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Review

Circular economy, data analytics, and low carbon concreting: A case for managing recycled powder from end-of-life concrete

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ABSTRACT

Reuse and recycling are vital practices in the circular economy. Despite progress in recycling aggregates from end-of-life concrete, the potential for reusing recycled concrete powder (RCP) as supplementary cementitious material (SCM) is now attracting research and commercial interest. Herein, a meta-analysis of concrete in which RCP was used as an SCM was conducted, and a multivariate regression model was developed for predicting strength from mix composition. The carbon footprint of alternative beneficiation strategies, including milling only, milling plus thermal treatment and milling plus CO_2 injection, were quantified and used with the regression model to investigate RCP-containing concrete's embodied carbon (eCO_2). The comparison was made with conventional SCMs and end-of-life scenarios of concrete from different cement types. The meta-analysis and regression model showed that 15% cement replacement by the non-beneficiated RCP caused a 40% reduction in 28-day compressive strength, and at 50% replacement, the strength reduction was 70%. Above 30% cement replacement, the RCP beneficiated through CO_2 injection reduced the concrete's eCO_2 per unit strength by 10–25%, while the thermally treated RCP had greater eCO_2 than conventional SCMs. Thus, circularising end-of-life concrete does not guarantee low carbon concrete production. Instead, treating RCP with waste CO_2 leads to a carbon-negative SCM, presenting the most promising route for low-carbon concrete in the circular economy.

1. Introduction

A circular economy (CE) is defined as an ecosystem that is always restorative and regenerative by design, such that materials and products remain at their highest value beyond the design life (Ellen MacArthur Foundation, 2013). Such an ecosystem offers a sustainable alternative for tackling the linear take-make-and-dispose model of production and consumption. The CE principles, which encompass the 3Rs - reduce, reuse, and recycle (Korhonen et al., 2018; Haas, 2015; Morseletto, 2020; Bocken et al., 2018) are being adopted in various industrial sectors, but uptake in the construction industry needs to catch up (Ghisellini et al., 2018). Adopting the CE principles can be pivotal in addressing some of the challenges around greenhouse gas emissions arising from the production and supply of construction materials (Pomponi and Moncaster, 2017). The concrete industry is under pressure to reduce its carbon footprint and become circular. Since cement production accounts for 5 – 9% of anthropogenic CO_2 (Hammond and Jones, 2008; Scrivener et al., 2018), various concrete roadmaps (CEMBUREAU 2019; GCCA 2021) recommend cement clinker substitution as a critical pathway toward decarbonisation.

Common SCMs, including Ground Granulated Blast furnace Slag (GGBS), and pulverised fuel ash (PFA), are in short supply due to environmental pressures on the coal and steel industries (Scrivener et al., 2018; Adu-Amankwah et al., 2022). Meanwhile, construction and demolition waste (CDW), including concrete, is amongst the significant waste streams in the EU, with more than 820 Mt generated annually (EEA 2020; Eurostat 2017). Concrete in CDW is routinely recycled into hardcore for road sub-bases or as aggregates for new concrete (Etxeberria et al., 2007; Evangelista and de Brito, 2007; Sagoe-Crentsil et al., 2001). However, a considerable volume of hardened cement powder is generated as a by-product during recycling for aggregates. Despite its similar elemental composition to conventional SCMs, the mineral-rich powder, designated as recycled concrete powder (RCP), is commonly downcycled or disposed in landfills (Ghisellini and Ulgiati, 2020;

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Gálvez-Martos et al., 2018). Recent studies have evaluated its feasibility as SCMs (Li et al., 2022; Lu et al., 2018; Topič et al., 2017; Wu et al., 2021), but there are challenges around efficient upcycling strategies and their impact on the process's carbon footprint.

Reuse of RCP as an SCM promotes resource conservation by diverting it from landfills. However, the environmental impact of repurposing techniques as levers for the CE must be quantified. For the RCP recovered from concrete to be used as SCM, beneficiation through thermal, chemical, or their combination (Skocek et al., 2020; Sui et al., 2020) is necessary due to its metastability (Lu et al., 2018; Kim and Choi, 2012; Ma and Wang, 2013). However, reported beneficiation strategies affect the mix design and performance (Lu et al., 2018; Wu et al., 2021; Bostanci et al., 2018; Xuan et al., 2017) and the eCO_2 of concrete incorporating the beneficiated RCP. Quantifying the eCO_2 must be underpinned by an awareness of RCP composition, mix proportioning of RCP-containing concrete, and performance, e.g., strength. Isolated parametric studies are insufficient for deriving such relationships due to sensitivity to local factors, e.g., cement composition, mixing and placing methods etc. Instead, a model recognising the inherent variability in RCP and concrete production is necessary. Consequently, this study performed a meta-analysis of the literature on RCP utilisation as an SCM and derived a regression model relating mix design variables to performance and, ultimately, the eCO_2 of RCP-containing concrete. In the following sections, recovery of the RCP and beneficiation techniques are reviewed as background to the meta and the eCO_2 analyses.

2. Recovery, reuse, and beneficiation of RCP for circularity

Recycled coarse aggregates (RCA) and fine recycled aggregates (FRA) have been investigated extensively (Etxeberria et al., 2007; Evangelista and de Brito, 2007; Bostanci et al., 2018; Xuan et al., 2017; Nedeljković et al., 2021; Fang et al., 2021; Khatib, 2005) but less so for the RCP. Typically, particles coarser than 5 mm are recovered as RCA and those between 100 μm and 5 mm as FRA (Martinez-Echevarria et al., 2020). The RCP comprises the residue finer than 100 μm and is dominated by hydrated and unreacted cement.

Several techniques for separating the aggregates from fines and the RCP in concrete have been reported (Kasai, 2006; Nagataki et al., 2004; Mulder et al., 2007; Shima et al., 2005; Akbarnezhad et al., 2011; Yonezawa et al., 2001) and reviewed in (Quattrone et al., 2014). The conventional method is the ordinary recycling process based on single-stage mechanical pulverisation with jaw crushers followed by sieving. A lower RCP recovery efficiency and reduced RCA quality were reported for this technique due to the attached mortar (Nagataki et al., 2004; Pedro et al., 2015). Meanwhile, eccentric rotor crushing (ERC) after Yonezawa et al. (2001) and screw abrasion crushing (SAC) (Kasai, 2006) are two-stage separation techniques. In the ERC, the pre-crushed concrete rubble is further rolled and sheared to maximise the removal of adhered mortar (Quattrone et al., 2014), whilst the SAC use friction in a circular tube to force contact between the aggregate particles. The compression and impact process (Nagataki et al., 2004) is a complex system comprising multiple crushers and sieving to reduce the concrete into various size classifications, with RCP the residue after recovering the aggregates.

The as-recovered RCP has limited application in the cement and concrete industry. Some studies examined the suitability of the RCP as a raw meal for cement clinker production (Schoon et al., 2015; Gastaldi et al., 2015; Krour et al., 2022). Kiln energy demand and quality of clinkers produced from RCP raw meal were influenced by recovery technique, gradation, and moisture contents. Liu et al. (2021) showed that as clinker raw meal, RCP required lower clinkering temperatures but the early age reactivity of the clinker reduced due to higher quartz inclusion and lower tricalcium/dicalcium silicate ratio (Diliberto et al., 2017) of the clinker. The as-recovered RCP has also been explored as SCM for new concrete (Diliberto et al., 2021; Cantero et al., 2020; Topič et al., 2017). For strength, optimum content ranged between 15 - 30% (Kim and Choi, 2012; Xiao et al., 2018), but durability reduced at contents exceeding 10% (Ma et al., 2020; Li et al., 2021). Hence, beneficiation is essential to increase the RCP content to compete as an SCM.

2.1. RCP beneficiation through size reduction

Comminution of RCP to finer or comparable gradation as cement clinker has been explored. Finer RCP has a larger surface area and would be expected to participate in the hydration process (Ma et al., 2020). Wu et al. (2019) investigated the fresh state and hardened properties of RCP of varying fineness. The finer RCP increased water demand and shortened setting time due to the larger surface area, but strength was only increased for substitutions not exceeding 7.5% of the cement. Zhang et al. (2019) examined resident time in the ball mill and grinding aids on RCP reactivity. Sodium sulphate as a grinding aid and a longer resident time improved reactivity, but further grinding beyond a threshold had marginal benefits. Horsakulthai (2021) reported lower strength activity indices when the RCP was ground finer than OPC. This implies that size reduction is an essential consideration in RCP beneficiation.

2.2. RCP beneficiation through thermal treatment

Above 100 °C, hydrated cement decomposes through dehydroxylation and decarbonisation reactions (Ramachandran et al., 2002). These weaken the cement matrix and promote the separation of the RCP. The relationship between treatment temperatures, size reduction, and reactivity of the beneficiated RCP was reported in (Wu et al., 2021; Sui et al., 2020; Shui et al., 2009; Larbi et al., 2000), with the 600 - 800 °C range suggested as optimal. In this temperature range, hydrated assemblages and any carbonates in the RCP decompose, while most calcium-bearing phases transform into quicklime. Higher beneficiation temperature promotes the formation of belite, a less reactive phase in cement. Mulder et al. (2007) proposed a heating and sorting process whereby crushed concrete was heated up to 700 °C and sorted into sizes. A similar treatment presented by Shima et al. (2005) involved heating the crushed concrete to 300 °C in a furnace. Microwave heating was explored in (Akbarnezhad et al., 2011; Quattrone et al., 2014) and found to reduce the mortar sticking to the aggregate significantly.

Techniques to improve RCP recovery efficiency were studied using the integrated drying and heating system (Gebremariam et al., 2020). In the composite air and heating classification system, heat was blown over circulating air at ~700 °C. Up to 20% of the hardened paste remained on the aggregate's surface, but according to the acid digestion test, 48% of the recovered RCP was reactive. Notwithstanding the improved performance of RCP or mixed FRA after thermal treatment (Shui et al., 2008), the energy required for heating to the 600–800 °C range is about half of the energy for cement production, which is still high, whilst CO_2 trapped in the concrete during its service life is released in the recommended temperature range.

2.3. RCP beneficiation through CO_2-injection

Carbonation can also be used to activate the RCP for reuse as an SCM. Reacting the RCP with CO_2 (i.e., carbonation treatment) destabilises the hydrated phases by converting the metastable assemblages into calcium carbonate, silica and alumina gels and gypsum, depending on the parent concrete's composition (Morandeau et al., 2015; Justnes et al., 2020). Like monolithic specimens, carbonation of the fine RCP is governed by CO_2 diffusivity into the powder, which depends on the RCP composition and water-to-binder (w/b) ratio, exposed surface area, CO_2 concentration, pressure, and relative humidity. Kinetics of carbonation of the various phase assemblages in OPC and composite cement, e.g., C-[A]-S-H, portlandite, ettringite and carboaluminates have also been studied extensively (Morandeau et al., 2015; Justnes et al., 2020; De Weerdt et al., 2019; Pu et al., 2012; Borges et al., 2010; Goñi et al., 2002; Gruyaert et al., 2013; Morandeau et al., 2014; Ngala and Page, 1997), and the underlying equations summarised in Appendix A5–1. The phase assemblages mentioned above tend to carbonate simultaneously (Castellote et al., 2009), but portlandite acts as a buffer, and its depletion accelerates carbonation of the other assemblages (Herterich et al., 2022). However, the cement type in a given concrete affects the nature and distribution of the phase assemblages (Adu-Amankwah et al., 2018; Adu-Amankwah et al., 2017; Lothenbach et al., 2008; Lothenbach et al., 2011) and their carbonation potential (Morandeau et al., 2015; Zajac et al., 2020; Adu-Amankwah et al., 2021; Juenger et al., 2019). This bears implications on the suitability of different cement types, e.g., composite cement beneficiation through CO_2-injection.

Carbonation treatment of RCP can be achieved by exposure to air (i.e., natural sequestration) or manufactured, e.g., CO_2 captured from industrial processes. Natural carbonation is slow, so its application to RCP beneficiation is limited. For example, after 28-days exposure at 65% relative humidity (RH) and atmospheric CO_2, Mehdizadeh et al. (2021) observed portlandite and calcium silicate hydrate (C-S-H) were still present in the matrix and measured low compressive strength even at moderate substitution levels. This aligned with the observations of Lu et al. (2019). Meanwhile, accelerated or enforced carbonation involving exposure to high concentration CO_2 at 50 – 70% RH (Lu et al., 2019; Fang and Chang, 2015; Li et al., 2019; Skocek et al., 2020) and wet carbonation have been used elsewhere (Zajac et al., 2020; Ben Ghacham et al., 2017). The carbonation protocol affects the phase assemblage from the process. For example, Zajac et al. (2020) found that the calcium carbonate precipitated primarily as calcite due to the water available during wet carbonation. Since the dominant hydrates in OPC-based cement are calcium-bearing, calcite precipitation was not sensitive to the nature of the cement used in the primary mix. Distinct from comminution and thermal activation, CO_2 injection into RCP presents unique opportunities to circularise RCP while lowering process emissions simultaneously.

3. Methods

The meta-analysis conducted in this study was based on secondary data collated from multiple primary research on RCP utilisation as an SCM. A schematic of the methodology is shown in Fig. 1. The PRISMA systematic review methodology (Moher et al., 2009) was followed to retrieve relevant published journal articles from the Scopus and Web of Science databases.

3.1. Literature search strategy

Research on RCP and its use as an SCM is still growing. Different terminologies have been used to describe this, e.g., hydrated cement paste, waste concrete fines, and waste concrete powder. Accordingly, the literature search was refined to capture all relevant publications. The query string was implemented with the Boolean operators, and the number of documents returned was included in Fig. 1.

The abstracts were scanned from the list of articles returned, and relevant papers were selected for detailed review. Specifically, RCP must have been used as an SCM, and the mix composition and performance must be reported to qualify an article for inclusion. A few studies that only investigated the microstructure and phase assemblages in RCP without strength data were excluded. From the selected articles, data relating to the RCP beneficiation strategy, mix compositions, and RCP mortar and concrete performance were extracted into an Excel spreadsheet. Once all documents were read, a snowball search was performed by looking through the references listed in each article with the same technique of assessing the suitability and extracting the data.

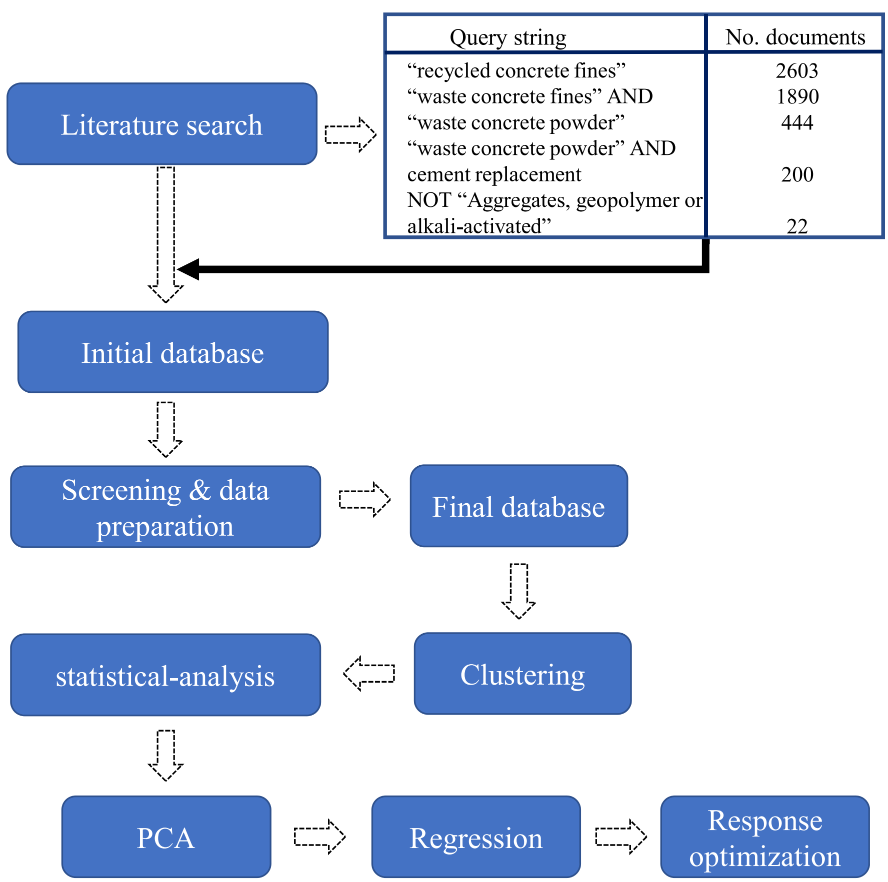

Fig. 1. Overview of methodology.

3.2. Preparation of the database

Variations in the primary data are implicit in the meta-analysis methodology. Herein, the origin of the RCP (i.e., end-of-life concrete, laboratory-made concrete, or idealised cement paste), the secondary material for which the RCP was utilised (i.e., in concrete, mortar or cement paste mixtures) and beneficiation techniques were observed sources of variation in the secondary data. Concerning the origin of the RCP, end-of-life concrete after demolition used in (Diliberto et al., 2021; Kim et al., 2014) was of interest. Meanwhile, RCP from laboratory-made concrete (Evangelista and de Brito, 2007; Cantero et al., 2020; Ben Ghacham et al., 2017) and idealised cement pastes (Fang et al., 2021; Zajac et al., 2020; Fang and Chang, 2015; Zajac et al., 2020) have also been reported. Classification based on the RCP origin and reuse scenario and the number of studies in these categories can be found in Appendix A1 and A2. The papers considered in this study, spanning between 2012 and 2022, are listed in Table 1.

The selected articles reported fresh and hardened properties of the RCP-containing concrete. The slump was the commonly reported fresh state property and compressive strength evolution with time, the hardened property. Statistical analysis of the secondary data was conducted to derive a generalisable relationship between mix design parameters (i.e., predictors) and compressive strength (i.e., response). This was a prerequisite for eCO_2 analysis of the concrete produced with RCP. The 28-day compressive strength was chosen because it is central to structural design and mix proportioning. It is also the functional unit for concrete eCO_2 analyses, e.g. (GCCA 2021; Purnell and Black, 2012).

3.3. Statistical analysis

There were 53 entries of extracted data on reusing RCP from end-of-life concrete. As expected, some of the entries had missing compositional or performance information. Incomplete data were excluded, leaving 44 cases for the analyses. This population size (>30) is sufficient to draw inferences (Wehrens, 2020).

Using Minitab, principal component and multivariate regression analyses were performed on the clustered data. The criteria for selecting the articles ensured that the secondary data was representative and randomised. Consequently, descriptive statistics, including each variable's statistical mean, variance and skewness, were initially assessed alongside a correlation test to identify multi-collinearity (Jobson, 2012). A significance level of 0.05 was chosen so that a p-value of less than this implied a non-zero correlation. Principal component analysis (PCA) was used to reduce the number of variables before deriving the strength versus mix composition model from the multi-regression analysis. The reader is referred to Appendix A3 for the equations supporting the statistical analyses.

3.4. Carbon accounting framework for beneficiated RCP as SCM

The LCA framework based on EN 15,978 (BSI 2011) was used to calculate the carbon footprint of RCP beneficiated by (i) milling of the as-recovered RCP only, (ii) thermal activation, and (iii) CO_2 injection, designated RCP-Un, RCP-T and RCP-C respectively. Potential in-service CO_2 uptake of the concrete before demolition was discounted to avoid double counting. Given that RCP recovery is not usually the primary reason for demolition, product-stage emissions from raw material

Table 1
Evidence table showing key publications included in the study and the range of variables.

Publication	Group	Treatment method	RCP mean particle size (D_{50}), µm	RCP Content,%	Tests conducted	Optimum replacement
Chen et al. (2019)	RC	Grinding	30	0, 10, 20, 30, 40	Compressive strength and slump	20%
Kim (2017)	RC	Grinding	90	0, 15, 30, 45	Compressive strength, tensile strength, and slump-flow	15%
Ma et al. (2020)	RC	Grinding	17.53 - 21.62	0, 15, 30, 45	Compressive strength	Up to 30%
Ma et al. (2019)	RC	Grinding	13.98 - 18	0, 15, 30, 45	Compressive strength and slump	<30%
Xiao et al. (2018)	RC	Grinding	4 - 115	0, 15, 30, 45	Compressive strength and slump	15 - 30%
Kim and Choi (2012)	RM	Grinding	14, 90,176	0, 15, 30, 45	Compressive strength, initial and final set	<15%
Li and Yang (2017)	RM	Grinding	154 - 1074	0, 17, 34	Compressive and flexural strength	
Liu et al. (2014)	RM	Grinding	10	0, 30	Compressive strength	30%
Oksri-Nelfia et al. (2016)	RM	Grinding	8.8	0, 25, 50, 75	Compressive strength and slump	Up to 25%
Sui et al. (2020)	RM	Grinding and thermal treatment	2.7	0, 30	Compressive and flexural strength	30%
Yu et al. (2019)	RM	Grinding	10	0, 12.5, 25, 50	Compressive and tensile strength	25%
Xue et al. (2016)	RM	Grinding	14.4 - 16	0, 20, 30, 40	Compressive strength, flexural strength and slump	30%
Nežerka et al. (2020)	RP	Grinding	60	0, 20, 40	Flowability	
Prošek et al. (2020)	RP	Grinding	8.2 - 106	0, 10, 20, 30, 40, 50	Compressive strength, flexural strength and flow test	Up to 30%
Qin and Gao (2019)	RP	Grinding; pastes carbonation cured	18.73 - 20.8	0, 10, 20, 30, 50	Compressive strength	20%
Topič and Prošek (2017)	RP	Grinding	16	0, 5, 10, 15, 20, 25	Compressive strength, flexural strength and slump	20–30%
Topič et al. (2017)	RP	Grinding	nd	0, 10, 20, 30, 40, 50	Compressive strength and flexural strength	20%
Diliberto et al. (2021)	RM	Grinding	7 - 25	0, 25	Comp strength	–
Likes et al. (2022)	RM, RC	Grinding	8 - 15	0, 20	Compressive strength, flow test	–
Duan et al. (2020)	RM	Grinding	9 - 66	0,10,20,30	Flow test, Compressive strength and flexural strength	
Li et al. (2022)	RM	Grinding	19, 32	0,10,30,50	Compressive and flexural strength, fluidity setting time	10%
Algourdin et al. (2021)	RM	Grinding and thermal treatment at 80 or 500 °C	20, 26	0,20 0,10,30,50	Compressive, flexural strength and freeze-thaw	

Note: RC, RM and RP denote demolished recycled concrete powder used in concrete, mortar or paste samples, respectively, as defined in Appendix A1.

extraction (A1) were discounted. Instead, emissions due to transportation (A2), crushing and separation of aggregates (A3), beneficiation (i.e. thermal or CO_2 injection treatment), and milling (A3) (BSI 2011) were considered.

A hotspot map of UK concrete demolishers and recyclers (Appendix A4) showed a ~40 km accessibility radius. These were assumed as RCP beneficiation sites. Transportation emission factors were adopted from the UK government's Department for Environment, Food, and Rural Affairs (DEFRA) database (DEFRA, 2022). The SAC separation technique was assumed for RCP extraction due to its efficiency and the process energy taken from (Quattrone et al., 2014). Energy coefficient for milling the beneficiated RCP to comparable fineness as cement was taken from (Tsakalakis and Stamboltzis, 2008). In addition to the RCP beneficiated by milling only, two beneficiation techniques via thermal treatment using a kiln operated up to 600 °C (Wu et al., 2021; Sui et al., 2020; Shui et al., 2009; Larbi et al., 2000) and (b) CO_2 injection (Lu et al., 2019; Li et al., 2019; Zhan et al., 2019; Liu et al., 2021) were investigated. For thermal treatment, the energy expended was estimated as half of that for cement clinker production (Ellis, 2004), assuming diesel-operated kiln and the fuel co-efficient taken from (DEFRA, 2022).

CO_2 injection provides a sink for removing atmospheric and industrial CO_2, as discussed in Section 2.3. CEN/TR 17,310:2019 (BSI 2019) provides a methodology for computing the CO_2 uptake potential of concrete based on the phase assemblages present. The cement type determines phase assemblages; hence two most common types, OPC and composite cement (CEM II), were considered. Typical assemblages in the RCP from both cements were reported in (Shen et al., 2022). The eCO_2 for RCP subjected to CO_2 injection was estimated using these and the detailed calculation framework shown in Appendix A5–1. Flue gas from the recycling plant was assumed as the CO_2 source. Consequently, emission factors of 0.049, 0.165, −0.261 and −0.083 eCO_2kg/kg of RCP for the milled-only RCP, thermally treated and the CO_2 injected CEM I and CEM II RCP were obtained respectively. Appendix A5–3 summarises these alongside those for GGBS, PFA and other constituents in concrete taken from (Hammond and Jones, 2008; GCCA 2021).

Precast concrete production was assumed, and 0.5% superplasticiser was added to compensate for reduced workability when using RCP. The cradle-to-concrete casting system boundaries were used for the eCO_2 analysis.

4. Results and discussion

4.1. Meta-analysis

4.1.1. Descriptive statistics of the data

Variations in RCP content and mix design parameters are readily seen in Table 2, showing the descriptive statistics and normality test (i.e., $Z_{skewness}$) results. The RCP mean fineness (D_{50}) and RCP content had greater spread, so their standard deviations were close to the variable mean. This warranted verification of the normality and linearity conformance in the data to establish suitability for the multivariate-regression analysis (Hair et al., 2019).

Normal distribution of variables is optimal for multi-regression analysis. The predictor variables were mostly symmetrical, so the skewness test provided a basis for identifying potential departures. Positive skewness in the RCP content, D_{50}, and OPC content indicated left-shifted distribution, while the reverse was seen in the aggregates content, w/b ratio and strength. However, a notable lack of normality was identified in the D_{50}, RCP and aggregates content, which showed higher peaks (Kurtosis) with less flat tails. Note that the non-normal distribution of variables arose from an unequal distribution of variance (i.e., heteroscedasticity), which would induce biases in the subsequent analyses. Transformation of the predictor variables was thus necessary (Hair et al., 2019). The original and transformed normal probability plots of the variables can be seen in Appendix A6. Notably, suitable transformation functions could not be found for the w/b ratio and the aggregates content; hence, their original data were used after scaling. This removed the size effect (i.e., units) of the variables. Thus, data points with zero magnitudes were equivalent to the variable means, whilst the scale variables' sign showed their importance relative to the mean, with larger variables being positive.

4.1.2. Correlations test on predictor variables

Linearity is also central to multivariate regression analyses, the conformance of which was verified from the correlation plots shown in Fig. 2. The correlation coefficients (R^2) obtained were generally weak and barely exceeded 0.5 in all cases. The trends, however, provide helpful insight into the relationship amongst the variables. It must be recognised that RCP was primarily used as SCM to replace a part of the cement; thus, an increase in its content decreased the OPC. The RCP content correlated strongly with its fineness (i.e., larger D_{50}), as the recycling process often led to finer particles (Liu et al., 2014), which increased water demand (Xiao et al., 2018; Duan et al., 2020), as reflected in the w/b ratio. Increasing the RCP and OPC contents led to a higher overall binder content and correspondingly reduced the volume of aggregates. The exceptions were the RCP content versus its mean particle size (D_{50}) and aggregates proportion versus D_{50}. The scatter in the data, where RCP increased without a corresponding decrease in OPC, represented instances where the former was added as filler, e.g., (Duan et al., 2020). At a 95% confidence interval, none of the correlations was deemed significant (i.e., $p > 0.05$). The conclusion from the above is that the predictors (mix design variables) would affect compressive strength independently. However, interpreting performance across five non-correlated dimensions is complex. A further test to identify underlying multi-collinearity was assessed from the PCA test.

4.1.3. PCA test

The PCA results in Table 3 show the principal components (PCs) alongside the eigenvectors. These reveal the extent to which the variables combine to explain the performance of the RCP-containing concrete. The RCP content and its fineness had the highest loading on PC1 and explained over 50% of the compressive strength. Meanwhile, OPC was the w/b ratio and aggregates content (PC3), each explaining 20% of the performance. This means that the compressive strength of RCP-containing concrete principally depends on three factors – (a) the RCP and its attributes, (b) the OPC content, and (c) non-cement constituents (i.e., aggregate and water). The loading scores for the three PCs agree with the literature, highlighting the importance of the RCP content and fineness on the performance of RCP-containing concrete (Kim and Choi, 2012; Xiao et al., 2018; Ma et al., 2019; Li and Yang, 2017) due to

Table 2
Descriptive statistics of the data collected for mortar samples made with RCP and normality test results at a 95% Confidence Interval.

Variable	N	Max	Min	Mean	Median	Normality	Skewness (Z)	Kurtosis	P-value (transformed)
D50, μm	44	153	2.7	39.8	15.7	0.268	1.60	1.25	<0.01 (0.012)
RCP, kg/m3	46	585	0	130.4	104	0.181	1.35	2.48	<0.01 (0.177)
OPC, kg/m3	46	913	125	377.9	315	0.17	1.02	0.44	<0.01 (0.042)
Aggregate proportion,%	46	80.4	60.0	75.9	77.6	0.206	−2.56	10.24	<0.01
w/b	46	0.71	0.16	0.46	0.5	0.288	−1.05	0.16	<0.01
Strength, MPa	46	55.3	4.2	33.6	34.4	0.075	−0.35	0.32	>0.15

Note: P-value after transformation shown in the bracket.

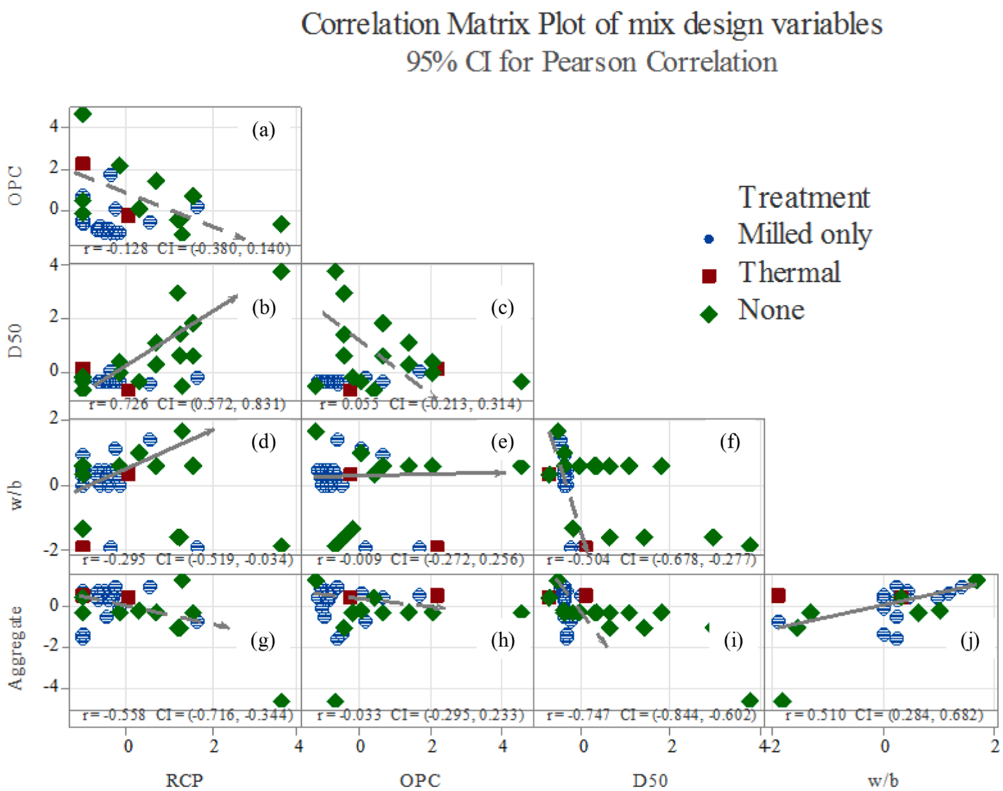

Fig. 2. Correlation matrix plots showing scatter correlations amongst the mix design variables: (a) RCP content vs OPC content; (b) RCP content vs RCP's D_{50}; (c) OPC content vs RCP's D_{50}; (d) RCP content vs w/b ratio; (e)OPC content vs w/b ratio; (f) RCP's D50 vs w/b ratio; (g) RCP content vs Aggregate content; (h) OPC content vs Aggregate content; (i) RCP's D50 vs Aggregate content; (j) w/b ratio vs Aggregate content. Note: The trend lines visually indicate potential correlations - dashed grey lines indicate positive trends, and solid grey lines, indicate negative trends. The colour code of the symbols denotes the beneficiation technique.

Table 3
Principal component analysis of the data in the RM category.

Variable	PC1	PC2	PC3	PC4	PC5
D50 (kg/m3)	**0.580**	−0.136	0.025	0.439	−0.672
RCP (kg/m3)	**0.586**	−0.077	0.099	0.319	0.734
OPC (kg/m3)	0.023	**−0.855**	0.393	−0.339	−0.014
Aggregate,%	−0.363	−0.489	**−0.599**	0.511	0.097
w/b	−0.433	0.078	**0.691**	0.574	0.011
Eigen-analysis of the Correlation Matrix					
Eigenvalue	2.475	1.130	0.897	0.3082	0.190
Proportion	0.495	0.226	0.179	0.062	0.038
Cumulative	0.495	0.721	0.900	0.962	1.000

Note: Factor loading in the first 3 PCs greater than 0.5 are bolded.

dilution arising from the non-beneficiated RCP (Lu et al., 2018). This shows that the first three PCs can explain over 90% of the compressive strength. Meanwhile, the negative correlations between the aggregates and RCP were only cross-loaded in PC4, which explained only 6% of the performance. This may be explained by the RCP-dominated matrix being weaker than the aggregates and the increase in RCP; hence, the binder content reduces the volume of aggregates.

4.2. Performance of RCP-containing concrete

4.2.1. Compressive strength

The compressive strength of concrete and its relationship with mix composition is an essential consideration in materials selection and structural design. Other design parameters such as the elastic modulus, dimensional stability and durability may be estimated from the compressive strength using normative design guidelines such as the Eurocode 2 (BSI 1992) and the Model Code (BSI, 2014). Understanding the strength versus mix composition relationship is imperative for eCO_2 analysis that considers the performance of concrete.

Fig. 3 shows the effect of changing the content of the non-beneficiated RCP simultaneously with each uncorrelated mix design variable on 28-day compressive strength. The RCP content had a more significant impact on strength than its fineness (Fig. 3a). The content also determined the extent of filler and dilution effects (Lothenbach et al., 2011) from the RCP and their impact on strength. At lower RCP content (i.e., $\leq$ 100 kg/m^3, equivalent to ~30% of the total binder content in normal-strength concrete), fineness did not affect compressive strength significantly because it was merely a filler. The filler effect would increase the effective w/b ratio (Lothenbach et al., 2011), thus sustaining cement hydration. Moreover, hydrates in the RCP, e.g. the C-S-H, are also known to provide favourable nucleation sites for hydration (CEB-FIP 2012). In RCP-containing ultra-high performance concrete (UHPC), where the total binder contents were much higher, e.g., (Yu et al., 2019), but RCP used a as filler, contents >150 kg/m^3 did not reduce strength.

Increasing RCP content at the expense of OPC (the most reactive constituent) caused strength reduction (see Fig. 3b). In standard mortars, 250 kg/m^3 of RCP would equate to >60% OPC replacement, and strength can be expected to decrease accordingly, as noted elsewhere (Oksri-Nelfia et al., 2016). Above 300 kg/m^3 OPC content (e.g., in UHPC), where the RCP was essentially a filler, strength was not affected by the RCP content (Oksri-Nelfia et al., 2016) since the reactive cement content was already high.

The effect of RCP content and w/b ratio or RCP and aggregate contents on strength, shown in Figs. 3c and 3d, indicate that at small RCP content, i.e., < 50 kg/m^3, a moderate increase in the w/b ratio or aggregate content had a negligible effect on strength. Conversely, when the w/b was below 0.4, and the aggregate proportion was < 70%, an increase in the RCP content changed the strength band significantly once the RCP content exceeded the 150 kg/m^3 threshold. RCP-containing concrete requires a higher w/b ratio to compensate for the loss in workability (Duan et al., 2020), but this was also beneficial for strength gain, especially at lower RCP content. Meanwhile, the increased aggregate content meant lower binder content, and substituting more of this with RCP led to a lower strength, as noticed in Fig. 3d.

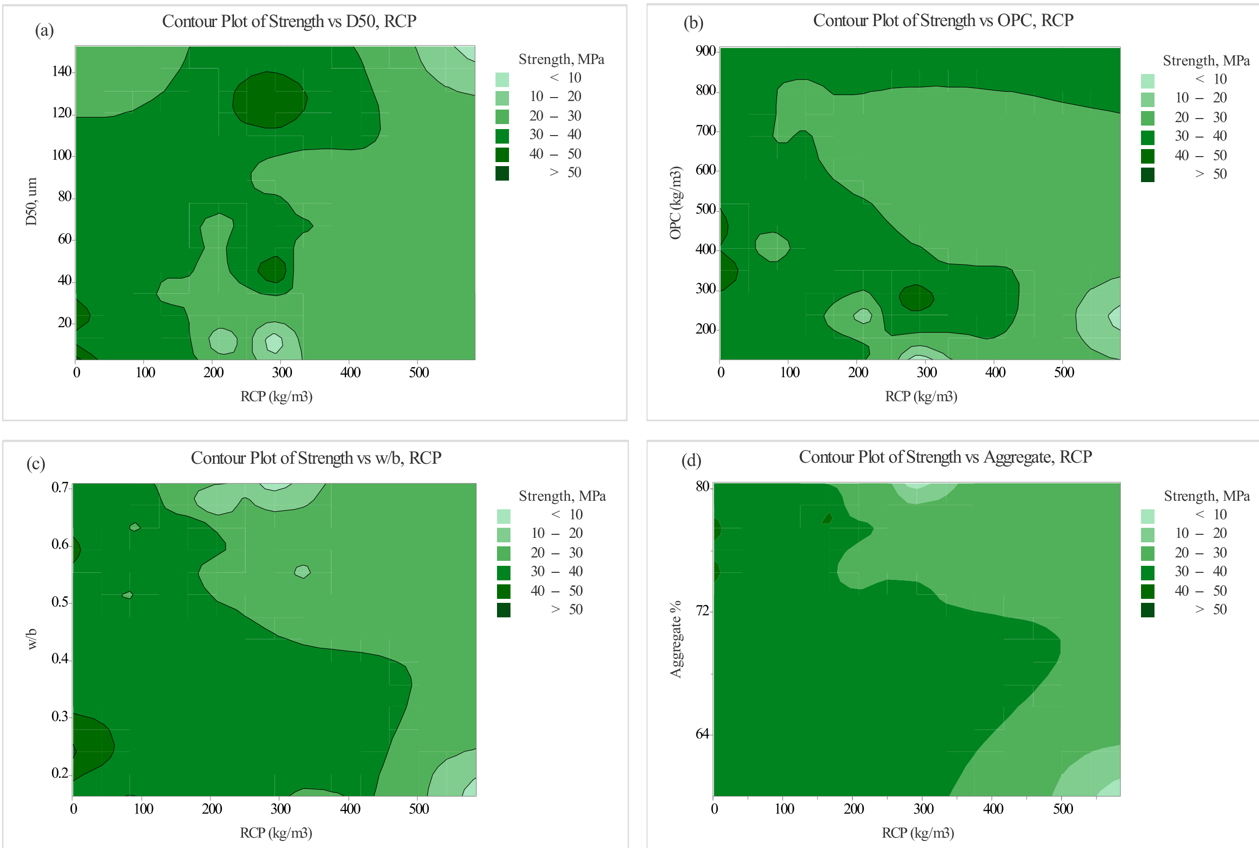

Fig. 3. Contour plots showing the effect of non-beneficiated RCP content and other mix design parameters on compressive strength (a) content and mean particle size of RCP vs strength, (b) RCP content and OPC content vs strength, (c) strength vs w/b ratio, (d) strength vs aggregate content.

4.2.2. Regression analysis

The preceding demonstrates that pairwise consideration of mix design variables provides valuable but limited insight into the effect on compressive strength because the relationships are multi-dimensional. Multivariate regression analysis provides a basis to quantify the contribution of each mix design variable to the compressive strength using the database created from Section 3.2. The ratio of independent mix design variables to the number of observations was ~1:15, and hence the data was deemed sufficient to model a generalised concrete mix composition versus strength relationship (Hair et al., 2019). The novelty of this approach is the strong statistical power derived from aggregating several independent studies as opposed to isolated experimental study, which has limited scope and generalizability.

Coefficients representing the change in compressive strength (S) associated with each mix design variable are shown in Eq. (1). The statistical significance of each factor can be found in Appendix A7.

$$S(MPa) = 79.8 - 0.068D_{50} - 0.053W_{RCP} + 0.005W_{OPC} - 0.237W, \%_{Aggregate} - 37.5w/b \quad (1)$$

Where W is the weight of the constituent in kg/m^3, and other variables as already defined.

The model's coefficient of determination, R^2 was 0.57, and the adjusted R^2 was 0.51, suggesting moderate predictive power of the model. The residual plots in the predicted (Appendix A8) show random distribution of about zero, which indicates a lack of bias in the prediction. However, there is a degree of overfitting (i.e., adj. $R^2 < R^2$), the model explains over 50% of the composition performance relationship, which is expected from a model based on several independently measured parameters as captured in the significant uncertainty factor.

In interpreting the predictor weightings, one must remember that most of the consulted literature used the non-beneficiated RCP (i.e., RCP-Un). Based on the *p-values*, RCP and OPC contents and the w/b ratio are the most influential factors affecting compressive strength. However, other factors which were not included as mix design variables, but were reflected in the intercept, play a critical role, albeit not statistically significant, $p > 0.05$ (see Appendix A8). Whereas the weighting of OPC increased strength, those of RCP content and the w/b ratio were inversely related to reduced strength, consistent with the literature (Topič et al., 2017; Kim and Choi, 2012; Prošek et al., 2020; Wyrzykowski et al., 2020). At 15% RCP content, the strength reductions ranged between 20 – 40% compared to the target strength achievable with GGBS and over 70% target strength reduction when 50% of the total binder content was RCP. These trends are consistent with the literature (Wu et al., 2019; Zhang et al., 2019; Horsakulthai, 2021). They can be explained by the dilution of effect (Lothenbach et al., 2011) arising from a higher fraction of unreactive constituents (i.e., the non-beneficiated RCP) in the cement matrix. Meanwhile, increasing the RCP's D_{50} (i.e., coarser RCP) and the aggregate content, although not statistically significant ($p > 0.005$), also reduced strength. Their low significance in the model stemmed from the multi-collinearity with RCP content and the w/b ratio, respectively, as identified in Table 2 and reflected in their higher variance inflation scores (see Appendix A7). The physical implication is that the RCP fineness would be influential only when it was reactive. Nevertheless, the RCP fineness may also affect the efficiency of the strategy, e.g., the resident time at a given temperature during thermal treatment or the exposed surface area during CO_2 injection.

4.3. Embodied carbon of beneficiate RCP and circular economy

The above-presented meta-analysis revealed that substituting OPC with the non-beneficiated RCP reduced compressive strength, the extent of which depended on the content (see section 4.2). Most of the reviewed articles used the ground RCP without further beneficiation (see Table 1). Meanwhile, thermal treatment (Wu et al., 2021; Shui et al., 2009; Bordy et al., 2017; Kalinowska-Wichrowska et al., 2020) and CO_2 injection (Lu et al., 2018; Skocek et al., 2020; Sui et al., 2020; Zajac et al., 2020) have both received considerable attention and have been shown to improve strength. The CO_2 injection treatment, on the other hand, depends on the phase assemblages (see Section 2.3), which are also dependant on the cement type. In this section, the embodied carbon assessment of concrete made with RCP in the three states – milled only (RCP-Un), milled and thermally treated (RCP-T), or milled and CO_2 injected (RCP-C) are compared to GGBS and PFA, two of the most commonly SCMs. The CO2 injection treatment for RCP derived from OPC (RCP-COPC) and composite cement (RCP-CCEM II) concrete was also considered. The RCP-$C_{CEM\ II}$ provided a basis to evaluate the beneficiation potential of emerging low-carbon cement.

Fig. 4 (a-c) compares eCO_2kg/m^3 per unit strength for reference CEM I and composite cement concretes in which 15, 30, or 50% of the OPC was replaced with GGBS, PFA, RCP-Un, RCP-T, or RCP-C (RCP-C_{OPC} and RCP-$C_{CEM\ II}$). Note that the 28-day compressive strength was chosen as the functional unit for the reasons explained in the preceding sections and consistent with (GCCA 2021; Purnell and Black, 2012). The strength of the RCP-Un was estimated from the model developed in Eq. (1), while those of the beneficiated RCP are expected to be comparable to GGBS and PFA (Xiao et al., 2018).

The results revealed the parabolic relationship between target compressive strength and the eCO_2 per functional unit strength. Up to ~30 MPa and above 70 MPa, a unit change in target strength was associated with a substantial increase in eCO_2 irrespective of the concrete's recipe. Between 30 – 60 MPa, the change in $eCO_2kg/m^3/MPa$ with increasing strength was marginal. Similar trends were reported in (Purnell and Black, 2012). This means that low-grade concrete (<30 MPa) and high-strength concrete, regardless of the recipe, had higher eCO_2 per unit strength, which deviates from recipe-based eCO_2 calculations available elsewhere (GCCA 2021). Due to design constraints, a lower strength class does not equate to a corresponding reduction in section, nor does a section of 25 MPa concrete perform the same function as that made with 50 MPa. Moreover, serviceability requirements, including fire resistance and durability, restrict the extent to which section size may be reduced. Therefore, there is a limit to the carbon reduction achievable through low-strength concrete. Outside the threshold, lower-strength concrete could have an even greater eCO_2. For applications, e.g., internal floor slabs, where strength above 30 MPa nor durability may be a critical functional requirement, up to 15% RCP content may be tolerated. Still, the benefit in terms of eCO_2 savings would be comparable to GGBS, albeit without the additional strength improvement. Conversely, a reduced w/b ratio and higher cement content per cubic metre of concrete would usually be required to

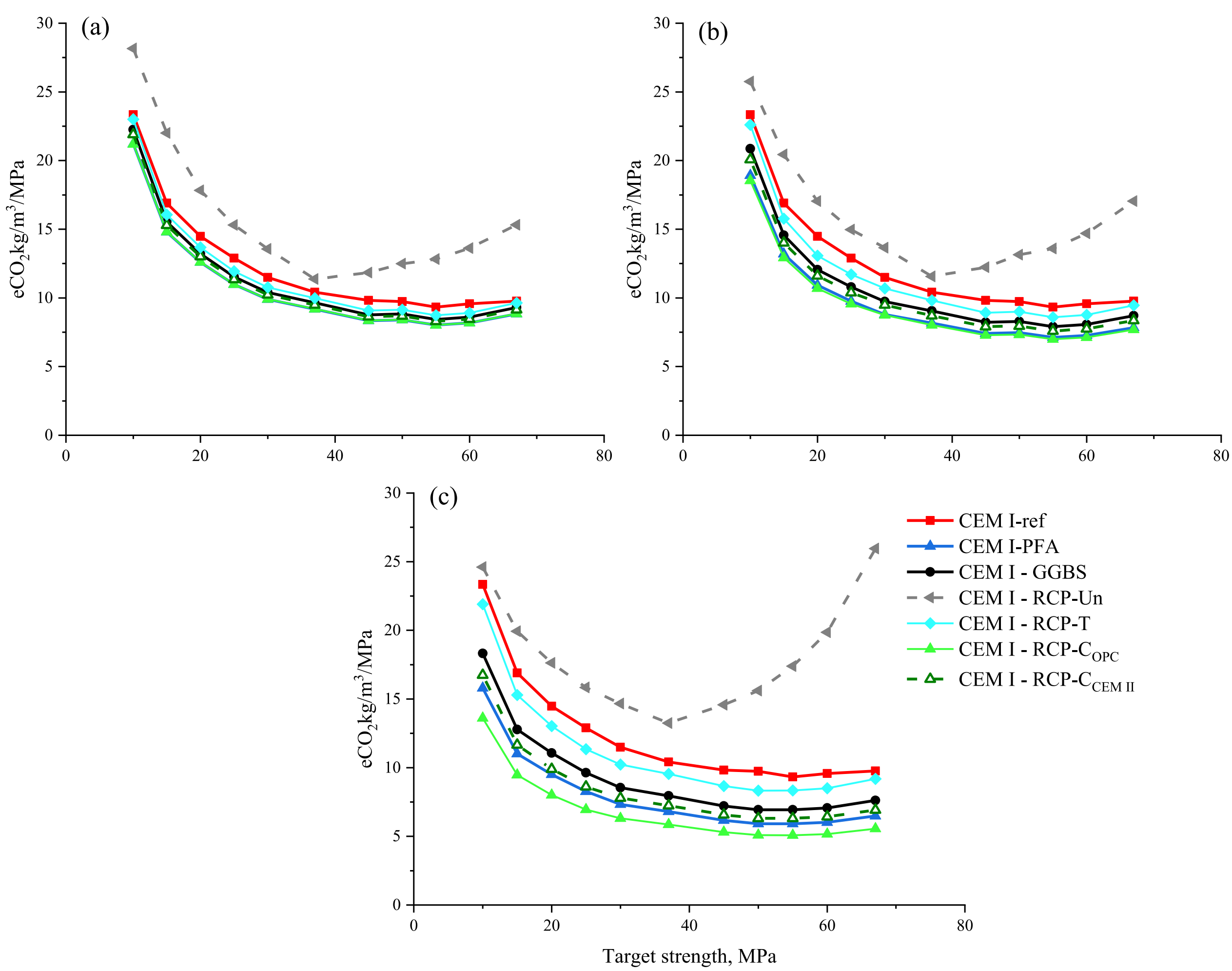

Fig. 4. Embodied eCO_2 of concrete in which non-beneficiated and beneficiated RCP replaced (a) 15%, (b) 30% and (c) 50% of CEM I as SCM in comparison to GGBS, PFA and CEM I. Note: RCP-Un is milled only RCP without additional treatment, RCP-T is milled and thermally treated RCP, and RCP-C is beneficiated through milling and CO_2 injection with the parent concrete containing OPC or CEM II composite cement.

produce high-strength concrete above 60 MPa, which explains the increased eCO_2. In the 30 – 60 MPa range, meanwhile, mix design optimisation, e.g., particle packing, high-range admixtures, etc., may be implemented to meet the strength requirement without necessarily increasing the cement content.

The eCO_2 of composite cement concrete depends on the SCM's embodied CO_2 and the attainable strength. Despite being circular, SCMs such as the non-beneficiated RCP do not necessarily have low carbon intensity. This is unsurprising, considering the grinding energy is comparable to GGBS, but the former attains lower strength at all replacement levels. Meanwhile, the thermally treated RCP, which can achieve comparable strength to PFA and GGBS at an equivalent mix design, has greater embodied CO_2 due to the energy for heating. This implies that using end-of-life concrete beneficiated through thermal treatment at the recommended 600–800 °C (Wu et al., 2021; Sui et al., 2020; Shui et al., 2009; Larbi et al., 2000), although circular, did not lead to a lower eCO_2 per unit strength of new concrete. The CO_2-injected RCP meanwhile yielded the least eCO_2 per functional unit at all contents, but this also depended on the cement used to prepare the parent concrete. At 15% cement replacement, the eCO_2 of the CO_2-injected RCP was just lower than those of conventional SCMs, with the benefit derived at higher replacement levels. The RCP from low-carbon cement concrete (RCP-$C_{CEM\ II}$) had ~4% higher eCO_2/MPa than the OPC-derived RCP. This increased to ~8 and 25% at 30 and 50% RCP content, respectively. This implies that beneficiating low-carbon cement concrete through CO_2 injection led to a lower eCO_2 than the thermally treated RCP.

Existing low-carbon concrete roadmaps are unanimous on carbon reduction associated with cement clinker (CEMBUREAU 2019). Others go a step further to suggest that lower-strength concrete corresponds to lower eCO_2 with the substitution of OPC with GGBS and other SCM at higher strength grades requiring a higher percentage of total cement content and increased global carbon emissions (GCCA 2021). The above findings provide further insight into the non-linear relationship between concrete strength class, SCM replacement and eCO_2 with even higher eCO_2 at lower strength grades. This demonstrates the importance of assessing the eCO_2 of concrete simultaneously with the functional unit instead of a recipe-focused approach (GCCA 2021). The latter risk circularising resources without considering their carbon intensity. This study showed that CO_2 injection into OPC-derived concrete was most beneficial in reducing eCO_2 while maintaining target strength. This benefit is reduced to a comparable range as PFA when the RCP was composite cement concrete derived, but both lower than GGBS and up to 40% lower than thermal treatment RCP.

5. Conclusion

Meta-analysis of the literature on recycled RCP as SCM and regression analysis for predicting the compressive strength of concrete containing the non-beneficiated RCP from the mix design recipe has been presented. Following, the embodied CO_2 of the ground RCP with or without beneficiation through thermal treatment and CO_2 injection was calculated together with the effect of the cement from which the parent concrete was made – OPC or CEM II. Results from the meta-analysis enabled the eCO_2 of concrete containing the non-beneficiated RCP to be linked to the function unit, which has been compared with the different beneficiation techniques (thermal treatment and CO_2 injection), cement type in the parent concrete, GGBS and PFA at different replacement levels. The regression model showed that replacing more than 15% of the binder with the non-beneficiated RCP reduced the target strength by up to 40% and up to 70% when the content was increased to 50%.

The study also demonstrates secondary treatment of the ground RCP through CO_2 injection or thermal treatment while enhancing reactivity, thus improving strength have different carbon footprints. When considering the eCO_2 of concrete, the non-beneficiated RCP is comparable to GGBS, but depending on the content, the former may be 30 – 70% weaker than a corresponding GGBS concrete. Ultimately, the non-beneficiated RCP became more carbon intensive when the eCO_2 was considered alongside the functional unit. The carbon footprint of concrete containing thermally treated RCP was higher. Still, due to the enhanced reactivity, its eCO_2 per functional unit was an improvement of the non-beneficiated RCP and lower than the reference CEM I. This might be improved significantly if waste heat from industrial processes such as that generated in the cement or steel plant were used for the beneficiation. Even then, the eCO_2 becomes comparable to that of GGBS. Above 30% replacement, RCP beneficiated through CO_2-injection reduced eCO_2 significantly, reaching 10 –25% of that achievable through GGBS. This also assumes atmospheric or industrial flue gas as the source for treatment. This presents the most promising route to leverage end-of-life concrete in the circular economy for low-carbon production. This, however, will require significant investment in the infrastructure, such as carbon capture technology at the industrial scale, alongside supply chains that use this for CO_2 injection treatment of RCP. Findings from this study contribute towards a better understanding of the relationship between mix design, eCO_2 and performance of concrete incorporating these. It justifies that the carbonation treatment of RCP is a viable circularity option for achieving the net-zero aspirations of the concrete sector.

The main limitation of this study remains the variations in the data available in the scientific literature and particular RCP composition. As several variables could affect the published data, including sample preparation and sources of constituent materials from different studies, some influential parameters were inevitably excluded from the analysis. The population size also limited the results of this research. For multivariate regression analysis, smaller sample sizes reduce the statistical power of predictions. With growing interest in using RCP, the literature on the subject will evolve, which can be used to improve the model as part of further research efforts. Further research is also required to evaluate the supply chain, resource and infrastructural requirements of treatment and use of RCP at an industrial scale as part of the concrete decarbonisation agenda.

CRediT authorship contribution statement

Ciara Martin: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Resources. **Emmanuel Manu:** Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Funding acquisition, Resources, Supervision. **Pengkun Hou:** Conceptualization, Methodology, Formal analysis, Investigation, Writing – review & editing, Resources, Supervision. **Samuel Adu-Amankwah:** Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Funding acquisition, Resources, Supervision.

Declaration of Competing Interest

The authors have no competing interests to declare.

Data availability

Data will be made available on request.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2023.107197.

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