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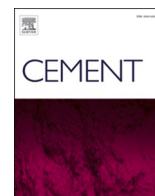To cite this publication, please use the final published version (if applicable).
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Downstream processing of End-of-Life concrete for the recovery of high-quality cementitious fractions

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A R T I C L E   I N F O

A B S T R A C T

The clinker in cement largely determines the environmental footprint of concrete. Therefore, concrete recycling should focus on retrieving high-quality cementitious fractions to replace clinker. This requires a shift from current traditional recycling techniques towards innovative recycling methods, enabling recovery of not only clean secondary aggregates, but also residual cementitious fines (RCF), potentially eliminating the carbon dioxide emissions associated with them. The production and upcycling of RCF offer new implementation routes that were previously deemed unfeasible. However, the properties of RCF may vary based on their origin, affecting their replacement and upcycling potential. Consequently, assessing the original concrete quality, with a focus on the binder type, before demolition is important. A handheld x-ray fluorescence technique appears promising for this purpose. To achieve effective separation of clean secondary aggregates from the original cementitious content, innovative crushing and separation techniques are needed. Additionally, electrostatic separation shows significant research potential for further optimizing RCF.

1. Introduction

The vast majority of concrete recycling research concentrates on the reuse of the aggregate fraction [1]. However, to significantly reduce the environmental impact of concrete, this emphasis should shift to the cementitious fraction and particularly its clinker content. This is because at least 80 % of the carbon dioxide (CO_2) emissions from concrete production stem from manufacturing the clinker in cement [2,3]. Prioritizing the acquisition of higher quality cementitious fractions that can partially or fully replace clinker is essential not only to lower environmental impact but also to reduce the use of primary raw materials.

Clinker constitutes 95 – 100 % of the total mass of Portland cement (CEM I), implying that fully recycling clinker could potentially reduce the CO_2 emissions of concrete by 80 % [2,3]. However, in other cement types, such as blended cements, a portion of the clinker (6 – 95 %) is substituted by other constituents [4]. For instance, García-Segura et al. [5] observed that blast furnace slag cement (CEM III), composed of 80 % blast furnace slag and 20 % Portland cement, only represents 30 % of the CO_2 emissions compared to pure Portland cement. Similarly, a cement type containing 35 % fly ash and 65 % Portland cement (i.e., CEM II/V)

accounts for 65 % less CO_2 emissions.

The impact of cement on emissions becomes more evident when considering its global warming potential. Various indicators contribute to determining the environmental impact of the material, with a primary focus on greenhouse gas emissions in this article. The global warming potential illustrates the potential contribution of these emissions to global warming [6]. Table 1 offers an overview of these values for the different components in a concrete mixture. It is evident that Portland cement exhibits the highest value, followed by limestone powder and ground granulated blast furnace slag (GGBS). Furthermore, the quantities of components used in a concrete mixture significantly influence their contribution to global warming potential. To illustrate this, three example mixtures are provided with the only difference being the type of cement. A comparison of these mixtures is presented in Fig. 1, indicating that Portland cement has the largest contribution in all scenarios. Even when Portland cement is partially replaced by GGBS or fly ash (FA), its contribution to the global warming potential remains the highest.

In addition to GGBS and FA, typical examples of constituents used to replace part of the clinker in blended cements are silica fume, limestone, and burnt shale [4]. These constituents may all be present in the retrieved residual cementitious fines and can influence the recycling

Table 1
Example concrete mixtures (strength class C30/37, exposure class XC2, slump class F4) with different cement types and the global warming potential of its components as indicated by specific suppliers (see references in the last column of the table), originally derived from EcoInvent 3.6. Portland cement has the largest global warming potential by one or two orders of magnitude per kg raw material.

Raw material	Portland cement based concrete (CEM I) [kg/m^3]	Blast furnace slag cement based concrete (CEM III/B) [kg/m^3]	Fly ash cement based concrete (CEM II/B-V) [kg/m^3]	Global warming potential [kg CO2 eq/kg raw material]	
Gravel 4–32 mm	1001	1001	1001	1.89E-03	[7]
Sand 0–4 mm	816	816	816	2.56E-03	[8]
Portland cement	340	102	238	8.48E-01	[9]
Ground granulated blast furnace slag (GGBS)	–	238	–	3.03E-02	[10]
Fly ash (FA)	–	–	102	1.33E-03	[11]
Limestone powder	25	25	25	3.11E-02	[12]

process and application potential (highest value) of the recycled binder fraction.

In addition to the influence of the cement fraction on the environmental impact, the increase in waste generation is also of importance. Construction and demolition waste (CDW) is recognized as one of the most important waste streams, because of its large volume and high potential for recycling and re-use [13]. In Europe, 35.9 % of the generated waste is related to CDW [14]. The European Commission estimated that this consists for approximately 60 – 70 % of concrete [13]. Generation of concrete waste is expected to grow, because of developing countries, global population growth and the related urbanization. Existing structures, such as bridges, building structures and concrete roads, are replaced or improved to accommodate the expanding population [15,16,17,18]. Additionally, a substantial number of structures in Europe were built after the second world war [19] and many of these will reach the end of their life in the near future, resulting in a significant increase in concrete waste [1]. In the Netherlands, it is expected that 60 % of the produced CDW consists of concrete, which

corresponds with approximately 15 million tons of concrete being available to be used in new concrete. This is an increase of 1.8 million tons compared to 2025 [20]. Concrete from End-of-Life structures in the Netherlands is primarily sourced from residential, commercial, and infrastructure buildings. The demolition process varies depending on the type of structure, largely due to differences in the presence of non-concrete materials. For example, viaducts are predominantly composed of concrete, whereas residential buildings contain significant quantities of other materials, such as wood, glass, gypsum and anhydrite. This results in a more complex demolition process for residential buildings to comply with the requirements set by EN 12,620 [21], which regulates the composition of recycled concrete fractions, limiting the inclusion of non-concrete components. Table 2 provides an overview of various concrete sources, along with their market share and expected strength classes. However, these strength classifications may vary, as they are typically determined by the structural engineer and also depend on the assigned exposure class [21,22]. The expected growth of concrete waste makes it possible to increase the amount of recycled concrete fractions, such as the residual cementitious fines, with the goal of fully reusing them in new concrete without loss of quality. It is essential, therefore, to identify the best methods for producing high-quality residual cementitious fines.

To achieve these higher quality cementitious fractions from recycled concrete, a transition from the current traditional recycling techniques towards more innovative recycling techniques is required. The current traditional concrete recycling process (Fig. 2) relies on a continuous input of primary raw materials and specifically of primary binder to meet the demand for new concrete. This is because the quality of the produced secondary coarse (>4 mm) aggregate fraction is insufficient, particularly due to contamination with residual and porous cement

Table 2
Sources of End-of-Life concrete in the Netherlands and expected most commonly used strength classes per source type [23,24,25,26,27,28].

	Residential	Commercial	Infrastructure	Agriculture	Other
Concrete market share	28.5 %	30 %	31.1 %	6.5 %	3.9 %
Strength class Examples	C20/25-C30/37 Buildings, driveways, garages	C30/37-C40/50 Office, retail, hotels	C35/45-C60/75 Highways, bridges, airports, dams	C25/30-C35/45 Buildings, paved surfaces	– –
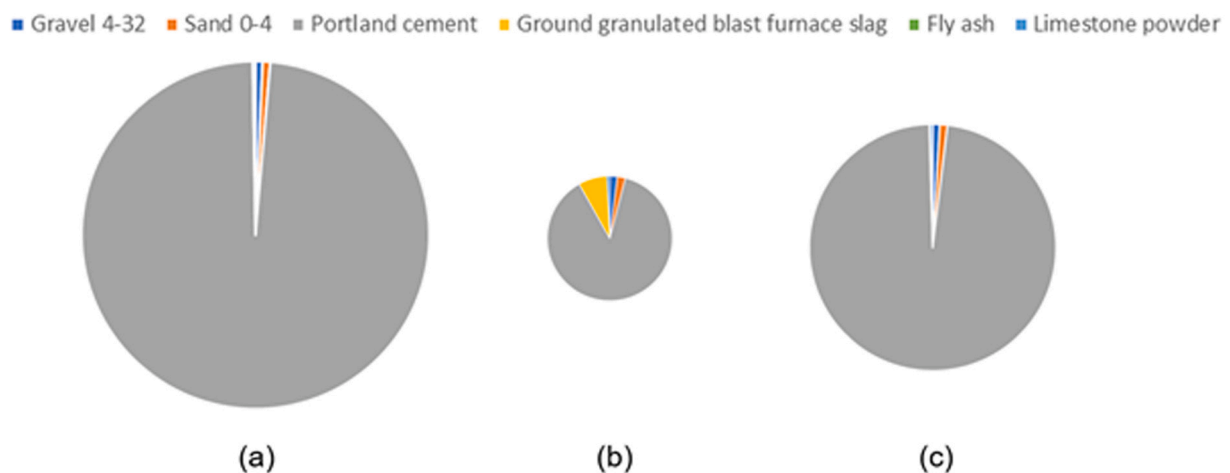
Fig. 1. Global warming potential of concrete based on raw material composition as indicated in Table 1 with (a) CEM I concrete (2.93E+02 kg CO2 eq/m^3 concrete), (b) CEM III/B concrete (9.85E+01 kg CO2 eq/m^3 concrete) and (c) CEM II/B-V concrete (2.07E+02 kg CO2 eq/m^3 concrete). For these three specific mix designs, CEM I concrete contributes respectively 1.4 and 3.0 times more to the global warming potential compared to CEM II/B-V and CEM III/B concrete.
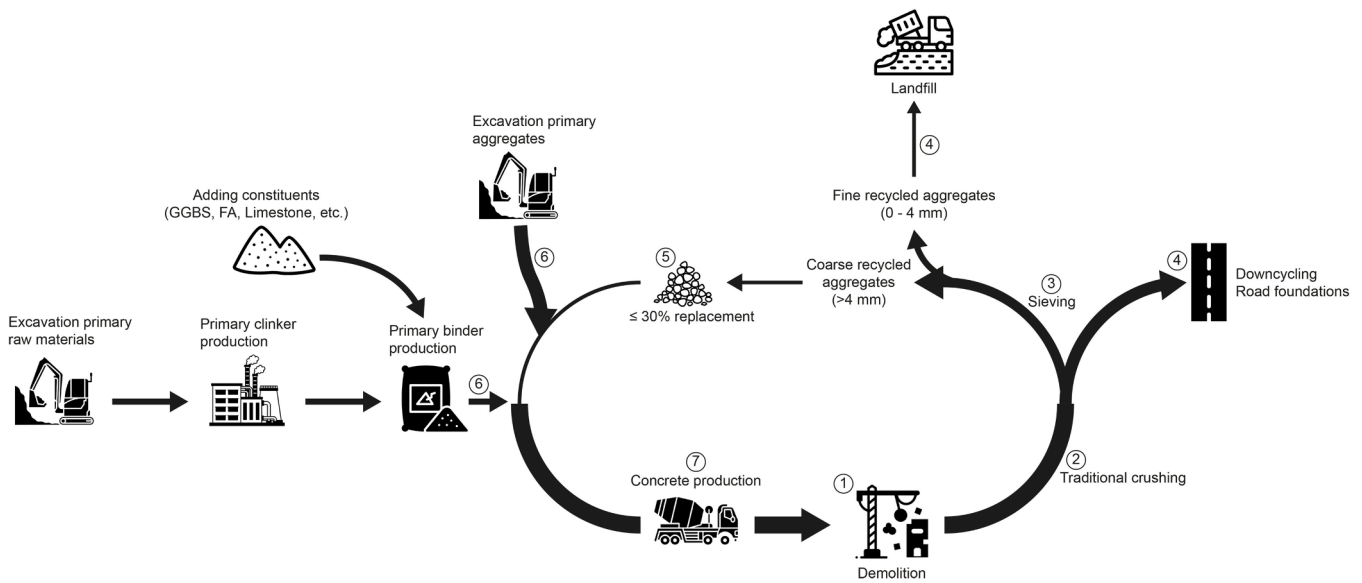
Fig. 2. Current traditional recycling process. Most of the obtained secondary coarse aggregate is downcycled, i.e. applied in lower grade applications, while the fine aggregate fraction also containing the cementitious binder is not reused at all.

stone, not enabling full replacement of primary coarse aggregate in new concrete. Consequently, this process is only partly circular, because the majority of the obtained recycled coarse aggregate is used in lower grade applications, essentially downcycling the aggregate, while the binder present in the retrieved 0–4 mm fine aggregate fraction is not reused at all. In contrast, the envisioned innovative recycling process in this study (Fig. 3) is fully circular, ensuring that materials remain within the concrete cycle and pursuing a material quality equal to that of the primary materials.

This paper therefore aims to provide insights into the downstream processing of End-of-Life (EoL) concrete to achieve high-value recovery of the cementitious fractions. In this context, downstream processing refers to the recovery and purification of a component from a heterogenous material such as concrete, with the objective of achieving high-quality reuse. Various process steps within the envisioned innovative recycling process are discussed, outlining characteristics and techniques aimed at enhancing the quality of the retrieved cementitious fractions. These steps will significantly improve the material quality in comparison to the current traditional recycling techniques.
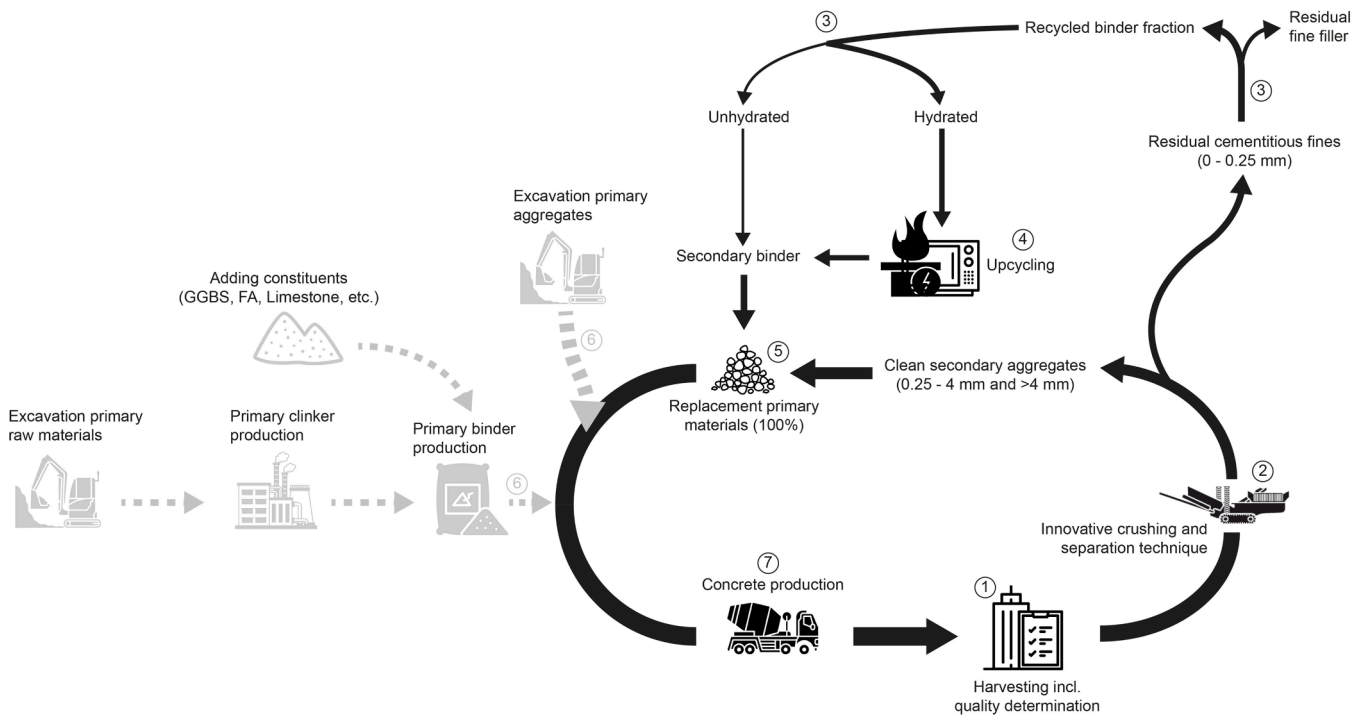
Fig. 3. Envisioned innovative recycling process. This process is fully circular and required material properties are ensured through harvesting (including quality determination) of the EoL concrete, using innovative crushing and separation optimisation of the residual cementitious fines and upcycling to obtain functional secondary binder.

2. Current state-of-the-art: the traditional approach

Traditionally, concrete recycling involves crushing concrete debris into granular products of specified particle sizes [29,30]. Typically, contaminants like wood, metals and plastics are present in the concrete debris. First generation recycling plants lacked the capability to remove these contaminants, prompting the development of second-generation processing plants. These plants employ dry or wet separation methods to eliminate foreign matter before crushing. The traditional concrete recycling process uses two types of crushers: a primary and secondary crusher. Initially, concrete debris measuring 0.5 – 1 meter arrives at the processing plant and undergoes crushing by a primary crusher, reducing it to a size of 40 – 50 mm [29,31]. Subsequently, a secondary crusher further reduces the particle size to <40 mm. Common examples of traditional concrete crushers include jaw crushers, impact crushers and cone crushers (Fig. 4) [29].

2.1. Traditional crushers

Traditional crushers operate by applying stresses to the concrete, resulting in strains that lead to breakage when they exceed the elastic limit of the material [33]. During this crushing process, no distinction is made between the different concrete components. The primary aim is to achieve the desired particle size, regardless of where the material breaks [34].

Jaw crushers are commonly used as primary crushers due to their ability to handle large pieces of debris [31,35]. They consist of two rigid plates known as jaws, with one fixed as a stationary crushing surface and the other moving to exert sufficient force to break the material [32,35]. Both plates are angled, gradually reducing the distance between them to crush the material until it reaches a particle size smaller than the gap at the bottom of the plates [32].

Impact crushers are suitable as both primary and secondary crushers [29]. They use hard steel blades attached to a rotating rotor to propel the material against impact plates. The high speed of the rotor generates high impact forces on the concrete debris [32]. However, this crusher type often has relatively high maintenance costs and typically yields lower quality coarse aggregates [29].

Cone crushers, suitable as secondary crushers, typically accommodate feed sizes up to 200 mm and use two cones to crush the material [29]. The outer stationary cone encases the inner cone, which is positioned around the eccentric axis. Crushing occurs as the space between the inner and outer cone changes continuously. Crushed material descends into the cone crusher until the desired particle size is attained [35].

2.2. Products and application

The current traditional recycling techniques produce two fractions, a coarse (>4 mm) and fine recycled aggregate (0–4 mm) (Fig. 5). The majority of these recycled concrete fractions are currently used for low-grade applications such as backfilling or road foundation [36,37]. The coarse recycled aggregate is often only partly used to replace primary gravel in concrete mix designs, while the fine recycled aggregate is not used at all to replace primary sand. The reason for this is that both coarse and fine recycled aggregates consist of gravel or sand with a certain amount of porous cement mortar attached to them. As the mortar content increases, the density of the aggregates decreases, while the porosity increases, and the water absorption rises. Consequently, recycled aggregates demand more water in concrete mixtures, leading to the use of more cement to ensure that the resulting concrete achieves the target strength [38]. However, adding cement is not preferred due to its significant contribution to the environmental impact of concrete. Additionally, the strength of the original concrete influences the quality of the aggregates, where a lower original strength results in a lower quality of the aggregates. Meaning that, to produce concrete with these aggregates a End-of-Life concrete source with higher strength is preferred [39]. Therefore, in practice, only a limited amount of secondary aggregates are used to replace primary raw materials, with most of the secondary aggregates being downcycled for other applications. When even the low-grade applications are not feasible, there is no alternative but to landfill the material [40].

The maximum replacement value for recycled aggregates (≥2100

Fig. 5. Two fractions produced by traditional recycling: coarse recycled aggregate (>4 mm; left) and fine recycled aggregate (0–4 mm; right).
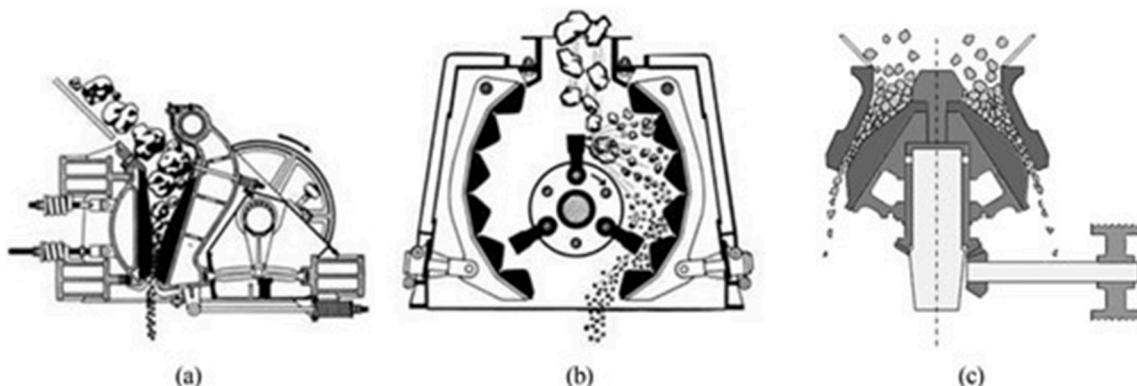
Fig. 4. Illustration of a (a) jaw crusher, (b) impact crusher and (c) cone crusher [32].

kg/m^3), as outlined in the European main standard for determining concrete composition EN 206, is shown in Table 3 [22]. Notably, the standard only addresses the replacement of coarse aggregates. Furthermore, the maximum substitution percentages vary according to the exposure classes. Currently, concretes from different origins are not recycled separately, resulting in obtained coarse aggregates being applicable only for replacing primary aggregates in specific exposure classes, namely X0, XC1-XC4, XF1, XA1 and XD1. For all other exposure classes no replacement is allowed, unless the recycled concrete is used for the exposure classes for which the original concrete was designed. When this is the case, a maximum replacement value of 30 % is allowed, indicating the need for separate recycling of concrete. However, replacement values may differ depending on additional standards adopted in different countries. For instance, in the Netherlands, the CROW-CUR recommendation 127 expands the replacement limit by volume to 60 % for fine and 100 % for coarse recycled aggregates, based on the total water absorption of the aggregates in the concrete. This recommendation is suitable for concrete ranging from compressive strength class C12/15 to C40/50 and all exposure classes mentioned in EN 206 + NEN 8005 [41]. Additionally, the threshold value for the allowed amount of absorbed water is correlated with the susceptibility of the concrete to shrinkage or creep. Both fine and coarse recycled aggregate must also adhere to the requirements of EN 12,620 + NEN 5905, including a density exceeding 2200 kg/m^3 and a composition conforming to Rc$_{90}$, Rcu$_{95}$, Rb$_{10-}$, Ra$_{1-}$, XRg$_{1-}$ and FL$_{2-}$. Regarding composition, concrete, concrete products, mortar and concrete masonry units should constitute at least 90 % by mass of the recycled aggregates (Rc$_{90}$). This proportion increases to at least 95 % when Including unbound aggregates, natural stone and hydraulically bound aggregates (Rcu$_{95}$). Clay and calcium silicate masonry units (such as bricks and tiles) and aerated non-floating concrete should be <10 % by mass (Rb$_{10-}$). Bituminous materials (Ra$_{1-}$) and glass and other materials (XRg$_{1-}$) both are only allowed to comprise 1 % by mass. Finally, the amount of floating material should be smaller than 2 cm^3/kg (FL$_{2-}$) [21, 42].

In 2023, EN 197–6 [43] was published, introducing new cement types as mentioned in Table 4 that allow the utilization of recycled concrete fines as a main constituent. The material in question has specific requirements regarding its total organic carbon ($\leq$ 0.8 % by mass), sulphate ($\leq$ 2.0 % by mass) content and clay (1.20 g/ 100 g) content. Table 4 shows the allowable quantity of recycled concrete fines allowed for various cement types.

3. Beyond state-of-the-art

To achieve high-quality recycling of both the obtained aggregates and the cementitious fine fraction, innovative recycling techniques are imperative. As previously noted, the traditionally obtained fractions are severely limited in their ability to substitute primary materials in concrete. Moreover, the demand for concrete in low-grade applications is declining in Europe [30,36,37], with some countries facing saturation of low-quality concrete aggregates in the market [36,37]. Additionally, concrete waste production is anticipated to grow due to increased demolition volumes, the replacement of existing structures and the

Table 3

Maximum replacement value coarse aggregates ($\geq$2100 kg/m^3, type A) by mass (*A maximum replacement value of 30 % is allowed for the other exposure classes when they are the same as the ones for which the original concrete was designed) adapted from EN 206 Table E.2.

Material	Exposure classes		
	X0	XC1 – XC4, XF1, XA1, XD1	All other exposure classes*
Recycled aggregate	50 %	30 %	0 %

Table 4

Amount of recycled concrete fines in percentages by mass and their corresponding cement types adapted from EN 197–6 Table 1 [43].

Main types	CEM II					CEM VI
Type	Portland-recycled-fines cement		Portland-composite cement			Composite cement
Notation	CEM II/A-F	CEM II/B-F	CEM II/A-M	CEM II/B-M	CEM II/C-M	CEM VI
Recycled concrete fines	6 – 20	21 – 35	6 – 14	6 – 29	6 – 20	6 – 20

development of new structures [15,16,19]. Consequently, there is a significant surge in concrete waste that ideally should be repurposed for high-grade applications [1]. This objective can be realized by transitioning the industry from traditional to innovative recycling techniques.

3.1. Innovative recycling techniques

Currently, innovative recycling techniques are being developed that produce materials of higher quality than the traditional process. These techniques prioritize the separation of concrete in its original constituents: gravel, sand and binder (residual cementitious fines), with the intention of utilizing them to replace primary materials in new concrete mixtures. Several techniques, such as the Smart Liberator [34], C2CA technology [44], CM-crusher [45] and Mangeler [46], have been specifically developed to enhance the concrete recycling process and the produced materials. Others, such as the Rhodax [47], Loesche mill [48] and Vertical Shaft Impactor (VSI) US7 [49], were originally designed for different purposes, but subsequently adapted for concrete recycling and the liberation of the initial components. Each of these techniques varies in methodology, approach, input and output fractions. Table 5 provides examples of these innovative recycling techniques and their respective characteristics.

3.2. Products and approach

The innovative recycling techniques can generate at least three fractions: secondary gravel (4+ mm), secondary sand (0.25 – 4 mm) and residual cementitious fines (0 – 0.25 mm) (Fig. 6) [45,67,68,69,70]. The gravel and sand fractions have the potential to completely substitute primary raw materials in concrete, as they are considered clean (free of attached mortar) and possess properties equivalent to those of natural aggregates. Consequently, factors such as the original concrete strength, as well as changes in density and water absorption no longer play a significant role. However, it is the residual cementitious fraction containing the old binder that is particularly interesting. This fraction holds significant potential for substantial reductions in CO$_2$ emissions, because it can lead to a decrease in primary cement usage and, potentially, achieve full binder replacement after upcycling.

To achieve this, the process must begin even before a structure is demolished by assessing the quality of the original components in EoL concrete. Concrete can comprise various materials that can influence the ultimately produced recycled binder fraction. Additionally, environmental factors affect the concrete composition [71,72]. These and other parameters that can affect the quality of the concrete will be discussed later in this article. Subsequently, by harvesting the different concrete qualities separately and processing them using innovative crushing and separation techniques, clean secondary aggregates and residual cementitious fines are produced. The clean secondary aggregates can directly replace the primary aggregates in new concrete mixtures. However, the residual cementitious fines require an additional separation step, because they consist of a combination of sand and

Table 5
Examples of innovative crushing technologies and their targeted output fractions.

Technology	Reason	Method	Input fraction	Fractions	Ref.
Smart Liberator	Improved concrete recycling to obtain the initial components	Selective crushing of cement stone. Adjustable crushing pressure, advanced screens and refiner for separation of fractions.	50 mm	Relatively clean gravel (> 4 mm) Relatively clean sand (0.25 – 4 mm) Filler (0.65– 0.25 mm) Retrieved binder (0 – 0.065 mm)	[34,50, 51,52]
C2CA technology	Improved concrete recycling to obtain the initial components	Advanced Dry recovery uses kinetic energy to obtain the coarse and fine fractions. Heating Air Classification heats the fine fraction to enhance the separation of the cementitious and sand fractions. Laser Induced Breakdown Spectrometry for quality assurance.	0 – 16 mm	Coarse fraction (4 – 16 mm) Fine fraction (0.25 – 4 mm) Ultra-fine fraction (0 – 0.25 mm)	[35,44, 53,54, 55,56]
CM-crusher	Increasing the quality and circularity of secondary aggregates	Pre-processing with a traditional crusher. Rubbing aggregates under pressure to remove attached cement.	> 16 mm	Gravel (8 – 16 mm) Gravel (4 – 8 mm) Sand (2 – 4 mm) Sand (0 – 4 mm) Sand (0 – 2 mm) Cemenstone powder (filler)	[45,57, 58,59]
Mangeler	Extract clean cement from concrete	Grinding concrete using a cylindrical tube with steel discs. A cem-shifter separates the cement from the sand fraction.	< 32 mm	Gravel (8 – 16 mm) Gravel (4 – 8 mm) Sand (2 – 4 mm) Sand (0.25 – 2 mm) Filler (0 – 0.25 mm) Partly reactive filler (0 – 0.125 mm)	[46,60]
Rhodax crusher	Improve limitations conventional crushers	Inertial cone crusher, uses unbalanced masses and interparticle crushing. Adjustable gap size wall-cone, rotation speed and static moment.	< 30 mm (smallest version) < 150 mm (largest version)	Gravel (> 4 mm) Sand (< 4 mm) Cementstone powder	[47,61, 62]
Loesche mill	Initially for fine comminution	Selective crushing of cement paste, adjustable settings for grinding. Airflow and overflow operations for separation of fractions.	< 20 mm (currently used) < 100 mm (largest version)	Gravel (8 – 20 mm) Gravel (2 – 8 mm) Sand (0 – 2 mm) Hardened cement paste (filler)	[48,63, 64]
Vertical Shaft Impactor	Consistent sand production, V7 process for cement removal from aggregates	Removing cement paste from aggregates by using impact members and an air screen system. Multiple (5) passes advised.	< 10 mm (V7) < 40 mm (RC7)	Coarse aggregate (5 – 20 mm) Fine aggregate (2.5 –.5 mm) Fine aggregate (1.2 – 2.5 mm) Sand (0.15 – 1 mm) Filler (0 – 0.15 mm)	[49,65, 66]

cementitious material. After separation, a recycled binder and residual fine filler are produced. The recycled binder fraction is divided into unhydrated and hydrated fractions. The unhydrated fraction possesses cementing properties and can therefore directly replace part of the primary cement as a secondary binder [73]. The hydrated fraction is envisioned to be used as a secondary binder after undergoing upcycling treatment, following further development of current and new upcycling techniques [74,75]. All fractions are then used to produce concrete, and the material cycle begins anew. Fig. 3 provides a schematic overview of the innovative recycling process.

3.2.1. Quality determination

Concrete from various sources can exhibit significant variations in material properties [29,76]. Throughout its lifespan, concrete may encounter diverse environmental conditions and associated degradation mechanisms, such as carbonation, chloride ingress, alkali-silica reaction and chemical attack, all of which impact its composition. Currently, there is no distinction made between concretes from different environments and origins during demolition, despite the fact that they may possess varying levels of quality or unwanted contaminants. In the process sequence of demolition, recycling and storage everything is typically mixed, despite potential deviations in properties that could

influence new concrete mixtures [71,72]. Additionally, conducting quality determination assessments before demolition could benefit the environment, because it has the potential to maximize the value from a structure while simultaneously reducing the amount of waste produced [77].

The predominant environmental impact of concrete originates from the clinker present in the cement. Therefore, there is a need to prioritize the recycling of the old binder in a manner that allows for clinker replacement, thereby reducing greenhouse gas emissions. This approach not only reduces the demand for raw materials in clinker production, but also decreases reliance on supplementary cementitious materials (SCMs). Ground granulated blast furnace slag (GGBS) and fly ash (FA) serve as examples of SCMs used in blended cement types. However, the availability of both materials is decreasing due to changes in the steel and coal-fired production industries [78]. Depending on the binder type, the potential applications for the obtained old binder fractions may vary. Differences in concrete composition and the formation of reaction products after treatment may lead to different implementation routes for the recycled fractions. Additionally, factors such as design requirements, age and the environment in which the structure is situated may results in differences in the quality of the recycled concrete fines. Therefore, it is important to identify the constituents and contamination present in EoL

Fig. 6. Innovative recycling produces at least three fractions: secondary gravel (left), secondary sand (middle) and residual cementitious fines (right).

concrete.

Quality determination is associated with all the parameters that influence the recycling and reuse potential of the recycled binder fraction. The following section focuses on the characteristics relevant for quality determination of the residual cementitious fines as part of a harvesting approach before demolishing a concrete structure. Concrete containing various binder qualities can then be recycled and implemented separately based on this distinction. Table 6 defines the quality before

Table 6

Envisioned quality definition. The binder type is the most important, followed by the presence of unreacted particles and the binder content (A, B,C, D and E refer to the priority of the characteristics, with A having the highest priority; I, II and III refer to quality indications with I being seen as the highest quality. By combining the priority and the quality a distinction can be made between material that needs to be recycled separately).

Characteristics	Priority	Type	Quality	Comments
Binder type	A	CEM I	I	Main quality
		CEM III/B	III	characteristic. CEM I is
		CEM II/B-V	II	seen as the highest quality due to its clinker usage.
Unreacted particles	B	Clinker	I	The presence of
		GGBS, FA	II	unreacted particles,
		Non/unknown	III	results in a higher quality. Although this is secondary to the binder type.
Binder content	D	>340 kg/m³	I	Higher binder content
		300 – 340 kg/m³	II	may mean a higher potential to obtain reusable clinker.
		<300 kg/m³	III	
Chemical contamination (Chlorides, sulfates, alkalis)	C	Yes	II	Required to know, but not leading. When present, obtained fines can be mixed with low/non contaminated fines.
		No	I	
Mixture components (aggregate)	E	River	I	Other aggregates than river aggregates may have different influences on the obtained residual cementitious fines.
		Other	II	

harvesting. A higher quality binder contains a larger amount of material that can serve as clinker replacement, thus having the potential to reduce the environmental impact of concrete. However, depending on the upcycling and application approach, the emphasis for certain types as mentioned in Table 6 may shift.

3.2.1.1. Age and environment. Over the decades, there have been developments in the number of available cement types that permitted use in concrete [79]. Historical data on the different cement constituents can help limit the possible binder components present in structures when the construction year is known. This is illustrated for Europe and specifically for the situation in the Netherlands.

The period between 1890 and 1910 marks the introduction of reinforced concrete in Europe, with Portland cement being deemed most suitable for this purpose [80,81]. This led in the Netherlands to the introduction of the first regulations in 1912, which served as a starting point for determining of the emergence of various cement types or cementitious components in history [82].

Table 7 provides an overview of the main binder constituents used in the European union and their historical application based on the publication year of the first Dutch regulations up to and including the

Table 7

Allowed binder main constituents based on the publication year of the first Dutch regulations up to and including the currently used regulations [4,86,87, 88,89,90,91,92,93,94,95,96,97,98,99,100,101,102,103,104,105]. Legend: not mentioned; -* not mentioned, but not forbidden; + allowed; o allowed under certain conditions; x not allowed; x* not allowed in reinforced concrete.

	1912 - 1918	1918 - 1930	1930 - 1962	1962 - 1986	1986 - 1995	1995 - 2012	2012 -current
Clinker (Portland cement)	+	+	+	+	+	+	+
Ground granulated blast furnace slag	–	x*	o	+	+	+	+
Fly ash	–	–	–	–	O	+	+
Limestone	–	–	–	–	–	o	o
Silica fume	–	–	–	–	–	–	o
Burnt shale	–	–	–	–	–	o	+
Trass	–	-*	–	o	O	–	–

currently used regulations in the Netherlands. The types mentioned can vary significantly between countries due to factors such as the available raw materials and technical requirements [79]. In the Netherlands blast furnace slag cement is the most commonly used cement type, followed by Portland and Portland – Fly ash cement. In other countries such as Austria, Finland, Portugal and Sweden, Portland – Limestone cement holds the largest market share [83]. Generally, this implies that clinker is the most used constituent in Europe, although limestone, slag and fly ash are commonly used as additions depending on the country. Silica fume and burnt shale, on the other hand, are only used to a limited extend [83,84,85]. By compiling country-specific historical overviews, insights can be gained into the types of cement applied during specific time periods in each country.

Throughout different historical periods, varying materials were used in concrete mixtures. Therefore, insight into the age or the year of construction of a structure can serve as an initial indication of the potential binder constituents present. For instance, structures erected between 1912 and 1930 predominantly consisted of Portland cement, as this was the sole permitted binder during that period. However, it is worth noting that adherence to regulations was not always strict, because instances of fraud were not unknown [81]. Nevertheless, knowing the age of a structure can provide valuable insights into the main binder constituents.

Another property of interest, influenced by the age of the material, is the fineness of the applied binder. As depicted in Fig. 7, the fineness of Portland cement has progressively increased over the years and continues to do so [106]. Coarser Portland cement tends to retain a higher proportion of unreacted clinker particles [107], implying that older structures are likely to contain a higher amount of unreacted cement.

The environment and in addition the exposure class of the structure is also of interest, because degradation mechanisms may lower the quality of the retrieved fines. In chloride-rich environments the chlorides react with the cement paste. With regards to durability, binding of the chloride to phases in the cement paste is favourable, because this slows down the chloride ingress [108]. Considering the recycling of cement fines this might be different depending on the amount of chlorides bound to the paste. Another example is the presence of sulfates in the environment. Together with chlorides, sulfates are one of the most aggressive chemicals influencing the durability of concrete. Reactions with the hydrated cement paste results in a reduction of strength, expansion, spalling and cracking of the concrete [109]. Due to its reaction the sulfates remain in the residual cementitious fines and need to be taken into account when producing new concrete containing recycled materials. Determining the type and amount of chemicals present in the environment, which can react with the binder in concrete, are therefore of interest to estimate the quality of the binder and its application potential in new concrete mixtures.

3.2.1.2. Concrete mixture components. Concrete generally consists of a mixture of aggregates (coarse and fine), binder and water. In addition, chemical admixtures and mineral additives may be present [110,111]. All these components may to a certain extent be present in the residual cementitious fines (RCF) depending on the recycling process. For example, when concrete is crushed, powders are produced consisting of not only binder components, but also crushed aggregates powders and mineral additives [74]. It is assumed that chemical admixtures such as plasticizers have minimal influence on the quality of RCF, because of the small amounts added to the concrete mixtures.

Approximately 75 % of the volume of concrete consists of aggregates [110,111,112]. The majority of aggregates consists of river sand and gravel, but crushed stones are also among the commonly used aggregate types [111]. Crushed rock aggregates can be divided into three main groups, namely sedimentary, igneous and metamorphic rocks. Examples of sedimentary rocks are limestone and sandstone. They are formed due to the consolidation of loose sediment. Igneous rocks form when molten or partly molten materials cool down and solidify. They can be formed within the Earth (slow cooling) or ejected onto the Earth (quick cooling). Examples are granite and basalt. Metamorphic rocks are rocks that underwent chemical, structural and mineralogical changes often as a result of heat and pressure treatment. An example is quartzite [112,113]. Table 8 shows the varying chemical composition of different aggregate types. During the recycling process fine powders related to the aggregates may end up in the RCF and influence not only the recycling process itself, but also the quality of the fines.

Mineral additives, such as GGBS, FA, silica fume and limestone powder [115,116], also vary in chemical composition. In general they are added to enhance the properties of the concrete [116]. Besides knowing the type of aggregates present in the concrete, the mineral
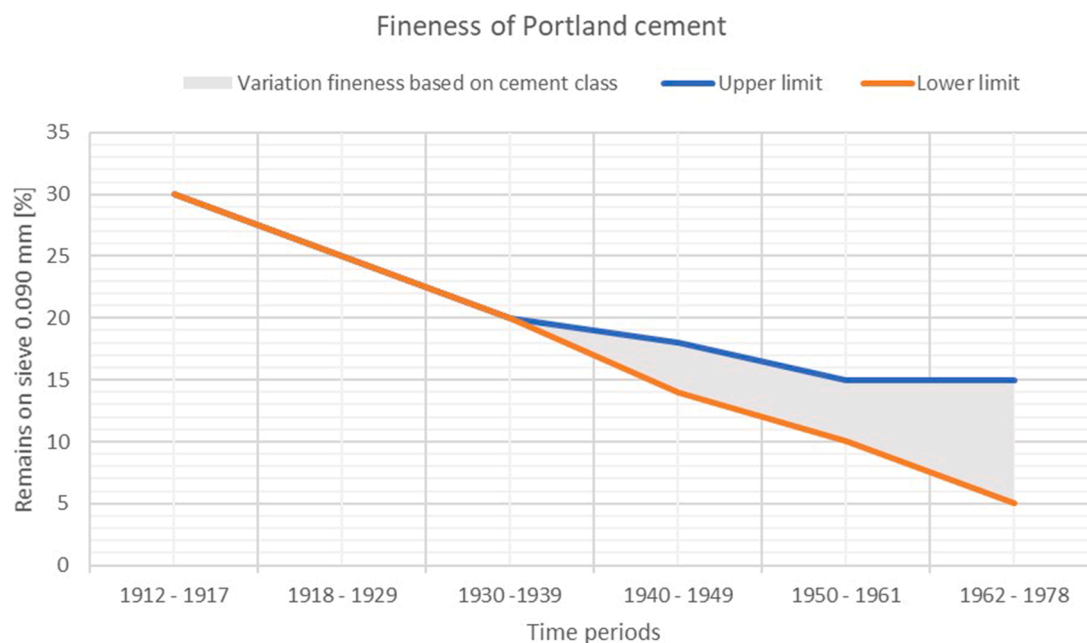
Fig. 7. Increasing fineness of Portland cement during the years. The upper limit is the class containing the coarser particles and the lower limit the class with the finer particles [86,87,88,89,90,91].

Table 8

Typical chemical composition of different aggregates types [71,112,113,114].

Chemical composition	River sand	Crushed rock aggregates				
		Limestone	Sandstone	Basalt	Granite	Quartzite
SiO_2	93 – 96	5 – 8	70 – 90	49 – 63	54 – 72	59 – 65
CaO	0.5 – 1.1	49 – 80	–	2 – 5	0 – 2	2 – 3
Al_2O_3	1.7 – 2.5	2 – 3	1 – 2	15 – 16	14 – 26	20 – 21
TiO_2	0.1 – 0.2	–	–	0 – 2	0 – 1	-
FeO	–	–	0 – 1	7 – 10	0 - 1	–
Fe_2O_3	0.5 – 0.9	0 – 1	1 – 2	4 – 7	1 – 2	9 – 10
MgO	0.1 – 0.2	1 – 2	0 – 1	2 – 6	0 – 3	2 – 3
K_2O	0.7 – 0.9	0 – 0.5	0 – 1	0 – 2	2 – 6	–
Na_2O	0.2 – 0.5	–	1 – 2	2 – 3	0 – 4	–
MnO	–	–	–	–	–	–
P_2O_5	–	–	–	–	0 – 0.1	–

additives are also of interest. The same reasoning as for the different aggregates can be used, but in this case the additives are of a smaller particle size and are expected to end up in the RCF. The implementation route may change depending on the type of mineral additives. This is also the case for the binder type, which is discussed in the next paragraph.

3.2.1.3. Binder type and content. The NEN-EN 197–1 mentions 27 types in the family of common cements, which differ in the amount of main constituents [4]. The most commonly used cement types in the Netherlands are Portland cement, Portland fly ash cement and blast furnace cement and each of them have their own specific properties and area of application [117]. Current concrete recycling processes do not take these deviations into account and all the concrete ends up on the same demolition pile [72]. For high-value retrieval, however, a distinction between the binder types should be made.

Mehdizadeh et al. [78] studied the influence of CO_2 treatment on the reactivity of residual cementitious fines containing GGBS and FA and found that the powders containing GGBS resulted in a higher carbonatation degree than those with FA. One of the reaction products formed during the carbonatation is a type of gel, which was found to contribute to the compressive strength development. Zajac et al. [118] also looked at the carbonation treatment of cement pastes, but compared Portland cement and blast furnace slag cement. The main reaction products after the treatment were calcium carbonate and alumina-silica gel. They found that a larger amount of GGBS has the potential to form more of the gel, because of the larger Si and Al contents. On the other hand Portland cement formed higher amounts of calcium carbonate. Besides the initial composition of the cement types, the degree of hydration is also of interest as a higher degree results in an increase in the amount of main reaction products.

The CO_2 treatment is an example of an implementation route in which the binder type in the EoL concrete influences the resulting product. For the formation of alumina-silica gel a blast furnace slag cement seems preferable although the degree of hydration may limit the amount. A lower carbonatation was observed for FA cements. When the goal is to store a large amount of CO_2 this binder type seems less preferable, whereas a Portland cement binds CO_2 the most due to its high degree of hydration [78,118]. For other implementation routes a comparable trend might be observed, where a certain binder type is preferable. More research is needed to determine the potential of certain applications for the different binder types. The importance of the presence of unreacted particles is discussed in paragraph 2.1.5, but this is also interesting in relation with the binder type. Depending on the binder type the amount of unreacted particles may differ as well as the type of unreacted particle itself.

Besides the binder type an indication of the content may also influence assigned quality before demolition. The EN 206 [22] mentioned the minimum cement content needed for the exposure classes of the concrete (Table 9). For both freeze/thaw attack and sea water the minimum

Table 9

Minimum cement content related to exposure class NEN-EN 206 [22].

	X0	XC 1	XC 2, XC 3	XC 4, XS 1, XD 1, XD 2, XF 1, XF 2, XA 1	XS 2, XD 3, XF 3, XA 2	XS 3, XF 4	XA 3
Minimum cement content [kg/m³]	–	260	280	300	320	340	360

cement content lies between 300 – 340 kg/m³. In an environment with chlorides other than from sea water the range is slightly smaller (300 – 320 kg/m³) and for carbonation induced corrosion the values are lower (260 – 300 kg/m³). The highest minimum cement content can be found for concrete in aggressive chemical environments (300 – 360 kg/m³). Other factors of influence are for example the type of structure and placement conditions. Bored and cast-in-place piles need a minimum amount of 325 kg/m³ in dry conditions, but in submerged conditions the amount has to increase ($\geq$ 375 kg/m³). For diaphragm walls the cement content also changes depending on the D_{max} value. The content for these type of structures varies between 350 – 400 kg/m³ [22]. The specific content or assumed range is important for the recycling process. Based on the initial cement content an assumption can be made of the amount of material in the different fractions. Because of the high environmental footprint of Portland cement and especially the clinker, a higher cement content means more cement present in the RCF and therefore a larger reduction of the environmental footprint.

3.2.1.4. Chemical requirements and contamination. The chemical composition of the RCF is directly related to its quality. Not only does concrete consist of different components, it is also exposed to different conditions during its lifetime. Additionally, requirements in the last century were different than the chemical requirements of today [119]. Table 10 shows an overview of chemical requirements from the first Dutch regulation until the currently used regulations. For requirements such as the insoluble residue, sulfate content and MgO content the limits were generally lower than or equal to the current day limits. Before the G.B.V 1918 only the sulfate content was mentioned and a limit to the chloride content was first referred to around the publication of the regulations in 1995. The contents of certain compounds before the establishment of the requirements may therefore exceed the currently stated limits.

The concrete composition also influences the chemical composition of the fines as stated before. For example, GGBS has a MgO content of 1 – 21 % [120], which may result in a MgO content exceeding the limit of 5 % for Portland cement. A further separation of binder types consisting of other components than Portland cement is needed to increase the recycling potential and meet the requirements for certain implementation routes.

Table 10
Chemical requirements Portland cement (* The maximum allowed sulfate content depends on the strength class) [4,86,87,88,89,90,91,92,93,94,95,96,97,98,99,100].

Property [%]	1912	1918	1930	1940	1950	1962	1979	1990	1995	2012
Insoluble residue	–	3.0	3.0	3.0	3.0	3.0	3.0	5.0	5.0	5.0
Sulfate content (as SO_3)	2.5	2.5	2.5	2.5	3.0	3.0	3.75*	3.75*	3.5*	3.5*
							4.0*	4.0*	4.0*	4.0*
MgO content	–	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0
Chloride content	–	–	–	–	–	–	–	–	0.1	0.1

Contamination of the cementitious fractions occurs as the concrete is exposed to different conditions and environments [77]. Nedeljković et al. [121] mentioned that contaminants such as sulfates (SO_4^{2-}), chlorides (Cl^-) and soluble alkalis accumulate in the finest fraction of the fine recycled concrete aggregates. An increased sulfate content can lead to delayed ettringite formation (DEF). This leads to expansion and cracking of the concrete [77,122,123]. High concentrations of chlorides should be prevented to avoid corrosion of the reinforcement [77]. Increased amounts of alkalis can results in expansive alkali-silica reactions. Siliceous aggregate reacts with the alkaline pore solution in concrete. Swelling of the formed gel increases stresses in the concrete and eventually leads to cracking [124]. It is of importance to determine the chemical contamination of the RCF to avoid the occurrence of deterioration reactions in new concrete.

3.2.1.5. Unreacted particles. The presence of unreacted binder particles in concrete is of great interest for the quality determination of the fines. Lu et al. [125] mentioned that concrete always contains a certain amount of unreacted cement particles. The amount is, among other things, related to the water to cement (w/c) ratio and the strength grade. A lower w/c ratio or a higher strength grade concrete results in the presence of more unreacted cement particles. An example is given where for a C50 concrete >25 % of the cement particles remained unreacted. According to Van Breugel [107] a w/c ratio of 0.4 does not result in complete hydration of the cement particles in practice, although theoretically this value is found to result in complete hydration. He mentioned that 30 % of the cement remains unreacted and this amount can be even higher in the case of coarser cement particles [107,126].

In the case of composite cement types containing SCMs such as ground granulated blast furnace slag (GGBS) or fly ash (FA), part of the GGBS or FA particles can also remain unreacted [71,126]. The hydration rate of SCMs is affected by the availability of portlandite and water. In case of insufficient amounts of portlandite part of the SCMs remain unreacted [127].

Unreacted cement particles have the ability to hydrate when they come in contact with water. The assumption is that retrieved unreacted particles are qualitatively equal to primary cement and can therefore replace it without further need for upcycling. This also applies to unreacted GGBS and FA in retrieved concrete, which are assumed to be equal to the initially used GGBS and FA. Therefore, the presence of unreacted particles in concrete elements is seen as valuable in the recycling process.

3.2.2. Potentially suitable techniques for the quality determination of the binder
Documentation related to concrete reaching the end of its lifetime can offer initial insights into quality determination characteristics. However, it is important to acknowledge that the documentation may not always align with the actual situation. Heinemann [119] noted the possibility of fraud, emphasizing the importance of research to determine the true characteristics of the material. Following a documentation review, the initial step in on-site quality determination involves visually inspecting the structure. This provides preliminary insights into potential degradation mechanisms that could affect the residual cementitious fines [128]. Additionally, visual inspection helps identify the presence and location of coatings or other surface layers on the concrete. Both visual inspection and documentation review serve as preliminary techniques, preceding a more detailed material analysis.

Because concrete recycling is done primarily at dedicated recycling plants, it is important that the used techniques are applicable in practice. A handheld x-ray fluorescence (HXRF) is therefore an interesting technique for the quality determination of EoL concrete. It is portable, non-destructive and provides rapid in situ or location based measurements [129]. It measures the elemental composition of a material [130,131]. The HXRF has already been used in research [130,132] to determine the chemical composition and mortar content of recycled aggregates as well as the concrete mixture proportions. In addition, research on the application of the HXRF for the characterization of EoL concrete in a laboratory setting showed promising results [133]. Question remains if this is still the case in practice and how to deal with surface contamination and the presence of coatings. A technique that gives similar results to the HXRF and is also portable is the laser induced breakdown spectroscopy (LIBS) [134,135,136]. However, it is micro-destructive, because a pulsed laser removes a small amount (1 µg) of the material surface [137]. This means that the same spot cannot be measured again. In addition, for certain situations (historic products, small sample size) even a small removal may not be desirable [138].

A technique that is often mentioned for the characterization of historic mortars [139] and is also well established as a technique for concrete research [140], is polarized and fluorescence microscopy (PFM). It is a destructive analysis technique, because it requires concrete samples taken from a structure to study under the microscope. However, it can give accurate insights in binder (unhydrated and hydrated) and aggregate types, degradation and contamination [141,142,143]. Although non-destructive techniques are preferred for characterizing structures for demolition, this technique may be necessary for a precise identification.

Another technique of interest is the X-Ray Diffraction (XRD). It is available in a portable setup, but requires powdered samples for the measurements [144]. The use of XRD to analyse cement phases in concrete is complicated due to possible overlapping peaks of the aggregates and binder. This can especially be the case for the poor crystalline phases of the calcium silicate hydrates [140]. For the crystalline phases however, this technique does allow for the differentiation between binder types [145,146]. XRD gives insight in both the unreacted binder components and some of the hydration products. Hydration products, such as portlandite and ettringite, are mostly crystalline just like the anhydrous cement phases and can therefore be recognized in the diffraction pattern [147].

Fourier Transform Infrared Spectroscopy (FTIR) is usually used to determine chemical groups in organic materials, but it is also used more frequently for the identification of construction materials [140]. It is able to distinguish both hydrated and unhydrated faces in individual samples. In hydrated binder types the identification is more difficult due to overlapping bands [148,149,150]. Although, using the first derivative of the spectra may give additional information for identification [151]. In addition, Tang [152] found that analysis of concrete was only possible for powders. Depending on the characteristics required, this technique may give complementary information in combination with the previously mentioned techniques.

Wet chemical analysis, such as selective dissolution or titration, is complementary to the previous techniques. It is traditionally used to

determine the content of the concrete mixture components [139] and uses acids to dissolve certain components in the material [153]. Depending on the type of acid, different characteristics, such as reaction degree, binder and aggregate type, can be determined. The reliability of this method is questionable, due to incomplete dissolution resulting in over- or underestimation of components [143,154,155,156]. However, from an environmental point of view, testing with acid is not preferred. In practice, it is therefore recommended to only apply this method if previously mentioned methods do not give the required results.

Thermogravimetric analysis (TGA) and Differential Scanning Calorimetry (DSC) serve as alternatives when previous techniques prove inconclusive. Both techniques may be of interest to gain insight in the binder type, based on the phase changes in the sample, or the degree of hydration [145,146,157,158,159]. However, they require a database of known materials or composition to determine a calibration curve. This curve can then be used to estimate the amount of a certain component or degree of hydration in unknown samples [160].

Techniques such as Nuclear Magnetic Resonance spectroscopy (NMR) and Scanning Electron Microscopy (SEM) are not preferred in practice, because these are often time consuming and cost intensive [161]. It is therefore recommended to only use these techniques if previous options do not give the required information. An overview of the potentially suitable techniques is shown in Table 11.

4. Separation optimization

Residual cementitious fines (RCF) is one of the fractions obtained after processing the concrete with traditional or innovative crushers. The composition of this fraction can deviate based on the original concrete, recycling process and storage conditions, but various studies [74, 162,163,164] mention that RCF still contains a certain amount of residual fine sand. For the high value reuse of binder an effective way to separate the cementitious material from the residual fine sand is needed [74]. Separation methods for the improvement of the quality of the recycled concrete components often focus on the removal or improvement of adhered mortar from the recycled concrete aggregates (RCA). The adhered mortar lowers the density and increases the water absorption and porosity. By removing or improving the characteristics of this mortar the quality of the RCA is improved. Methods for removing the adhered mortar can be divided into mechanical, thermal, chemical or a combination of these treatments. For the improvement of the characteristics of the adhered mortar polymer treatment, surface coating or carbonation are mentioned [165,166,167]. In the context of circularity, reducing the use of raw materials and the environmental footprint of concrete, high value reuse of the clinker fraction in the concrete is of interest. This paragraph focuses on methods for the separation of the recycled binder fraction from the residual fine sand.

4.1. Acid pre-soaking

An option to improve the quality of RCA is pre-soaking the aggregates in acid [165,166]. Different types of acid can be used, but Tam et al. [168] found that HCl and H_2SO_4 showed the best results for the water absorption and density of the RCA. This also indicated that more mortar had been removed. Scrivener et al. [156] mentioned the use of selective dissolution as a measurement tool for the determination of the reactivity of SCMs. Because the clinker and hydrated binder phase are dissolved, only the unreacted SCMs remain. These unreacted SCMs might be able to replace the primary SCMs. Acid treatment or selective dissolution is not preferrable when looking at circularity and the contribution of clinker to the environmental footprint of concrete. Both hydrated and unreacted clinker phases are dissolved with this method and therefore can no longer be recycled.

Table 11

Potentially suitable techniques for the identification of the binder type in End-of-Life concrete (O: orienting, –: not suitable in practice, -: low suitability, +/-: comparable or complementary to other techniques, +: suitable, ++: high suitability, ND: non-destructive, MD: micro-destructive, D: destructive).

Technique	Suitability	Type	Portable	Comments
Documentation	O	ND	–	Indication of the used materials, but this may deviate from the in practice situation.
Visual	O	ND	–	Insight in deterioration, presence coatings and surface contamination.
Wet chemical analysis	+/-	D	No	Questionable reliability and environmentally taxing. May give complementary information.
X-Ray Fluorescence	++	ND	Yes	Measures chemical composition. Insight in contamination, cement and aggregate type.
Laser Induced Breakdown Spectroscopy	+/-	MD	Yes	Comparable to the HXRF, but less preferable due to its micro-destructivity.
X-Ray Diffraction	+	D	Yes	Allows for binder identification (unhydrated and hydrated) for the crystalling phases. Portable XRD uses powder samples.
Polarized and Fluorescence Microscopy	++	D	No	Accurate insights in binder (unhydrated and hydrated) and aggregate types, degradation and contamination.
Scanning Electron Microscopy	–	D	No	Not preferred in practice. Time consuming and cost intensive.
Thermogravimetric analysis	–	D	No	Requires a database and calibration curve of known samples for identification.
Differential Scanning Calorimetry	–	D	No	Requires a database and calibration curve of known samples for identification.
Fourier Transform Infrared Spectroscopy	+/-	D	Yes	Complementary. Sample has to be ground to a powder. First derivative of the spectra is of interest. Preferably used in combination with other techniques.
Nuclear magnetic resonance spectroscopy	–	D	Yes	Not preferred in practice. Time consuming and cost intensive.

4.2. Mechanical treatment

Mechanical treatment often refers to mechanical forces breaking the attached mortar from the aggregates. Removed mortar is often collected through sieving of the mechanically treated material [165,166]. Shaban et al. [166] mentioned an eccentric-shaft rotor or mechanical grinding can be used during this treatment. The grinding approach has the benefit that the particle shape is improved. A drawback of mechanical treatment regarding the quality of the aggregates is the occurrence of micro-cracks

due to the forces applied on the material. Furthermore, fines produced may not only be binder related powders, but may also originate from the aggregates. The fine powders comprising of binder and aggregate relate components still needs further separation.

4.3. Gravity separator

The primarily separation mechanism of a gravity separator relies on differences in material densities [169]. Factors such as particle shape, size, and weight also affect the separation process [170]. Both water and air can serve as separation mediums, with air gravity separators gaining popularity due to water conservation concerns and the elimination of the drying requirement post-application, a common practice with wet processes [171,172]. The choice of medium is also influenced by the desired separation efficiency, with water requiring less power to achieve a certain degree of separation due to its higher density compared to air [173].

Ulsen et al. [174] investigated heavy liquid separation and elutriation for separating cement paste and aggregates (> 0.15 mm), observing reduced cement paste content for fractions with higher density and increased content for fractions with lower density. This was particularly notable in smaller particle sizes due to enhanced liberation of cement paste from aggregates [174]. Petit and Irassar [175] explored dry classification for dust removal from manufactured sand, yielding three fractions based on particle size: manufactured sand (6 mm - 300 μm), dust/fine fraction (300 - 45 μm), and ultra-fine fraction (150 - 0 μm). Cepuritis et al. [176] studied air classification to treat the fines from crushed sand, because of their influence on concrete rheology. They separated fines (250 - 0 μm) from sand (2 – 0 mm), obtaining varied particle distributions for micro-proportioning.

A fluidized bed separator, employing a vibrating deck, separates particles based on their density, with lighter particles rising and heavier ones falling [177]. Hu et al. [178] achieved a concrete purity of 95 wt.% using this approach to separate concrete and brick. Effective liberation of the binder from aggregates is paramount for enhancing the efficiency of this separation method [174]. This is especially of importance when materials possess identical sizes and shapes but differing densities, leading to formations of high, low, and middle density layers. The middle layer consists of a combination of the high and low densities and needs further treatment for separation [177]. Considering concrete, the middle layer may contain incompletely liberated binder-aggregate particles or other binder-related particles in composite binder types. Particle shape and size can significantly influence separation, with irregular shapes exhibiting higher settling velocities than spherical ones, lowering the separation degree [179]. Additionally, wet processes are not preferred due to their influence on unreacted cement particles that remain within the material.

4.4. Electrical treatment

Sonic pulses and electric pulses are both utilized in treatments for concrete recycling. Sonic pulses propagate through water, generating pressure and stresses upon encountering interfaces of different densities within the concrete mixture. This stress weakens the binder matrix, causing cracks primarily at the interface of different densities without damaging the aggregates, thus facilitating separation of the binder from the aggregates [180,181,182]. Linß and Mueller [181] noted that sonic pulse treatment reduces the attached mortar content of recycled aggregates, with higher efficiency observed in binder types with lower portlandite content. Katz [183] used ultrasonic cleaning, varying treatment duration based on concrete qualities, with coarser particles experiencing higher mortar removal.

In electric pulse treatment, plasma formation within concrete air voids leads to shockwaves and increased pressure, resulting in cracking at constituent interfaces and mortar removal from aggregates [182,184, 185,186,187]. Touzé et al. [184] investigated electrical fragmentation,

finding higher liberation rates in concrete with larger aggregate sizes and minimal influence of aggregate mineralogical composition on treatment efficiency. Narahara et al. [188] observed that higher numbers of electrical pulse discharges resulted in increased mortar removal from aggregates, leading to higher density and reduced water absorption, with the optimal discharge number estimated between 40 and 60.

However, both sonic and electric pulse treatments involve water, which hydrates any unreacted cement particles present in the concrete. This hydration poses a challenge as unreacted cement can potentially replace primary cement directly. Therefore, exposure to water is not preferred to ensure the availability of these particles for hydration.

4.5. Thermal treatment

Thermal treatment focusses on the expansion of concrete to remove the attached cement stone from the aggregates. Thermal stresses result in expansion of the materials. Aggregates and attached binder have different expansion coefficients, which results in delamination. The aggregate and cement stone quality influence the treatment temperature and should therefore be known before starting this treatment [165,166]. The main techniques for thermal treatment are traditional and microwave heating. These are high temperature treatments, but low temperature treatment, such as freeze-thaw cycles, is also mentioned [166, 182].

Traditional heating involves placing the material in a furnace and heating it to a specific temperature for several hours, exploiting the difference in expansion between aggregates and attached mortar to weaken the bond strength and facilitate mortar reduction [166,182]. External heating from the furnace environment initially affects the material its surface before spreading inward [189]. Studies by Pawluczuk et al. [190] investigated various treatment temperatures (300 °C, 600 °C, and 900 °C) with a residence time of 3 h, resulting in reduced attached mortar, albeit with some remaining and damaging of the aggregates at higher temperatures. Additional research by Despotović [191] noted potential aggregate damage above 350 °C, with internal stresses occurring between 400 °C and 600 °C, and microcracks forming between 600 °C and 800 °C. Akbarnezhad [165] highlighted the dependency of the traditional heating approach on aggregate type and mortar strength, typically recommending heating for 2 h between 300 °C and 600 °C. Shima et al. [192] demonstrated that heating the concrete to 300 °C dehydrates the cement paste, rendering it brittle, after which a mill was used to remove the cement paste. Pre-treatment techniques, such as water saturation and quenching in cold water, enhance efficiency by inducing pressure during evaporation and generating stresses for further mortar removal [165,166,182].

Microwave heating uses electromagnetic radiation to heat materials with dielectric properties, affecting aggregates and attached mortar differently due to their varying properties [165,189,193]. The higher heating rate of mortar leads to thermal expansion discrepancies, resulting in stress gradients and subsequently delamination [165,194]. Studies by Wei et al. [195] observed decreased bond strength between aggregate and mortar, with cracks forming around aggregates and spreading to mortar upon heating. Increased microwave powder and aggregate type may influence separation efficiency, with basalt and granite exhibiting separation under high microwave power (4 and 5 kW), while limestone and coal sandstone show weakened bond strength without direct separation [195]. Microwave treatment decreases bond strength, as noted by Bru et al. [196], especially when used as pre-treatment before impact crushing. Additionally, mechanical rubbing may further enhance mortar removal during this process.

Freeze-thaw cycles involve treating soaked aggregates to remove attached mortar, where liquid in pores freezes, increasing internal pressure and eventually causing cracks between aggregates and mortar [180,182,197]. This method, though time-consuming, reduces attached mortar, particularly with post-treatment approaches, albeit with the risk

of damaging aggregates [180,182]. The efficiency of this treatment also depends on the original concrete composition and strength [197].

Thermal treatment has been researched to enhance aggregate quality, but a finer fraction often remains. In freeze-thaw cycles, the use of water or chemicals impacts binder particle quality, as seen in other wet treatment methods. Both traditional and microwave heating alter binder composition. Dehydration occurs depending on the attained temperature, potentially affecting the recycling process.

4.6. Magnetic separation

Magnetic separation, initially for tramp iron removal and ferrous ore beneficiation, has expanded in applications over time to include iron recovery, mineral beneficiation, waste treatment, chemical processing and removing paramagnetic particulate impurities [198]. Magnetic susceptibility refers to the ability of a material to become magnetic and shows to what extend a material responds to a weak magnetic field [199]. Gopalakrishnan [200] found anhydrous cement to have a magnetic susceptibility of 91.028×10^{-6} emu/g (11.4×10^{-7} m^3/kg), showing its weak attraction to magnets due to impurity ions, mainly iron. Gołuchowska [201] examined factors affecting the susceptibility of cement dust, highlighting additives, fuels, raw materials used during the production and the process type itself, with iron content being of importance. As the iron content increases, the magnetic susceptibility also increases. Additionally, dry kilns produce dust with higher susceptibility. This suggests that it is important to know the origin of the binder within the concrete to optimise the separation.

Ulsen et al. [174] noted differences in recycled sand and cement paste susceptibility, making it a useful characteristic for separation. They investigated materials ranging from 0.3 to 0.6 mm in size under various magnetic field strengths and showed that using a Frantz barrier field separator to remove magnetic phases decreased cement paste content in the recycled sand fraction, with higher magnetic intensities enhancing separation. Carriço et al. [163] used high-intensity magnets to separate concrete constituents in the 125 μm – 1 mm range, because smaller particles are largely influenced by air convection and adhesion. The fractions were washed and dried before entering the separator. Initially, most high-quality (non-magnetic) aggregate was separated from the (magnetic) aggregate with more mortar. After adjusting belt speed and divider opening size, the magnetic aggregate underwent three separator passes to extract cement from the aggregates. They found approximately 40 wt% of the total cement content could be retrieved from the concrete after two passes of the magnetic aggregate. This percentage was influenced by the removal of the fraction below 125 μm and the liberation of attached mortar from the aggregates, indicating the need for additional liberation steps. To minimize environmental impact, handling fractions below 125 μm is important. These fractions likely contain most of the cement and possibly some aggregate particles from the crushing process. Another drawback is washing and drying of the material to remove the fine fraction and the required monolayer feeding, which reduces capacity on an industrial scale.

4.7. Electrostatic separation

Multiple characteristics are of importance when analysing a powder. Malvern instruments [202] states that the most important physical characteristics are:

- Particle size
- Particle shape
- Surface properties
- Mechanical properties
- Charge properties
- Microstructure

The charge of particles is one of the fundamental properties that is used to gain insight in the dispersion stability [202], but is also an interesting property for separation. Electrostatic separation is a dry separation process that takes into account the difference in charging behaviour of materials. Frictional contact between a material and a charging wall leads to electron transfer and subsequently charging of the material. In an electric field this material is attracted to either the negative or positive electrode based on its polarity (negative or positive) [203,204].

Dötterl et al. [205] gives a list of materials ordered based on the charging they obtain during friction. Quartz is placed at the bottom of the list, which means that it will become negatively charged. Binkhorst and Cornelissen [206] established that cement moves towards the negative electrode after charging. In total three methods were tested to charge the particles, namely conductive charging, contact charging and ion bombardment. The results showed that contact charging was the separation technique of interest. This charging approach is mentioned as a possibility for the separation of binder and aggregates. Although, further research is necessary to gain insight in the application potential of the technique for the realisation of the separation of quartz and cement.

The expectation is that, compared to gravity or magnetic separation, the separation becomes easier especially for smaller particle fractions. Therefore, electrostatic separation is a method of interest and should be studied to determine its potential for optimizing the separation of the cementitious fraction and the siliceous aggregate fines. An overview of the treatments methods is shown in Table 12.

5. Conclusion and outlook

As the demand for higher quality recycled concrete fractions is increasing [30], the recycling process must be designed in such a way that quality improvement is achieved. This article focused on the downstream processing of EoL concrete to recover high-value cementitious fractions. The main conclusions drawn from this study are as follows:

1. The concrete recycling industry needs to shift from the current traditional recycling techniques towards innovative recycling techniques, because these not only produce clean separated gravel and sand fractions, but more importantly they produce residual cementitious fines. These fines specifically, have the potential to fully replace primary clinker, which could eliminate the environmental impact associated with its current production and significantly reduce CO_2 emissions from concrete production.
2. To understand the potential of the residual cementitious fines after recycling (output), it is necessary to determine the quality of the concrete (input), particularly its binder, before a structure is demolished. Quality assessment primarily involves determining the binder type. A handheld x-ray fluorescence technique appears to be the most promising for rapid in-situ binder type identification.
3. Residual cementitious fines, often contaminated with sand derived silica, should be further optimized and separated to remove non-cementitious materials and enhance its application potential. For this purpose, electrostatic separation should be further researched, as most other optimization techniques focus on improving recycled concrete aggregates, often neglecting residual cementitious fines, which can lower their quality or make them unusable.

Based on the conclusions outlined in this article and aiming to achieve future sustainability goals, it is recommended to prioritize forthcoming research endeavours towards optimization of the residual cementitious fines and its subsequent recycled binder fractions. This calls for the global adoption of innovative recycling techniques, as the current traditional recycling process (Fig. 2) is only partially circular, leading to downcycling. The breakthrough in using innovative recycling techniques (Fig. 3) lies in the potential for fully reusing and upcycling

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