0.125 mm) particles from contaminated fractions. Somi et al. (Lotfi and Rem, 2016, 2018) highlighted the viability of HAS technology for the recovery of the fine particles and their conversion into valuable products, such as clean sand for new concretes, by extracting the ultrafine particles (<0.125 mm), moisture, and contaminants. They also stated, in line with Shui et al. (2009), that thermal treatment at 500 °C produces ultrafine particles with phases that are mainly composed of calcium-silicate-hydrate (C-S-H), CaO, and non-crystalline dehydrated phases that displayed rehydration properties when in contact with water. The above underlines the potential reactivity of the ultrafine recycled particles from the treatment of concrete waste for use as cementitious material in the manufacture of new circular cement-based products. This conclusion supports the study of ultrafine recycled concrete particles for use as SCM in new circular cement-based products. It is, likewise, supported by recent research in this area that is recovering waste materials for alternative additions to such products (Juenger and Siddique, 2015; Yu and Shui, 2014; Bouchikhi et al., 2019; Aprianti, 2017).

In that context, further studies have been performed as part of the European VEEP Project (EU project) for the design of a HAS prototype at pre-industrial scale (3 t/h) and its construction. Their remit has at all times been to demonstrate the economic and the technical viability of the VEEP project at TRL 6-7 for its optimization and eventual commercialization. In this study, our aim will be to describe the working principles and the operating parameters of the HAS process<sup>1</sup> and to analyze the potential of the ultrafine recycled particles that are obtained as cementitious particles for new circular cement-based products. To do so, the contaminated fine fractions of two EoL concretes will be introduced into the HAS setup and then exposed to a flow of hot gas, while the ultrafine particles (<0.125 mm) will be classified and raised by the hot-air flows up into the cyclone. The chemical and the physical properties of the ultrafine recycled concrete particles obtained in that way will then be assayed. Their cementitious properties and their potential use as SCM will be evaluated through two approaches; an initial assessment of physico-chemical effects (Kumar et al., 2017) will be performed, by studying the reaction with the clinker phases, the hydration kinetics, and the mechanical properties will then be studied, by employing cement pastes composed of a commercial Portland clinker blended with different quantities (up to 10% by weight of clinker) of the HAS ultrafine recycled concrete particles. Then, their potential use as Supplementary Cementitious Materials (SCMs) added to cement-based products, which could also contribute to a reduction in cement content, will be described in the following sections. The SCMs will be evaluated through the study of both the fresh and the hardened properties of standardized mortars manufactured with a commercial Ordinary Portland Cement (CEM II) that will be blended (by replacing 10% of the cement) with ultrafine recycled concrete particles treated with HAS technology.

Finally, a screening of the LCA will be performed, to evaluate the environmental viability of using the ultrafine recycled concrete particles as SCM, produced at a pre-industrial scale with HAS technology. The environmental potential of the ultrafine particles used in substitution of cement in new circular cement-based products will be evaluated, in the light of those results, thereby laying the basis for further optimization of the process parameters and subsequent scaling up for industrial production and commercialization.

#### 2. Materials and methods

#### 2.1. Materials and equipment

#### 2.1.1. EoL concrete

In this research, the HAS-processed EoL concrete waste was produced using ADR technology (De Vries, 2017). The ADR processing unit was fed with crushed EoL wet concrete wastes (0-12 mm) from two different sources, representing the two most widely used concrete types in Europe. The EoL concrete wastes were collected from two European locations:

- EoL siliceous concrete waste (EoL-SCW) from building demolition in the Netherlands: the original concrete had been manufactured with natural siliceous aggregates.
- EoL limestone concrete waste (EoL-LCW) from building demolition in Spain: the original concrete had been manufactured with natural limestone aggregates.

ADR technology produced two main fractions. The coarse recycled concrete fraction between 4 and 12 mm, of sufficient quality for use in construction applications, as reported in previous research works (Lotfi et al., 2015). The fine contaminated fractions (0-4 mm), mainly composed of hydrated cement paste, impurities, and a high moisture content when compared to the coarse recycled concrete fraction. HAS technology is designed to offer a cost-effective quality enhancement of the fine recycled concrete (0.125–4 mm) fractions. In the process, large quantities of cement paste adhering to the fine aggregates are released and ultrafine recycled fractions (<0.125 mm), free of moisture and organic contaminants, are recovered. The complete recycling set-up, which combines both ADR and HAS, is illustrated in Fig. 1.

The main properties of the two fine contaminated fractions, from siliceous concrete waste (EoL-SCW 0–4 mm) and limestone concrete waste (EoL-LCW 0–4 mm), which form the input into the HAS setup, are presented in Table 1. The moisture content of the HAS feed was determined by drying the material in an oven at 105 °C, for 24 h.

#### 2.1.2. Heating-air classification system (HAS)

The HAS setup at a pre-industrial scale (Fig. 2) consists of a combination of heaters and incoming fine aggregate (0-4 mm)

classification systems. Its production capacity amounts to 3 tons of contaminated aggregate per hour. The HAS applies heating both to evaporate inherent moisture and to burn out the organic contaminants and it applies air flow as a means of separating and classifying both the size and the form of the product.

The technology is based on a gravitational-counter flow zone, in which hot air rising within a vertical chamber is used to classify the particles. The process begins when the burner zone has a regulated heating temperature of 600 °C (5). At that point, the temperature in the top part of the heating separation chamber (7) rises to approximately 250 °C. The wet contaminated fine fractions are then fed in from the top (1) and are exposed to heating for a period of approximately 25–40 s in the pre-industrial scale HAS setup. The drying process mainly takes place in the heating chamber as the feed continues.

Once in the chamber, the drag force will run contrary to the force of gravity. Thus, the classification process will take place wherever the drag force exceeds the force of gravity. Particle classification takes place in the separation zone (7) where the particles are interacting with the hot-air flow. Fine fractions (0.125–4 mm) will continue to fall downwards (2), while ultrafine particles (<0.125 mm) and the vapor will be dragged by the hot air into the cyclone (4) where the ultrafine particles will be separated. The exhaust gases that rise through the cyclone are partially recirculated into the HAS setup, helping to economize on energy and to avoid heat loss. Gas recirculation is controlled by a compressor equipped with a pressure sensor that maintains a vacuum pressure within the cyclone. Temperature sensors are installed within the top compartment near the HAS burner, and within the bottom compartment near the recycled fine fraction output gate. Temperature sensors help to control the drying process and the gas temperature is always maintained above 120 °C, to prevent any condensation of vapor. A typical temperature profile during operation is shown below, in Figure 3. The sensor measures the temperature of the outgoing gas stream before it enters the cyclone. A drop-in temperature indicates an exchange of temperature between the wet input materials and the hot gas stream. The input is halted whenever the temperature at the top of the HAS section falls to below 130 °C and is then restarted when the temperature rises to 230 °C. Flow and relative humidity are frequently monitored to guarantee an effective drying cycle.



Fig. 1. Material flow in the recycling process.

Unlike conventional air classifiers, the unique feature of HAS

# Table 1

Main properties of the HAS feed.

Test	Standard	EoL SCW 0–4 mm	EoL LCW 0–4 mm
Saturated Surface Dry Density (g/cm3) Water absorption at 24 h (%)	UNE-EN 1097–6:2014 UNE-EN 1097–6:2014	2.28	2.39
Moisture content (%)	_	8.2	7.1



Fig. 2. Diagram of HAS and working principles (Left). A pilot scale HAS lab setup at the Delft University of Technology (Right): 1) Aggregate feed; 2) Recycled fine particles; 3) Recycled ultrafine particles; 4) Cyclone; 5) Burner; 6) Blower; 7) Separation chamber.

technology is the presence of horizontally staggered tubes within the vertical chamber (7). Their function is to increase the treatment period of fine aggregates for efficient heat/mass transfer between the wet aggregate and the hot gas (5). Furthermore, any minuscule wooden and plastic shards that can at times be found in EoL concrete waste will be carbonized while in the heating zone.

#### 2.2. Output of ultrafine recycled particles and test method

Based on the above configuration and the operating conditions of the HAS process, two major products are produced through two output streams: the ultrafine particles (<0.125 mm) collected from the cyclone; and, the fine fraction (0.125–4 mm), collected at the bottom, as shown in Fig. 4.

The ultrafine recycled concrete particles (Fig. 4 (3)) collected from the cyclone compartment are a subject of great interest in this



Fig. 3. Temperature profile at the top compartment of the HAS during feeding at 3 t/h.

study for further physical and chemical characterization and for the study of their potential use as SCM in new circular cement-based products. The two SCM are:

- Ultrafine recycled siliceous concrete addition (URSCA) from EoL-SCW;
- Ultrafine recycled limestone concrete addition (URLCA) from EoL-LCW.

The viability and the potential of the ultrafine recycled particles as SCM was assessed in three complementary processes:



Fig. 4. (1) HAS input. (2&3) HAS recycled fractions.

- Firstly, a preliminary physical and chemical characterization was completed for the ultrafine recycled particles following HAS treatment.
- Then, preliminary assessments of the physico-chemical effects and the cementitious properties were performed by studying the hydration kinetics (calorimetry) and the mechanical properties (compressive strength) of cement pastes, prepared with a commercial clinker blended with different combinations (up to 10%) of ultrafine recycled particles following HAS-treatment.
- Finally, the potential use of the ultrafine recycled concrete particles as SCM and the reduction in cement content of the new cement-based materials were both evaluated. Standardized mortars were prepared with a commercial CEM II Portland cement blended with different combinations (up to 10% of cement replacement) of ultrafine recycled particles from the HAS for the evaluation of their hydraulic activity. At this stage, a CEM II was selected, as this research work is framed within an innovation project (EU project) where the recycled concrete fractions are used for the manufacturing of precast concrete panels. The European precast concrete industry mostly uses this type of cement for its products. The optimum cement replacement rate and the effects of the ultrafine recycled concrete particles on the mortars were investigated, in order to assess the potential reduction of cement content and the effect on the mechanical properties of the new green circular cement-based products.

#### 2.2.1. Characterization of the ultrafine recycled concrete particles

#### - Particle size distributions through Laser Diffraction:

A Mastersizer 3000 laser diffraction particle size analyzer was used to assess particle distribution. It delivers rapid, accurate particle size distribution measurements of both wet and dry samples, by measuring particle sizes over the nanometer to millimeter range. Reliable data were recorded on the small particle footprints of all the samples.

When a laser beam passes through a particulate sample previously dispersed (in alcohol), the angular variation in the intensity of the diffracted light can be measured. Large particles scatter light at acute angles in relation to the incident beam and small particles scatter light at obtuse angles. Thus, the angular diffraction intensity can be analyzed, from which the particle size is determined by laser diffraction spectroscopy. The particle size is reported as a volume equivalent sphere diameter. In this study, the optical parameters selected for the ultrafine recycled concrete particle analysis were IR = 1.544 and IAbs = 0.001.

#### - Density and specific surface area:

The density and the specific surface area of the materials were measured with the Blaine permeability test as per standard UNE-EN 196–6:2010 (E (2010)E-196–6:2, 2010).

#### - Chemical composition and Loss on Ignition:

The chemical composition of the materials was determined and quantified by X-ray fluorescence (XRF) spectroscopy, which was complemented by the Loss On Ignition (LOI) test results. The latter technique consists of heating ("igniting") a sample of the material to the point of ignition at a specified temperature, allowing volatile substances to escape, until its mass remains constant. The volatile materials usually consist of "combined water" (hydrates and labile hydroxy-compounds), organic substances, and carbon dioxide from carbonates.

### - Mineralogy:

The mineralogical composition of the materials was quantified using X-ray diffraction (XRD) spectroscopy, employing MoK $\alpha$ 1 radiation in a BRUKER diffractometer; model D8 Advance. The optical system consists of a primary monochromator and the LYNXEYE XE detector system. The measurements were taken from 3° to 40° (2 $\theta$ ) at 50 kV and 50 mA, while the sample was rotated, in order to increase the particle statistics. MoK $\alpha$ 1 radiation was used, to prevent fluorescence interference, due to the presence of Fe in the materials, which is a source of error when CuK $\alpha$  radiation is employed. The samples were mixed with the crystalline standard (ZnO) and, following the identification of the phases; quantification was completed using the TOPAS software, version 4.2.

#### 2.2.2. Assessment of cement-paste hydration and hardening

The physico-chemical effects of the ultrafine recycled concrete particles processed through the HAS were firstly evaluated by studying their influence on the cement pastes. Cement pastes composed of a commercial clinker blended with different quantities of ultrafine recycled concrete particles from the HAS were prepared. A standard clinker used for the commercial production of CEM I was employed. It was supplied without a setting regulator by the Spanish company FyM, a subsidiary of the Heidelberg Group, and was selected to determine the effect, at early ages, of the ultrafine recycled concrete particles mixed with the pure phases of the clinker. Possible interference from binary and ternary cementitious matrixes (gypsum, limestone, fly ashes, etc.) was therefore avoided. The ultrafine recycled concrete particles were employed in partial substitution of the clinker at rates of 3%, 5%, and 10% by weight of clinker (Table 2). Prismatic specimens of  $1 \times 1 \times 6$  cm were prepared. 30 g of distilled water was added to each blend, yielding a constant water-to-solid ratio of 0.30 for all the tests. This ratio, commonly between 0.25 and 0.60, was adjusted, to obtain an optimal cement-paste consistency and to prevent physical and chemical problems due to excess water (Chaussadent, Baroghel-Bouny, Hornain, et al.). Once demolded, the prisms were stored in mains water for 6 h, 1, 2, 7, and 28 days.

Cement-paste hydration and hardening were assessed with a calorimetry study (hydration kinetic) at 24 h and compressive strength tests (mechanical behavior at different ages: 6 h, and 1, 2, 7 and 28 days), respectively.

#### - Calorimetry:

Clinker paste hydration was evaluated by calorimetry employing a Q2000 TA Instruments calorimeter to determine the heat flow of the pastes from the end of the mixing period up until 48 h thereafter, at a constant temperature of 25 °C. The heat flow was normalized to the weight of the clinker employed in each sample.

#### - Compressive strength:

Table 2	
Cement pastes employed for the study of hydration and harden	iing.

Cement Pastes	Clinker (g)	Ultrafine recycled concrete particles (g)
Clinker - Ref. Clinker - 3% URSCA	100 97	0 3
Clinker - 3% URLCA	05	-
Clinker - 5% URSCA Clinker - 5% URLCA	95	5
Clinker - 10% URSCA Clinker - 10% URLCA	90	10

Compressive strength tests were performed with an AUTOTEST 200/10-SW hydraulic press, equipped with a clamp for  $1 \times 1 \times 6$  cm prismatic specimens, at 6 h and at 1, 2, 7 and 28 days.

#### 2.2.3. Effect of the recovered SCM in new circular mortars

Standardized mortars were mixed as per standard UNE-EN 196-1 (E (2018)E-196–1:2, 2018), in order to study the effect of the ultrafine recycled particles employed as SCM. The mortars were performed in the laboratory under controlled conditions using a commercial OPC cement (CEM II 42.5), widely used in the European precast industry. Those controlled conditions are required to certify the commercial cements according to the European standard UNE-EN 197–1:2011 (E (2011)E-197–1:2, 2011) for common cements.

The mixing process was performed with a planetary mixer as per standard UNE-EN 196-1:

- Addition of water;
- Addition of solid constituents;
- 15 s of mixing at low speed;
- 75 s of mixing at normal speed.

The materials consisted of a commercial OPC cement (CEM II/A-LL 42.5R from the Heidelberg Group), distilled water, standardized siliceous sand (0–2 mm) as per UNE-EN 1015-2, and the ultrafine recycled concrete additions that were obtained from the HAS and used as SCM. This specific cement was selected, because it is the object of study in the framework of the VEEP European project (EU project) and it is widely used for the production of pre-cast components. The potential reduction of cement content and the effect on the mechanical properties of the new green circular cementbased products could therefore be assessed. This cement type is also of interest, as it only contains inert ground limestone (between 6 and 20 wt%), so the analysis is unaffected by pozzolanic admixtures, such as fly ash, which would complicate the discussion of the results. The water to binder (cement + SCM) ratio was maintained constant at 0.5. Three substitution rates of cement (3, 5 and 10%) were tested with the following formulas (Table 3):

The mortar mixes were studied in both the fresh state and the hardened state, in order to assess the effect of the recovered SCM on the commercial cement and to establish the reduction in cement content.

#### - <u>Slump test</u>

Determination of mortar consistency on the shaking table, as per standard UNE-EN 1015-3, involves a test procedure in which the mold (60 mm in height, internal diameter: base 100 mm - top 70 mm) is placed in the center of the flow table and filled with two successive layers, each of which is tamped ten times with a tamper. The mold must be held firmly in place during this operation. Any excess mortar is wiped from the top of the mold with a palette knife and the area around the base of the mold is cleaned with a cloth. A period of approximately 15 s elapses before the mold is removed and the table is then jolted 15 times at a rate of one jolt per second. The diameter of the mortar spread is measured with calipers in two directions at right angles to each other and both results are noted.

#### - Flexural strength

The flexural strength of the hardened prismatic mortar samples was determined in three-point loading tests. The compressive strength was determined on each half of the prism, following the failure by breakage of each specimen.

The flexural and compressive strengths of the prismatic specimens with dimensions of  $40x40 \times 160$  mm were measured as per standard UNE-EN 1015-11. A total of 3 specimens were tested at three different ages (1, 7 and 28 days).

#### - Compressive strength

After the flexural strength test, the two parts of the broken specimen were recovered, and a compression test was performed (UNE-EN 1015-11) on each part: 6 specimens were tested at each age (1, 7 and 28 days), for greater test accuracy.

#### 2.3. Environmental assessment

#### 2.3.1. Goal and scope definition

Life Cycle Assessment (LCA) was selected to evaluate the environmental viability of employing the ultrafine recycled concrete particles obtained with HAS technology as SCM in new circular cement-based products. LCA methodology was performed as per standard ISO 14040-44 (O 14044:2006 (2006) Ges, 1404; O 14040:2006 (2006)A, 1404).

To this end, in a first stage, the environmental impact was assessed for a commercial cement (CEM II 42.5R) and the recovered ultrafine particles from two streams, siliceous concrete waste and limestone concrete waste (URSCA and URLCA). Two energy systems for the HAS were assessed, firstly, diesel fuel on which the HAS system runs at present, and secondly biomass fuel, in the event of future technological upgrades. Then the environmental impacts of the CEM II 42.5R, and the same cement blended with the recovered ultrafine particles, were compared.

For that purpose, four scenarios, intended to assess the environmental impact of the blended cement with a replacement rate of 5%, reflecting the optimal results of this research work, were compared with the commercial CEMII 42.5R from a cradle-to-gate perspective.

- S1.1–5% of CEMII 42.5R was replaced by URSCA with the HAS consuming diesel fuel.
- S1.2–5% of CEMII 42.5R was replaced by URSCA with the HAS consuming biofuel.
- S2.1–5% of CEMII 42.5R was replaced by URLCA with the HAS consuming diesel fuel.

#### Table 3

Standardized mortar mixtures.

Mortar Pastes	Cement II/A-LL 42.5R(g)	Sand(g)	Recovered SCM(g)	Water(g)
CEMII - Ref.	450	1350	0	225
CEMII - 3% URSCA	436.5		13.5	
CEMII - 3% URLCA			13.5	
CEMII - 5% URSCA	427.5		22.5	
CEMII - 5% URLCA			22.5	
CEMII - 10% URSCA	405		45	
CEMII - 10% URLCA			45	

• S2.2–5% of CEMII 42.5R was replaced by URLCA with the HAS consuming biofuel.

For the recovered products generated by HAS, the input and output flow of the three technologies (crushing, ADR and HAS) were considered within the system boundaries, including the transport of the ADR and HAS equipment to the demolition site as ADR and HAS were designed for transportability and on-site production to save the cost of transporting huge amount of EOL concrete to the recycling plants. The boundaries of the CEM II system run from the extraction of raw materials to the manufacture of the final product. The system boundaries of the blended cement are shown in Fig. 5.

So that the LCA was comparable, 1 ton of each product (CEMII, recovered ultrafine fractions from URSCA and URLCA and the two blended cements) was selected as the functional unit.

#### 2.3.2. Life cycle inventory (LCI)

Material flows and energy consumption within the HAS, to recover the recycled products (URSCA and URLCA), were monitored with the data from earlier experiments within the framework of the VEEP project. These primary data were mainly supplied by the partner responsible for each technology (crushing, ADR and HAS) (Zhang et al., 2019a) (see Table 4 and Fig. 6). The data on the background processes (electricity, water, fuel, etc.) were taken from the European Life Cycle Database (ELCD) (ELCD EPLCA, 2015), except for the production of CEMII 42.5R that were taken from the Eco-invent database, as the CEMII production data were not available in the ELCD. More details on the processing system are available in the Appendix section (Table 8).

Although the foreground processes in this analysis were gathered from primary data and the background processes come from reliable LCI databases, the following key assumptions were considered to perform the LCA:

- The distance of the ADR and HAS equipment transported from the storage depot to the demolition site was set at 50 km. Transport back to the storage depot was also considered.
- The environmental impact of transportation of the mobile ADR and HAS was calculated, considering that the demolition of a typical building will produce around 15,000 tons of EoL concrete (C2project -Grant Ag, 2651). Therefore, the environmental impact from the transport of equipment was allocated based on the amount of concrete for disposal.
- All processes were assumed to consume average European energy values. The exception was for the biomass energy, which is assumed to use the Netherlands consumption mix.



Fig. 5. System boundaries of blended cement production.

Table 4				
Primary data for the three technologies	(crushing	ADR	and	HAS

	Crushing	ADR	HAS
Productivity (t/h)	300	50	3
Diesel (MJ/t)	5.07	-	216
Water (L/t)	0.7	0.7	_
Electricity (kWh/t)	-	0.46	0.01
Mass of equipment (t)	-	25	7.5

- The energy consumption of the blending process of the ultrafine fraction and the CEMII was considered null, because it was assumed that the blended cement could easily be prepared at the construction site when all the concrete components are poured into the mixer without increasing the energy consumption of the mixer. In addition, the distance to the construction sites from the cement plant and the demolition site was supposed to be identical and was not considered in the LCA as it had no effect on the goal of the study.

The technological system to recover the ultrafine particles (URSCA and URLCA) is multifunctional as different particle sizes fractions are obtained. In this case, the ISO 14040 (O 14044:2006 (2006) Ges, 1404) recommends avoiding allocation, either by dividing the process or by expanding the system boundary. Based on the data, expansion of the system boundary was selected. Therefore, some unintended co-products for this analysis, were also produced (see Fig. 6).

#### 2.3.3. Life-cycle impact assessment

The software OpenLCA 1.7 along with the CML impact assessment method (Guinée et al., 2002) was used to calculate the environmental impact. The impact categories were selected as per standard EN 15804:2012 (E-15804 (2013)E-E, 1580), including Global warming potential (GWP-kg CO<sub>2</sub> eq.), ozone depletion (ODP-kg CFC<sup>11</sup> eq.), acidification (AP-kg SO<sub>2</sub> eq.), eutrophication (EP-kg (PO<sub>4</sub>)<sup>3-</sup> eq.), photochemical ozone creation (POCP-kg Ethene eq.), depletion of abiotic resources-elements (ADP-E-kg Sb eq.) and depletion of abiotic resources-fossil fuels (ADP-F-MJ). To express the different impact scores on a common scale, the environmental impact results were normalized according to the normalization factors given for the European emission per persons emission unit for the year 2000 proposed by the CML (Guinée et al., 2002).

#### 3. Results and discussion

#### 3.1. Characterization results

#### 3.1.1. Particle size distribution, density and specific surface area

The PSD of the HAS outputs (URSCA and URLCA) were analyzed together with the commercial clinker and the commercial cement employed for this study (Fig. 7).

The density, the specific surface area and the characteristic sizes of the PSD are shown in Table 5.

No relevant differences related to the PSD, density and specific surface area were observed between the two ultrafine recycled particles. However, in agreement with previous studies, both types of ultrafine recycled particles showed lower specific surface areas than the clinker and the CEMII (Oey et al., 2013). For the preliminary physico-chemical study, the PSD results also suggested that the use of clinker (instead of CEMII) blended with the ultrafine recycled particles should be more suitable, because of the lower difference of the PSD, improving the packing density of the cement pastes (Gallias and Bigas, 2002). The higher content of hydraulic phases can be also considered as an advantage offered by the use of



Fig. 6. Mass balance for the production of 0.05t of URSCA and URLCA from EoL-SCW and EoL-LCW, respectively.



Fig. 7. Particle size distribution of ultrafine limestone and siliceous aggregates after the HAS treatment (URSCA and URLCA), clinker and CEMII.

Table	5
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Density and specific surface area of ultrafine aggregates.

Sample	Specific surface area (cm <sup>2</sup> /g)	Density (g/m <sup>3</sup> )	D10 μm	D50 μm	D90 µm
CEMII 42.5	9170	3.2	3.8	15.5	37.6
Clinker	4020	3.15	6.8	24.3	59.2
URSCA	2980	2.5-2.6	10.3	57.9	117.8
URLCA	2620	2.7	10.2	49.8	121.4

clinker instead of the binary cement.

#### 3.1.2. Chemical composition, LOI and mineralogy

The chemical compositions, the LOI, and the mineralogy of all the ultrafine recycled concrete particles (both URSCA and URLCA) were compared with the contaminated fine fractions (EoL-SCW and EoL-LCW) prior to input into the HAS (see Table 6). The difference in the elemental composition between both materials corresponded to the changes induced by the HAS. Considering the impact of the technology on the treated materials, in terms of the components present in the cement pastes, the output materials should have a richer chemical and mineralogical composition than the input materials. In addition, according to the characteristics of the technology (heating up to 600  $^{\circ}$ C), a lower LOI will be expected, due to the reduction of organic impurities.

A comparison between the percentile results of the chemical characterization will show that the amounts of the main oxides CaO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and Fe<sub>2</sub>O<sub>3</sub> in the 0-0.125 mm fraction revealed a

higher composition of those elements that are normally found in pure cement pastes (Vegas et al., 2006). The ultrafine particles had higher quantities of those main oxides when compared to contents sourced from pure hydrated cement, as they are diluted with particles of quartz, calcite and albite (for the URSCA) and particles of dolomite and calcite (for the URLCA). In a similar way to the chemical composition, the mineralogy presented an increase in the amorphous content containing hydrated phases of cement pastes (C–S–H), a lower quartz content in the primitive siliceous natural aggregates and a lower dolomite content in the primitive limestone natural aggregates, demonstrating that the ultrafine recycled concrete particles enriched the hardened cement paste. Those results were consistent with the results of Lotfi et al. at lab scale (Lotfi and Rem, 2016, 2018), who observed that an EoL concrete from siliceous concrete waste processed with HAS technology at lab scale was enriched with CaO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and Fe<sub>2</sub>O<sub>3</sub> components. It was therefore demonstrated that the observations at lab scale were also replicated at a pilot scale. As expected, the LOI decreased in both cases, implying a reduced quantity of organic impurities.

The PSD, chemical, and mineralogical results obtained in this section, will help to explain the physico-chemical effects of the ultrafine recycled concrete particles in the blended cement pastes.

#### 3.2. Assessment of cement paste hydration and hardening

Different physico-chemical effects were observed following the addition of minerals to cement matrices, which accelerated hydration rates and enhanced the mechanical properties, depending on the nature and the characteristics of the mineral addition, i.e.:

- The <u>filler effect</u> in cement, due to the physical presence of mineral additions, is known to accelerate the hydration of the clinker component. That reaction is attributed to the larger surface area of the filler that provides nucleation sites for C–S–H, as there is a clear dependence on the specific surface area of the filler particles. It has also been demonstrated that Supplementary Cementitious Materials (SCM) from natural and synthetic sources (fly ashes, silica fume, nanoparticles, etc.) have a filler effect during cement hydration, improving both mechanical and rheological properties while reducing the overall environmental impact (Diezgarcia et al., 2017; Lothenbach et al., 2011; Papadakis and Tsimas, 2002).

- C–S–H formation mechanisms have been described in nanolevel simulations (Dolado et al., 2013). Those simulations also describe the formation of C–S–H gels, in terms of rate dependent nucleation of C–S–H nanoparticles, autocatalytic growth, in a 
 Table 6

 Chemical composition LOL and mineralogy of the ultrafine recycled concrete additions

Chemical Compound	EoL-SCW 0-4 mm (wt%)	URSCA (wt%)	EoL-LCW 0-4 mm (wt%)	URLCA (wt%)
SiO <sub>2</sub>	66.92	55.91	13.03	14.54
CaO	13.40	20.50	30.92	32.35
Al <sub>2</sub> O <sub>3</sub>	4.62	6.04	2.52	3.00
Fe <sub>2</sub> O <sub>3</sub>	1.75	2.30	1.47	1.71
MgO	1.35	2.10	13.08	11.78
Na <sub>2</sub> O	0.60	2.14	0.221	0.03
K <sub>2</sub> O	0.99	1.06	0.59	0.67
SO <sub>3</sub>	0.94	1.61	1.37	1.39
TiO <sub>2</sub>	0.32	0.41	0.15	0.17
P <sub>2</sub> O <sub>5</sub>	0.32	0.09	-	-
MnO	0.08	0.12	0.03	0.04
Others	_	0.30	0.54	0.46
LOI	8.71	7.42	36.30	33.89
Mineral	EoL-SCW 0-4 mm (wt%)	URSCA (wt%)	EoL-LCW 0-4 mm (wt%)	URLCA (wt%)
Dolomite	_	_	64.1	56.3
CaMg(CO <sub>3</sub> ) <sub>2</sub>				
Amorphous content	28.8	37.5	14.9	17
Calcite	5.8	8.4	13.4	18.3
CaCO <sub>3</sub>				
Quartz	59.7	48.7	7.6	7.7
SiO <sub>2</sub>				
Albite	3.2	2.3		
NaAlSi <sub>3</sub> O <sub>8</sub>				
Orthoclase	2.5	2.1	_	0.7
KAlSi <sub>3</sub> O <sub>8</sub>				
Others	-	0.1	-	-

hierarchical manner, of C–S–H nanoparticles, to form C–S–H clusters, and the aggregation of these growing clusters. It is well established that the artificial increase of C–S–H nuclei, due to the addition of C–S–H nanoparticles (seed effect) during the mixing, will greatly accelerate C–S–H formation and a concomitant strength gain in the mechanical properties. In this context, it must be highlighted that both URSCA and URLCA, respectively, contained significant amounts, 37.5% and 17%, of amorphous matter (C–S–H gel).

- The presence of hardened cement in the ultrafine recycled concrete particles and the high temperatures applied during the HAS process can induce the regeneration of the cementing activity of the hardened cement paste powder by dehydration process (Serpell and Lopez, 2013a). Chemical transformations occur in the hydrated paste at high temperatures, leading to unhydrated compounds with cementitious characteristics. Various authors (Shui et al., 2009; Serpell and Lopez, 2013a; Hu YJH, 2007; Alonso et al., 2004) studied the effect of temperature on the dehydration of hydrated cement paste and concluded that the dehydration process required to produce reactivated cementitious materials involves much lower temperatures than those required to produce new Portland cement. Poorly crystallized Calcium Silicate Hydrates (C–S–H) were also shown to decompose gradually at over 300 °C to produce modified C–S–H ( $\beta$ -C<sub>2</sub>S), CaO, and dehydrated C–S–H (nesosilicate) (Alonso et al., 2004; Okada et al., 1994) which rehydrates upon contact with water to produce new C-S-H. They therefore displayed cementitious behavior similar to that of the calcium silicates present in Portland cement, developing strength at advanced curing ages and thus potentially enabling the production of construction materials (Shui et al., 2008). The increase in CaO and the amorphous phases that are shown above, in Table 5, are consistent with this observation.

In the framework of this research, the physico-chemical effects

resulting from the addition of the ultrafine recycled concrete particles from siliceous concrete waste (URSCA) and limestone concrete waste (URLCA) were studied during the hydration and hardening of cement pastes combining different amounts of clinker and ultrafine recycled particles.

#### 3.2.1. Hydration kinetics

The study of hydration kinetics was focused on the acceleration period of the main peak of the heat flow during setting which can be described by its slope. Fig. 8 shows the slope value as a function of the replacement level for both types of ultrafine recycled particles that were employed. The curves were normalized by dividing the results by the weight of the clinker, the only reactive component, providing insight into the influence of the ultrafine particles on the hydration of the cement pastes. The hydration kinetics at an early age will help to explain the influence of filler, its effect at an early age and the seed effect.

An acceleration of the hydration process was observed when employing both ultrafine recycled particles. The main peak was higher and the acceleration slope steeper as higher ratios of ultrafine recycled particles were used, regardless of the nature of the particles. A higher amount of particles provided further nucleation sites for C–S–H, directly related to the filler effect.

Comparing both materials, with the same replacement rates, the URSCA showed slightly steeper acceleration curves, which shifted towards earlier ages when compared to the URLCA. The filler effect was directly influenced by the PSD and the specific surface area of the mineral addition (Berodier and Scrivener, 2014). In this case, as the PSD and specific surface of both URSCA and URLCA were similar, the differences between both materials might be attributed to the larger amounts of amorphous matter (C–S–H gel) in the URSCA sample than in the URLCA sample (Table 6), which increased the seed effect.

Subsequently, the results showed that accelerated hydration, at 24 h, was related to both the filler and the seed effect. The filler effect contributed to a steeper initial slope and to an increase in the



Fig. 8. Normalized heat flow of cement pastes.

Left: employing URSCA materials from HAS Right: employing URLCA materials from HAS.

main peak, enhancing the heat release at an early age. This effect appears to be more relevant when employing URLCA (limestone). In turn, the seed effect steepened the slope still further, especially in relation to the acceleration of the hydration process, which shifted the hydration process without increasing the heat release value, linked particularly to the URSCA particles.

A study of the mechanical properties of the cement pastes was performed, in order to clarify the effect of the ultrafine recycled particles on hardening at more advanced ages.

#### 3.2.2. Compressive strength

The compressive strength results of the cement pastes blended with different quantities of ultrafine recycled particles are presented in Fig. 9, below.

The analysis of the results for compressive strength, both at 6 h and at 1 day of curing, yielded similar conclusions to those for the hydration kinetics, i.e. substitutions of up to 10% of ultrafine recycled particles enhanced the hydration kinetics regardless of the nature of the additions. Slightly higher compressive strength was observed for the URLCA, confirming the results of the literature for the limestone additions related to the filler effect and the higher dissolution of the limestone particles at an early age. The results of previous studies (Oey et al., 2013; Berodier and Scrivener, 2014; Marie and Berodier, 2015) have reported that limestone is more effective than quartz as an accelerant of clinker hydration that stimulates C–S–H nucleation, due to the lower specific C–S–H/ limestone interface energy in comparison with C–S–H/quartz interfaces. The studies also revealed that limestone additions have a higher accelerating effect on hydration, due to the dissolution of the



Fig. 9. Compressive strength of cement pastes.

limestone and the favorable surface structure, providing a "template" for C–S–H precipitation. However, at substitution rates of up to 10% no difference between URSCA and URLCA was noticeable at 6 h, showing that the seed effect was the main factor behind the enhanced hydration of the URSCA at an early age.

At more advanced ages (2, 7 and 28 days), the effect of the URLCA on the compressive strength of the cement pastes was more relevant than the URSCA, at similar levels of replacement. When employing low (3%) and moderate (5%) replacement rates of URSCA, the compressive strength is similar or slightly higher than the reference, meanwhile it is significantly higher than the reference for the URLCA. The use of the higher rate (10%) slightly penalized the resistance of the cement pastes in almost all cases, while hydration at early ages was related more to the seed and filler effects. It should be noted that the effects of lower proportions of both URSCA and URLCA continued throughout the curing time. In related research works (Shui et al., 2009; Serpell and Lopez, 2013b), significant gains in the strength of cement pastes at intermediate (7 days) and advanced ages (28 days) were caused by the incorporation of thermally treated hydrated cement pastes with regenerated cementitious activity. Those results are consistent with the literature and confirmed the cementitious activity of the ultrafine recycled concrete particles, due to the concentration of chemical components and mineral phases present in the original cement and the presence of modified C–S–H ( $\beta$ -C<sub>2</sub>S) and dehydrated C–S–H (nesosilicate) (Alonso et al., 2004; Okada et al., 1994) (Alonso et al., 2004; Okada et al., 1994) (Alonso et al., 2004; Okada et al., 1994) (Alonso et al., 2004; Okada et al., 1994), which rehydrates upon contact with water producing new C-S-H and displaying cementitious behavior.

Broadly speaking, the ultrafine recycled concrete particles from HAS technology presented better mechanical behavior than the reference clinker. This is due to the filler effect at early ages and better regeneration of cementitious activity. The best results were obtained with low-to-moderate substitution rates of 3 and 5%, thereby providing new perspectives for the design of new blended circular systems and demonstrating the potential benefits of the HAS ultrafine products when employed as SCM in new circular cement-based products.

#### 3.3. Effect of SCM on new circular mortars

The ultrafine recycled concrete particles employed as SCM and their effects on the quality of new circular cement-based products were finally assessed and the best replacement rate determined through the study of the fresh (slump test) and the hardened properties (flexural and compressive strength) of the standardized cement mortars. The use of standardized mortars yielded highly reliable test results, in tests that are normally applied to certify commercial cements, as per standard UNE-EN 196–1:2018 (E (2018)E-196–1:2, 2018).

#### 3.3.1. Fresh properties

The workability of the mortars with different contents and with both types of ultrafine recycled concrete particles are presented in Fig. 10, below.

The workability of a cement-based material with mineral additions greatly depends on the particle size, specific surface area, particle shape, and replacement level. In general, smaller particle sizes and higher specific surface areas of the mineral additions implies that the water uptake of the concrete will be higher. The authors of one work, Ullah Khan et al. (2014), concluded that mineral additions may be categorized into two groups: chemically active mineral additions and microfiller mineral additions. Chemically active mineral additions decrease concrete workability and setting times, although they increase hydration heat and reactivity. In contrast, the addition of minerals as a microfiller will increase concrete workability and setting times, but will decrease hydration heat and reactivity. The authors stated that mineral additions with low reactivity and a moderate filler effect helped to maintain workability and even increased it at times.

As can be seen in Fig. 10, the workability of the mortars is not significantly affected by the use of ultrafine recycled concrete particles. With low replacement rates, the workability is slightly increased revealing a low reactivity and filler effect. As both the particle size and the specific surface area were higher than the same values for cement, the mortars had a lower water uptake than the reference specimen. Although the substitution rate makes the mortars slightly stiffer, we can conclude that workability was maintained confirming the results of the literature and the low reactivity and the moderate filler effect of the ultrafine recycled concrete particles.

The siliceous or limestone nature had no influence on workability. As previously stated, the topology of the additions had a great influence on workability. Both types of ultrafine recycled concrete particles employed in this study have similar particle size distributions, specific surface areas, and morphologies (EU project), however the URLCA slightly improved workability, due to the higher filler effect. In that sense, the high performance of HAS technology leads to a homogeneous and constant flow of ultrafine recycled concrete particles, regardless of the original source of the EoL concrete.



Fig. 10. Consistency of mortar pastes with ultrafine recycled concrete particles.

#### 3.3.2. Mechanical properties

Finally, the ultrafine recycled concrete particles used as SCM and the effects of their mechanical properties on the mortars were tested on days 1, 7, and 28. The results of the compressive and flexural strengths are presented in Fig. 11.

Both the compressive and the flexural strength of the samples were maintained and even increased when the cement was partially replaced with low (3%) and intermediate (5%) amounts of recovered SCM. A drop in performance that was higher than the substitution rate was observed with replacements of 10%, confirming the observations made with the cement pastes. An observation that is aligned with other studies dealing with fine recycled concrete aggregates and recycled cement paste (Gastaldi et al., 2015; Aprianti, 2017; Kumar et al., 2017; Singh, 2018) when employed as SCM in mortar pastes and where, Consequently, depending on the nature and properties of the SCM, it was found an optimal replacement rates of up to 10% for similar SCM (limestone and silica-based).

Contrary to the cement pastes, the influence of SCM from siliceous sources (URSCA) was more important in this case than in the limestone-based samples (URLCA). This observation might be due to the type of cement (CEM II/A-LL 42.5R), the content of which already had high rates of natural limestone of up to 20% of the total mass. The effect of the URLCA as SCM can be distorted by the presence of homologous limestone additions, adding a greater quantity of limestone powder that reached amounts of up to 30% for this kind of cement. This distortion is one of the reasons why the physico-chemical effects of the mineral additions is commonly studied with clinker or cement type I pastes, avoiding other potential interference from other binary or ternary cementitious matrixes (gypsum, limestone, fly ashes, among others). Different studies (Nehdi et al., 1996; Gudissa and Dinku, 2010) have demonstrated the effect of moderate and high replacement rates of cement by limestone filler on the strengths of mortars, concluding



Fig. 11. Compressive and flexural strength of mortar pastes blended with ultrafine recycled concrete additions.



Fig. 12. LCIA relative results. Comparison between 1 ton of recovered ultrafine concrete particles and 1 ton of CEM II 42.5R.

than moderate rates of around 15% had no effect on the strength at all ages, although the use of limestone filler did cause significant strength losses when employing replacement rates higher than 15%.

All in all, gains or maintenance of compressive strengths are



Fig. 13. Normalized life cycle environment results.

pieces of evidences at replacement rates of 3 and 5% of cement by URSCA and URLCA, thereby, revealing the positive contribution of such additions. Furthermore, our research team is currently engaged in studies, with the aim of determining the influence of the heterogeneity of diverse concrete waste samples, to provide a better understanding of the paths that lead to higher hydration and hardening, and to compare various recycling processes, as well as durability issues.

#### 3.4. Environmental impact assessment

The environmental impact of the HAS ultrafine recycled concrete particles in substitution of the commercial cement (CEMII 42.5R) was evaluated, in order to determine the environmental feasibility of their use as SCM at the optimum replacement level (5%) based on the findings of this study.

In order to analyze the contributions of the recovered ultrafine particles in blended cements, the relative environmental performance of the production of 1 ton of recovered particles (URSCA and URLCA) for the two energy systems of the HAS was firstly compared to the production of 1 ton of the commercial CEM II 42.5R (Fig. 12). The results revealed important benefits, higher than 93.5% in most of the impact categories for both URSCA and URLCA, compared to the commercial cement, with the exception of depletion of fossil fuel resources (ADP-F) and photochemical ozone creation (POCP) when diesel fuel is used as energy source of the HAS. Both impacts showed more modest reductions of about 54.4% and 85.6% respectively compared to the commercial cement, this is due to the consumption of diesel fuel from the HAS process to heat air. In fact, when the fossil fuel is replaced by biomass, it can be seen a reduction of all impacts, especially of the most affected, ADP-F and POCP. As expected, the thermal energy consumption of HAS technology is one of the hotspots for the environmental improvement in the treatment process of the EoL concrete. On the other hand, slight differences were found in the environmental impacts between URSCA and URLCA, mainly due to the lower flows of recycled material when the EoL-SCW was processed (see Fig. 6).

Regarding the global warming potential (GWP), the relative impact of the recovered particles compared to the commercial cement is practically negligible, obtaining benefits greater than 98.3%. This fact is due to the simplicity and relative low energy consumption of the HAS technology, no matter the energy system, compared to the cement manufacturing process, which is very energy intensive. It is estimated that more than 50% of the CO<sub>2</sub> emitted by the cement sector are released only from the calcination of limestone to produce Portland clinker (Lehne and Preston, 2018). In this sense, the research in clinker-lowering technologies is of great importance for reducing the emissions of the cement sector, i.e. processes and products that lower the share of Portland clinker in cement and concrete like the HAS technology and the use of recovered particles as SCM.

The effect of the recovered SCM in the novel blended cements was evaluated in a second step. Table 7 shows the environmental impact results of the partial replacement of up to 5% of the commercial cement by the ultrafine recycled concrete particles (<0.125 mm), employing both diesel fuel and biomass fuel. The results revealed reductions up to 5% in most of the environmental impacts categories that were assessed when using URSCA and URLCA in partial replacement of cement. This observation is directly related with the negligible impact of the HAS process compared to the cement production process, as demonstrated above. However, and always in line with what was previously discussed, the environmental impact on ADP-F presents lower reductions (2.7–2.8%) when diesel is the energy source of the HAS since the impact of cement production is only reduced by about 45% Table 7

Final results of life-cycle impact assessment. Comparison between the reference CEMII 42.5 and the blended cement employing 5% replacement of CEMII 42.5 by HAS-recovered SCM.

	Ref. CEMII 42.5 (1t)	S1.1 95% CEMII 42.5 + 5% URSCA (Diesel fuel) (1t)	S1.2 95% CEMII 42.5 + 5% URSCA (Biofuel) (1t)		S2.1 95% CEMII 42.5 + 5% URLCA (Diesel) (1t)		S2.2 95% CEMII 42.5 + 5% URLCA (Biofuel) (1t)		
	Impact	Impact	Impact Difference	Impact	Impact Difference	Impact	Impact Difference	Impact	Impact Difference
GWP [kg CO2 eq]	8.12E+02	7.72E+02	-4.92%	7.72E+02	-4.97%	7.72E+02	-4.93%	7.72E+02	-4.98%
ODP [kg CFC 11 eq]	2.25E-05	2.14E-05	-4.77%	2.14E-05	-4.81%	2.14E-05	-4.84%	2.14E-05	-4.88%
AP [kg SO2 eq]	1.44E + 00	1.37E+00	-4.68%	1.37E+00	-4.89%	1.37E+00	-4.72%	1.37E+00	-4.93%
EP [kg (PO4)3- eq]	4.00E-01	3.80E-01	-4.92%	3.80E-01	-4.98%	3.80E-01	-4.93%	3.80E-01	-4.99%
POCP [kg Ethylene eq]	5.45E-02	5.22E-02	-4.28%	5.19E-02	-4.87%	5.22E-02	-4.34%	5.19E-02	-4.92%
ADP-E [kg Sb eq]	3.02E-04	2.88E-04	-4.90%	2.87E-04	-4.94%	2.88E-04	-4.91%	2.87E-04	-4.95%
ADP-F [MJ]	2.88E+03	2.80E+03	-2.72%	2.74E+03	<b>-4.77%</b>	2.80E+03	<b>-2.80%</b>	2.74E+03	<b>-4.86%</b>

compared to the production of ultrafine particles. Therefore, the technological upgrade of HAS to be powered by biofuel seems to be a promising alternative to improve even more the environmental performance.

Finally, Fig. 13 shows the normalized results to provide an overview of the relative magnitude for the different impact categories. As can be observed, GWP presents the greatest magnitude for all the assessed systems considering the value of the standardized baseline. This greater magnitude, together with one of the greatest impact reductions, makes the use of the blended cements, studied in this work, particularly interesting in the field of the prevention of greenhouse gas emissions.

In the struggle to limit climate change, technological progress is essential for the reduction of greenhouse gas emissions in all economic sectors. The use of HAS technology has been shown to be effective for the production of alternative recovered SCM for their use in novel blended cements, contributing to lowering the share of Portland clinker and therefore the reduction of CO<sub>2</sub> emissions (GWP) in the cement industry by 5%, equivalent to 41 kg CO<sub>2</sub> eq./ton of cement. The results are consistent with recently published results on parallel eco-efficiency assessments of holistic EoL concrete sorting technologies (Zhang et al., 2019b) but it should be noted that the results of the impact values cannot be directly compared, as the scope considered in each study (technology (Zhang et al., 2019b) versus product in this study) and the impact assessment methodologies and impact categories all differed. Thus, the advantages of HAS technology are that it offers the means to reduce both environmental and economic impacts. In a world where the cement industry produced 4200 million tons of cement in 2017, the reduction could have implied savings of up to 80 million tons of CO<sub>2</sub> eq./year.

#### 4. Conclusions

In view of the growing commitment to combat climate change and the consumption of natural resources, the construction sector is improving technologies, materials and manufacturing processes, contributing significantly to the reduction of both greenhouse gas emissions and the consumption of raw materials.

This research work presents the operating parameters and the working principles of a novel pilot recycling technology based on a Heating-Air classification System (HAS) designed to improve the most problematic fractions of concrete waste: the fine fractions < 4 mm. These fractions usually present problems for their recovery, due to their high levels of moisture, absorption and impurities that make them unsuitable for use in new cement-based products. The physico-chemical behavior and the environmental

impact of the ultrafine recycled concrete particles (<0,125 mm) recovered from EoL concrete have been studied, in order to assess their potential use as SCM in new circular cement-based products.

Physical, chemical and mechanical characterizations have led us to conclude that the ultrafine recycled particles from two EoL concretes show properties that make them suitable for their use as SCM in new eco-products, providing a technically and environmentally viable solution for a complete closed-loop of this waste stream. HAS technology provides a solution to the technical barrier that up until now existed for fine concrete waste fractions.

HAS technology is designed to remove both water content and impurities (reduction in LOI), while enriching the materials with CaO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub> components, and with amorphous content containing hydrated phases of cement pastes (C–S–H). These improvements provide the product with physico-chemical properties that mean it can be employed as SCM in new cement-based products for the reduction of up to 5% of the cement content. A combination of the seed effect, the filler effect and cementing activity, regenerated through the dehydration process of the initial C–S–H following HAS heat treatment, can improve the hydration and the mechanical properties both at early and advanced ages, when employed in partial replacement of up to 5% of commercial CEMII Portland cement.

The environmental assessment of the ultrafine recycled concrete particles, processed with HAS technology and converted into SCM, emphasizes a reduction of as much as 5% of the environmental impact, for all the categories under assessment, when producing new blended cements with 5% of ultrafine concrete particles. This reduction is related with the negligible impact of the HAS technology compared to the cement production process. The high potential of HAS technology to contribute to reductions in the CO<sub>2</sub> emissions of the cement industry is clear. A 5% cement replacement in new circular cement-based products would be equivalent to reductions of 41 kg CO<sub>2</sub> eq./ton of cement. In an increasingly alarming context of global warming, in which the global cement industry is responsible for 8% of all CO<sub>2</sub> emissions, producing 4200 million tons of cement in 2017, such reductions would imply savings of 80 million tons of CO<sub>2</sub> eq./year.

This novel technology provides new perspectives for the design of new blended circular products with contents of up to 5% of HASrecycled SCM, contributing both to the struggle against climate change and to the sustainability of the construction sector, by reducing CO<sub>2</sub> emissions, natural resource extraction and waste landfilling. New research work continues for the semi-industrial upscaling and the improvement of the technology through the use of biomass fuel for HAS processing of CDW.

Table 8

LCI dataset used for the LCA methodology.

Type of flow	Process	Database source
Diesel	Diesel, consumption mix, at refinery, from crude oil, 200 ppm sulphur - EU-15	ELCD v3.2
Water	Drinking water, production mix, at plant, water purification treatment, from groundwater - RER	ELCD v3.2
Electricity	Electricity Mix, consumption mix, at consumer level, AC, 1 kV-60 kV - EU-27	ELCD v3.2
CEMII	Cement production, alternative constituents 6–20%   cement, alternative constituents 6–20%   Cutoff, U - Europe without Switzerland	Ecoinvent v3.4
Biomass	Biomass (solid) for bioenergy, consumption mix, to consumer, technology mix - NL	ELCD v3.2

#### Author contribution statement

**J. Moreno-Juez**: Conceptualization, Methodology, Investigation, Writing- Original Draft preparation and Editing, Visualization.

**Iñigo J. Vegas**: Conceptualization, Methodology, Validation, Writing- Review and Supervision.

**Abraham. T. Gebremariam**: Investigation, Software, Resources, Writing – Original Draft preparation.

**V. García-Cortés**: Formal analysis, Data Curation, Writing-Original Draft and Visualization.

F. Di Maio: Writing- Review and Supervision.

#### 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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#### Appendix

LCI data from Ecoinvent and the European Life Cycle Database (ELCD) are specified in Table 8:

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