## A review on re-activation of hardened cement paste and treatment of recycled aggregates

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## ABSTRACT

The generation of construction and demolition waste (C&DW) worldwide keeps on escalating due to growth in infrastructure and the related refurbishment or replacement of existing structures. Numerous researches have been conducted towards utilizing C&DW materials for production of new concrete, from which it has been well-established that their use in concrete mixtures generally leads to inferior mechanical properties of the resulting concrete. As such, the general acceptance and conventional utilization of C&DW in the construction industry, is undermined. In this paper, the concept of triple layered interfacial transition zone (3ITZ) is suggested to be the main reason for the inferior mechanical properties exhibited by recycled aggregate concrete. The present paper reviewed past researches on re-activation of hydrated cement paste and on improving the quality of recycled aggregates by using different treatments. It is shown that a combination of mechanical grinding and thermal treatment at temperatures of 500 to 800°C, is an effective means of activating the cementitious properties of hardened cement paste. Adhered mortar removal and mortar fortification are the main approaches for enhancing the properties of recycled aggregates. Mechanical abrasion and/or thermal treatment at a temperature of 500°C, are the most effective techniques for improving the properties of recycled aggregates.

Keywords: Cement paste, temperature-related and thermal effects, aggregates

#### **Glossary of abbreviations**

- C&DW Construction and demolition waste
- RFA Recycled fine aggregates
- RCA Recycled coarse aggregates
- HCP Hardened cement paste
- DCP Dehydrated cement paste
- O-DCP Original dehydrated cement paste
- D-DCP Dispersed dehydrated cement paste
- OPC Ordinary Portland cement
- GGBS Ground granulated blast-furnace slag
- RCF Recovered concrete fines
- CH Calcium hydroxide
- CSH Calcium silicate hydrate
- CS Calcium silicate
- FA Fly ash
- SF Silica fume
- ITZ Interfacial transition zone
- 3ITZ Triple layered interfacial transition zone
- RA Recycled aggregate
- RAC Recycled aggregate concrete
- NCA Natural coarse aggregate
- RMA Recycled mortar aggregates
- MCP Microbial carbonate precipitation
- GO Graphene oxide
- PVA Polyvinyl alcohol
- PCE Polycarboxylate ether
- w/c Water / cement
- w/b Water / binder

#### **1. Introduction**

The total annual concrete production worldwide has been estimated to be more than 10 billion tons (Meyer, 2009). However, this enormous consumption of concrete for modern infrastructure construction also leads to depletion of the natural resources used for its production. Meanwhile, the generation of construction and demolition waste (C&DW) continues to grow as a result of replacement or refurbishment of existing structures. It is estimated that about 450 million tons of construction waste is generated each year in Europe (De Bito and Saikia, 2013), of which only 28% is recycled while 72% is disposed-off (Matias et al., 2013). The construction industry in South Africa alone is estimated to generate C&DW of about 5 to 8 million tons each year (Benjamin, 2004). Presently, most C&DW materials are dumped in landfills or used in low quality applications, for example, as road base materials.

Numerous researches have been conducted on utilization of C&DW in concrete. Khatib (2005) reported that concrete made using 25% and 100% recycled fine aggregates (RFA) gave compressive strength reductions of 15% and 30%, respectively. Xiao (2012) concluded that compressive strengths of concrete made using recycled coarse aggregates (RCA) were generally lower than those of the control. They reported that strength decreased with increase in RCA content. Experimental work carried out by (Zega and Di Maio, 2011) showed that the 28-day compressive strength of concrete made at a 0.45 water/cement (w/c) ratio, reduced from 43.6 MPa for control to 41.4 MPa when 30% of the fine aggregate was substituted with RFA, i.e. a strength decrease of about 5%. Similarly, Bester et al. (2017) reported that as the replacement of natural aggregate with recycled demolition aggregate increased, there was a drastic reduction in compressive strength of the 0.67 w/c ratio concrete from 45 MPa for control to 5 MPa, when 100% of the natural aggregate was substituted with recycled aggregates. Ekolu et al. (2012) used different types of recycled aggregates including RCA obtained from C&DW and from crushing of laboratory prepared hardened concrete. They found that the extent of compressive strength reduction due to use of RCA, was dependent on the mix design. Concretes made with RCA obtained from C&DW gave 35.2% and 21.8% strength reductions for 32 MPa and 50 MPa concretes, respectively. Such relatively inferior results

obtained for concretes made with recycled aggregates, give a negative connotation which undermines the potential for conventional use of C&DW in concrete construction.

During crushing of waste concrete, fine particles of sizes less than  $150 \,\mu m$  are typically generated. Hardened cement paste (HCP) is a major constituent of these fine particles. Considering the need to promote environmental sustainability through recycling or re-use of waste materials, rejuvenating the cementitious properties of HCP is a subject of major interest in concrete research (Marcela, 2007; Yu and shui, 2014). It has been shown that HCP transforms into dehydrated cement paste (DCP) when subjected to high temperatures of 600 to 800°C for several hours. In the literature, DCP has been shown to exhibit some recovery of hydration characteristics, albeit not to the level of original Portland cement (Alonso and Fernandez, 2004; Castellote et al., 2004). However, the particle sizes of HCP that is separated from waste concrete, are typically coarser than those of cement. Hence, pulverizing of HCP or DCP to levels of cement fineness is necessary, prior to its use as a cementitious material. Also, DCP particles have a tendency to attach onto the ball mill and to form micro-agglomeration during grinding. This behaviour reduces grinding efficacy and produces coarser sizes of ground DCP particles, relative to those of cement particles. When ground DCP is used as a cementitious material, its drawbacks include a relatively short setting time, high water demand, and low strength (Shui et al., 2009), which altogether undermine its potential utilization in concrete.

## 2. Economical aspects of recycling concrete to produce aggregates

Studies have shown that the cost of RA's is significantly lower, being typically about 60 to 80% the cost of natural aggregates (Silva et al., 2017; Dhir et al., 1998; Eckert and Oliveira, 2017). Hameed (2009) evaluated the cost of RCA produced under three different scenarios. Case 1 involved crushing of demolished concrete on site to produce RCA and using it as a base material at the same site. Case 2 involved disposal of demolished concrete at a landfill then purchasing natural aggregate from a quarry, while Case 3 comprised disposal of demolished concrete for crushing at a recycling plant then purchasing the produced RCA for use on site. It was reported that the total production costs for Case 1, Case 2, and Case 3 were \$40,824, \$104,275, and \$96,766 per kiloton of demolished

concrete, respectively. Evidently, the production and/or use of recycled aggregates exhibits relatively lower costs.

#### 3. Significance

There is high research interest towards enhancing the properties of C&DW so as to promote its potential re-use in concrete and the construction industry, generally. The objectives of this paper were to identify and highlight the available promising techniques that could be used to regenerate HCP and to improve the quality of waste aggregates for use in sustainable concrete construction. The potential for recovery of HCP's cementitious properties using thermal treatment is reviewed based on the available literature.

Researches on *removal* of adhered mortar from recycled aggregates by mechanical abrasion, thermal treatment or by soaking in acids, are discussed. Also discussed are the researches relating to *fortification* of the adhered mortar using polymer impregnation, carbonation treatment, pozzolan slurry coatings, and biodeposition treatment. A comparative discussion on the effectiveness, benefits, advantages and disadvantages of the various techniques, is also presented. Overall, this study could contribute towards better general understanding for improvement of C&DW characteristics, with a view of promoting waste utilization in the construction industry. Use of C&DW at industrial scale would lead to preservation of natural resources, reduction of CO<sub>2</sub> generation from cement production, reduction of waste disposal to landfills, and mitigation of environmental pollution, generally. The present work also aimed at determining the extent of, hitherto, established scientific knowledge on this subject to which future researches may improve upon.

# 4. Thermal activation of hardened cement paste for re-use as a cementitious material

Shui et al. (2008) used laboratory crushed concrete to prepare DCP. In their study, crushed particle sizes smaller than 5 mm were obtained from the concrete and heat-treated in a furnace at a temperature of 800°C. DCP was obtained by grinding the heat-treated crushed particles, followed by sieving through a 75  $\mu m$  sieve size. Paste cubes of 20 mm size were prepared at a water/DCP ratio of 0.4 and cured in water. It was reported that compressive

strengths of 7.8 MPa and 8.0 MPa were obtained after 7 and 28 days of curing, respectively. The observed strength developed could be attributed to the rehydration of phases in the DCP. Clearly, the 28-day strength of the DCP pastes was quite low and similar to the early age strength. This observation may be related to the generation of hydration heat by DCP, which occurred mostly at the early hydration stages (Zhang et al., 2018). These results imply that the residual or unreacted cement compounds in DCP were perhaps mainly  $C_3S$  and  $C_3A$ , with little or no  $C_2S$ .

Yu and Shui (2014) also conducted experimental research to evaluate the potential reactivation of HCP. They prepared hardened paste powder by crushing and grinding samples of HCP fragments in a ball mill for 30 minutes. The resulting powder material passing 75  $\mu m$  was collected and subjected to thermal treatment at 650°C for 4 hours. The heat-treated HCP was designated as original DCP (O-DCP). Some O-DCP was dispersed using an ultrasonic device and designated as dispersed DCP (D-DCP). Paste cubes of 40 mm size were prepared using a binder comprising blends of O-DCP and fly ash (FA) or of ordinary Portland cement (OPC) and D-DCP. Pastes were prepared at a water/binder (w/b) ratio of 0.3 and cured in a fog room for 28 days. It was reported that a high compressive strength of 61 MPa was obtained using a mixture containing 55% FA and 45% O-DCP.

Zhang et al. (2018) carried out research involving the use of ground granulated blastfurnace slag (GGBS) to enhance the mechanical properties of DCP. In the investigation, waste concrete obtained from an old concrete structure in a local city was crushed to produce RCA, RFA, and HCP. The HCP was heat-treated in a muffle furnace at 600°C for 3 hours to form DCP. GGBS was blended with DCP at mass ratios of 3:1, 2:1, 1:1, 1:2, and 1:3 GGBS to DCP, then ground in a ball mill for 15 minutes. During grinding, an amount of natural gypsum equal to 5% DCP, was added. Mortar samples were then prepared at a water/DCP binder ratio of 0.5 and sand/binder ratio of 3:1. It was reported that the cumulative heat of hydration for 100% OPC binder paste of grade CEM I 42.5 N and for 100% DCP binder pastes were 203.37 J/g and 445.94 J/g, respectively. It is interesting to note that DCP paste gave a higher heat of hydration than plain OPC paste. For 1:3 GGBS/DCP mixture, the cumulative heat of hydration was 360.34 J/g while the mixture of 1:1 GGBS/DCP gave a cumulative heat of hydration for 237.12J/g, a reduction of 46.8% relative to the hydration heat of 100% DCP. The hydration heat and compressive strengths of the GGBS/DCP mixtures continued to decrease with increase in the content of GGBS. After 3 days of curing, the 1:3 GGBS/DCP paste gave a compressive strength of 30.8 MPa which was higher than the corresponding 27.6 MPa obtained for the plain OPC paste. At 28 days, however, the plain OPC paste gave a much higher strength of 48.2 MPa relative to 37.5 MPa of the 1:3 GGBS/DCP binder paste.

A study on activation of recovered concrete fines (RCF) was done by (Florea et al., 2014) in a manner similar to the foregone investigations conducted using HCP or DCP. They prepared the RCF from laboratory-made concrete samples by first crushing the concrete to particle sizes finer than 2 mm, followed by sieving the particles through a 150  $\mu m$  sieve. The RCF obtained was heat-treated at 500, 800, 1100°C, and used to prepare mixtures. Various waste materials comprising FA, untreated RCF-20 (RCF-20), 500°C treated RCF (RCF-500), 800°C treated RCF (RCF-800), were used to partially replace OPC at proportions of 0, 10, 20, and 30% RCF. Mortar mixtures consisting of 1350 g sand, 450 g cement, and 225 g water, were prepared and used to cast 50 mm cubes and 40 × 40 × 160 mm prisms, which were then cured in water for 28 days. It was reported that the 30% RCF-800/OPC mortar gave the least flexural /compressive strengths of 4.8 /28.5 MPa. The 10% RCF-20/OPC and 10% FA/OPC mortar specimens achieved flexural /compressive strengths of 7.8 /47.5 MPa and 7.5 /46.3 MPa respectively, which were comparable to 8.3 /50.0 MPa of the reference OPC mortar. Evidently, incorporation of 30% RCF as OPC replacement gave significant reductions in flexural and compressive strengths.

Bordy et al. (2017) investigated the potential re-use of HCP without thermal activation. In their study, HCP was made by first casting cement paste at a w/b ratio of 0.3 using OPC of grade CEM I 52.5 N. After 89 days of curing the paste in water, the HCP was removed from curing water and broken into fragments which were then ground into powder using a laboratory ball mill. The powder obtained was sieved through the 80  $\mu m$  sieve and incorporated as a cement replacement material in proportions of 0, 10, 20, 30, 40, 50, 75, and 100% HCP. Mortar mixtures of 0.45 water/HCP binder ratio and 2.32: 1 aggregate/binder ratio were used to cast  $40 \times 40 \times 160$  mm specimens, which were then cured in water for 90 days at room temperature. It was reported that the total porosity of specimens increased with increase in the HCP content. This observation may be attributed to the decrease in total clinker content with increase in HCP content. HCP contains

relatively much less clinker than original OPC. As such, the amounts of hydration products formed, comprising calcium hydroxide (CH) and calcium silicate hydrate (CSH) are relatively much less in systems containing HCP, which leads to relatively higher porosity in HCP mixtures. As expected, compressive strengths decreased with increase in HCP content of the mortar mixtures. The 90-day compressive strength of mortars containing 100% non-treated HCP was 7.29 MPa compared to 62 MPa for the 100% OPC mortar mixture.

Unlike Bordy et al's. (2017) study which evaluated non-activated HCP, thermally treated HCP was investigated by (Serpell and Lopez, 2015). In their study, cement paste slabs of size  $500 \times 500 \times 40$  mm thick were cast at a w/b ratio of 0.5, using OPC of ASTM type I. The paste slabs were cured in water for 28 days, then broken into fragments of 40 mm maximum size. The fragments were oven-dried at 105°C and reduced to fine HCP sizes by crushing and grinding using a jaw crusher and a ball mill. The ground paste particles were sieved through a 75  $\mu m$  sieve to obtained fine powder that was then heattreated at temperatures of 659 to 941°C for 150 minutes to form DCP. The binder was prepared by replacing DCP with 0, 40, 50% silica fume (SF) or with 0, 10, 20, 30, 40, 50, 60, 70% FA. Mortar mixtures of 0.75 to 1.05 w/b ratios were prepared using SF/DCP or FA/DCP as binders. Mortar cubes of 50 mm size were cast and cured in water then tested at 7 and 28 days. It was reported that both the proportion of DCP in the binder and the reactivation temperature, did significantly affect the compressive strength of the DCP mortars. As expected, the compressive strength of mortars increased as the SF content increased. The highest 28-day strength was 26 MPa, obtained with DCP mixtures containing 40% SF. For mortars made using FA/DCP as the binder, the 28-day compressive strength decreased as the proportion of FA increased. It was also observed that increasing the HCP's treatment temperature from 660 to 800°C, led to increase in compressive strength of the DCP mortars. However, further increase in the treatment temperature to the range of 800 to 940°C, led to decrease in compressive strengths.

Like other investigations discussed in the foregone, Shui et al. (2009) prepared HCP using OPC of ASTM type I, at a w/b ratio of 0.5. The paste samples were cured at 20°C for 30 days, then crushed into paste fragments of less than 25 mm size. The fragments were oven-dried at 80°C for 48 hours, then ground in a ball mill to produce HCP powder passing

75  $\mu m$  sieve size. The powder was heat-treated for 2.5 hours at 300, 400, 500, 600, 700, 800 or 900°C to produce DCP. Paste cubes of 20 mm size were prepared at a water/DCP binder ratio of 0.5 then cured in water. The Loss On Ignition (LOI), which is a measure of the non-evaporable water content, was considered as an estimate of the degree of rehydration for DCP paste. The non-evaporable water content was determined as the relative mass loss upon firing of DCP paste powder in a furnace at temperatures of 105°C and 1050°C. It was reported that the rehydration of DCP was quite rapid, attaining 70% rehydration within 24 hours. The degree of rehydration then increased gradually to 80, 85, and 95% at 3, 7, and 28 days, respectively. As usual with cementitious systems, the compressive strength of the DCP paste samples increased with curing age. The DCP paste that had been treated at 800°C gave 12.5 MPa and 17.5 MPa after 3 and 28 days of curing, respectively. Also, it was observed that the compressive strength of pastes increased as the treatment temperature increased up to 800°C. For example, the 3-day strength of the DCP paste increased from 2 MPa at a treatment temperature of 300°C to 12.5 MPa when treated at 800°C. However, when the treatment temperature exceeded 800°C, strength decreased to 8.4 MPa at 900°C. This behaviour may be attributed to the loss of chemically bound water when HCP is subjected to temperatures exceeding 900°C (Florea et al., 2014). Such high temperatures may also break down the primary bonds of hydration products and can cause fusion of unreacted cement compounds present in the HCP. These factors tend to diminish the hydraulic reactivity of the resulting DCP.

## 5. Approaches and techniques for improvement of recycled aggregate properties

## **5.1 Properties of recycled concrete aggregates**

Generally, recycled aggregates (RA's) comprise 65 to 70% original aggregate and 30 to 35% original cement mortar, by volume. As such, RA's are mostly inhomogeneous, less dense, and more porous than natural aggregates (Zhang et al., 2015). RA's are different from natural aggregates in that they contain two components, i.e. the natural aggregates and the cement mortar which is adhered to the surface of aggregates (Juan and Gutiérrez, 2009). Figure 1 shows these two components of RA's. The present authors herein propose that when RA's are used in concrete, a triple layered weak zone (3ITZ) will form which consists of the existing or old interfacial transition zone (ITZ) together with its adhered

mortar, and a newly formed ITZ. It is postulated that the 3ITZ is thicker and much weaker than the single layered ITZ found in concretes made with natural aggregates. Figure 2 gives an illustration of the difference between ITZ and 3ITZ. Owing to the presence of 3ITZ, the mechanical properties of concretes made with RA's are relatively inferior.

As mentioned earlier, it is well-established in the literature that RA's exhibit properties that are inferior to those of natural aggregates. Table 1 gives some basic properties of RA's as reported in various literatures. It is evident that the water absorption values of RA's are in the range of 5 to 7%, which are much higher than the typical values of 1 to 5% for natural aggregates (Gomez-Soberon, 2002; Katz, 2004). The characteristically high water absorption tendency of RA's is attributed to the presence of mortar which is adhered to the surfaces of aggregates. The other properties such as specific gravity, density, crushing value etc. are also different for RA's but remain comparable to those specified for natural aggregates (BS 882, 1992).

#### 5.2 Enhancement upon the properties of concretes containing recycled aggregates

Two approaches are typically employed to enhance the relatively inferior properties of concretes containing RA's. The approaches comprise, (i) incorporation of nanoparticles into RAC and (ii) treatment of RA's prior to their use in concrete. The present study focusses on the latter, as discussed throughout the later sections of the paper from Subsection 5.3 to Section 6.0. In this section, however, a brief discussion is given on the potential use of nanomaterials to enhance RAC properties. The superfine particle sizes and extremely large surface areas of nanomaterials, allow effective infilling of pores and also promote rapid hydration. Nanoparticles may also form nucleation sites for secondary or additional formation of CSH (Wang et al., 2016).

NanoSiO<sub>2</sub> is perhaps the most widely studied nanomaterial towards enhancement of RAC properties, as indicated by the numerous literatures available on the subject (Luo et al., 2018; Mukharjee and Barai, 2017; Zhang et al., 2015; Shaikh et al., 2018; Li et al., 2017; Singh et al., 2018; Shaikh et al., 2015). Hosseini et al. (2009) incorporated 0.0, 1.5, and 3.0% nanoSiO<sub>2</sub> by mass of cement, into RAC made using CEM I 42.5 N at a 0.40 w/c. It was reported that the RCA concrete showed an increase in compressive strength from 28.1 MPa for the control to 35.3 MPa for the mixture containing 3.0% nanoSiO<sub>2</sub>, a strength

gain of 25.6%. In a similar study of RAC made at 0.48 w/c, into which 0.4, 0.8, and 1.2% nanoSiO<sub>2</sub> by mass of cement was incorporated, Younis and Mustafa (2018) reported a 28-day strength increase of 17% for the mixture containing 1.2% nanoSiO<sub>2</sub>. Mukharjee and Barai (2014) reported a 14% strength increase in RAC mixtures of 0.40 w/c containing 3% nanoSiO<sub>2</sub>.

Other types of nanomaterials that have been employed in researches to enhance the RAC properties include nanoTiO<sub>2</sub>, nanoCaCO<sub>3</sub> and graphene oxide (GO) (Luo et al., 2018; Long et al. 2018; Lei et al., 2016). Sharma and Arora (2018) studied the influence of GO on OPC/fly ash mortars made using RA's. Mortar mixtures containing additions of 0.0, 0.05 and 0.10% GO by weight of binder, were prepared at a 0.40 w/b. A strength increase of 37% was reported for the mixture containing 0.05% GO. In a similar study conducted by Fang et al. (2017) using mortar mixtures containing 0.0, 0.05, 0.10, and 0.20% GO by mass of cement, it was reported that the mixture containing 0.20% GO gave a 41% increase in flexural strength.



Figure 1: Components of recycled concrete aggregates (Juan and Gutiérrez, 2009).



Figure 2: The interfacial transition zone in concretes made with natural or recycled aggregates.

Author	Recycled aggregate size (mm)	Specific gravity	Bulk density (kg/m <sup>3)</sup>	Water absorption (%)	Fineness modulus	Crushing value (%)
Kim et al. (2016)	4.75	-	2270	6.56	3.40	-
Qiu et al. (2014)	20	-	2440	7.10	-	-
Zhang et al. (2015)	2.5	2.53	-	8.06	-	18.60
Kong et al. (2010)	20	-	1290	5.50		16.50
Al-Bayati et al. (2016)	19	2.30	-	5.91	-	27.42
Xuan et al. (2016)	20	-	2605	6.10	-	27.80
Katz (2004)	19	2.48	1356	5.10	-	-
Pandurangan et al. (2016)	20	2.45	1360	4.58	-	33.23
Saravanakumar et al. (2016)	20	2.47	1390	6.80	-	-
Mukharjee and Barai (2014)	20	2.46	1321	-	-	31.52
Ismail and Ramli (2013)	20	2.33	-	4.44	-	29.15
Zhan et al. (2014)	20	-	2639	6.58	-	-
De Brito et al. (2016)	20	-	1248	4.66	-	-
Kou and Poon (2010)	20	-	2423	6.23	-	-
Kou et al. (2011)	20	2.35	-	7.42	-	-
Shi et al. (2018)	4.75	-	2490	5.3	2.45	-
Song et al. (2015)	25	2.32	-	7.4	-	-
Etxeberria et al. (2006)	20	-	2430	4.44	-	-
Gokce et al. (2013)	25	2.29	-	5.9	-	-
Paine and Dhir (2010)	20	-	1360	5.5	-	-
De Brito et al. (2011)	16	-	2300	5.8	-	-

Table 1: Properties of recycled concrete aggregates.

#### 5.3 Treatment methods for improving the quality of recycled aggregates

The physical and mechanical properties of concretes made using RA's can be enhanced by using various types of treatments. Basically, there are two approaches that are used to treat RA's, namely the *removal* of adhered mortar and *fortification* of the adhered mortar. Subsequently discussed are the different methods that have been employed in various literatures to treat RA's.

#### **5.3.1** The adhered mortar removal approach

Mechanical abrasion, thermal treatment, soaking of aggregates in acid or combinations of these methods, are the commonly used techniques for removal of adhered mortar from RA's.

## (a) Soaking in acid

An acid solution tends to soften the cement hydration products found in HCP, making it potentially effective in removing the adhered mortar and thus improving the quality of RA's. Tam et al. (2007) conducted research on removal of adhered cement mortar from RA's by soaking the aggregates in hydrochloric acid (HCL), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), and phosphoric acid (H<sub>3</sub>PO<sub>4</sub>) solutions of 0.1 M concentrations. RCA samples of sizes 10 mm and 20 mm, were used in the study. The samples were soaked for 24 hours in the various types of acids and then washed with water to remove residual acid along with corroded mortar from the surfaces of the treated aggregates. They reported that the most effective mortar removal was obtained using the treatments done with H<sub>2</sub>SO<sub>4</sub> and HCL, which respectively gave 10.3% and 12.2% reductions in water absorption values. Akbarnezhad et al. (2011) suggested that complete removal of adhered mortar using acid treatment, could be achieved by soaking of RA's in H<sub>2</sub>SO<sub>4</sub> of concentrations greater than 2.0 M, over long durations of at least 5 days.

Saravanakumar et al. (2016) also carried out an experimental study in which HCL, nitric (HNO<sub>3</sub>), and  $H_2SO_4$  acids of 10% normality, were used to treat RCA. During the treatments, samples of aggregates were initially soaked in the three acid solutions for 24

hours at room temperature. Afterwards, the treated aggregates were thoroughly washed with distilled water to remove the residual acid solvents and loose particles. The HCL-treated aggregates were further treated by soaking the samples in SF slurry for 24 hours at room temperature. It was reported that the treated RCA showed improvement in density from 1390 kg/m<sup>3</sup> before treatment to 1472 kg/m<sup>3</sup> after treatment. Also, the treated RCA had 30.7% lower water absorption relative to the non-treated RCA. The improved results were attributed to effective removal of adhered mortar from the RCA using the HCL treatment.

It should be considered that while the acid treatment technique can be effective in removal of adhered mortar, it may not be suitable for weak or friable aggregates such as limestone etc. Such weak aggregates typically dissolve under acid attack (Juan and Gutierrez, 2009).

#### (b) Mechanical abrasion with or without thermal treatment

Mechanical abrasion is a common method of removing cement mortar from the surfaces of aggregates by applying vigorous physical wear using an abrasion machine. Although this method can significantly decrease the amount of the adhered cement mortar, it has the potential to cause fracturing of particles when used to treat friable aggregates (Tam et al., 2008; Juan and Gutiérrez, 2009; Akbarnezhad et al., 2013).

Basically, the mechanical abrasion method can be conducted with or without heat treatment. The method is conducted by applying rolling and vibratory motions at high speed in a rotary mill. This process tends to fracture and fragment the adhered mortar. Use of high speeds during mechanical abrasion runs, tends to improve the peel-off efficacy of the mortar layer, thereby producing a better quality of the treated RCA (Shi et al., 2016). Crushed recycled concrete may also be heat-treated at around 300°C to desiccate the adhered mortar, making it fragile before subjecting it to abrasion in a rotary mill. The ease with which the adhered mortar can be removed increases with rise in the temperature of heating. However, the quality of RCA may be adversely affected if the heating temperature exceeds 500°C. Experimental research carried out by (Tateyashiki et al., 2001) demonstrated that high quality treated RCA can be obtained by combining thermal treatment with mechanical abrasion.

## (c) Combined mechanical abrasion and acid wash

This method was employed by (Kim et al., 2016) to treat RFA of particle sizes less than 5 mm. In their treatment set-up, the fine aggregates were placed in an abrasion machine, then acid water that was made by diluting  $H_2SO_4$  was fed into the machine. The treatment process was carried out at varied abrasion time durations of 5 to 15 minutes. It was reported that the oven-dried density of RFA increased from 2270 kg/m<sup>3</sup> before the treatment to 2480 kg/m<sup>3</sup> after the treatment. Also, the water absorption of RFA reduced from 6.56% before the treatment to 2.90% after the treatment, i.e. a significant decrease of about 56%.

#### (d) Thermal treatment followed by soaking in water

Juan and Gutierrez (2009) employed this method to study the removal of adhered mortar from RCA of size fractions 4 to 8 mm and 8 to 16 mm. In their experiment, the aggregates were dried in a muffle furnace at 500°C for 2 hours, then immediately immersed in cold water. The resulting sudden cooling causes differential stress movements between the adhered mortar and the substrate aggregate, which then leads to cracking of the adhered mortar, making it to detach from the aggregate surface. It was reported that the treatment led to a 15% increase in density values and 70% reduction in water absorption values of the RCA. This method can be used for various types of aggregates including friable or weak aggregate types.

#### (e) Thermal treatment followed by soaking in acid

Kumar and Minocha (2018) investigated the effectiveness of using a combination of thermal treatment and soaking of RA's in acid, to treat RFA. In their study, RFA samples were soaked in water for 24 hours, then subjected to thermal treatment at 300, 400, 500, and 600°C. The treated samples were then soaked in 0.1, 0.4, and 0.7 M HCL for 24 hours. The treated RFA samples were further immersed in water for 24 hours to wash out the residual acid from the surfaces of the aggregate particles. It was reported that the samples that were heat-treated at 600°C, then soaked in 0.1, 0.4 and 0.7 M HCL showed 54.1%,

68.7%, and 69.2% reductions in water absorption, while their specific gravity values correspondingly increased by 25.8%, 34.7%, and 39.8%. Evidently, the use of higher concentrations of acids, led to greater removal of the adhered mortar.

## (f) Soaking in acid followed by thermal treatment

This procedure is a reverse of the method discussed in the foregoing Section 5.3.1(e). Al-Bayati et al. (2016) carried out an experiment to evaluate the physical properties of 19 mm RCA by first soaking the aggregates in acid before subjecting the samples to thermal treatment. In their experiment, the RCA samples were washed thoroughly and oven-dried at 105°C for 24 hours, then cooled and soaked for 24 hours in acid solutions of 0.1M HCL or 0.1M acetic acid ( $C_2H_4O_2$ ). Afterwards, the samples were immersed in distilled water to wash out residual acid from the aggregates before again oven-drying the samples at 105°C for 24 hours. Finally, the RCA was heat-treated for one hour at 250, 350, 500 or 750°C.

It was reported that the treatment process generally removed a significant amount of adhered mortar from the RCA. For samples that were treated at 350°C, the water absorption reportedly decreased from 5.91% before the treatment to 4.29% after the treatment. However, the treated samples that were subjected to the higher temperatures of 500 and 750°C gave poor water absorption results. These adverse results could be attributed to possible microcracking of the aggregate particles, as a result of thermal expansion under high temperatures (Al-Bayati et al., 2016). It is also possible that wet-dry cycles consisting of oven-drying followed by soaking of samples in acid or water, generated differential internal stresses within the aggregate particles, which initiated or exacerbated microcracking. Considering these adverse effects of the treatment method, this procedure which involves applying the treatment in a number of wet-dry cycles, does not appear to be suitable for treating RA's.

#### (g) Special techniques

#### (i) Ultrasonic water cleaning

By soaking RCA in water, some impurities found at surfaces of crushed concrete aggregates can be removed. Katz (2004) repeatedly applied ultrasonic water cleaning on 19 mm RCA until clear water was obtained. The procedure reportedly caused the removal

of weak adhered mortar layers from RCA. The study showed that RA's from low grade concrete required more cleaning cycles than the aggregates recovered from higher grade concrete. This observation may be attributed to the weak matrix of the low grade concrete, which releases relatively higher quantities of fine particles more easily, compared to the stronger matrix of higher strength concrete. In the experiment by (Katz, 2004), the ultrasonically washed RCA were used with OPC to prepare concretes of 0.57 w/b ratio. It was reported that the concretes made using the treated RCA gave a strength increase of 7%.

## (ii) Soaking in acid followed by soaking in calcium silicate

Ismail and Ramli (2013) treated 10 mm and 20 mm RCA by soaking the aggregates in acid solutions followed by soaking the samples in calcium silicate (CS). During the first stage of the treatment, RCA samples were immersed in 0.1 M and 0.5 M HCL for 24 hours, in order to remove the adhered mortar. The aggregates were removed from acid solutions and washed with distilled water, then drained. They were then oven-dried at 105°C for 24 hours and soaked in CS solution for 24 hours. Finally, the treated RCA samples were removed from the the apparent particle density of the treated RCA slightly improved, while water absorption significantly reduced by 20%.

Figure 3 shows a schematic diagram of the different treatment methods that have been employed in the literatures, for removal of adhered mortar. It can be seen that a variety of method combinations can be employed to achieve effective removal of adhered mortar. From the foregoing discussion, it is clear that the method(s) that may be considered for treating any given RCA have to be cautiously selected. Mechanical abrasion and acid treatment methods may not be suitable for treating friable aggregates. It also seems that treatments that apply repeated wet-dry cycles of oven-drying and soaking in acid or water, may induce microcracking of aggregate particles. While thermal treatment is shown to be effective, use of high temperatures exceeding 500°C can have adverse effects, resulting in poor properties of the treated aggregates (Al-Bayati et al., 2016; Shi et al., 2016).



Figure 3: Treatment methods for removal of adhered mortar.

#### 5.3.2 The adhered mortar fortification approach

In this approach, attempt is made to fill the existing voids within the adhered cement mortar, thereby strengthening the ITZ. Polymer emulsions are commonly used as fortifying agents. The treatment process involves vacuum impregnation of aggregates with or immersion of the samples into, an emulsion. The various mortar-fortifying treatment methods that have been employed in various literatures, are subsequently discussed.

#### (a) Polymer impregnation

Silicon-based polymers are water repellents which can be used to reduce the water absorption of porous materials such as RCA. Polymer molecules tend to fill pores within the adhered mortar, when RCA are immersed in the polymer emulsion (Shi et al., 2016). Spaeth and Tegguer (2013) investigated the effect of polymer treatment on properties of 20 mm RCA. The polymer solutions used in the investigation were sodium silicate,

siloxane, and silane emulsion. The treatment procedure consisted of impregnating the RCA with the polymer solutions, followed by air-drying of the samples at room temperature for 24 hours before oven-drying at 50°C for another 24 hours. It was reported that the treatments were beneficial in reducing the water absorption of RCA, depending on the type and concentration of the polymer solution used. For example, use of 30% sodium silicate solution reduced the water absorption of RCA from 4.5% before the treatment to 2.1% after the treatment.

Kou and Poon (2010) investigated the viability of improving the physical properties of 10 mm and 20 mm RCA by treating the aggregates using polyvinyl alcohol (PVA). The polymer solution was prepared by adding 120, 160, 200, and 240 g of PVA powder to 2 litres of boiling water, giving concentrations of 6%, 8%, 10% and 12% PVA, respectively. The RCA samples were soaked in the polymer solution by applying a vacuum suction of 920 mbar for 6 hours in a desiccator. After stoppage of the vacuum, the samples were left to soak for a further 18 hours before tests were done. It was reported that the PVA treatment led to increase in density of the 20 mm RCA samples from 2423 kg/m<sup>3</sup> to 2566 kg/m<sup>3</sup>, while water absorption decreased significantly from 6.23% before the treatment to 2.39% after the treatment.

#### (b) Carbonation treatment

CSH and CH are the principal hydration products present in adhered mortar which typically contains HCP. Carbon dioxide (CO<sub>2</sub>) can easily penetrate into the pores of adhered mortar and react with these compounds under partially dry conditions (Castellote et al., 2009). Thiery et al. (2007) reported that carbonation of CH and CSH led to 11.5% and 23.1% volume increases, respectively. CH is generally present at the pore walls and within the HCP. CO<sub>2</sub> reacts with the CH to form CaCO<sub>3</sub>, which in turn fills existing pores and voids within the mortar, causing densification (Ekolu, 2016; Ekolu, 2018). Accordingly, carbonation reduces the permeability of the adhered mortar.

Zhang et al. (2015) reported an improvement in physical properties of RA's following carbonation treatment of the aggregates. In their study, 2.5 mm RFA samples were exposed to 20% CO<sub>2</sub> concentration in a controlled carbonation chamber maintained at 20°C and 60% RH. During progress of the experiment, the extent of carbonation of the RFA samples

was monitored periodically. At specific time periods, samples were taken out from the carbonation chamber then ground and sprayed with 1% phenolphthalein alcohol solution. It was reported that a duration of 7 days was required for complete carbonation of the samples to occur. Also, the water absorption values of the carbonated gravel RFA and carbonated crushed RFA were found to be respectively 28% and 22% lower than the corresponding values of the non-carbonated samples. The apparent density and crushing value of the carbonated gravel RFA were found to be 5% and 9% higher than those for the non-carbonated samples. The carbonated RFA samples were also mixed with OPC of grade CEM I 42.5 N, to prepare mortar cubes of 40 mm size at a w/b ratio of 0.50. The cubes were cured in lime-saturated water for 90 days. It was reported that the 90-day compressive strengths of the carbonated gravel RFA and carbonated granite RFA mortars were 18% and 8% higher than those for the corresponding non-carbonated samples, respectively.

The mechanical properties of concretes containing carbonated and non-carbonated RCA of 10 mm or 20 mm sizes were also investigated by (Xuan et al., 2016). The carbonation treatment consisted of pre-conditioning the samples in a drying chamber maintained at 25°C and 50% RH before placing inside a 100 L cylindrical steel chamber, which was then filled with 100% CO<sub>2</sub> gas under a vacuum suction of -0.6 bar. After 24 hours of carbonation, the treated RCA samples were removed from the chamber and tested for water absorption and crushing value. It was reported that the carbonation treatment led to a reduction of 16.7% in water absorption and an increase of 25.9% in crushing value of the RCA, while the 10% fines value increased by 4%.

Shi-Cong et al. (2014) also reported an enhancement in properties of recycled mortar aggregates (RMA) upon CO<sub>2</sub> treatment of the aggregates. In conducting the treatment, 4 kg of crushed RMA were placed inside a 33 L air-tight steel cylindrical chamber, then a vacuum suction of -0.5 bar was applied before the chamber was filled with 100% CO<sub>2</sub> gas supply. The CO<sub>2</sub> treatment was sustained over various durations of 6, 12, 24, 48, and 72 hours. Results showed an increase in density and in 10% fines value from 2326 kg/m<sup>3</sup> and 96 kN before the treatment, to 2349 kg/m<sup>3</sup> and 111 kN after the treatment. Also, the water absorption of RMA reduced from 11.8% before the treatment to 10.1% after the carbonation treatment. It was reported that a CO<sub>2</sub> treatment duration of at least 24 hours was needed for substantial improvement in properties of the RMA to be attained.

#### (c) Coating using pozzolanic materials

Studies have shown that the relatively inferior properties of concretes made using RA's can also be improved by coating the aggregate particles using pozzolanic materials such as FA, SF etc. (Shannag, 2000). These coatings tend to fill some existing cracks and voids within the adhered mortar. The pozzolans can also react with some CH that may be present within the pore walls of the adhered mortar, forming secondary hydration products (Ekolu et al., 2006; Kong et al., 2010). Such a pozzolanic reaction may in turn improve the microstructure of ITZ, thereby enhancing the strength and durability properties of the RCA concretes.

A study on use of carbonation treatment or pozzolan slurry coatings, to treat 4.75 mm RFA, was conducted by (Shi et al., 2018). The pozzolan slurries used in their study were prepared using SF, FA and nanoSiO<sub>2</sub> materials whose specific surface areas were 18500, 427, and 160000 m<sup>2</sup>/kg, respectively. The nanoSiO<sub>2</sub> slurry was made at a water to solid ratio of 20:1, while the SF and FA slurries were of 10:1 water to solid ratio. RFA samples were oven-dried at 60°C for 48 hours, before mixing with fresh slurry for 30 minutes and leaving the samples to soak for a further 60 minutes. The treated RFA samples were then removed from the slurry, drained, and oven-dried at 100°C for 2 hours, then tested. The treatment of RFA using CO<sub>2</sub> was done in a carbonation chamber maintained at 20°C and 60% RH, filled with 20% CO<sub>2</sub> for 3 days.

It was reported that both the carbonation and pozzolan slurry treatments had the effects of reducing water absorption and increasing the apparent density of the RFA. However, the carbonation treatment seemed to be slightly more effective than the pozzolan slurry treatments, giving a water absorption value of 26.4% compared to corresponding values of 24.5%, 20.7%, 20.7% for FA-slurry, SF-slurry, nano-SiO<sub>2</sub> slurry treatments, respectively. As already explained in Section 5.3.2(b), the effectiveness of the carbonation treatment is attributed to the reaction of CO<sub>2</sub> with hydration products within the adhered mortar. This reaction forms CaCO<sub>3</sub> that fills empty spaces within the capillary pores of the adhered mortar (Cakır, 2014; Zhang et al., 2015). In contrast, the pozzolan slurry forms a thin layer of coating at surfaces of RA's and may not easily penetrate deeper into the adhered mortar,

unlike the case with  $CO_2$  ingress. However, both techniques can significantly improve the quality of ITZ and enhance the properties of RA's, as already discussed.

#### (d) Biodeposition treatment

The biodeposition concept is based on the ability of bacteria to precipitate CaCO<sub>3</sub> at the outer surface of the cell wall, due to occurrence of negative zeta potential of adequate strength (Grabiec et al., 2012). Sporosarcina pasteurii cell (Sp. cell) can attract Ca ions (Ca<sup>2+</sup>), which then react with carbonate ions (CO<sub>3</sub><sup>2-</sup>) originating from urea (CO(NH<sub>2</sub>)<sub>2</sub>) hydrolysis. Simultaneously, ammonia (NH<sub>4</sub><sup>+</sup>) increases the pH value of the surrounding medium, which in turn improves calcite precipitation (Shi et al, 2016).

Wang et al. (2017) carried out an experiment to investigate the influence of biogenic CaCO<sub>3</sub> treatment on the properties of RA's. In their study, 20 mm RCA were subjected to four different treatments. In treatment A, the RCA samples were immersed in a grown culture for 24 hours. The samples were then removed and transferred to a deposition medium consisting of 0.5 M urea and 0.5 M Ca-nitrate. In treatment B, the RCA samples were submerged in a highly concentrated bacterial suspension of  $10^9$  cells/ML for 24 hours. Afterwards, the samples were removed and transferred to the same deposition medium as that of treatment C was similar to treatment A, the main difference being the incorporation of a yeast extract of 5 g/L into the deposition medium. In treatment D, the RCA samples were immersed in a grown culture for 48 hours, then transferred to the same deposition medium as that of treatment A. It was reported that the most effective method was treatment D, which gave the lowest water absorption value of the treatments A, B, C and D, respectively.

Biodeposition treatment may also be referred to as microbial carbonate precipitation (MCP). As already explained, the treatment refers to the process of employing microorganisms, particularly bacteria, to stimulate CaCO<sub>3</sub> crystal precipitation (Qiu et al., 2014; Zhan et al., 2019). The formation of CaCO<sub>3</sub> is controlled by four main factors, i.e. the Ca<sup>2+</sup> ion concentration, dissolved inorganic carbon, pH, and availability of the crystal nucleation site (Hammes and Verstraete, 2002). While most bacteria can modify all the four factors, their key function in stimulating precipitation is achieved by increasing the pH through metabolic processes (De Muynck et al., 2010) and by attracting metal ions such as  $Ca^{2+}$ , to form crystal nucleation sites (Morita, 1980). An investigation by Qiu et al. (2014) found that an MCP treatment which was done on 20 mm RCA samples using a bacterial concentration of  $10^8$  cell/mL, gave 1.03% increase in weight and 15% reduction in water absorption of the treated samples.

Grabiec et al. (2012) conducted a study of biodeposition treatment in which 19 mm RCA were dried at 78°C for 24 hours before submerging the samples in a liquid culture of Sp. cell for 24 hours. Water absorption tests were done on RCA samples before and after the biodeposition treatment. It was reported that water absorption reduced by 8% and 15% when bacterial concentrations of  $10^6$  and  $10^8$  cell/mL were used in the treatments.

At the end of Section 5.3.1 (Figure 3), some considerations on the selection of methods for treatment of RA's were discussed. Table 2 further attempts to compare the different treatment methods, on the basis of their effects on water absorption and density of RA's. Also, given in the table are the advantages and disadvantages of the different treatment methods. It can be seen that the mechanical abrasion method is one of the most effective treatment techniques for RA's, along with the thermal treatment method. However, thermal treatment can be costly due to its high energy requirements. Both techniques are non-hazardous, unlike the acid soaking method and the carbonation treatment technique, which can pose high health risks when applying the treatments. The other techniques such as polymer impregnation and biodeposition etc., require extensive use of chemicals and bacterial cultures, which renders these methods to be quite expensive.

	Effect on		
Treatment method	24-hour water	Density	Advantages and
	absorption (%)	$(kg/m^3)$	disadvantages
(A). Adhered mortar removal approach			
<ol> <li>Combined mechanical abrasion and acid wash (Kim et al., 2016).</li> </ol>	↓ down to 56%	↑ up to 9%	Mechanical abrasion is effective, simple, direct, not costly, non-hazardous. Not suitable for friable aggregates.
2. Soaking in acid (Tam et al., 2007).	$\downarrow$ down to 12%	-	Can be hazardous, not suitable for weak aggregates
3. Thermal treatment (Juan and Gutierrez, 2009).	$\downarrow$ down to 70%	↑ up to 15%	Effective but costly due to high energy requirement.
4. Pre-soaking in acid followed by thermal treatment (Al-Bayati et al., 2016).	$\downarrow$ down to 27%	-	The use of acid can be hazardous.
5. Thermal treatment followed by soaking in acid (Kumar and Minocha, 2018).	$\downarrow$ down to 69%	-	Can be costly. The use of acid can be hazardous.
6. Special technique: soaking in acid followed by soaking in calcium silicate (Ismail and Ramli, 2013).	↓ down to 20%	-	Expensive due to the high cost of chemical agents.
(B). Adhered mortar fortification approach			
1. Polymer impregnation (Kou and Poon, 2010; Spaeth and Tegguer, 2013).	$\downarrow$ down to 53%	↑ up to 6%	Expensive due to the high cost of chemical agents.
2. Carbonation treatment (Zhang et al., 2015).	$\downarrow$ down to 28%	$\uparrow$ up to 8%	Simple but can be hazardous
3. Biodeposition treatment (Grabiec et al. 2012)	$\downarrow$ down to 15%	-	Costly and time consuming.

Table 2: Comparison of different methods for treatment of recycled aggregates.

## 6. Cost implications of the various treatment methods for recycled aggregates

The authors of the present paper conducted a survey by contacting the supervisors of various aggregate quarries in Gauteng, South Africa. The survey showed that the average cost of natural aggregate and RCA in the province is about \$24.7 and \$20.8 per ton, respectively. As expected, the application of treatments on RCA generally increases the cost of the treated aggregates. Tables 3a,b,c give detailed considerations on the cost inputs associated with each of the various treatment methods. It can be seen that the average cost

of the non-treated RCA increases from \$20.8 per ton before treatment to \$26.2, \$27.3, \$28.1, and \$28.6 per ton after application of the abrasion, ultrasonic water cleaning, carbonation, and thermal treatments, respectively. Evidently, the treatments elevate the cost of RCA by \$1.5 to \$3.9 above that of natural aggregates, depending on the treatment method. However, this cost increment is small considering the enormous benefits of treated RA's including the improvement of engineering properties, sustainability of resources and environmental impacts.

Treatment	Cost (\$1000)	Note*
Thermal treatment		
Cost of muffle furnace	5.0	Muffle furnace machine costs about \$50,000,
		i.e. \$5,000 per year for 10 years
Working capital	1.5	15% variable operating cost of about \$1,500
		per unit per year for the muffle furnace
Equipment maintenance	1.6	Cost of maintenance per year is about \$1,600
Labour	8.9	One person at about \$8,869 per year
Power (electricity)	2.7	Cost of power is about \$2,748.3 per year
Total cost of the treatment	19.5	About 2,500 tons per year at \$7.8 per ton
Soaking in acid		
Cost of acid	3.5	Average cost of HCL, H <sub>2</sub> SO <sub>4</sub> or H <sub>3</sub> PO <sub>4</sub>
		solution of 0.1 M for treating one ton of
		recycled aggregates is about \$3,500
Labour	0.2	Cost of labour for treating one ton
		of aggregates is about \$200
Water	0.005	Cost of water for treating one ton of
		aggregates is about \$5
Total cost of the treatment	3.7	About \$3,705 per ton
Thermal treatment followed by	3.7	About \$3,712.8 per ton
soaking in acid		
Pre-soaking in acid followed by	37	About \$3,712.8 per ton
thermal treatment	5.7	
Abrasion method		
Cost of abrasion machine	0.5	Abrasion machine cost is about \$5,000, i.e.
		\$500 per year for 10 years
Working capital	1.0	15% variable operating cost of about \$1000
Equipment maintenance	1.0	Cost of maintenance per year is about \$1,000
Labour	8.9	One person at about \$8,869 per year
Power (electricity)	2.1	Cost of power is about \$2100 per year
Total cost of the treatment	13.5	About 2500 tons per year at \$5.4 per ton
Combined abrasion and acid		
treatment	3.7	About \$3,710.4 per ton

Table 3a: Cost implications of thermal treatment with /without soaking in acid and of the abrasion method.

\*Data obtained from a survey conducted with industry experts and suppliers of treatment chemicals and machinery.

Treatment	Cost (\$1000)	Note*
Ultrasonic water cleaning		
Cost of the ultrasonic machine	2	Ultrasonic machine cost is about \$20,000 i.e.
		\$2,000 per year for 10 years
Working capital	1.7	15% variable operating cost of about \$1,700
		per unit per year for the ultrasonic machine
Equipment maintenance	1.8	Cost of maintenance per year is about \$1,800
Labour	6.8	One person at about \$6,800 per year
Power	1.9	Cost of power is about \$2,900 per year
Water	2.0	Cost of water is about \$2,000 per year
Total cost of the treatment	16.2	About \$2,500 tons per year at \$6.5 per ton
Polymer impregnation treatment		
Cost of the drying oven	2.0	Drying oven cost is about \$20,000
		i.e. \$2,000 per year for 10 years
Working capital	1.6	15% variable operating cost of about \$1,600
		per unit per year for the oven
Equipment maintenance	1.7	Cost of maintenance per year is about \$1,700
Labour	8.6	One person at about \$8,600 per year
Power (electricity)	2.5	Cost of power is about \$2,500 per year
Cost of (only) using the oven	16.5	About 2,500 tons per year at \$6.6 per ton
Cost of polymer	3.0	Average cost of silane, siloxane or sodium
		silicate solution for treating one ton of
		aggregates is about \$3,000
Labour	0.5	Cost of labour for treating one ton
		of aggregates is about \$500
Cost of (only) applying the polymer	3.5	About \$3,500
Total cost of the treatment	3.5	Cost of the treatment is about \$3,507 per ton

Table 3b: Cost implications for ultrasonic cleaning and polymer impregnation treatment methods.

\*Data obtained from a survey conducted with industry experts and suppliers of treatment chemicals and machinery.

Treatment	Cost (\$1000)	Note <sup>*</sup>
Carbonation treatment		
Cost of the carbonation	4.0	Carbonation chamber cost is about \$40,000
chamber		i.e. \$4,000 per year for 10 years
Working capital	2.0	15% variable operating cost of about \$2,000
		per unit per year for the chamber
Equipment maintenance	1.9	Cost of maintenance per year is about \$1,900
Labour	8.5	One person at about \$8,500 per year
Cost of the CO <sub>2</sub>	1.8	Cost of CO <sub>2</sub> is about \$1,800 per year
Total cost of the treatment	18.2	About 2,500 tons per year at \$7.3 per ton
Pozzolanic materials treatment		
Cost of drying oven	2.0	Drying oven cost is about \$20,000
		i.e. \$2,000 per year for 10 years
Working capital	1.6	15% variable operating cost of about \$1,600
		per unit per year for the oven
Equipment maintenance	1.7	Cost of maintenance per year is about \$1,700
Labour	8.6	One person at about \$8,600 per year
Power (electricity)	2.5	Cost of power is about \$2,500 per year
Cost of (only) using the	16.5	About 2,500 tons per year at \$6.6 per ton
oven		
Cost of pozzolanic materials	0.008	Average cost of SF, FA or nanoSiO <sub>2</sub> for
_		treating one ton of recycled aggregates is
		about \$8.0
Labour	0.1	Cost of labour for treating one ton
		of aggregates is about \$100
Water	0.0006	Cost of water for treating one ton of
		aggregates is about \$0.6
Cost of (only) applying the	0.1	About \$108.6 per ton
pozzolanic materials		-
Total cost of the treatment	0.1	Cost of the treatment is about \$115.2 per ton

Table 3c: Cost implications of the treatments done using carbonation and pozzolanic materials.

\*Data obtained from a survey conducted with industry experts and suppliers of treatment chemicals and machinery.

#### 7. Conclusions

This review paper has identified and highlighted the available promising techniques that may be used to regenerate cementitious properties from hardened cement paste and to improve the properties of recycled aggregates. It was found that a combination of mechanical grinding and thermal treatment techniques, is an effective means for activating the cementitious properties of hardened cement paste. Considering that the relatively inferior properties of recycled aggregates are mainly attributed to the presence of adhered mortar at the surfaces of aggregates, a wide range of techniques are categorized into the *adhered mortar removal treatments* and the *adhered mortar fortifying methods*. The important mortar removal techniques consist of mechanical abrasion and/or thermal treatment with or without soaking of the aggregates in acid or water. Polymer impregnation, carbonation treatment, pozzolanic slurry coating and biodeposition, are the main mortar fortifying techniques. From findings in the literature, the following specific conclusions have been reached.

- (a) Hardened cement paste (HCP) can be transformed into dehydrated cement paste (DCP) by subjecting it to high temperatures, causing it to recover some hydration capacity. The recommended range of temperatures for the thermal treatment of HCP is 500 to 800°C. Use of higher temperatures is detrimental.
- (b) For HCP to be used as a recycled cementitious material, it is necessary for its fineness to be high enough to pass 75  $\mu m$  sieve. Relative to ordinary Portland cement, DCP gives a short setting time, has a higher water demand, and generally gives lower strength.
- (c) Recycled aggregates typically exhibit high water absorption values of 5 to 7%, owing to the presence of adhered mortar at surfaces of the aggregates. When recycled aggregates are used in concrete, the adhered mortar and the associated existing interfacial transition zone (ITZ) along with the new ITZ, form a triple layered weak zone (3ITZ). It is suggested that this relatively weaker and thicker 3ITZ is majorly responsible for the inferior properties of concrete made with recycled aggregates.

- (d) Mechanical abrasion and/or thermal treatment methods are the most effective techniques for improving the properties of recycled aggregates. These techniques also possess the merit of simplicity, are least costly and non-hazardous. Thermal treatment of recycled aggregates at temperatures exceeding 500°C can be detrimental to properties of the treated aggregates.
- (e) While the cost of non-treated recycled aggregates is lower than that of natural aggregates, treatment of the former tends to elevate its cost to levels somehow above those for the natural aggregates. However, a majority of the methods including the abrasion and thermal treatment techniques, remain competitive and feasible.

## Acknowledgement

The study presented in this paper was supported by the National Research Foundation (NRF) of South Africa, IPRR Grant No. 96800. The authors are grateful for the financial support given by NRF.

## References

- Akbarnezhad A, Ong K, Tam C, Zhang M (2013) Effects of the parent concrete properties and crushing procedure on the properties of coarse recycled concrete aggregates. *Journal of Materials in Civil Engineering* 25: 1795–1802.
- Akbarnezhad A, Ong KCG, Zhang MH, Tam CT, Foo TWJ (2011) Microwave-assisted beneficiation of recycled concrete aggregates. *Construction and Building Materials* 25: 3469–3479.
- Al-Bayati HKA, Das PK, Tighe SL, Baaj H (2016) Evaluation of various treatment methods for enhancing the physical and morphological properties of coarse recycled concrete aggregate. Construction and Building Materials 112: 284–298.
- Alonso C, Fernandez L (2004) Dehydration and rehydration processes of cement paste exposed to high temperature environments. *Journal of Material Science* 39: 3015– 3024.
- Benjamin K (2004) Performance of concrete made with commercially produced recycled coarse and fine aggregate in Cape Peninsula Master's Thesis, Department of Civil Engineering, University of Cape Town
- Bester J, Kruger D, Miller B. (2017) South African construction and demolition waste procedure and its sourced material effects on concrete. *MATEC Web of conferences* **120**, 02008; 2017.
- Bordy A, Younsi A, Aggoun S, Fiorio B (2017) Cement substitution by a recycled cement paste fine: role of the residual anhydrous clinker. *Construction and Building Materials* 132: 1–8.

- BS 882 (1992) Specification for aggregate from natural sources for concrete. British Standards Institution (BSI), London, UK.
- Çakır O (2014) Experimental analysis of properties of recycled coarse aggregate concrete with mineral additives. *Construction and Building Materials* **68** (3): 17–25.
- Castellote M, Alonso C, Andrade C, Turrillas X, Campo J (2004) Composition and microstructural changes of cement pastes upon heating, as studied by neutron diffraction. *Cement and Concrete Composites* **34**: 1611–1644.
- Castellote M, Fernandez L, ndrade C, Alonso C (2009) Chemical changes and phase analysis of ordinary Portland cement pastes carbonated at different carbon dioxide concentrations. *Materials and structures* 42(4): 515-525.
- De Bito J, Saikia JN (2013) Recycled aggregate in concrete: use of industrial, construction and demolition waste, Springer.
- De Brito J, Bizinotto MB, Ferreira L (2011) influence of the pre-saturation of recycled coarse concrete aggregates on concrete properties. *Magazine of Concrete Research* **63(8):** 617 627.
- De Brito J, Ferreira J, Pacheco J, Soares D, Guerreiro M (2016) Structural, material, mechanical and durability properties and behaviour of recycled aggregates concrete, *Journal of Building Engineering* **6**: 1–16.
- De Muynck W, De Belie N, Verstraete W (2010) Microbial carbonate precipitation in construction materials: a review. *Ecological Engineering* **36(2)**: 118–136.
- Dhir R.K., Henderson N., and Limbachiya M.C. (1998) Use of recycled concrete aggregate, Thomas Telford publishing, United Kingdom, 1998.
- Eckert M. and Oliveira M. (2017) Mitigation of the negative effects of recycled aggregate water

absorption in concrete technology. Construction and Building Materials 133: 416-424.

- Ekolu SO (2016) A review on effects of curing, sheltering, and carbon dioxide concentration upon natural carbonation of concrete. *Construction and Building Materials* **127**: 306–320.
- Ekolu SO (2018) Model for practical prediction of natural carbonation in reinforced concrete: Part 1-formulation. *Cement and Concrete Composites* **86**:40-56.
- Ekolu SO, Makama LN, Shuluuka WP (2012) Influence of different recycled aggregate types on strength and abrasion resistance properties of concrete. *Proceedings 3<sup>rd</sup> International Conference on Concrete Repair, Rehabilitation and Retrofitting*. Cape Town, South Africa 193-198.
- Ekolu SO, Thomas MDA, Hooton RD (2006) Studies on Ugandan volcanic ash and tuff. Proceedings 1<sup>st</sup> International Conference on Advances in Engineering and Technology. Entebbe, Uganda 75-83.
- Etxeberria M, Vazquez E, Mari A (2006) Microstructure analysis of hardened recycled aggregate concrete. *Magazine of Concrete Research* **58**(**10**): 683 690.
- Fanga C, Long W, Weib J, Xiaoc B, Yand C (2017) Effect of graphene oxide on mechanical properties of recycled mortar. *Materials Science and Engineering* (274) 012144.
- Florea MVA, Ning A, Brouwers HJH (2014) Activation of liberated concrete fines and their application in mortars. *Construction and Building Materials* **50**: 1–12.
- Gokce HS, Simsek O (2013) The effect of waste concrete properties on recycled aggregate concrete properties, *Magazine of Concrete Research* **65**(14): 844 854.

- Gomez-Soberon J (2002) Porosity of recycled concrete with substitution of recycled concrete aggregate: an experimental study. *Cement and Concrete Res*earch **32**: 1301-1311.
- Grabiec AM, Klama J, Zawal D, Krupa D (2012) Modification of recycled concrete aggregate by calcium carbonate bio-deposition. *Construction and Building Materials* 34: 145–150.
- Hameed M (2009) Impact of transportation on cost, energy, and particulate emissions for recycled concrete aggregate. Master's Thesis, Department of Building Construction, University of Florida.
- Hammes F, Verstraete W (2002) Key roles of pH and calcium metabolism in microbial carbonate precipitation. *Reviews in Environmental Science and Bio/Technology* **1**:3–7.
- Hosseini P, Delkash M, Booshehrian A, Ghavami S, Zanjani MK (2009), Use of Nano-SiO<sub>2</sub> to improve microstructure and compressive strength of recycled aggregate concretes, Book: *Nanotechnology in Construction*, 3, Pages 215 221.
- Ismail S, Ramli M (2013) Effect Surface treatment of recycled concrete aggregate on properties of fresh and hardened concrete. *IEEE Business Engineering and Industrial Applications Colloquium* 651-656.
- Ismail S, Ramli M (2013) Engineering properties of treated recycled concrete aggregate for structural applications. *Construction and Building Materials* **44**: 464–476.
- Juan DMS, Gutiérrez PA (2009) Study on the influence of attached mortar content on the properties of recycled concrete aggregate. *Construction and Building Materials* 23: 872–877.
- Katz A (2004) Treatments for the improvement of recycled aggregate. *Journal of Materials in Civil Engineering* **16(6)**: 597–603.
- Katz A (2009) Properties of concrete made with recycled aggregate from partially hydrated old concrete. *Cement and Concrete Research* **33**: 703-711.
- Khatib JM (2005) Properties of concrete incorporating fine recycled aggregate, *Cement and Concrete Research* **35** (4): 763–769.
- Kim H, Park S, Kim H (2016) The optimum production method for quality improvement of recycled aggregates using sulfuric acid and the abrasion method. *International Journal of Environmental Research and Public Health* 13: 769–783.
- Kong D, Lei T, Zheng J, Ma C, Jiang J, Jiang J (2010) Effect and mechanism of surface-coating pozzolanics materials around aggregate on properties and interfacial transition zone microstructure of recycled aggregate concrete. *Construction and Building Materials* 24: 701– 708.
- Kou SC, Poon CS (2010) Properties of concrete prepared with PVA-impregnated recycled concrete aggregates, *Cement and Concrete Composites* **32**: 649–654.
- Kou SC, Poon CS, Etxeberria M (2011) Influence of recycled aggregates on long term mechanical properties and pore size distribution of concrete. *Cement and Concrete Composites* **33**: 286–291.
- Kumar GS, Minocha AK 92018) Studies on thermo-chemical treatment of recycled concrete fine aggregates for use in concrete. *Journal of Material Cycles and Waste Management* 20: 469–480.

- Lei B, Zou J, Rao C, Fu M, Xiong J, Wang X (2006) Experimental study on modification of recycled concrete with graphene oxide. *Journal Building Structures* (**37**): 103–108..
- Li W., Long C., Tam V.W., Poon C-S, and Duan W.H (2017) Effects of nano-particles on failure process and microstructural properties of recycled aggregate concrete. *Construction and Building Materials* 142:42–50.
- Long W-J., Zheng D., Duan H-b, Han N. and Xing F. (2018) Performance enhancement and environmental impact of cement composites containing graphene oxide with recycled fine aggregates. *Journal of Cleaner Production* **194**, 193-202
- Luo Z., Li W., Tam V.W.Y., Xiao J. and Shah S.P. (2018) Current progress on nanotechnology application in recycled aggregate concrete *Journal of Sustainable Cement-Based Materials*, **0(0)**, 1–18.
- Marcela F (2007) Hydration of belite cements prepared from recycled concrete Residues, *Ceramics-Silikaty* **51**: 45–51.
- Matias D, De Brito J, Rosa A, Pedro D (2013) Mechanical properties of concrete produced with recycled coarse aggregates influence of the use of superplasticizers. *Construction and Building Materials* **44**: 101–109.
- Meyer C (2009) The greening of the concrete industry. *Cement and Concrete Composites* **31**: 601-605
- Morita RY (1980) Calcite precipitation by marine bacteria. *Geomicrobiology Journal* **2(1)**: 63–82.
- Mukharjee B.B, and Barai S.V. (2017) Mechanical and microstructural characterization of recycled aggregate concrete containing silica nanoparticles. *Journal of Sustainable Cement-Based Materials* **6**(1): 37–53.
- Mukharjee B.B. and Barai S. (2014) Influence of nano-silica on the properties of recycled aggregate concrete, *Construction and Building Materials* **55**, 29–37.
- Mukharjee BB, Barai SV (2014) Influence of Nano-Silica on the properties of recycled aggregate concrete, *Construction and Building Materials* **55**: 29–37.
- Novoselov KS, Geim AK, Morozov SV, Jiang D, Zhang Y, Dubonos SV, Grigorieva IV, Firsov AA (2004) Electric field effect in atomically thin carbon films, *Science* **306** (**5696**): 666–669.
- Paine KA, Dhir RK (2010) Recycled aggregate in concrete: a performance-relate approach, *Magazine of Concrete Research* 62(7): 519 – 530.
- Pandurangan K, Dayanithy A, Prakash SO (2016) Influence of treatment methods on the bond strength of recycled aggregate concrete. *Construction and Building Materials* 120: 212–221.
- Qiu J, Tng DQS, Yang EH (2014) Surface treatment of recycled concrete aggregates through microbial carbonate precipitation. *Construction and Building Materials* **57**: 144–150.
- Saravanakumar P, Abhiram K, Manoj B (2016) Properties of treated recycled aggregates and its influence on concrete strength characteristics. *Construction and Building Materials* 111: 611 – 617.

- Serpell R, Lopez M (2013) Reactivated cementitious materials from hydrated cement paste wastes, *Cement and Concrete Composites* **39**: 104–114.
- Serpell R, Lopez M (2015) Properties of mortars produced with reactivated cementitious materials, *Cement and Concrete Composites* **64**: 16–26.
- Shaikh F, Chavda V, Minhaj N, and Arel H.S (2018) Effect of mixing methods of nano-silica on properties of recycled aggregate concrete. *Structural Concrete* **19**(**2**): 387-399
- Shaikh FUA, Odoh H, Than AB (2015) Effect of nanosilica on properties of concrete containing recycled coarse aggregate. Proceedings of the Institution of Civil Engineers – Construction Materials 168(2): 68 – 76.
- Shannag M (2000) High strength concrete containing natural pozzolan and silica fume, *Cement and Concrete Composites* 22(6): 399 406.
- Sharma S, Arora S (2018) Economical graphene reinforced fly ash cement composite made with recycled aggregates for improved sulphate resistance and mechanical performance. *Construction and Building Materials* **162**: 608–612.
- Shi C, Wu Z, Cao Z, Ling TC, Zheng J (2018) Performance of mortar prepared with recycled concrete aggregate enhanced by carbon dioxied and pozzolan slurry. *Cement and Concrete Composites* 86: 130–138.
- Shi C, Yake LY, Zhang J, Li W, Chong L, Xie Z (2016) Performance enhancement of recycled concrete aggregate a review, *Journal of Cleaner Production* **112**: 466–472.
- Shi-Cong K, Bao-jian Z, Chi-Sun P (2014) Use of a carbon dioxide curing step to improve the properties of concrete prepared with recycled aggregates. *Cement and Concrete Composites* 45: 22–28.
- Shui Z, Xuan D, Wan H, Cao B (2008) Rehydration reactivity of recycled mortar from concrete waste experienced to thermal treatment. *Construction and Building Materials* **22**:1723–9.
- Shui ZH, Xuan DX, Chen W, Yu R, Zhang R (2009) Cementitious characteristics of hydrated cement paste subjected to various dehydration temperatures. *Construction and Building Materials* 23: 531–537.
- Silva R.V., de Brito J., and Dhir R.K. (2017) Availability and processing of recycled aggregates within the construction and demolition supply chain: a review. *Journal of Cleaner Production*, 143: 598-614.
- Singh L., Bisht V., Aswathy M., Chaurasia L. and Gupta S. (2018) Studies on performance enhancement of recycled aggregate by incorporating bio- and nano-materials. *Construction and Building Materials* 181:217–226.
- Song X, Qiao P, Wen H (2015) Recycled aggregate concrete enhanced with polymer aluminium sulfate, *Magazine of Concrete Research* **67**(10): 496 502.
- Spaeth V, Tegguer AD (2013) Improvement of recycled concrete aggregate properties by polymer treatments. *International Journal of Sustainable Built Environment* **2**: 143–152.
- Tam VM, Tam CM (2008) *Re-use of construction and demolition waste in housing developments*, 1<sup>st</sup> Edition; Nova Science Publishers: New York, NY, USA.
- Tam VWY (2008) Economic comparison of concrete recycling: A case study approach. *Resources, Conservation and Recycling* **52**: 821–828.

- Tam VWY, Tam CM, Le KN (2007) Removal of cement mortar remains from recycled aggregate using pre-soaking approaches. *Resources, Conservation and Recycling* 50(1): 82-101.
- Tateyashiki H, Shima H, Matsumoto Y, Koga Y (2001) Properties of concrete with high quality recycled aggregate by heat and rubbing method. *Proceedings of Japan Concrete Institute* **23**(2): 61-66.
- Thiery M, Villain G, Dangla P, Platret G (2007) Investigation of the carbonation front shape on cementitious materials: effects of the chemical kinetics, *Cement and Concrete Research* **37**(**7**): 1047–1058.
- Wang J, Vandevyvere B, Vanhessche S, Schoon J, Nico BN, De Belie N (2017) Microbial carbonate precipitation for the improvement of quality of recycled aggregates, *Journal of Cleaner Production* **156**: 355 366.
- Wang LG, Zheng DP, Zhang SP, Cui HZ, Li DX (2016) Effect of Nano-SiO<sub>2</sub> on the hydration and microstructure of Portland cement. *Nanomaterials* 6: 241.

Xiao JZ (2012) Recycled Concrete. Chinese Building Construction Publishing Press, Beijing;

- Xuan D, Zhan B, Poon CS (2016) Assessment of mechanical properties of concrete incorporating carbonated recycled concrete aggregates. *Cement and Concrete Composites* 65: 67–74.
- Younis K, Mustafa (2018) Feasibility of using nanoparticles of  $SiO_2$  to improve the performance of recycled aggregate concrete. *Advances in Material Science and Engineering* **18**:1 11.
- Yu R, Shui Z (2014) Efficient reuse of the recycled construction waste cementitious materials. *Journal of Cleaner Production* **78**: 202–207.
- Zega CJ, Di Maio AA (2011) Use of recycled fine aggregate in concretes with durable requirements, *Waste Management* **31**: 2336–2340.
- Zhan B, Poon CS, Liu Q, Kou S, Shi C (2014) Experimental study on carbon dioxide curing for enhancement of recycled aggregate properties. *Construction and Building Materials* 67: 3–7.
- Zhan M, Pan G, Wang Y, Fu M, Lu X (2019) Recycled aggregate mortar enhanced by microbial calcite precipitation. *Magazine of Concrete Research*, https://doi.org/10.1680/jmacr.18.00417.
- Zhang H., Zhao Y., Meng T., and Shah S. (2015) The modification effects of a nano-silica slurry on microstructure, strength, and strain development of recycled aggregate concrete applied in an enlarged structural test. *Construction and Building Materials* **95**: 721–735.
- Zhang J, Shi C, Li Y, Pan X, Poon CS, Xie Z (2015) Influence of carbonated recycled concrete aggregate on properties of cement mortar. *Construction and Building Materials* 98(15): 1–7.
- Zhang J, Shi C, Li Y, Pan X, Poon CS, Xie Z (2015) Performance Enhancement of Recycled Concrete Aggregates through Carbonation. *Journal of Materials in Civil Engineering* 29: 1-7.
- Zhang L, Ji Y, Huang G, Li J, Hu Y (2018) Modification and enhancement of mechanical properties of dehydrated cement paste using ground granulated blast-furnace slag, *Construction and Building Materials* 164: 525–534.