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USING FORCED CARBONATION OF RECYCLED CONCRETE FRACTIONS TO IMPROVE THE PERFORMANCE OF NEW CONCRETE

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ABSTRACT

Reuse of recycled concrete aggregate is often restricted to coarse aggregates, albeit with slight loss in performance. There is growing interest in valorisation techniques to improve the properties of coarse recycled concrete aggregate. One such treatment is accelerated carbonation treatment, which is also being considered for fine recycled aggregates and recycled concrete powder, so maximising reuse potential. This study has evaluated the effect of carbonation on all size fractions arising from crushing end-of-life concrete. End-of-life concrete was crushed in a jaw crusher and then subjected to forced accelerated carbonation. Following sieve analysis, each size fraction was characterised to determine its physical properties (particle size, attached mortar, absorption, and specific gravity). Concrete mixes containing all size fractions of recycled aggregates were subsequently prepared to assess the efficacy of up to 100% recycled concrete aggregate both pre-and post-carbonation treatment. Carbonation significantly improved the characteristics of all size fractions of recycled concrete aggregate, and consequently improved the performance of the resultant concrete prepared with it. Concrete prepared with 100% recycled coarse and fine aggregate outperformed a reference mix prepared with natural aggregates.

1. INTRODUCTION

Growing global consumption of cement and concrete over the past 50 years, has led to increased production of construction and demolition waste. Each year the EU produces 350 million tonnes of C&D waste. Of this, about 35% is end-of-life concrete (Joseph et al., 2022). Options for the treatment of such material are limited and much is either landfilled or downcycled in low-value applications such as backfilling (waste, 2022, May 11).

It is possible to reuse recycled concrete aggregates (RCA) in new concrete. But the presence of adhered mortar can cause problems. Firstly, this adhered mortar is more porous than natural aggregate, increasing water absorption and the LA Abrasion value (Kisku et al., 2017). The water absorption of RCA has been reported as 2.3–4.6 times more than that of natural aggregates (NAs) (Suryawanshi et al., 2015, Parthiban and Mohan, 2017). Secondly, RCA inclusion in concrete leads to the presence of three different interfacial transition zones, as shown in Figure 1. Consequently, the inclusion of RCA in concrete can diminish mechanical performance and raise concerns over long-term durability.

There has been considerable focus on managing the adhered mortar content and its properties, with many studies looking at either removing the adhered mortar (Tam et al., 2021) or strengthening this critical component (Gholampour et al., 2024). While many studies have been successful, there remain certain limitations associated with this approach, namely elevated energy consumption, substantial costs, and adverse effects on reinforced concrete structures, such as the occurrence of micro-cracks and the introduction of harmful ions (Shaban et al., 2019).

One of the most promising approaches is accelerated carbonation (or enhanced carbonation). This technique is a viable approach for treating RCAs, since it not only enhances their overall quality but also provides a means for permanent carbon dioxide (CO_2) sequestration (Monkman and Shao, 2010). Forced carbonation of RCA can significantly improve RCA performance, with a reduction in water absorption by 30% and 22% for 5-10 mm and 10-20 mm aggregate respectively, concurrent with an increase in apparent density of 4.8% and 3.2% respectively (Lu et al., 2019). This is due to the precipitation of crystalline calcium carbonate within cracks and pores on the RCA surface (Pu et al., 2021).

While the replacement of natural aggregate with RCA typically leads to a slight loss in performance, with reduced workability and loss in strength, this is not the case with carbonated RCA (cRCA). Indeed, in many instances, carbonated recycled aggregate concrete (cRAC) shows significantly improved performance compared to non-carbonated recycled aggregate concrete (RAC), for example, Lu et al. reported a 32% increase in compressive strength when using 100% cRCA compared to RCA (Lu et al., 2019). The reduced water absorption of cRCA compared to RCA leads to reduced water demand, which allows a lower effective water-to-cement ratio (w/c) in the recycled aggregate concrete, and so improves compressive strength (Wang et al., 2020). However, Luo et al. (Luo et al., 2018) observed a slight reduction in strength when using cRCA compared to NA, but still an improvement compared to using non-carbonated RCA. Hence, the implementation of accelerated carbonation for treating RCA not only leads to a substantial enhancement in the performance of recycled aggregate concrete but also shows potential for more widespread adoption of RCA in place of relying on natural aggregates.

To date, most studies into the use of RCA have focused on the reuse of coarse recycled aggregates. This is understandable given that they constitute the majority of the reclaimed material; and given the preclusion of recycled fine aggregates from many standards. Nevertheless, over the past twenty years, there has been a threefold increase in the demand for sand, resulting in significant environmental consequences (Gallagher and Peduzzi, 2019). Global consumption of sand for use in concrete has been estimated to range from about 3 billion tonnes (Gavriletea, 2017, Zhang et al., 2023) to about 10 billion tonnes per year and this figure has been predicted to increase by between about 50 and 100% (Sverdrup et al., 2017) by the middle of this century. Meanwhile, the recycling rate for sand has been estimated at only 5% (Sverdrup et al., 2017). This recycling rate is so low for several reasons, not least its potential impact on concrete performance. Aggregate water absorption capacity increases with decreasing aggregate size as the proportion of adhered mortar increases (Duan et al., 2020b, Frías et al., 2020, Moreno-Juez et al., 2020, Moreno-Juez et al., 2021). For fine recycled aggregates this poses a significant problem, with water absorption values over 10% being reported for some fine aggregates (Duan et al., 2020b, Frías et al., 2020, Moreno-Juez et al., 2020, Moreno-Juez et al., 2021, Nedunuri et al., 2021).

However, given the benefits of carbonation in enhancing CRCA performance, it has also been applied to FRCA (Shi et al., 2016, Zhan et al., 2019). Sub 2.5 mm FRCA has been carbonated and used effectively to produce recycled aggregate mortar (Zhan et al., 2019). Carbonation increased aggregate density, while water absorption and crushing value were both reduced. This led to improved mortar performance regarding drying shrinkage, flowability, and compressive strength. Pan et al. (Pan et al., 2017) pre-soaked FRCA in a calcium hydroxide solution prior to employing gas-solid carbonation to enhance its efficacy. The processed FRCA demonstrated improved crushing value, water absorption, and subsequent mortar strength. Li et al. (Li et al., 2018a) examined sub-5 mm aggregates obtained from crushed laboratory concrete. These were carbonated in pure, dry CO_2, leading to increased micro-hardness in both the aged interfacial transition zone (ITZ) and the aged mortar. Moreover, the compressive strength and modulus of the concrete produced using carbonated recycled concrete aggregate were markedly enhanced.

The final product arising from the crushing of end-of-life concrete is recycled concrete powder. This fraction, with sub-millimetres dimensions, primarily comprises hardened cement paste, with a fraction of pulverised aggregate. While there has been some interest in reusing this material directly (Duan et al., 2020b, Nedunuri et al., 2021, Ozcelikci et al., 2024, Singh and Kapoor, 2024), more recent work has focused on the valorisation of hardened cement paste via forced carbonation (Ma et al., 2024, Skocek et al., 2025, Skocek et al., 2020, Zajac et al., 2020a, Zajac et al., 2020b, Zhao et al., 2024). Here, the emphasis is not on consolidating the physical properties of the paste, as with the carbonation of the aggregates, but rather on exploiting the carbonation-induced chemical and mineralogical changes within the cement paste. Carbonation converts portlandite within the cement paste to calcium carbonate, often calcite. In addition, the aggressive conditions of forced carbonation leads to abstraction of calcium from the C-S-H within the cement paste to an amorphous aluminosilicate gel in addition to calcite (Herterich, 2017, Zajac et al., 2020a). The aluminosilicate gel is an effective pozzolan (Zajac et al., 2020b), while the calcium carbonate provides nucleation sites and contributes to later-age hydration (Zajac et al., 2020b). As such, the resultant carbonated cement paste can be used as a supplementary cementitious material, replacing a proportion of cement clinker within fresh cement (Herterich, 2017, Duan et al., 2020b, Moreno-Juez et al., 2020, Al-Janabi et al., 2023).

With growing quantities of C&DW being produced each year, and with large quantities of this being concrete, the successful processing of all size fractions of end-of-life concrete offers an opportunity to mitigate primary aggregate extraction and hence mitigate the degradation of natural ecosystems. Most prior research focused on enhancing the qualities and performance of a single component of recycled concrete products, and in certain instances, two of the three products derived from recycled concrete were combined.

However, this study aims to provide a pathway for the reuse of end-of-life concrete by examining the effect of incorporating all size fractions (coarse, fine, and powder) obtained from crushing end-of-life concrete. Furthermore, it examines the impact of forced carbonation on water capacity and specific gravity of these materials as a means of improving their performance and that of the resultant concrete compressive strength.

2. Materials and methodology

2.1 Recycled concrete products, crushing, and autogenous cleaning

The recycled aggregate used in this study was obtained by crushing laboratory concrete from an earlier study. The concrete, with a mix design as shown in Table 1 had been cured for 28 days in a fog room, whereupon it had a 52 MPa 28-day mean compressive cube strength. This was followed by ambient curing for a further 5 months.

Table 1: Parent concrete mix proportion.

Component	Quantity (kg/m^3)
CEM I 52.5 N	422
Water	177
Fine Aggregate*	754
Coarse (20 mm) Aggregate**	1024
w/c ratio	0.42

* quartz sand ** 20mm mixed quartzite crushed and gravel aggregate

The concrete was manually crushed and passed twice through a jaw crusher, set at 30 and then 15 mm. The first input was used to weaken the adhered mortar, and the second was used to achieve the required maximum RCA size. The recycled concrete aggregate was exposed to a rubbing friction action for 30 minutes, created by a vertical-axis concrete mixer, adopted from the autogenous cleaning technique (Pepe, 2015), but without subsequent washing.

2.2 Aggregate sieve analysis

The crushed and tumble-cleaned RCA was then subjected to a sieve analysis according to BS EN 933-1:2021 (Institution, 2021). Each size fraction was taken and weighed, with the recycled concrete powder being considered to be that material passing through a 150-micron sieve.

2.3 Cement and mortar contents

The RCA was then classified according to particle size. Coarse RCA comprised 5-10, 10-14, and 14-20 mm fractions, while the fine RCA was categorized as; 150-300 µm, 300-600 µm, 600 µm-1.18 mm, 1.18-2.36 mm, and 2.36-5 mm.

For each size fraction, the adhered mortar content was determined by acid washing. Coarse RCA was washed in 5 molar hydrochloric acid (l/s = 5) for 8 hours and then 16 hours in a fresh solution. The residual aggregate was washed in water over a 150-micron sieve and then sieved through a 5 mm sieve to differentiate between coarse aggregate and fine aggregate previously embedded within the attached mortar.

Fine RCA was also acid washed, immersing each size fraction in 5 molar hydrochloric acid for 48 hours and then washed over a 150-micron sieve.

Following the washing process, samples were oven-dried and the adhered mortar content was determined by calculating the proportional mass loss, as shown in equation 1 below:

$$Cement/mortar\ content = \left(\frac{M_{initial} - M_{washed}}{M_{initial}}\right) \times 100 \ldots\ldots\ldots\ldots\ldots\ldots 2.1$$

2.4 Water absorption and specific gravity

The method mentioned in BS EN 1097-6:2013 (EN, 2013) for determining the water absorption of natural coarse aggregate can also be applied to coarse recycled concrete aggregate, but modified with longer immersion to account for slower ingress into adhered mortar, and with drying at temperatures below 75 ºC (Zhou and Glasser, 2001). Consequently, samples were immersed in water for 72 hours then towel dried, (mass “B”). Samples were also immersed in water at 23ºC in a basket and then weighed in water (mass “C”). The wet samples were then dried in a controlled oven at 50 °C and 20% relative humidity for 48h to a constant weight. The samples were allowed to cool before being weighed (mass "A"). From these weights, the following properties were determined:

$$Relative\ density\ (Specific\ gravity\ (oven\ dry) = \frac{A}{B - C} \ldots\ldots\ldots\ldots\ldots\ldots 2.2$$

$$Absorption, \% = \frac{B - A}{A} \ldots\ldots\ldots\ldots\ldots\ldots 2.3$$

For the fine RCA, water absorption was determined by centrifugation (Li et al., 2018b), avoiding the variability seen when using the standard methods. The method attains the saturated-surface-dry state after centrifugation at 2,000 rpm for 10 minutes. At this point the water content can be considered as absorbed water. The centrifuged samples were then oven-dried at 40 ºC to constant weight. The relative density of every FCRA particle size was found by helium pycnometry.

2.5 Accelerated carbonation treatment

Forced carbonation of the recycled aggregate was conducted with aggregate samples spread across trays within a carbonation chamber. The temperature was fixed at 35 ºC (Li et al., 2017, Lu et al., 2019, Wang et al., 2020, Xuan et al., 2016), and the CO_2 concentration at 20% (Cui et al., 2015, Fang et al., 2017). This concentration is approximately the concentration of flue gas from cement kilns (Simoni et al., 2022). The relative humidity was fixed at 65% via the use of a saturated sodium bromide solution (Greenspan, 1977). Samples were removed periodically and tested for the extent of carbonation. By 8 days, the adhered mortar was found to have been completely carbonated, and so aggregates were exposed for this duration.

The recycled concrete powder meanwhile was exposed under identical conditions, but was found to have reached a steady extent of carbonation by 2 days, so was exposed for just this duration. It is known that CO_2 concentrations above 3% can change the nature of cement carbonation, leading to C-S-H decalcification, hence the choice of such aggressive conditions (Castellote et al., 2009).

2.6 Mineralogical tests for RCP and RCA:

Thermal analysis (TG-DTG) was used to determine the nature of the recycled concrete powder (RCP) and adhered cement mortar, including determination of bound water contents, $Ca(OH)_2$ and $CaCO_3$ contents and to aid, via the decomposition onset temperature, both identification of the carbonate polymorph and whether carbonate originated from carbonation of portlandite or

C-S-H (Thiery et al., 2007, Herterich, 2017), Testing was conducted under nitrogen on a Thermo Plus EVO2 TGA, at a heating rate of 10 °C/min from 20 °C to 1000 °C.

Qualitative analysis was also performed by Fourier Transform Infrared FTIR spectroscopy, using a PerkinElmer – Spectrum Two. Spectra were recorded accumulating 16 spectra over the range 400-4000 cm^{-1} with 4 cm^{-1} resolution.

Surface morphology and confirmation of calcium carbonate precipitation following carbonation was followed by scanning electron microscopy (SEM) in secondary electron mode (SE), with a Zeiss Evo 15 scanning electron microscope operating at 20kV accelerating voltage. Polished cross-sections were also examined from small fragments of concrete prepared with RCA. Samples were fixed in resin before aggregates, pastes and ITZs were exposed by polishing, before viewing using a backscattered electron detector.

2.7 Concrete mixes

The RCA, pre- and post-carbonation was incorporated into various concrete mixes using a CEM I 52.5N binder. The control mix was prepared with a w/b ratio of 0.58 and an approximately 3:1 mixture of 20 mm and 10 mm quartzite gravel aggregate (from the same source as used to prepare the RCA). Various mixes were then prepared incorporating up to 100% recycled aggregate (both carbonated and non-carbonated), simultaneously replacing both coarse and fine aggregate.

In addition, with the use of RCA widely reported reduce workability (Quattrone et al., 2016), mixes prepared with 100% RCA were then also prepared with the addition of 0.2 or 0.4% superplasticizer by weight of binder, sufficient to bring the slump to that of the control mix.

Finally, to investigate the potential for maximizing the utilization of all size fractions, a further four mixes were prepared. These contained 100% RCA (carbonated and non-carbonated) but with 10% of the CEM I binder replaced with recycled concrete powder. This powder was either as-received (RCP) – which is non-reactive with a strength activity index of 0.6 [38] or carbonated (cRCP) – which is known to show pozzolanic behaviour and have a strength activity index of 0.86 [38]. The choice of 10% replacement was based on preliminary studies and on published work (Oliveira et al., 2023). Table 2 gives the quantities of components used in each concrete mixture.

Table 2: Concrete mixes proportions.

Mix name	W*	Binder		F*		G (10 mm)*		G (20 mm)*	
		C*	RP*	NA*	RA*	NA*	RA*	NA*	RA*
Control mix	174	300	0	630	0	310	0	940	0
33% RAC	174	300	0	422.1	207.9	207.7	102.3	629.8	310.2
67% RAC	174	300	0	207.9	422.1	102.3	207.7	310.2	629.8
RAC	174	300	0	0	630	0	310	0	940
33% cRAC	174	300	0	422.1	207.9	207.7	102.3	629.8	310.2
67% cRAC	174	300	0	207.9	422.1	102.3	207.7	310.2	629.8
cRAC	174	300	0	0	630	0	310	0	940
SP-RAC	174	300	0	0	630	0	310	0	940
SP-cRAC	174	300	0	0	630	0	310	0	940
RCP-SP-RAC	174	270	30	0	630	0	310	0	940
RCP-SP cRAC	174	270	30	0	630	0	310	0	940
cRCP-SP-RAC	174	270	30	0	630	0	310	0	940
cRCP-SP cRAC	174	270	30	0	630	0	310	0	940

W*: water content in kg, C*: cement in kg, RP*: Recycled concrete powder in kg, F*: Sand in kg, G(10mm)*: Gravel with max. size 10mm in kg, G(10-20mm)*: Gravel with size 10-20 mm in kg, NA*: Natural aggregate, RA*: Recycled aggregate, RAC: Recycled Aggregate Concrete, cRAC: Carbonated-Recycled Aggregate Concrete, SP: Superplasticizer (0.4% bwb when using RCA and 0.2% bwb when using cRCA), RCP: recycled concrete powder, cRCP: carbonated recycled concrete powder.

To decrease the negative impact of the RCA's high water absorption, mixes were prepared by blending the dry ingredients for 30 seconds before introducing half of the total water and mixing for 1 minute. After this the remaining water was added and mixed for 1 ½ minutes (Tam et al., 2005).

Workability was determined using the slump test; according to BS EN 12350-2 (2019) (12350-2:, 2019). Compressive strength was measured in triplicate at 7 and 28 days using 100 mm cubes, according to BS EN 12390:2009 (12390-3, 2019). Density was also determined in triplicate.

3 Results and discussion

3.1 Efficiency of Carbonation

3.1.1 Recycled Concrete Aggregate

Figure 2 shows cross-sections through 20 mm RCA before and after carbonation, after spraying with phenolphthalein. The deep purple colour indicates the presence of alkaline cement paste stuck to the surface of the original aggregate particles. After carbonation there was very little purple colouration, indicating carbonation of the adhered mortar.

Calcium carbonate formation was confirmed by viewing samples by scanning electron microscopy (SEM). Images were captured from both carbonated and non-carbonated aggregates. The non-carbonated sample (Figure 3a) showed a surface covered with nondescript hydrated cement paste. However, the carbonated sample (Figure 3b) was covered in angular calcium carbonate crystals with some nondescript hydrated cement paste still visible.

To examine carbonation-induced changes in the adhered mortar, small quantities of recycled concrete powder before and after carbonation were characterized by FTIR spectroscopy and thermal analysis.

The FTIR spectra are shown in Figure 4. The spectrum from non-carbonated RCP (RCP) exhibited a significant silicate stretching band at ~970 cm-1, typical of C-S-H, accompanied by a distinct shoulder at ~1060 cm^{-1} due to quartz (Horgnies et al., 2013). This tallies with the mortar comprising hardened cement paste and quartz sand. The spectrum also showed a moderately-sized asymmetrical carbonate stretching band from ~1400 to 1500 cm^{-1}, characteristic of calcite. This is due to the calcite present in the original anhydrous cement, plus potential slight carbonation during processing. However, carbonation was incomplete, as evidenced by the weak OH stretching band at ~3620 cm^{-1} due to portlandite (Horgnies et al., 2013). This agrees with the pink colouration seen when spraying non-carbonated RCP with phenolphthalein.

Forced carbonation of RCP (RCPc) led to the disappearance of the OH stretching band, with a corresponding increase in the carbonate band. There was also growth in the out-of-plane band (v^2) band at 875 cm^{-1}, with the appearance of a second such band at 858 cm^{-1}. This suggests the presence of calcite together with another carbonate species, possibly vaterite (Horgnies et al., 2013). Significantly, there was also a significant shift in the silicate stretching band to a higher wavenumber, with the maximum shifting to 1034 cm^{-1} and increased intensity at ~1150 cm^{-1}. These changes indicate the formation of highly polymerized silicate phases (Herterich et al., 2014), corresponding to C-S-H decalcification, plus residual quartz sand.

Meanwhile, Figure 5 shows thermal analysis data from the same samples. The non-carbonated RCP sample contained about 5% portlandite and just under 4% calcium carbonate. The calcium carbonate decomposed at 650ºC, identifying it as calcite (Herterich et al., 2014). Carbonation led to the consumption of about 70% of the portlandite. However, there was more than a 10% increase in carbonate content. This supports the FTIR data suggesting carbonation by abstraction of calcium from the C-S-H. This was also supported by the significant reduction in the carbonate decomposition onset temperature to ~580ºC, as observed previously upon carbonation of immature high slag content blends (Herterich et al., 2014).

3.2 Recycled Concrete Aggregate Grading & Properties

Table 3 presents the results of the sieve analysis. It also shows adhered mortar contents for each size fraction, plus how much of that was cement paste, i.e. acid soluble, and how much was finer aggregate embedded within a cement matrix. The percentage mass increase upon carbonation was theoretical mass changed calculated based on the extent of carbonation of portlandite and C-S-H in the cement paste determined by thermal analysis. This was then used to determine CO2 uptake by multiplying by the adhered cement paste content, and subsequently to determine a theoretical particle size distribution accounting for the change in mass upon carbonation.

Table 3: Size distribution of RCA, plus adhered cement and mortar contents. * Adhered aggregate is defined by the amount of coarse aggregate passing given sieve size but retained on a 5mm sieve, and by the amount of fine aggregate retained on a 150-micron sieve

Size range (mm)	Amount (%)	Adhered mortar content (%)	Adhered cement paste (%)	Adhered aggregate (%)	Proportion of total cement paste adhered on given size fraction (%)	% mass increase upon paste carbonation	CO_2 uptake into size fraction (% of total concrete mass)	Post-carbonation amount (%)
14-20	39.80	20.67	11.25	9.42	21.98	1.369	0.545	39.37
10-14	24.03	24.41	12.61	11.8	14.88	1.535	0.369	23.81

5-10	15.26	33.9	24.13	9.77	18.07	2.937	0.448	15.32
2.36-5	6.19	34.09	28.01	6.08	8.52	3.409	0.211	6.25
1.18-2.36	3.39	36.98	32.8	4.18	5.46	3.992	0.135	3.44
0.6-1.18	2.83	37.44	33.84	3.6	4.71	4.118	0.117	2.88
0.3-0.6	2.54	43.91	36.01	7.9	4.49	4.382	0.111	2.59
0.15-0.3	0.51	52.53	38.1	14.43	0.95	4.637	0.024	0.52
<0.15	5.45	-	78.33	-	20.95	9.533	0.519	5.82

Recycled coarse aggregate comprises almost 80% of the total mass, a significant increase from the 43% coarse aggregate in the original concrete, and due to almost half of the cement paste being found adhered to the 5-20 mm size fractions. Despite this, there is a gradual increase in adhered mortar content with decreasing particle size. It is this adhered mortar which can have a detrimental impact on aggregate properties (Etxeberria et al., 2007) and precludes the use of fine recycled concrete aggregates in new concrete within many standards.

That cement paste is predominantly found within the coarse aggregate and sub-150 micron recycled concrete powder fraction suggests that these fractions show the greatest potential for carbon we only have compressive strength data recorded from each sample in triplicate. As such, in-depth statistical analysis would not be very robust. Instead, we have revised our discussion of the results playing down the slight strength increases, and instead focussing on how our results differ from the oft-reported loss in strength seen when replacing natural aggregates with recycled ones. (line 477-481)
ration upon forced carbonation.

While adhered mortar contents themselves are not a criterion for consideration in current standards, adhered mortar contributes to RCA's increased water absorption (which is a criterion), and so lower adhered mortar contents are what enable reuse of coarse RCA in new concrete. So, despite not being an explicit consideration in many standards, both de Juan et al. (De Juan and Gutiérrez, 2009) and the Building Contractors Society of Japan [88], recommend that RCA can be reused when the proportion of adhered mortar is less than 40% by volume. While our data are presented as percentage by weight, 40 vol% is about 35 wt% (assuming densities of 2.35 and 2.65 g/cm^3 for cement paste and quartz respectively). As such, all of the size fractions larger than 2.36 mm meet this criterion.

Also of interest is the distribution between adhered cement paste and aggregate embedded within that paste. Almost half of the adhered mortar on the 14 and 20 mm RCA comprised fine aggregate particles embedded within the cement paste, while for the fine RCA the figure was considerably lower; between 10 and 30%.

Based on the sieve analysis, plus the adhered cement and mortar contents, it was possible to estimate the flow of components upon crushing, tumble-cleaning and subsequent forced carbonation (Figure 6). The figure shows that, despite the coarse aggregate having the lowest adhered mortar content, the overwhelming coarse RCA content meant that this fraction comprised the largest source of hardened cement paste. This provides justification for considering coarse RCA as a suitable medium for valorisation through forced carbonation of paste within recycled concrete.

3.3 Water absorption and specific gravity

Table 4 presents the specific gravity and water absorption values of the natural aggregate plus the RCA both before and after carbonation. The specific gravity of the natural aggregates showed little dependence on size, being approximately 2560 kg/m^3 for coarse aggregate and ~2650 kg/m^3 for fine aggregate, reflecting the slight changes in mineralogy. The density of the recycled aggregate was less than that of the equivalently-sized natural aggregate due to the aforementioned adhered mortar. This can perhaps best be seen in Figure 7 and Figure 8, where the distinction between coarse and fine aggregate is clear. Figure 7 shows how the deviation between natural aggregate and coarse RCA increased with decreasing aggregate size as the proportion of adhered mortar increased, and this dependence on adhered mortar content is more clearly demonstrated in Figure 8.

Carbonation led to densification of the adhered mortar and so an increase in the relative density of the RCA. De Juan et al. (De Juan and Gutiérrez, 2009) examined the suitability of recycled aggregates in new concrete, determining a minimum specific gravity for RCA of 2.160 for adequate concrete performance. All of the size fractions in this study met this limit.

Table 4: Water absorption and specific gravity of each size fraction of each aggregate.

	Relative density		Water absorption (%)	
		Recycled Aggregate		Recycled Aggregate

Aggregate size (mm)	Natural Aggregate	before carbonation	after carbonation	Natural Aggregate	before carbonation	after carbonation
14-20	2.565	2.471	2.500	0.78	2.63	2.10
10-14	2.575	2.443	2.472	1.24	3.27	2.55
5-10	2.561	2.371	2.392	2.13	4.63	3.06
2.36-5	2.690	2.485	2.492	2.96	6.99	5.24
1.18-2.36	2.712	2.475	2.488	2.67	8.33	6.04
0.6-1.18	2.672	2.468	2.476	2.92	9.35	7.56
0.3-0.6	2.649	2.459	2.468	2.37	10.02	8.15
0.15-0.3	2.650	2.446	2.467	3.12	11.56	10.23

Water absorption of natural aggregate varied little with size, with everything over 300 microns diameter meeting the 3% water absorption limit defined by BS EN 1097-6:2013 (EN, 2013). However, the adhered mortar significantly increased RCA water absorption, with an increase seen with decreasing particle size (Zhou and Glasser, 2001) such that only the 20 mm coarse RCA fell under the 3% limit in BS EN 1097-6:2013 (EN, 2013). However, all coarse fractions met the 8% limit identified by both de Juan et al (De Juan and Gutiérrez, 2009) and the Building Contractors Society of Japan (Hansen, 1986) as giving acceptable performance. However, all of the fine fractions exceeded the 3% limit, and only the 2.36-5 mm fraction met the 8% limit.

Carbonation led to a reduction in RCA water absorption for all size fractions, but the 10-14 mm RCA was the only size fraction brought within the 3% limit stipulated in BS EN 1097-6:2013. But all bar the 600 microns size fraction met the aforementioned 8% limit. Carbonation is known to reduce the porosity of concrete (Dubina et al., 2013), with the conversion of portlandite to calcite or other calcium carbonate polymorphs being expansive (Dubina et al., 2013) and therefore filling pores. It has also been suggested that the precipitation of calcium carbonate will fill pores and fissures within recycled aggregates, thus lowering water absorption (Pu et al., 2021). In most field-exposed samples, or any conditions where the relative humidity exceeds 80%, calcite is the predominant carbonate polymorph (Dubina et al., 2013). However, at lower relative humidities, aragonite and vaterite can form (Peter et al., 2008). Carbonation of the hardened cement paste will also form an aluminosilicate gel (Bertos et al., 2004, Phung et al., 2015). This formation of this aluminosilicate gel is associated with a slight volume reduction, but this is far less than the expansion associated with calcium carbonate formation. Thus, the overall volume change is positive, with a positive impact on RCA properties.

While previous studies have examined the relationship between aggregate properties and adhered mortar content, it is actually the cement paste within the mortar that contributes to the increased water absorption. With this in mind, the relationship between water absorption and the amount of adhered cement paste on each size fraction is shown in Figure 9: Water absorption versus the square root of the adhered cement content for RCA and carbonated RCA. Note that the dashed grey lines represent the 3% and 8% limits recommended by BS EN 1097-6:2013 and de Juan [29] respectively.Figure 9. For the coarse and the fine RCA there is an approximately linear relationship between the water absorption and the square root of the adhered paste content, suggesting that the water absorption is determined by the thickness of the paste layer coating the RCA.

3.4 Fresh and hardened concrete

Table 5 shows the slump and hardened density, plus 7- and 28-day compressive strengths for each of the mixes studied here, while Figure 10 shows the compressive strength data graphically.

Table 5: Compressive strength and slump test results.

Mix name	Slump (cm)	Hardened Density (kg/m^3)	7-days Compressive strength (MPa)	7-days Standard deviation (%)	28-days Compressive strength (MPa)	28-days Standard deviation (%)
Control mix	14	2420	32.47	1.60	43.66	2.06
33% RAC	14	2345	31.93	1.82	43.15	2.15
67% RAC	11	2320	32.96	1.53	43.95	2.18
RAC	8.5	2295	34.79	1.49	44.84	2.20
33% cRAC	13.5	2360	33.09	1.64	43.55	2.18
67% cRAC	13	2330	33.00	1.61	44.01	2.13
cRAC	12.5	2264	34.14	1.52	48.77	2.10
SP-RAC	14	2242	37.54	0.50	49.20	0.89
SP-cRAC	17	2280	38.63	0.74	50.91	0.81
RCP-SP-RAC	14	2293	30.06	1.50	40.08	1.26
RCP-SP cRAC	14.5	2284	33.39	1.35	45.75	1.08
cRCP-SP-RAC	14.5	2264	31.80	1.16	44.17	1.16
cRCP-SP cRAC	15.5	2298	39.35	1.21	50.46	0.98

Moderate replacement (33%) of natural aggregates with non-carbonated RCA led to minimal change in slump. But increasing replacement led to a significant reduction in slump, namely dropping from 14 cm to 8.5 cm upon 100% replacement. This is due to the rough nature of the non-carbonated RCA compared to the gravel natural aggregate, plus water uptake into the adhered mortar. However, the reduced water absorption capacity of carbonated RCA, plus the possibility of fissures in the aggregates being filled with calcium carbonate, meant that slump only dropped from 14 cm to 12 cm when replacing all of the aggregate with carbonated RCA.

The high water absorption values of RCA have previously been reported to have a notable impact on workability, while carbonated RCA is reportedly affected to a lesser extent (Lu et al., 2019, Luo et al., 2018, Wang et al., 2020). This was repeated here, despite over 60% of the non-carbonated and nearly 40% of the carbonated RCA exceeding the 3% water absorption threshold specified in the British Standard (EN, 2013). However, over 90% of the non-carbonated and nearly 97% of the carbonated RCA met the 8% water absorption limit considered acceptable according to the literature (De Juan and Gutiérrez, 2009, Japan, 1981), suggesting that the British Standard might be somewhat conservative.

The addition of a superplasticizer improved performance. The non-carbonated RCA mix required 0.4% PCE superplasticizer to bring the workability to that of the control mix. Meanwhile, the addition of just 0.2% superplasticizer was sufficient to regain workability when using carbonated RCA.

The hardened densities of all the recycled aggregate concrete mixes were less than the reference mix. In general, all RACs are typically classified as normal concrete due to their densities above 2200 kg/m^3 (Neville and Brooks, 1987).

Replacing natural aggregate with RCA led to a slight increase in strength with increasing RCA content. This contradicts many of the findings in the literature, whereby the various interfacial transition zones between the original aggregate the adhered mortar and the new cement paste are known to be points of weakness resulting in lower strengths (Lu et al., 2019, Luo et al., 2018, Wang et al., 2020, Duan et al., 2020a, Padmini et al., 2009). In this study there are a number of factors which may explain this difference in behaviour. Firstly, the RCA was not saturated surface dry, so may have absorbed some water during mixing. Secondly, the original aggregate was a rounded quartzite gravel, while the RCA was more angular, and the strength

of recycled aggregate concrete is boosted by the rough surface of the attached mortar , which strengthens the bond between the aggregate and the fresh paste. However, the carbonated RCA gave a higher strength than the non-carbonated RCA. Since the carbonated material had a lower water absorption capacity than the non-carbonated material, the water uptake during mixing cannot explain this. We suppose that carbonation improved the strength of the adhered mortar, thus improving the integrity of the interfacial transition zones. Indeed, when viewing polished cross-sections in the electron microscope, the effects of aggregate carbonation could be seen. The concrete prepared with non-carbonated RCA showed a typical interfacial transition zone. The micrograph in
Figure 11a shows cement paste between three large aggregate particles, with the slightly darker regions surrounding the aggregates being the weak, interfacial transition zone. Meanwhile,
Figure 11b shows a cross-section of concrete prepared with carbonated RCA. Here the aggregates are coated with an intact, thin, densified layer of carbonated adhered mortar. Additionally, the reactivity of carbonated adhered mortar may have provided additional pozzolanic material to the mix, improving performance (Duan et al., 2020b, Li et al., 2018a, Moreno-Juez et al., 2020, Nedunuri et al., 2021, Ozcelikci et al., 2024, Singh and Kapoor, 2024, Zhang et al., 2023).

The improved performance of carbonated RCA was seen even more clearly when a superplasticizer was used, with the mix with 100% replacement giving a 28-day strength of nearly 51 MPa, compared to 43.7 MPa for the reference mix. Again, this is a combination of the more angular RCA and potential reactivity of the adhered mortar with the fresh cement paste.

The use of recycled concrete powder was investigated to attempt maximum replacement with recycled material in fresh concrete. Replacement of cement with recycled concrete powder led to a reduction in compressive strength of about 3 MPa when using non-carbonated or carbonated RCA. This is in agreement with what has been reported previously (Chen et al., 2021, Topič et al., 2017).

However, carbonated RCP performed better than non-carbonated RCP, such that the strength was comparable to mixes prepared with CEM I. This reflects the reactivity of the carbonated RCP, as has been reported previously (Mehdizadeh et al., 2021, Al-Janabi et al., 2023), and attributed to the combined reactivity of small quantities of calcium carbonate plus an aluminosilicate gel.

4 Conclusions

This study has examined how forced carbonation of recycled concrete aggregates can improve their performance. Key in the production of high-quality carbonated RCA was the production of good-quality RCA by crushing end-of-life concrete followed by tumbling it in a horizontal pan mixer. The process produced a more angular aggregate, but all materials complied with size limits.

The tumbling action removed much of the adhered mortar from the RCA, but some remained on all fractions, with the amounts increasing with decreasing particle size. However, over two-thirds of the cement paste was stuck to the coarse aggregate. Thus, this fraction shows potential for total CO_2 uptake.

Forced carbonation of RCA improves the properties of all size fractions, from coarse aggregate to RCP. Calcite precipitation and the subsequent densification of the mortar significantly reduces water absorption capacity of all size fractions. It is known that forced carbonation of RCP leads to the production of a reactive supplementary cementitious material. Similarly, carbonation of recycled aggregates leads to the adhered mortar comprising a reactive mixture of precipitated calcium carbonate and an aluminosilicate gel. This improves the interfacial transition zone, known to be a point of weakness in recycled aggregate concrete. As such, the use of carbonated RCA can show significant improvement in concrete performance over non-carbonated RCA and a slight improvement over natural aggregate. This improved performance could also be due to the more angular nature of the RCA and water absorption by the RCA.

While there is growing interest in carbonated recycled cement powder to produce a novel supplementary material, the presence of about two-thirds of the original cement paste stuck to the coarse aggregate means that forced carbonation should be considered as a viable means of valorisation, improving its performance in the process and aid any transition to a more circular approach to concrete.

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Figure Captions

Figure 1: Schematic of natural aggregate (NA) and recycled concrete aggregate (RCA). In RCA there are three interfacial transition zones (ITZ1, ITZ2 and ITZ3), while RA-OM and RA-VG refer to recycled aggregate original mortar and virgin aggregate respectively. (Zhang and Zhao, 2015)

Figure 2: RCA (left) and carbonated RCA (right) after spraying with phenolphthalein.

Figure 3: SEM micrographs from non-carbonated and carbonated RCA (Fields of view: 45 μm).

Figure 4: FTIR spectra for nnon-carbonated RCP (black) and carbonated RCP (cRCP red).

Figure 5: Thermal gravimetric analysis traces from non-carbonated RCP (black) and carbonated RCP (cRCP, red).

Figure 6: Sankey diagram showing the partitioning of coarse aggregate, fine aggregate, and cement paste upon crushing, tumble-cleaning, and forced carbonation of end-of-life concrete.

Figure 7: Plot of specific gravity versus aggregate size for natural aggregate, recycled aggregate and carbonated recycled aggregate.

Figure 8: Plot of specific gravity versus adhered mortar content for recycled aggregate and carbonated recycled aggregate. (Note that black data points indicate typical specific gravities for natural aggregates.

Figure 9: Water absorption versus the square root of the adhered cement content for RCA and carbonated RCA. Note that the dashed grey lines represent the 3% and 8% limits recommended by BS EN 1097-6:2013 and de Juan [29] respectively.

Figure 10: Compressive strength test results.

Figure 11: SEM micrographs of polished concrete cross-sections prepared with non-carbonated and carbonated RCA. (Fields of view 280 μm). The carbonated adhered mortar is indicated by the white arrows.

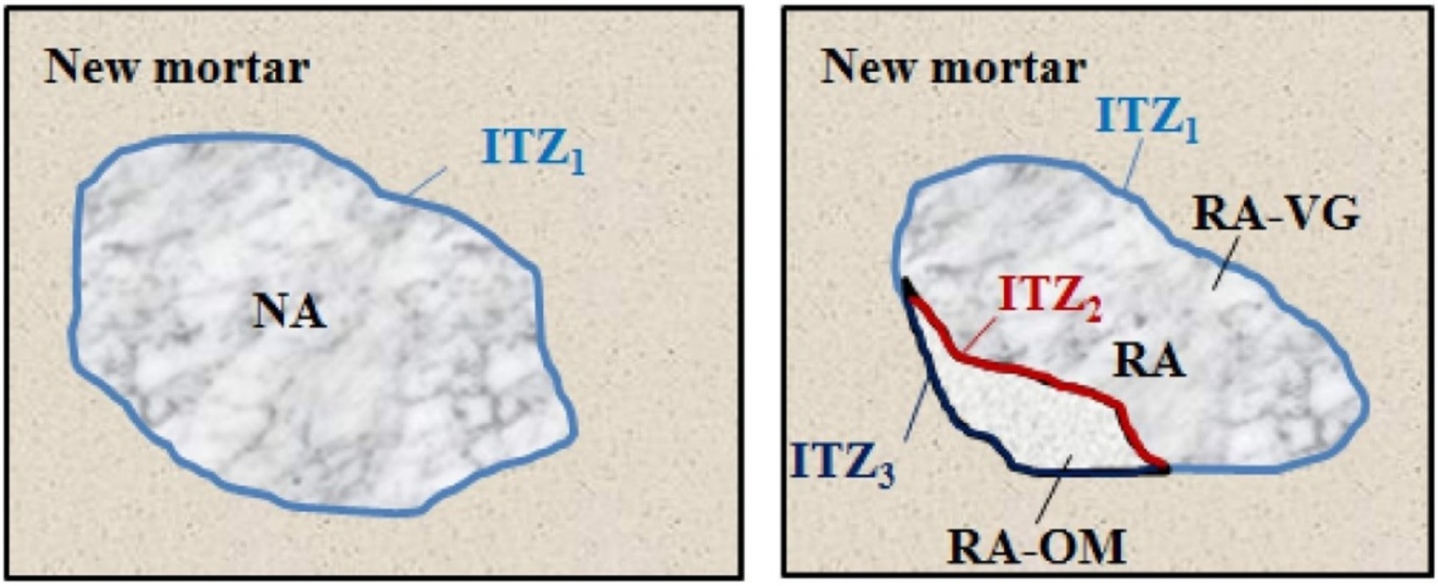
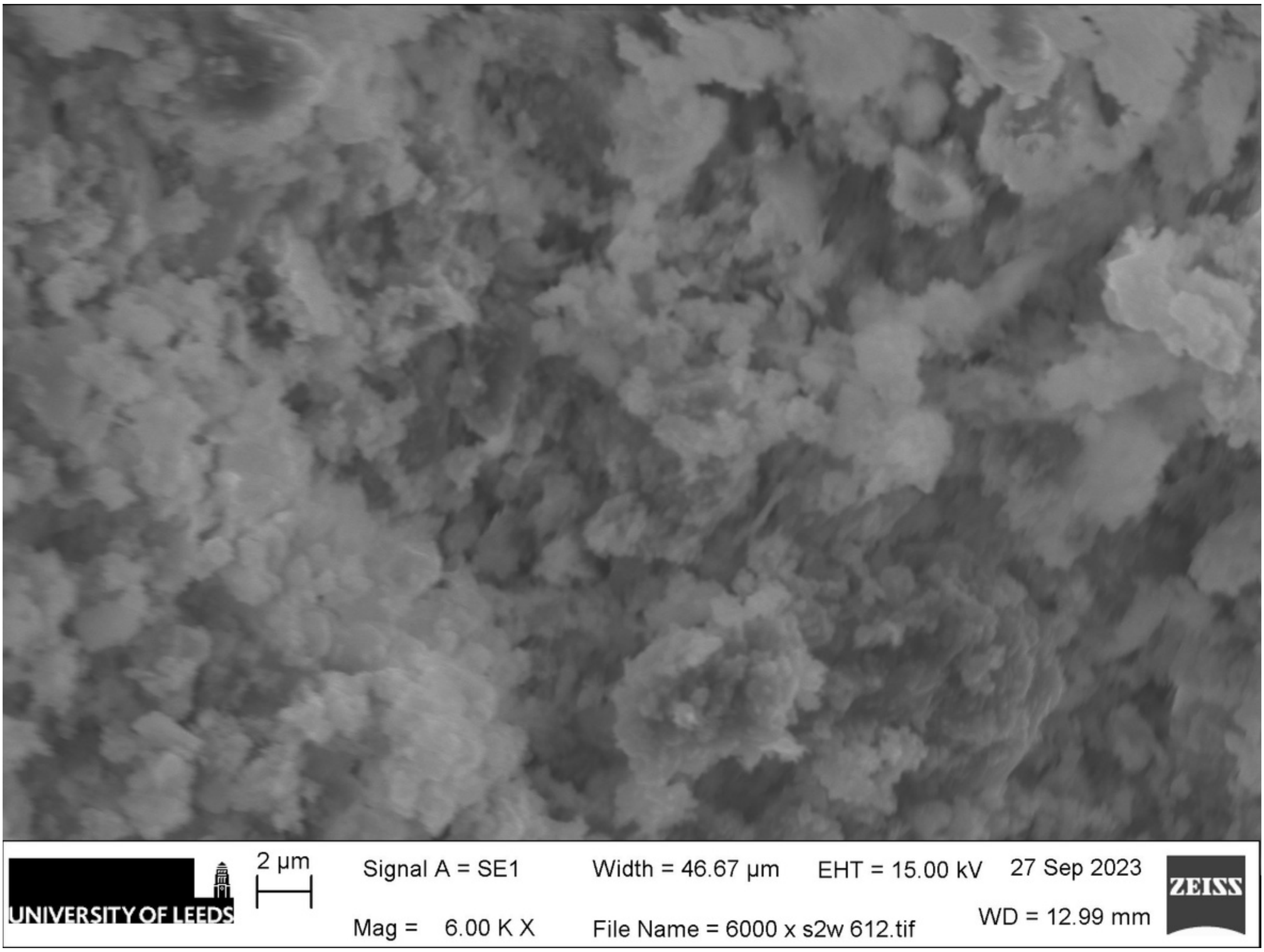

New mortar
ITZ$_1$
NA
New mortar
ITZ$_1$
RA-VG
ITZ$_2$
RA
ITZ$_3$
RA-OM

Figure-2.jpg Figure


UNIVERSITY OF LEEDS
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Mag = 6.00 K X
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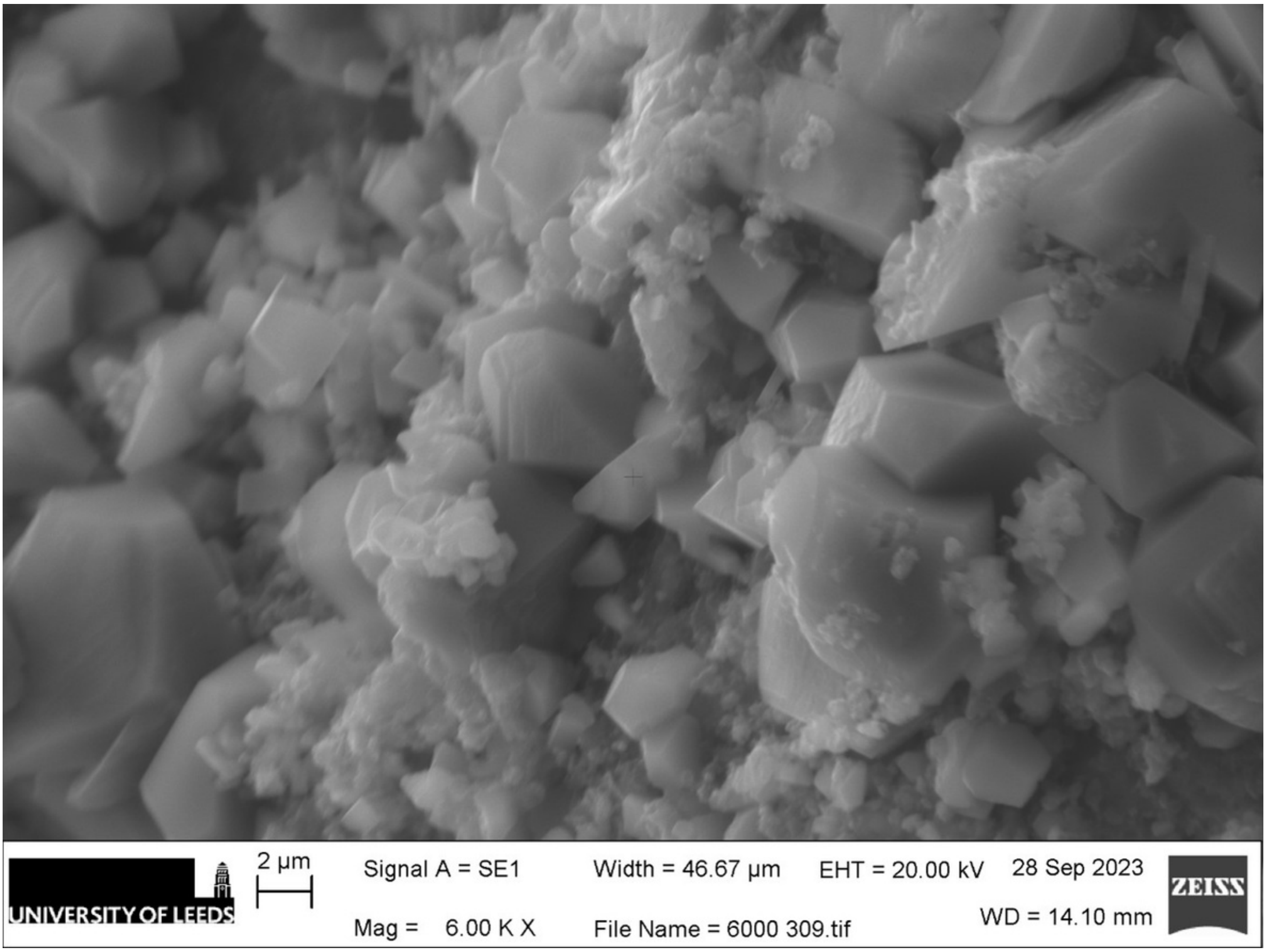
EHT = 15.00 kV
27 Sep 2023
WD = 12.99 mm
ZEISS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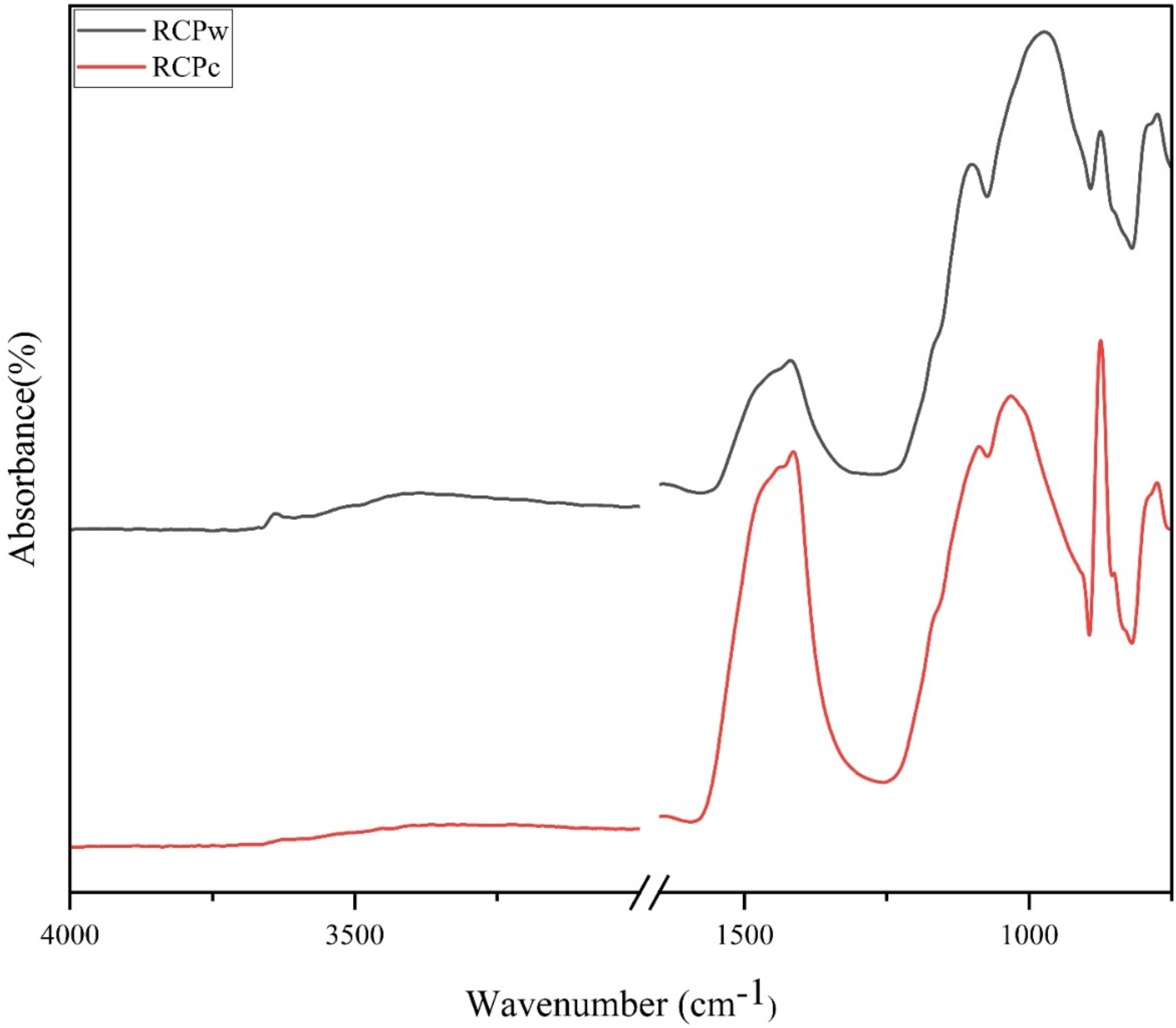
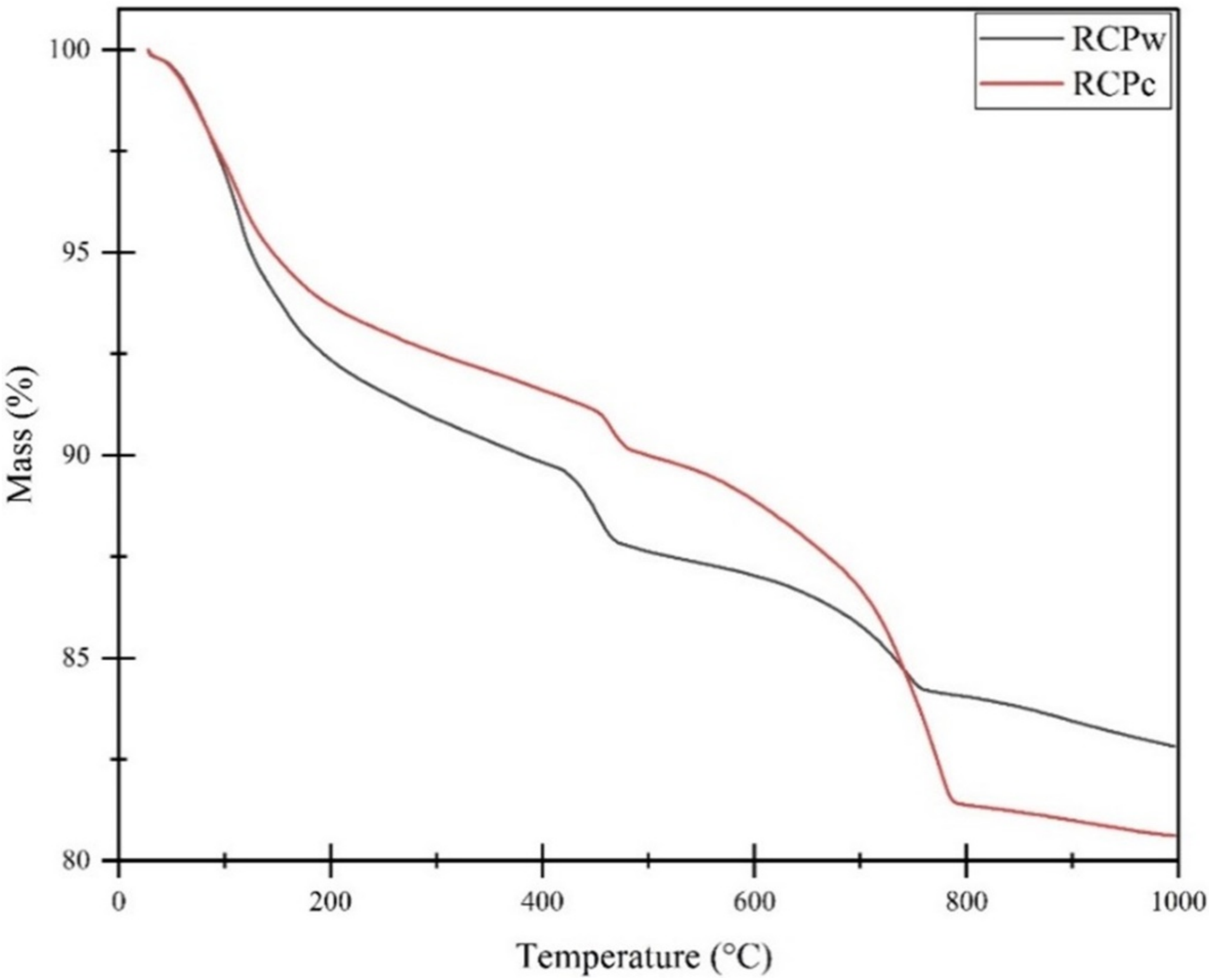

RCPw
RCPc
Absorbance(%)
4000
3500
1500
1000
Wavenumber (cm-1)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RCPc
Mass (%)
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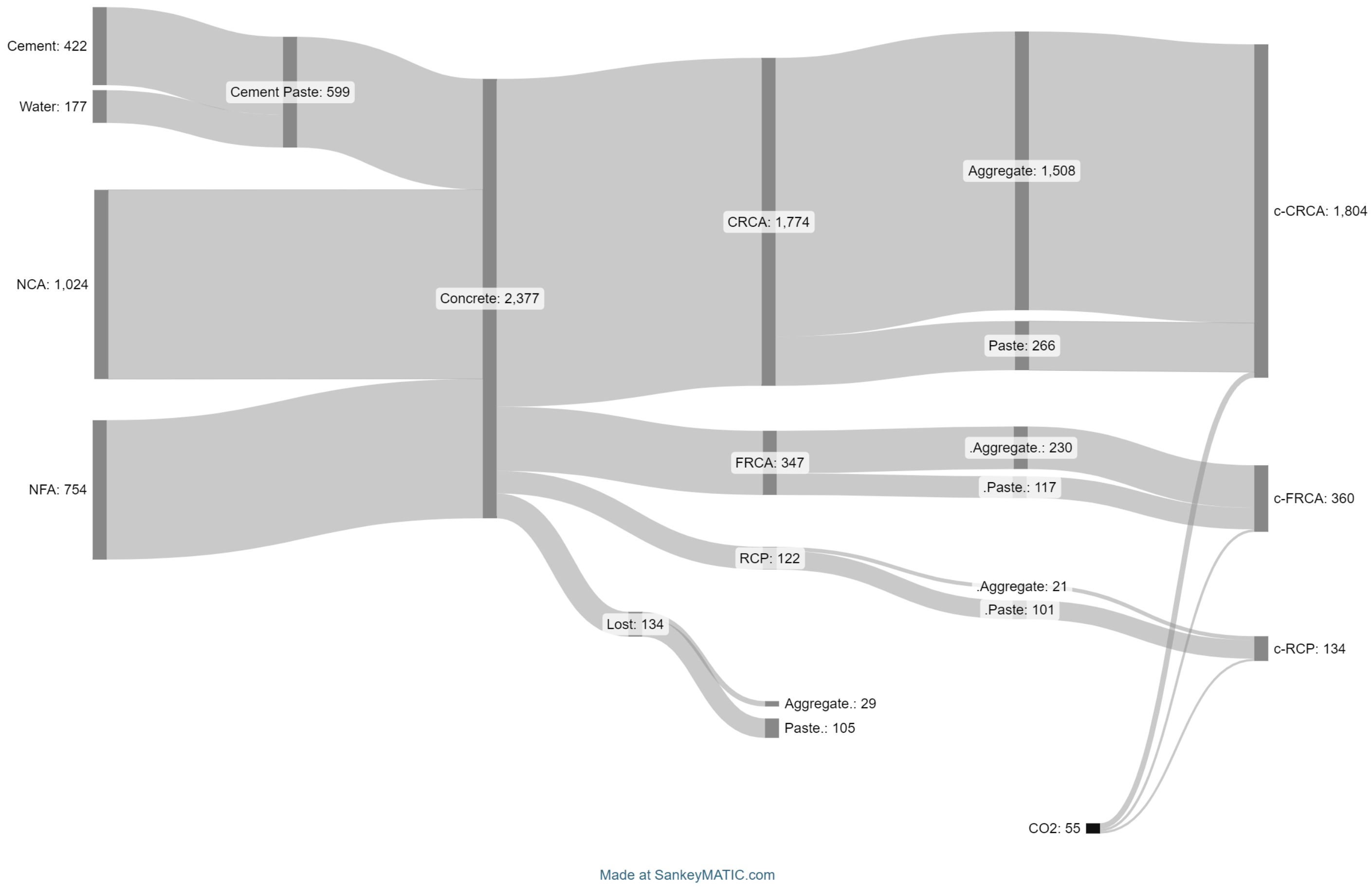
95
90
85
80
0
200
400
600
800
1000
Temperature (°C)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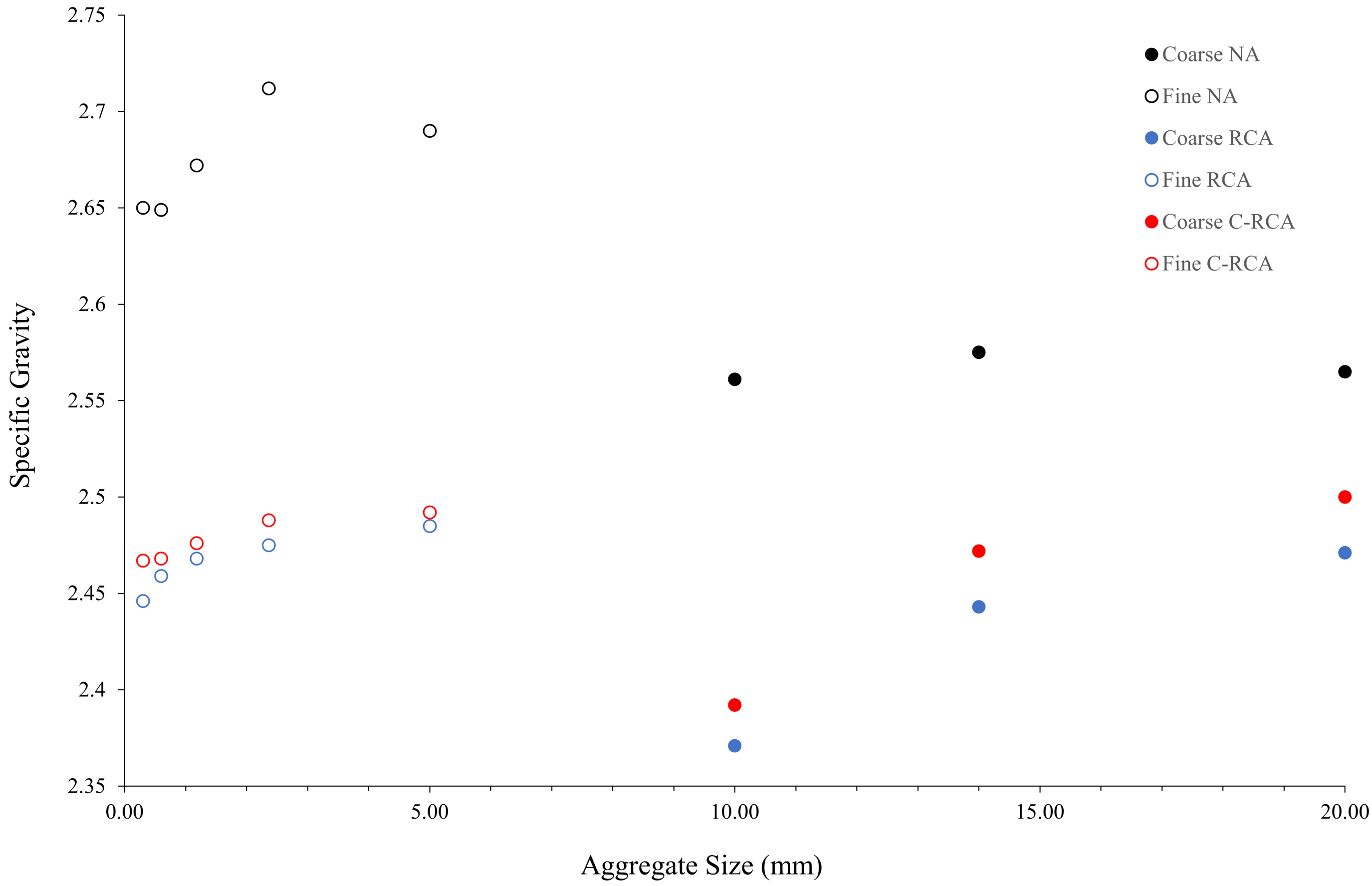
Water: 177
Cement Paste: 599
NCA: 1,024
NFA: 754
Concrete: 2,377
CRCA: 1,774
FRCA: 347
RCP: 122
Lost: 134
Aggregate: 1,508
Paste: 266
.Aggregate.: 230
.Paste.: 117
.Aggregate: 21
.Paste: 101
Aggregate.: 29
Paste.: 105
c-CRCA: 1,804
c-FRCA: 360
c-RCP: 134
CO2: 55
Made at SankeyMATIC.com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2.7
2.65
2.6
2.55
2.5
2.45
2.4
2.35
0.00
5.00
10.00
15.00
20.00
Specific Gravity
Aggregate Size (mm)
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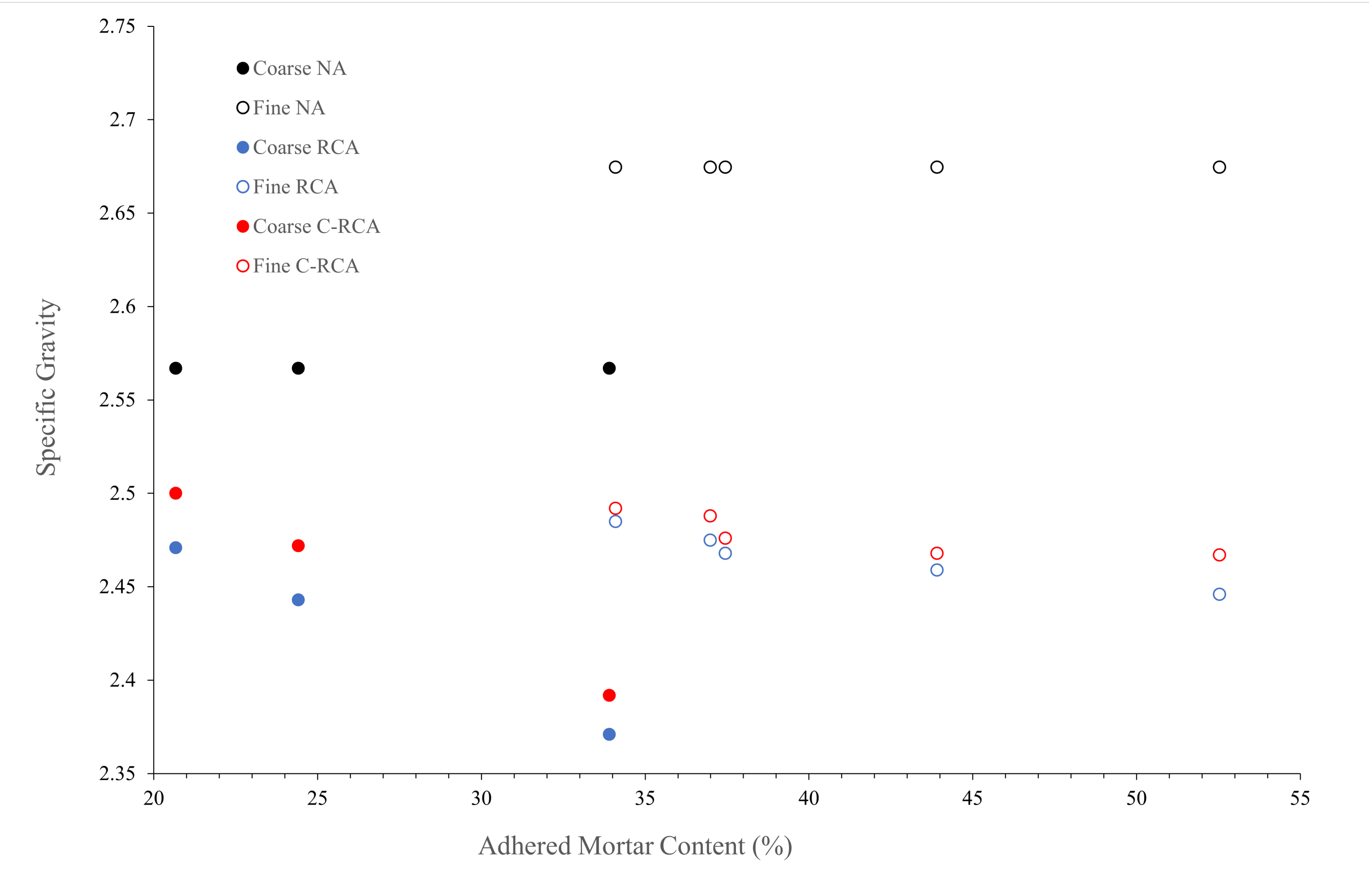
Fine NA
Coarse RCA
Fine RCA
Coarse C-RCA
Fine C-RCA


Coarse NA
Fine NA
Coarse RCA
Fine RCA
Coarse C-RCA
Fine C-RCA
Specific Gravity
Adhered Mortar Content (%)
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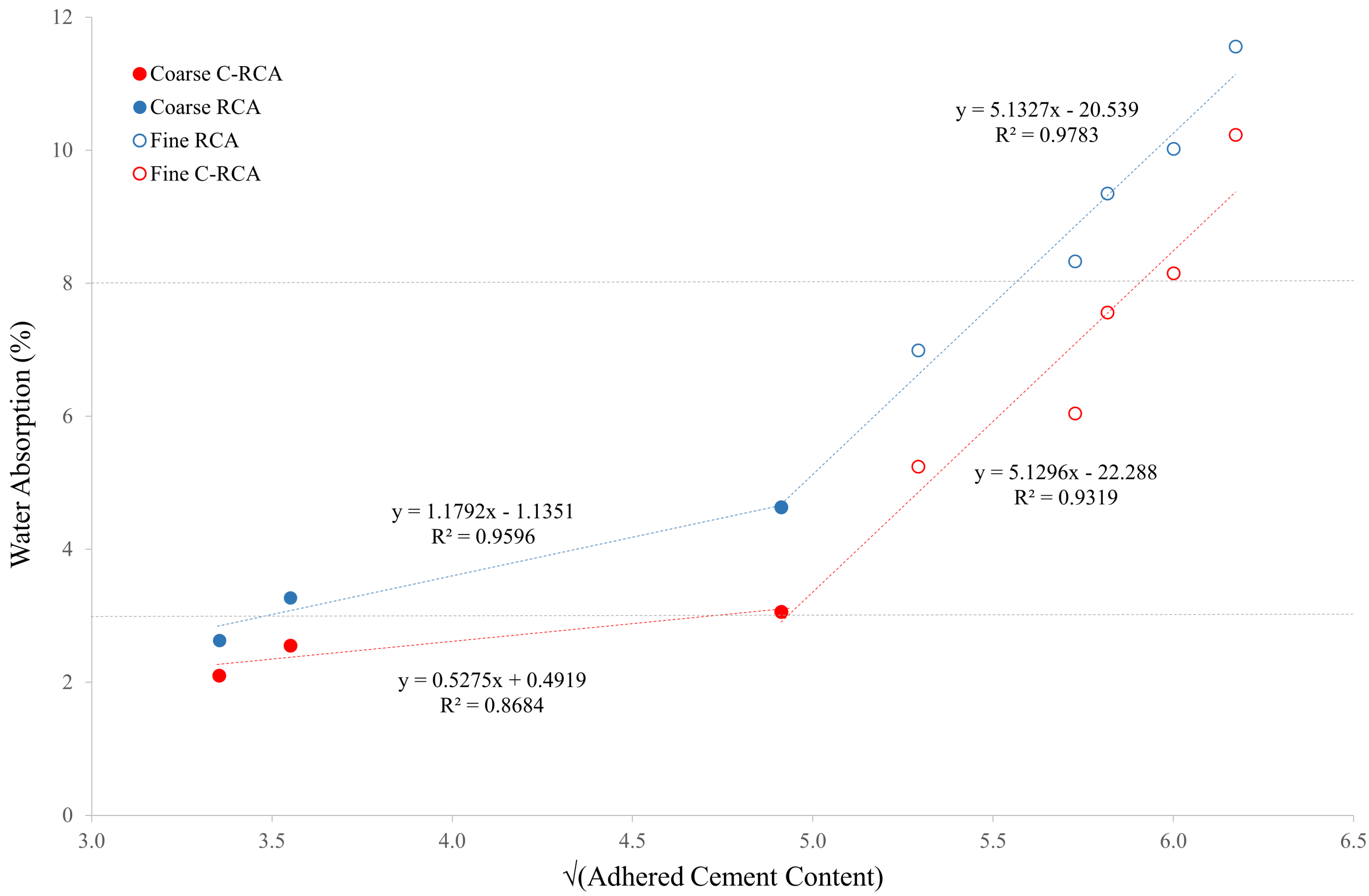
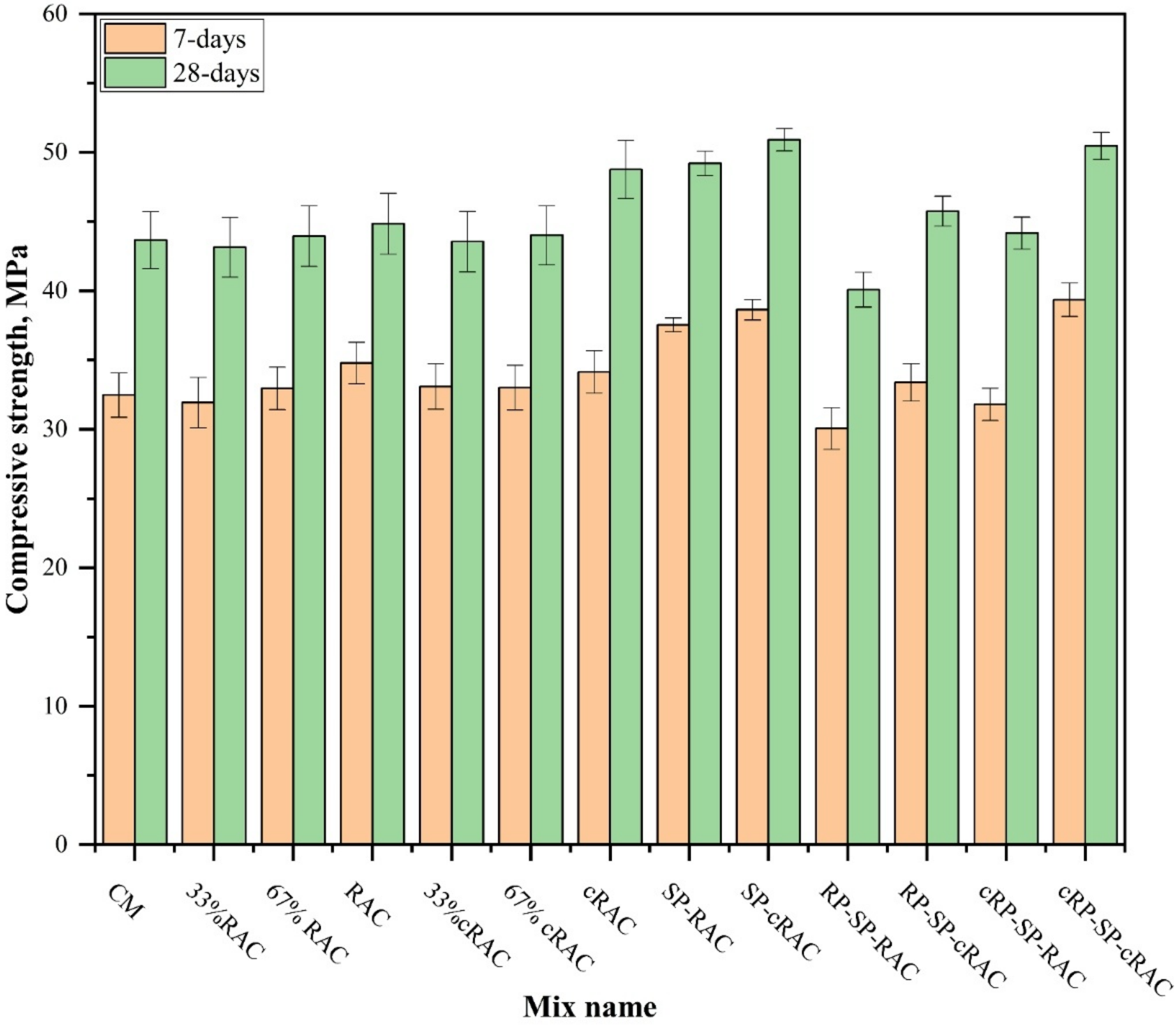
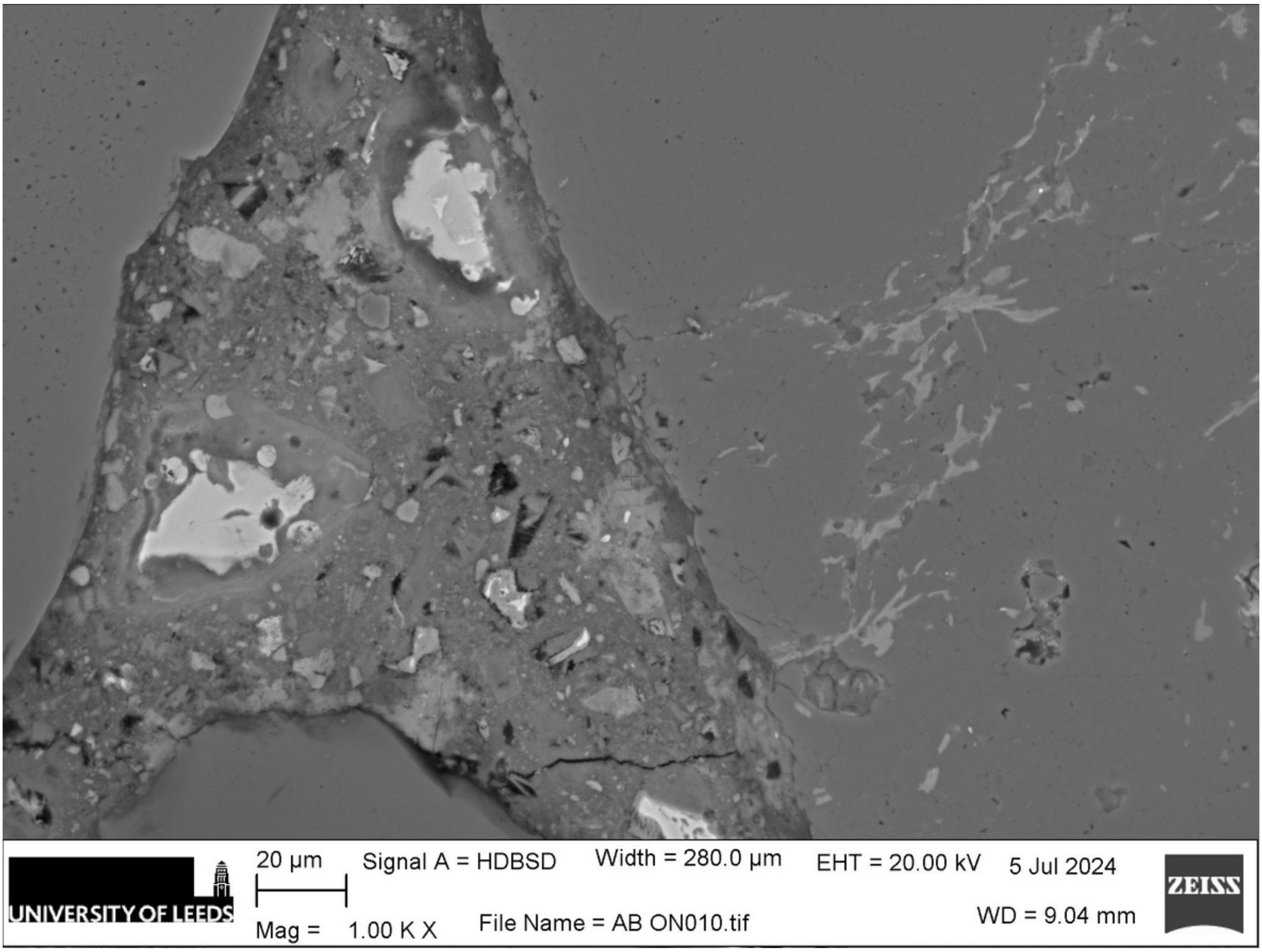
2.7
2.65
2.6
2.55
2.5
2.45
2.4
2.35
20
25
30
35
40
45
50
55

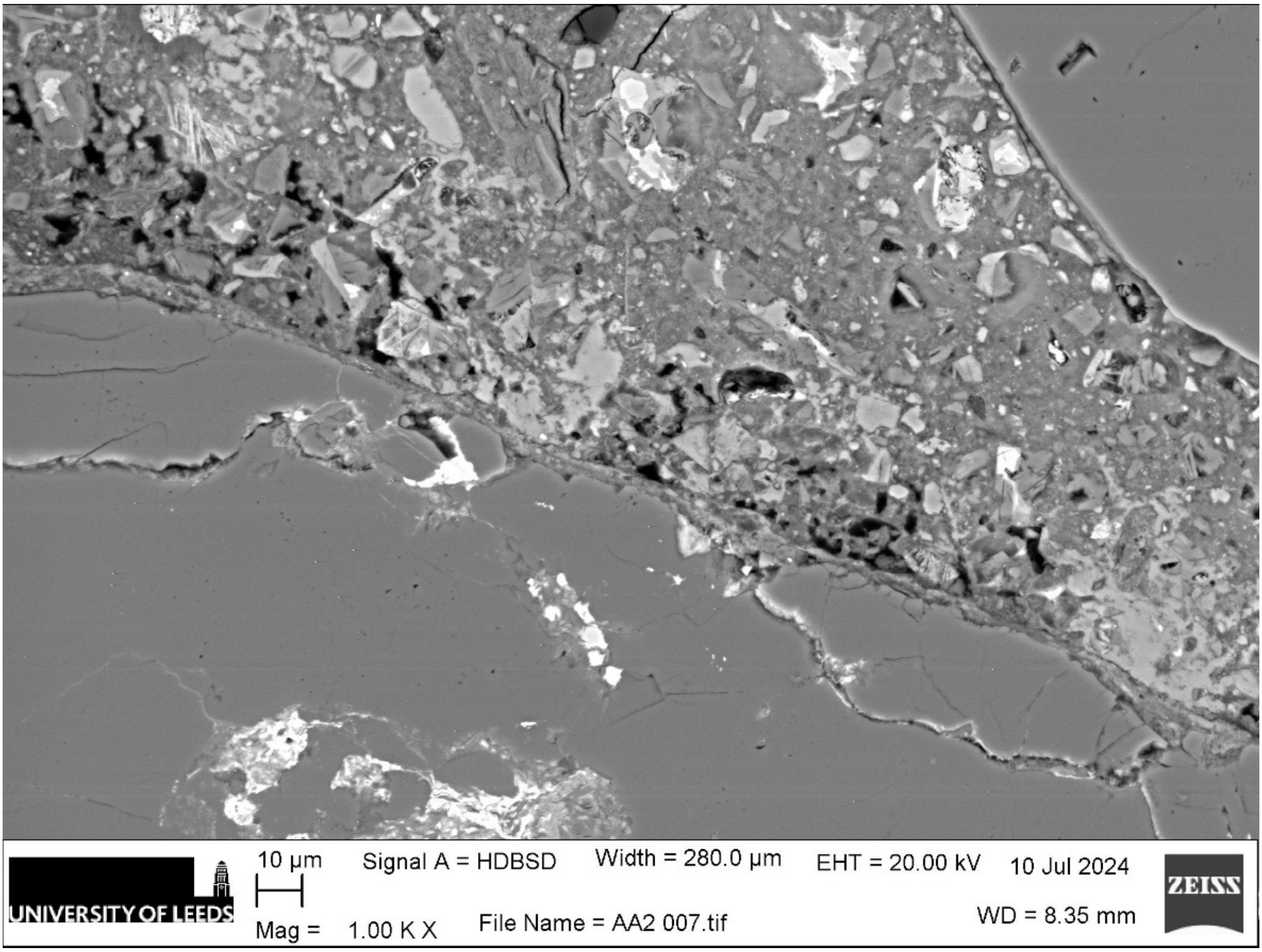


12
10
8
6
4
2
0
Coarse C-RCA
Coarse RCA
Fine RCA
Fine C-RCA
y = 5.1327x - 20.539
R² = 0.9783
y = 5.1296x - 22.288
R² = 0.9319
y = 1.1792x - 1.1351
R² = 0.9596
y = 0.5275x + 0.4919
R² = 0.8684
Water Absorption (%)
3.0
3.5
4.0
4.5
5.0
5.5
6.0
6.5
√(Adhered Cement Content)



7-days
28-days
Compressive strength, MPa
0
10
20
30
40
50
60
CM
33%RAC
67% RAC
RAC
33%cRAC
67% cRAC
cRAC
SP-RAC
SP-cRAC
RP-SP-RAC
RP-SP-cRAC
cRP-SP-RAC
cRP-SP-cRAC
Mix name


20 µm
Signal A = HDBSD
Width = 280.0 µm
EHT = 20.00 kV
5 Jul 2024
UNIVERSITY OF LEEDS
Mag = 1.00 K X
File Name = AB ON010.tif
WD = 9.04 mm
ZEISS


10 µm
Signal A = HDBSD
Width = 280.0 µm
EHT = 20.00 kV
10 Jul 2024
UNIVERSITY OF LEEDS
Mag = 1.00 K X
File Name = AA2 007.tif
WD = 8.35 mm
ZEISS