OPTIMIZING CARBONATION CONDITIONS FOR LOW-QUALITY RECYCLED CONCRETE AGGREGATES AND SUSTAINABLE VALORIZATION IN CEMENT-BASED SYSTEMS

DÜŞÜK KALİTELİ GERİ DÖNÜŞÜM BETON AGREGASININ OPTİMUM KARBONATLAŞTIRMA KOŞULLARININ BELİRLENMESİ VE ÇİMENTO BAĞLAYICILI SİSTEMLERİN SÜRDÜRÜLEBİLİRLİĞİNE ETKİSİ

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ABSTRACT

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This study aims to examine the parameters affecting the accelerated carbonation performance of low-quality recycled concrete aggregates (RCA), obtained from end-of-life buildings, with low mechanical and chemical properties, and to determine the effects of the accelerated carbonation process on the engineering performance of cement-based systems through a series of performance tests performed on mortar samples produced with carbonated RCA (CRCA), uncarbonated RCA and natural aggregate (NA) under the optimum parameters determined in the first section. The accelerated carbonation process was implemented using a newly designed laboratory-scale carbonation reactor that provides a dynamic carbonation process. Relative humidity (50-70-90%), ambient pressure (1-2-3 bar) and ambient temperature (50-90ºC) were determined as the main parameters for the accelerated carbonation process. Carbonation treatments were applied to low-quality RCAs with five different particle size ranges: <0.85, 0.85-2.00, 2.00-4.75, 4.75-9.50 and 9.50-14.00 mm. The effects of carbonation durations (2-4-6-12-24-48-72-96-120 hours) on the carbonation performance of RCAs were also investigated. Thermogravimetric analysis (TG/DTA) was applied to measure the CO_2 uptake rate of low-quality RCAs, and the water absorption capacities of CRCAs and RCAs were measured to obtain information about precipitated calcium carbonate. As a result of examining all parameters, the optimum carbonation condition was determined as 70% relative humidity, 1 bar ambient pressure, 50°C ambient temperature, 48 hours carbonation duration. The highest CO_2 uptake rate occurred in the particle size range of 2.00-4.75 mm. In the continuation of this study, flowability, mechanical strength, water absorption capacity, capillary water absorption (sorptivity), freezing-thawing and chloride permeability properties of mortar samples produced with fine (0.85-4.75 mm) RCA, CRCA and NA were determined. The results showed that the losses in engineering properties observed in mortars when using RCA can be significantly eliminated by the accelerated carbonation method. The main reason underlying the gains brought by the use of CRCA is the improved chemical and mechanical properties of the high-void, high-permeable, low-density mortar residue and the interfacial transition zone (ITZ) on the RCA surface. Results showed that CRCA, rather than remaining only as a low-quality construction and demolition waste (CDW), can be actively used in cement-based systems to reduce their negative environmental impacts. In this way, the sustainability of cement-based systems produced with CRCA can be contributed.

Keywords: Construction and Demolition Waste, Recycled Concrete Aggregate, Accelerated Carbonation, Mechanical Performance, Durability Performance.

ÖZET

DÜŞÜK KALİTELİ GERİ DÖNÜŞÜM BETON AGREGASININ OPTİMUM KARBONATLAŞTIRMA KOŞULLARININ BELİRLENMESİ VE ÇİMENTO BAĞLAYICILI SİSTEMLERİN SÜRDÜRÜLEBİLİRLİĞİNE ETKİSİ

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Bu çalışma, ömrünü tamamlamış binalardan elde edilen, düşük mekanik ve kimyasal özelliklere sahip, düşük kaliteli geri dönüşüm beton agregalarının (GDA) hızlandırılmış karbonatlaştırma performansını etkileyen parametrelerin incelenmesine, belirlenen optimum parametreler altında karbonatlaştırılmış GDA (KGDA), işlenmemiş GDA ve doğal agrega (DA) ile üretilen harç numuneleri üzerinde gerçekleştirilen bir dizi performans testi ile hızlandırılmış karbonatlaştırma prosesinin çimento bazlı sistemlerin mühendislik performansına olan etkilerinin belirlenmesine odaklanmaktadır. Hızlandırılmış karbonatlaştırma prosesi, dinamik bir karbonatlaştırma işlemi sağlayan laboratuvar ölçekli bir karbonatlaştırma reaktörü kullanılarak uygulanmıştır. Hızlandırılmış karbonatlaştırma prosesi için bağıl nem (%50-70-90), ortam basıncı (1-2-3 bar) ve ortam sıcaklığı (50-90^{o}C) ana parametreler olarak belirlenmiştir. <0.85, 0.85-2.00, 2.00-4.75, 4.75-9.50 ve 9.50-14.00 mm olmak üzere beş farklı parçacık boyutu aralığına sahip düşük kaliteli GDA'lara karbonatlaştırma işlemleri uygulanmıştır. Karbonatlaştırma sürelerinin (2-4-6-12-24-48-72-96-120 saat) GDA'ların karbonatlaşma performansı üzerindeki etkileri de araştırılmıştır. Düşük kaliteli GDA'ların CO_2 yakalama oranını ölçmek için termogravimetrik analiz (TG/DTA), çökelmiş kalsiyum karbonat hakkında bilgi edinmek için de GDA'ların su emme kapasitesinin belirlenmesi yöntemleri uygulanmıştır. Tüm parametrelerin incelenmesi sonucu optimum karbonatlaştırma koşulu %70 bağıl nem, 1 bar ortam basıncı, 50^oC ortam sıcaklığı, 48 saat karbonatlaştırma süresi olarak belirlenmiştir. En yüksek CO_2 yakalama oranı 2.00-4.75 mm tane boyut aralığında gerçekleşmiştir. Bu çalışmanın devamında, ince (0.85-4.75 mm) GDA, KGDA ve DA ile üretilen harç numunelerinin akma, mekanik dayanım, su emme kapasitesi, kılcal su emme, donma-çözülme ve klorür iyonu geçirgenlik özellikleri belirlenmiştir. Sonuçlar, GDA kullanımında harçlarda gözlenen mühendislik özelliklerindeki kayıpların hızlandırılmış karbonatlaştırma yöntemi ile önemli ölçüde giderilebileceğini göstermiştir. KGDA kullanımının getirdiği kazanımların altında yatan temel neden, GDA yüzeyindeki yüksek boşluklu, yüksek geçirgen, düşük yoğunluklu harç kalıntısının ve arayüzey geçiş bölgesinin (ITZ) geliştirilmiş kimyasal ve mekanik özellikleridir. Çalışma sonuçları, GDA'nın, yalnızca düşük kaliteli bir inşaat ve yıkım atığı (CDW) olarak kalması yerine, çimento bazlı sistemlerde, bu sistemlerin olumsuz çevresel etkilerini azaltmak için aktif olarak kullanılabileceğini göstermiştir. Bu sayede KGDA ile üretilen harç veya betonların sürdürülebilirliğine katkı sağlanabilir.

Anahtar Kelimeler: İnşaat ve Yıkıntı Atıkları, Geri Dönüşüm Beton Agregası, Hızlandırılmış Karbonatlaştırma, Mekanik Performans, Durabilite Performansı.

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SYMBOLS AND ABBREVIATIONS

Symbols

CO_2-e - Carbon Emission Value

CH - Calcium Hydroxide

CSH - Calcium Silicate Hydrate

CC - Calcium Carbonate

Abbreviations

PC - Portland Cement

NA - Natural Aggregate

RA - Recycled Aggregate

RCA - Recycled Concrete Aggregate

CRCA - Carbonated Recycled Concrete Aggregate

CDW - Construction and Demolition Waste

CCS - Carbon Capture and Storage

CCU - Carbon Capture and Utilization

CCUS - Carbon Capture, Utilization or Storage

CLC - Chemical Looping-based Combustion

CLR - Chemical Looping-based Reformation

SCM - Supplementary Cementitious Materials

PVC - Polyvinyl chloride

LOI - Loss of Ignition

RAC - Recycled Aggregate Concrete

NAC - Natural Aggregate Concrete

ITZ - Interfacial Transition Zone

SEM - Scanning Electron Microscopy

TG/DTA – Thermogravimetric and Differential Thermal Analysis

XRD - X-ray Diffraction Analysis

EDX - Energy Dispersive X-ray

RH - Relative Humidity

1. INTRODUCTION

1.1. Problem Definition

Concrete is the most widely used building material in the construction industry worldwide, and Portland Cement (PC) and natural aggregate (NA) are the main ingredients used for its production. Globally, concrete production increases with a growing need for shelter/infrastructure/superstructure. Although the usage of concrete is beneficial regarding the quantity produced, and cost yet has an undeniable environmental burden. Its detrimental effects can be mainly classified under two headings, namely PC production-related significant amount of CO_2 emission and overuse of natural sources (e.g., clinker, aggregate, gypsum, etc.). In total, cement production accounts for about 5-8% of global anthropogenic CO_2 emissions because of both fossil fuel combustions and the conversion reaction of $CaCO_3$ to CaO (Metz et al., 2005; Kajaste and Hurme, 2016). As the main aspect, concrete production requires different-size aggregates whose production process is also highly energy-intensive, creating significant amounts of waste next to the depletion of natural resources (Pandurangan et al., 2016). Given the negative effects of concrete production, special attention requires to pay on reducing concrete production related-CO_2 footprint and protecting natural resources.

Negative impacts of concrete are coupled with insufficient longevity of traditional structures, having a high deterioration tendency under combined mechanical/environmental effects, demanding demolition/reconstruction activities, all of which create waste concrete/masonry/glass collectively termed as “Construction and Demolition Waste” (CDW). These wastes, which are stored in empty clean lands outside the city, have harmful effects such as emitting harmful substances, polluting the groundwater, and disrupting the relevant ecosystem (European Commission, 2011). CDW industry is one of the largest global solid waste producers and accounts for 30–40% of the total urban waste generation (Wang et al., 2017). Worldwide, the amount of annually produced CDWs accounts for more than 10 billion tons (Wu et al., 2019). Therefore, the generation of CDW can be regarded as a global issue requiring a relatively high demand for proper handling.

In general, CDW has a heterogenous composition and consists of different types of material, including concrete, ceramic, plastic, metal, glass, brick, and timber (Zheng et al., 2017). However, concrete waste has a large share of the CDWs, which accounts for 40–85% of the total amount of CDW, varying on the type of demolished structures (Rimoldi, 2010). Currently, a high portion of generated concrete waste is used for low-value-added applications, such as land fillings of road bases or non-structural filling. To valorize the waste concrete in high-value-added applications, researchers made great efforts to use concrete waste as a recycled concrete aggregate (RCA) in concrete production after several processes (e.g., crushing, sieving, etc.) (Nguyen et al., 2021; Rahal and Hassan, 2021; Zou et al., 2021). RCA, obtained by subjecting construction demolition waste to various grinding processes, has been used in cementitious composites without any process in literature and its various properties have been determined. In addition, they are also used in some engineering applications such as non-structural building elements, minor embankments or highway embankments. When used as in the production of structural elements, RCA cause some problems. Hydrated mortar residues remaining on the surfaces of RCA cause strength losses or excess water requirement in the final product.

For this reason, some studies in the literature show that studies have been carried out on adding these aggregates to the mixture after being processed. Examples of methods are listed below.

- pre-wetting (González-Fonteboa et al., 2011)
- mechanical abrasion (Yanagibashi et al., 2005)
- heating-abrasion (Kuroda et al., 2005)
- microwave heating (Choi et al., 2014)
- acid/sodium silicate solution-based treatments (Shi et al., 2016; Mistri et al., 2020)
- microbial-induced treatment (Sonmez et al., 2021)

Although a certain amount of improvement has been achieved with these methods in general. It is obvious that some negative effects such as high energy need, additional concrete dust etc. are also observed. Therefore, it is necessary to seek improvement with an alternative method. For this purpose, in this thesis study, it is aimed to use the method also known as accelerated carbonation (active carbonation, carbonation cure) to improve RCA.

1.2. Scope and Objectives

Increasing anthropogenic CO_2 emissions, generation of CDWs, and the need for natural sources are the main factors that lead to find solutions for managing CO_2 emissions and procurement of secondary raw materials via upcycling of CDW-based RCA. It is estimated that millions of buildings will be affected by the urban renewal law in Turkey. In addition, as a result of the devastating earthquakes that took place in Kahramanmaraş, Turkey on February 6, 2023, many buildings were destroyed and a significant amount of CDW was released. For this reason, in this thesis, a number of experimental studies were carried out for the purposes of first evaluating the CO_2, which is released from Portland cement production to the atmosphere and second reusing of CDW as RCA arising from urban transformation activities and natural disasters (Figure 1.1).

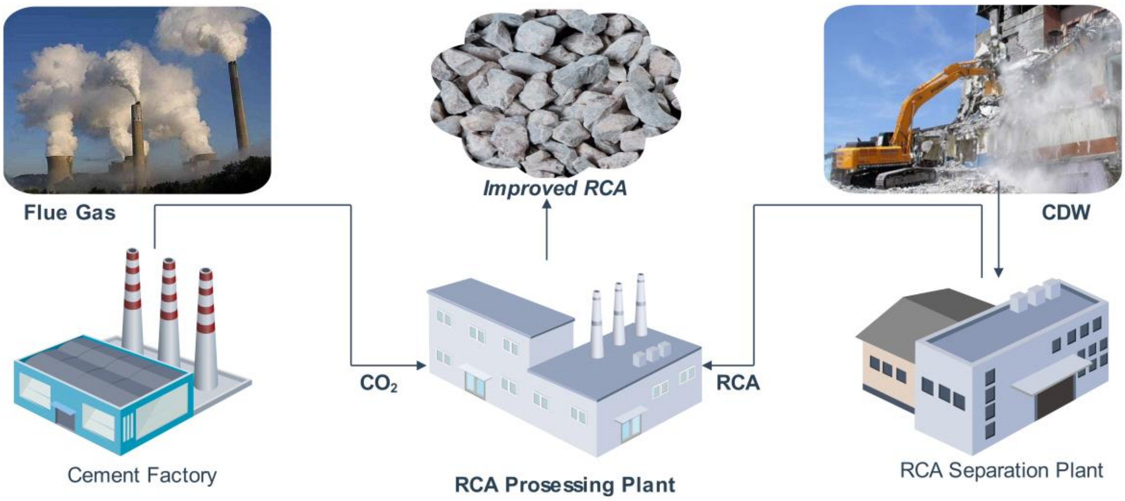


Figure 1.1 Shematic view of objective of thesis study

Also, large amounts of concrete waste having a compressive strength of less than 10 MPa will be released. This causes the RCA obtained from these wastes to be of low quality.

Improving the characteristics of the RCA from low quality concrete wastes has been one of the objectives of the thesis study. As seen in the Figure 1.1, the thesis study represents the concept of improving the properties of the low quality RCA coming from the RCA separation plant, as a result of the use of CO_2 with accelerated carbonation in the RCA processing plant.

Thus, this study focused two main targets:

1. Comprehensive investigation of parameters affecting the efficiency of accelerated carbonation of low-quality RCA from end-of-life buildings was carried out. Relative humidity (50%-70%-90%), temperature (50 ^oC and 90 ^oC), ambient pressure of carbonation chamber (1, 2, 3 bar), particle size (<0.85, 0.85-2, 2-4.75, 4.75-9.5, 9.5-14 mm), and curing duration (2h, 4h, 6h, 12h, 24h, 48h, 72h, 96h, and 120h) were determined as variables for the carbonation process implemented with a newly designed rotary carbonation reactor. It was aimed to increase the efficiency of carbonation procedures and enhance the properties of low-quality RCA through accelerated carbonation.
2. An answer was sought whether the disadvantages caused by RCA in terms of engineering performance could be eliminated by improving RCA with the accelerated carbonation method. Studies in the literature that comprehensively address multiple mechanical and durability effects are very limited. In one of them, it was found that there were very few studies examining the durability effects on samples produced with fine carbonated RCA (CRCA) and coarse CRCA; in fact, it has been stated that there are no studies that combine the two (Liang et al., 2019). It has been emphasized that there are very limited studies in the literature with different results in terms of durability, especially gas and chloride ion permeability, and therefore detailed and comprehensive studies should be carried out in terms of durability (Li et al., 2022). On the other hand, it is understood that the effect of carbonation has more impact on durability properties rather than mechanical properties (Zhan et al., 2014). For these reasons, this study also focused on effects in terms of durability along with mechanical effects.

1.3. Thesis Outline

The thesis study consists of five main chapters as summarized below.

In the first chapter, the scope of the subject covered in the thesis, the definition of the problem and the points where the subject differs from the literature are summarized.

In the second part, a summary of the literature related to the subject of the thesis is discussed. The subject of the thesis is presented from general to specific with examples from the literature. In this context, starting from the topic of carbon capture, utilization and storage, remarkable information from the literature was shared on the carbon footprint of PC, and CDW. In the following, RCA and their general mechanical and durability performances are mentioned. Then, in this chapter, the techniques found in the literature to improve the properties of RCA are mentioned and this chapter is completed by summarizing the studies in the literature on the subject of accelerated carbonation discussed within the scope of the thesis.

The third chapter covers the descriptions of the raw materials, sample preparation methods, test setups and techniques used within the scope of the thesis. In this section, details of how RCA is obtained and the subsequent accelerated carbonation process are given.

In the fourth chapter, the results of the study on determining the optimum carbonation conditions of recycled aggregate, which is the first stage of the thesis study, are given. Next, the results of the experiments carried out to determine the various engineering properties of mortar samples produced with CRCA under optimum conditions are examined.

In the fifth and last chapter, the remarkable results of the experiments discussed within the scope of the thesis are summarized and the prominent findings are highlighted.

2. LITERATURE REVIEW

2.1. Carbon Capture, Utilisation and Storage Techniques

It is estimated that the consumption of fossil fuels, one of the biggest factors causing global climate change, will continue for at least 50 years and therefore the climate change will worsen (EIA, 2015). In the coming years, fossil fuel-induced climate change is expected to deepen further as a result of this use, with the expected increase in the world population (Al-Mamoori et al., 2017). Increasing CO_2 emissions due to factors such as economic growth and population growth of countries is proven by measurements of annual CO_2 emissions and global warming values. Therefore, one of the greatest needs of the current century is to capture and utilize industrially produced CO_2 before it is released into the atmosphere. This requirement is among the popular and the most challenging topics addressed in scientific studies. When looked at industrial scale, it seems that the integration of large-scale facilities or power plants that are responsible for CO_2 emissions is not yet suitable for the revision of CO_2 capture (Andrew et al., 2010).

Intergovernmental organizations are also taking action to prevent increasing global warming and resulting climate changes. In this context, the Paris Climate Agreement was signed in 2015 within the scope of the United Nations Framework Convention on Climate Change (UNFCCC). In accordance with this agreement, countries participating in the agreement are expected to reduce CO_2 emissions and finance studies in this direction starting from 2020. The prepared agreement was signed by 197 UNFCCC member countries and came into force in 2016 (European Commission, 2018).

In order to eliminate CO_2 emitted from large-scale production facilities such as power plants, cement and steel factories, the most applicable and common methods in the context of climate change and carbon management are the capture, utilization or storage of CO_2 (CCUS). One of these methods, the carbon capture and storage (CCS) method, involves capturing the produced CO_2 at its source, bringing it to a supercritical state after separating it from other waste gases, and storing it in geological formations such as oceans and drained oil and natural gas wells (Boot-Handford et al., 2014). It was also stated in the report published by the International Energy Agency (IEA) in 2013 that CCS techniques will make a great contribution to reducing CO_2 emissions resulting from industrial production. On the other hand, although storing CO_2 through various technological processes seems to be an effective and shortcut solution, the possible negative effects of disposing of CO_2 with this method on water resources and other underground resources remain unclear. In addition, bringing CO_2 to a supercritical state as a result of various processes creates a very intense energy need (Andrew et al., 2010). Additionally, another problem encountered in the CCS method is the need for storage space (IPCC 2005).

Carbon capture and utilization (CCU) techniques involve converting CO_2 resulting from industrial production into value-added products or utilizing it as raw material, instead of storing it in deep areas in different geological formations (Aresta 2010; Bilgen 2016). In recent years, great interest has been shown in CCU techniques, which offer a more environmentally friendly approach than CCS. CO_2 is used directly in areas such as water purification, food production and preservation, the beverage industry, polymer synthesis processes, urea production or fire extinguishing (Figure 2.1) (Boot-Handford et al., 2014; Aresta, 2003). However, considering the amount of CO_2 produced, the CO_2 need of sectors where CO_2 is used directly remains at low levels. In fact, it is seen that approximately 1% of the amount of CO_2 produced as waste is used in these sectors (Hu et al., 2013; Aresta et al., 2014).

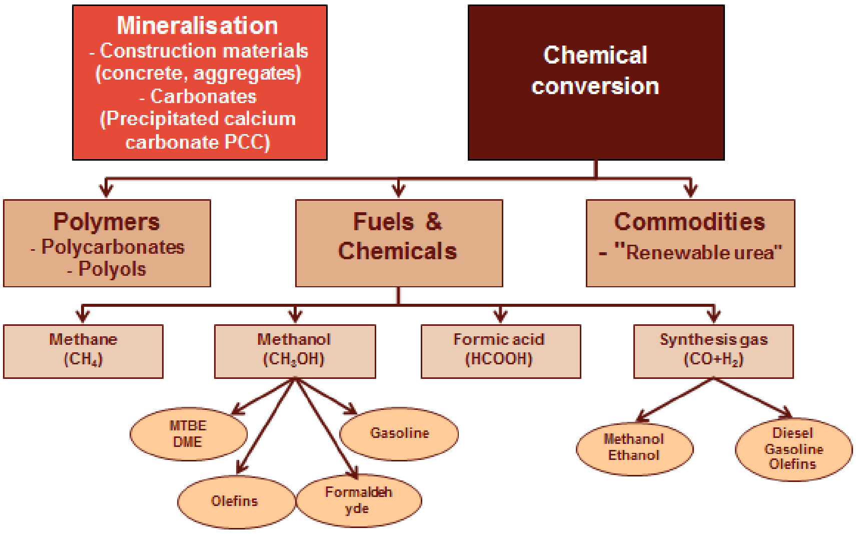


Figure 2.1 Potential CCU areas and techniques (European Commission, 2018)

Figure 2.2, which compares the global CO_2 emissions reached in 2017 with the targeted CO_2 emissions by 2050, shows the current and long-term predicted CCU rates (European Commission, 2018). In the long term, it is predicted that approximately 1-2 Gt/y of the targeted 8 Gt/y emission rate can be eliminated with CCU. Likewise, it is understood that current utilization rates are far below this prediction (Aresta et al., 2013; Assen et al., 2016). As a result, the proliferation of CCU techniques appears to be due to the increase of large-scale applications involving chemical and biological processes that directly require CO_2. (Andrew et al., 2010). The subject discussed in this thesis study proposes a new application of CCU techniques and has the potential to contribute to the widespread use of CCU.

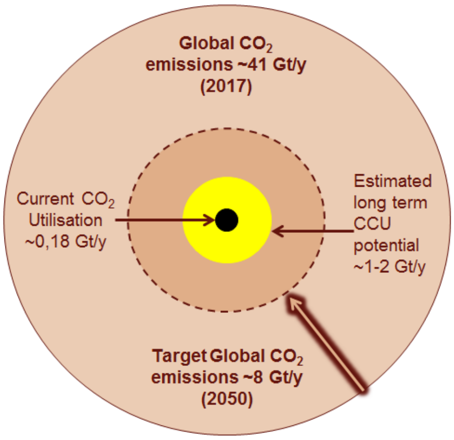


Figure 2.2 Current status of global CO_2 emissions and CCU (European Commission, 2018)

Although CCU has a more environmentally friendly potential than CCS, it is very difficult to capture CO_2 in high purity, convert it through various chemical processes, and use it due to its stable thermodynamic nature. Chemically, CO_2 appears to be a very thermodynamically stable molecule. In order to react with other chemicals, a certain amount of energy may be required under different conditions (Styring et al., 2011). In terms of turning CO_2 into a sustainable raw material, the source of waste CO_2, the applied capture technology and the diversity of the sector that produces CO_2 as waste are important factors (Al-Mamoori et al., 2017). The CO_2 capture process requires energy needs at different levels from sector to sector. For example, since there is a relatively low concentration of CO_2 in different waste gases in cement production or thermal power plants, more energy can be spent to capture CO_2. On the other hand, since the waste CO_2 generated in sectors such as ethanol production is of relatively higher purity, the energy consumption of the capture process in these processes is lower (Abotalib et al., 2016).

2.1.1. CO_2 Capture Processes

In both CCS and CCU techniques, various capture processes are primarily applied. These vary depending on the source where CO_2 will be captured and the area of use to be adapted afterwards.

2.1.1.1. Absorption-based CO_2 Capture:

This method, which can be applied both before and after combustion, is generally used widely in the chemical and petroleum sectors. In this method, physical and chemical solvents are used for CO_2 separation (Markewitz et al., 2012). Amine solvents such as monoethanolamine, aqueous ammonia and N-methyldiethanolamine can be used for pre-combustion, and alkaline solvents such as NaOH and $Ca(OH)_2$ are used in chemical absorption processes for post-combustion stages, especially in sectors such as energy, steel, petroleum and cement (Kong et al., 2016; Arstad et al., 2008; Builes et al., 2015; Geoppert et al., 2011). Although the absorption performed in this method has high efficiency, the energy requirement is high (Markewitz et al., 2012).

2.1.1.2. Adsorption-based CO_2 Capture

In some sectors, CO_2 is adsorbed and retained by porous solid materials. In this method, two types of processes are used: pressure (PSA) and temperature (TSA) oscillation. The efficiency and preferability of these processes depend on parameters such as the number of beds, physicochemical properties, cycle time, operating temperature or pressure of the adsorbent (Ruthven, 1984; Hauchhum and Mahanta, 2014). In the PSA-based approach, the process is low-cost and offers low energy requirements (Agarwal et al., 2010). However, this is a disadvantage of the PSA method as CO_2 recovery remains at low levels (Hauchhum and Mahanta, 2014). TSA is generally preferred to capture CO_2 after the combustion process (Merel et al., 2008; Chaffee et al., 2007; Marx et al., 2016). However, TSA creates high energy requirements, requires a long cooling phase, and therefore its use on a large scale cannot be widespread. Although it has some advantages over absorption techniques, adsorption applications are still in the research and development phase due to their high cost (Li et al., 2009).

2.1.1.3. Membrane-based CO_2 Capture

Membrane-based techniques, which offer more environmentally friendly and efficient separation and capture compared to previous methods, can be used in gas separation processes. In this process, which aims at absorption by creating a pressure difference, it is necessary to obtain a stable environmental condition. For this reason, the material, morphology, operating conditions and configuration of the membrane used in the process directly affect the efficiency of the CO_2 capture process (Al-Mamoori et al., 2017).

Since flue gases are generally released at low pressure after combustion, the membrane-based separation method does not provide sufficient efficiency. Therefore, they are more suitable for use after the combustion process that takes place under high pressure. The disadvantages of this method include the need to work in multiple stages and multiple cycles. In addition, the energy consumed to provide a sufficient pressure environment and the long process make this method a difficult method (Al-Mamoori et al., 2017).

2.1.1.4. Chemical Looping-based CO_2 Capture

In the chemical loop capture method, two processes are applied: combustion (CLC) and reformation (CLR). In this method, CO_2 and H_2O can be separated from flue gases naturally and NO_x formation can be minimized. It also offers an economical option with low energy loss (Hossain and Lasa, 2008). The general challenge in this process is the high pressure environment requirement. However, if CCS is aimed after this capture, this situation is no longer a disadvantage. This method, designed especially for energy production facilities, has not yet been implemented on a large scale and is in the research and development phase (IEA, 2013).

2.1.1.5. CO_2 Capture by Hybrid Processes

The different methods outlined above can be used in pairs or multiple ways to obtain a more economical and sustainable capture process. In these applications, which are described as hybrid models, the optimum situation is achieved by combining the methods that are most suitable in terms of cost and efficiency in series or parallel. For example, the PSA method and membrane technology are combined and the high pressure achieved by PSA can be used as a driving force in the permeability of the membrane (Al-Mamoori et al., 2017).

2.1.2. Capture and Storage of CO_2 by Mineralization

Capturing free CO_2 in the atmosphere and forming bonds with calcium and magnesium through mineralization is a thermodynamically spontaneous and natural process. However, this naturally occurring carbonation process is a very slow process (Boot-Handford et al., 2014). The concentration of CO_2 in the atmosphere has remained in balance throughout the life of the earth, and carbonation has played an important role in natural CO_2 absorption (Styring et al., 2011). Beaulieu et al. (2012) estimate that 300 Mt of CO_2 gas is absorbed from the atmosphere as a natural process every year as a result of mineral carbonation. Mineral carbonation is based on the reaction of minerals such as magnesium and calcium in rocks with CO_2. This process, which is currently quite slow, takes place on suitable silicate rocks as in the equation below (Hills et al., 2020):

$$(Ca,Mg)SiO_{3\,(s)} + CO_{2\,(g)} \rightarrow (Ca,Mg)CO_{3\,(s)} + SiO_{2\,(s)} \qquad (1)$$

Calcite ($CaCO_3$) and magnesite ($MgCO_3$), whose formation is shown in the equation above, are used in the manufacture of many products such as ink, rubber, detergent, explosives, medicine and construction materials (Cuéllar-Franca ve Azapagic, 2015). National Academies of Sciences, Engineering and Medicine (NASEM) (2019) states that the carbonation mechanism is a natural mechanism that is safe in terms of storing CO_2 with the product it creates and has positive health and environmental benefits.

The natural mineralization process can also be carried out by an industrial, artificial, and faster process. CO_2 captured under suitable conditions can be converted into carbonated compounds in carbonation facilities, utilized and indirectly stored. Mineral carbonation helps capture CO_2 by transforming minerals, which are raw materials, into carbonated compounds as a result of being processed with CO_2. The minerals taken into this process industrially are first ground, increasing the reactive surface area and accelerating the mineralization process (Baena-Moreno et al., 2019).

During the naturally occurring mineralization process, CO_2 and rocks enter into solid-gas interaction, which prolongs the process considerably. To accelerate this process, solid, gas and liquid phases are brought together in appropriately designed reactors. CO_2 dissolved in aqueous medium is converted into an acidic form with the formula HCO_3. In this environment, Mg and Ca minerals used as input precipitate with HCO_3 (Baciocchi et al., 2013; Lombardi et al., 2012).

With mineral carbonation, CO_2 emissions are captured and stored in a permanent, healthy and environmentally friendly approach. Therefore, in a way, this method is also seen as a CCS method (Pan et al., 2015). Environmental risks do not occur as in classical CCS applications. It stands out as an accelerated version of naturally occurring CO_2 absorption. Due to these features, mineral carbonation appears as an interesting and effective capture and storage technique to significantly reduce CO_2 emissions (Baena-Moreno et al., 2019). For this reason, in this thesis study, the utilization of CO_2 with the mineral carbonation technique is discussed.

Since mineral carbonation reactions occur exothermically, the energy requirement decreases considerably. In addition to this advantage, CO_2 can take part in the mineralization reaction without separating other gases such as SO_x and NO_x in the flue gases (Metz et al., 2005). In this way, the effort, energy and cost spent on capturing high purity CO_2 in other capture methods are eliminated. In addition to the advantage of capturing CO_2 effectively and at low cost, it also contributes to the properties of the new product in which it is stored. For example, it is possible to obtain high early strength by carbonating building elements produced with Portland cement with CO_2. As a result, 7-12% CO_2 capture may be possible (Monkman and Shao, 2006).

A sustainable, environmentally friendly and economic benefit can be achieved with innovative approaches that can be established for the on-site disposal of CO_2 released after the combustion process. Having the raw material suitable for carbonation and the source of CO_2 emissions in the same place can offer good opportunities for the use of this CO_2 without the need for high technology and energy (Hills et al., 2020).

This thesis study was designed in the light of the advantages listed above. In the designed setup, CO_2 in the flue gas of a representative cement factory was subjected to mineral carbonation with RCA without being separated from other impurities. In this way, it is aimed to improve the properties of RCA for use as new aggregate in new building elements, and to contribute to the reduction of carbon emissions by capturing and utilization CO_2 in the flue gases through these reactions.

2.2. Carbon Footprint of Portland Cement and Concrete

Within the scope of the Paris Climate Agreement signed in 2016, it is aimed to reduce CO_2 emissions from various sources causing global warming to zero by 2050, thus to limit the increase in global warming to 1.5°C. European Union member countries also aim to reduce carbon emissions to zero by 2050, in line with the Green Deal targets that came into force in 2019.

Some measures began to be taken years ago in order to reduce global warming. It is stated in the Green Deal that CO_2 emissions could be reduced by 23% between 1990 and 2018. However, it is estimated that this rate can only reach 60% in 2050 if current practices continue. For this reason, it is accepted by many circles that more serious actions should be taken (European Commission, 2019). In addition to the recommended actions, the commission recommends preventing waste and recycling the waste of products produced by industries with high carbon emissions and turning them into a secondary raw material market.

One of the main sectors responsible for carbon emissions that accelerates global warming is the cement sector. In addition, transportation-related and other industrial production facilities with high energy consumption also have a serious share in global warming (IPCC, 2014). At this point, the necessary technological transformations are expected to be made in all sectors by 2050, and governments are providing funding towards this goal. In this regard, many studies are carried out to calculate the carbon footprint in sectors that have a significant share in carbon emissions. Carbon footprint is an index obtained by measuring the unit CO_2 produced by a product, industry or country over a certain period of time (Goodier, 2010). According to the report published by IPCC, the sectors with the highest carbon footprint on an annual basis are the iron and steel industry, chemistry, cement and wastewater treatment sectors (Figure 2.3). As can be seen from here, the cement and concrete sector stands out as one of the sectors where major measures should be taken regarding carbon emissions.

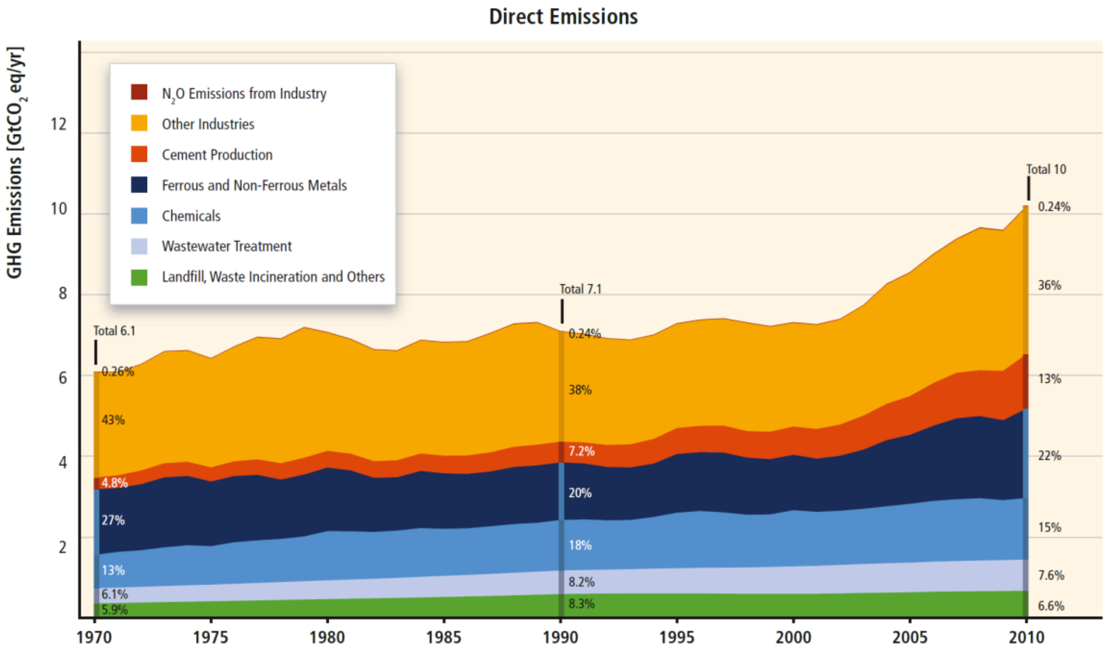


Figure 2.3 Total annual greenhouse gas (GHG) emissions by different industries, 1970 – 2010 (Gt CO_2/yr) (IPCC, 2014)

Concrete is the most commonly used building material in many construction activities such as housing and infrastructure, whose raw materials can be easily accessed all over the world. Developing economies around the world and the increasing need for modern living have increased the need for concrete, a durable and easily applicable building material. Concrete, which generally uses PC in its production, is produced around 1 ton per person every year. The production of cement, which is the main raw material of concrete and the most produced material in the world, is also at a serious level. According to the IPCC report, there has been a significant increase in cement production compared to other industrial products from 1970 to 2014 (Figure 2.4). It has been reported that the amount of PC produced worldwide is 4.1 billion tons in 2022 (Cembureau, 2023). An increase of 12-23% in cement production is estimated by 2050 due to the fact that structures built with concrete have a certain lifespan, the increase in the world population, modern architectural searches and new infrastructure needs for development (Kızıltan and Doğan, 2021).

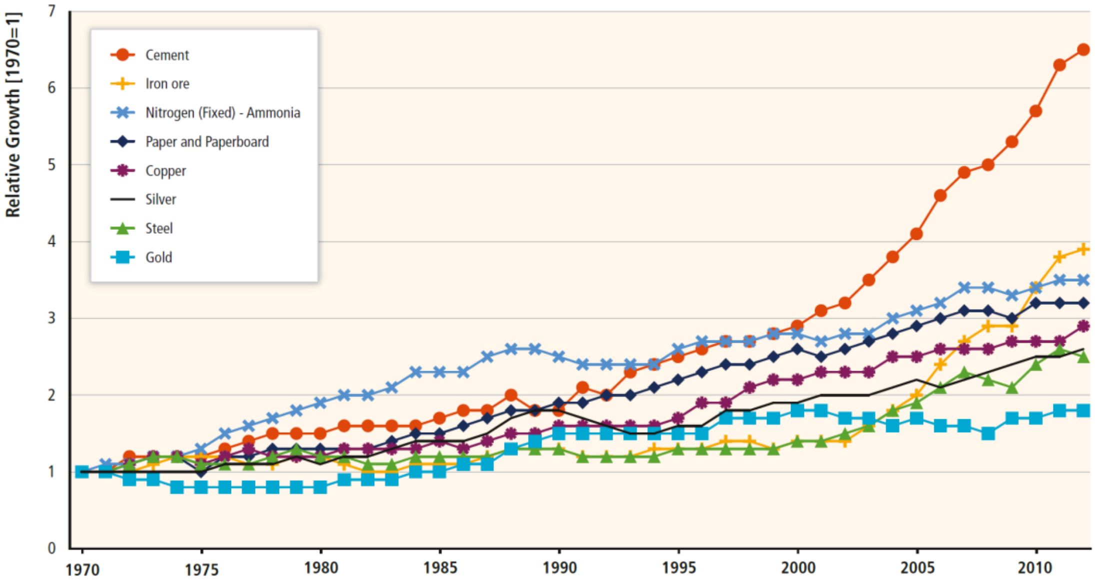


Figure 2.4 Change in production volumes around the world (1970 = 1) (IPCC, 2014)

PC, one of the main ingredients used in concrete, is responsible for approximately 8% of annual global carbon emissions (Lehne and Preston, 2018). There are also sources where the proportion of PC in the entire global carbon emission reaches 10% (IPCC, 2014). The PC sector is one of the sectors with the highest carbon emissions, along with sectors such as energy production and iron and steel industry. For this reason, it is one of the main sectors where precautions should be taken in line with carbon zero targets. Although PC is a necessary material for creating sustainable and durable cities, it creates many negative environmental impacts. In order to avoid serious environmental consequences caused by these effects in the future, it is important to reduce the CO_2 emissions that occur in PC production.

During PC production, various wastes such as CO_2, nitrogen, sulfur and dust are produced, which affect global warming and environmental pollution. Similar wastes also occur in different industries (Figure 2.5). However, in PC production, since the proportion of CO_2 in these wastes is quite high, its impact on carbon emissions is higher than in many sectors. Limestone burned with fossil fuels contains minerals such as sulfur, and after combustion, other gas emissions such as NO_x and SO_x occur from this process (Hägg et al., 2017).

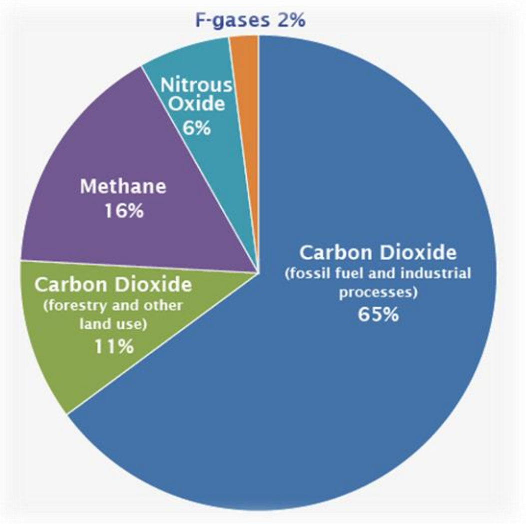


Figure 2.5 Types of Global Greenhouse Gas Emissions (IPCC, 2014)

CO_2 from PC production is formed in two main processes. The first of these is that limestone, which is subjected to heat treatment in rotary kilns as a raw material, decomposes under high temperatures and releases CO_2. The other factor is the fossil fuels burned in the heating process to bring the rotary kilns to a temperature of approximately 1400^{o}C. Approximately 100 kg of fossil fuel must be used for each ton in PC production (CEMBUREAU, 2023). It is known that the CO_2 released at the end of the entire production process is approximately 900 kg CO_2/ton clinker. This process does not include the energy spent to grind the clinker and make it ready for use. When such final processes are added, the CO_2-e value of PC increases even more (Türkçimento, 2023).

Roughly speaking, PC, aggregate, water and steel reinforcement are used in concrete production. In addition to these materials, some chemicals and supplementary cementitious materials (SCM) called Pozzolan can also be used in concrete production. The CO_2 and carbon footprints resulting from the production of these materials have been examined in many studies (Orhon and Altın, 2012). Of these materials, PC has the most obvious carbon footprint. In a study (Gürsel, 2012), It has been determined that the carbon emission value (CO_2-e) of PC produced in Turkey is 1165 kg CO_2/ton. The CO_2-e values of other components in concrete are quite low compared to PC. For example, the CO_2-e value calculated in the process of obtaining aggregates is calculated as 3 kg CO_2/ton for Denmark (Nielsen, 2008). On the other hand, the carbon footprint of steel reinforcement used in building construction along with concrete is considerable. However, these reinforcements can also be manufactured from recycled steel. While the CO_2-e value of raw steel is 2,750 kg CO_2/ton, it can be reduced to 430 kg CO_2/ton with the use of recycled steel (Hammond and Jones, 2008). Mineral admixtures that can be used as substitutes for PC generally have a much lower carbon footprint than PC. In a study where the CO_2-e value of PC was calculated as 860 kg CO_2/ton, blast furnace slag reached 79 kg CO_2/ton and fly ash reached 0.1 kg CO_2/ton (Leese and Casey, 2012).

CO_2-e values of concrete are calculated in many studies, from the production of materials used in concrete production to the activities in the construction of buildings (Vieira and Horvath 2008, Xinga et al., 2008). In the study conducted by Collins (2010), raw material acquisition, production, construction activities, lifespan of the building, demolition of the building, recycled materials and secondary construction activities were shown in a diagram and what kind of carbon emissions could occur at each step were expressed (Figure 2.6). As can be seen from this figure, there are many factors that increase carbon emissions in the production of concrete and the final product, buildings. A CO_2-e value is calculated by taking all these factors into account. However, local factors such as the source of raw materials, costs of local energy resources, construction, and transportation distances of the regions where raw materials are brought can also play an important role in estimating emission rates.

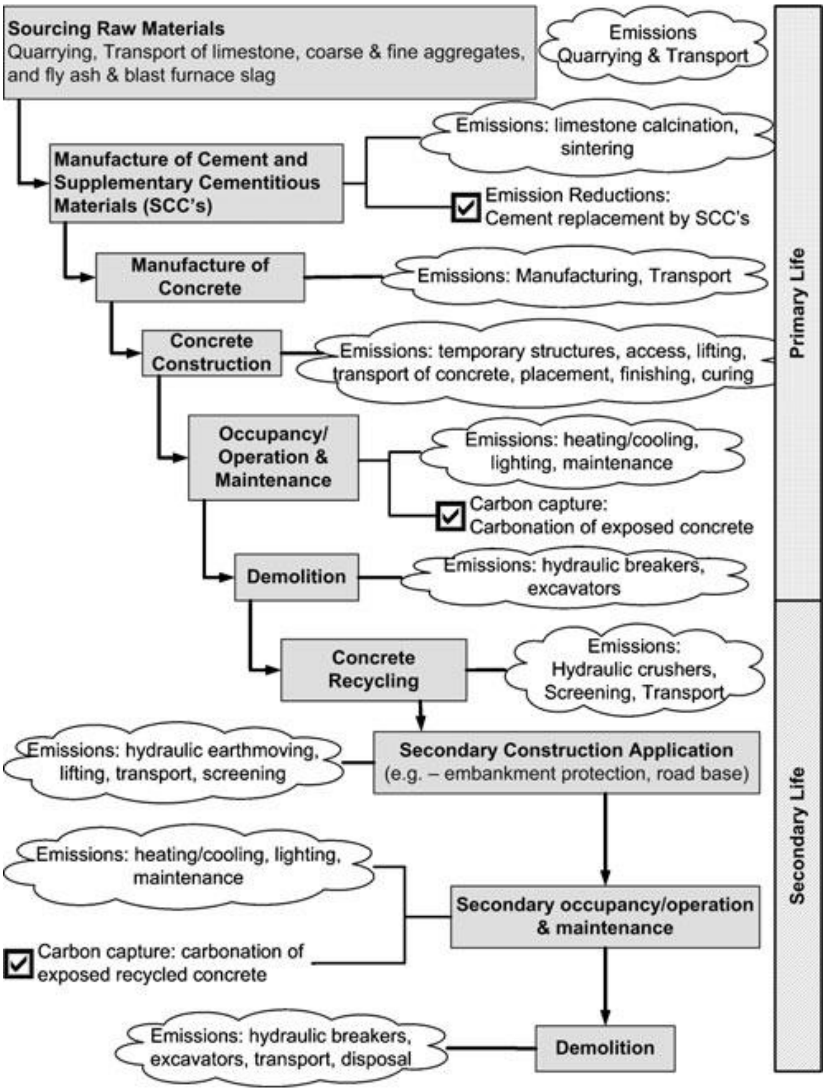


Figure 2.6 Life cycle aspects of CO_2 emissions and carbon sequestration in concrete (Collins, 2010)

PC, the material with the largest carbon footprint among the concrete produced, stands out as a material that is constantly needed. Therefore, it is possible to reduce its effects instead of giving up its production. The Global Cement and Concrete Association (GCCA) (2021) has prepared and published the "2050 Net Zero Road Map" document for the goal of limiting global warming to 1.5^{o}C in the Paris Climate Agreement. This road map shows the revisions that companies operating in the sector can make in line with this goal. Measures that can be taken in this direction can be summarized as follows:

- The use of clinker-based PC can be reduced by using SCM, thus achieving the expected performance of the final product.

- Alternative fuels can be used instead of fossil fuels with high CO_2 emissions during the clinker production phase.
- Innovative technologies that produce lower carbon emissions can be developed as an alternative to rotary kilns that use high temperatures.
- CCUS technologies that are effective in reducing carbon emissions can be adapted.

The use of mineral additives, which is the first of the recommended measures, has a significant potential in reducing the carbon footprint of concrete. PC, which has a significant share in the CO_2-e value of concrete, can be replaced with some industrial wastes as SCM and the amount of PC in concrete production can be reduced. It is observed that these materials can replace cement at levels of 50% or more, depending on their type. Since the carbon footprint of SCM is very low compared to PC, PC-related carbon emissions can be reduced by half as a result of their use instead of PC at 50% levels.

In the report they published in 2011, the European Commission stated that the construction sector is the largest consumer of natural resources. The demolition waste of buildings that have completed their lifespan is still stored in landfills without being adequately recycled. It is recommended to use building materials efficiently during the initial construction process and after the demolition of the buildings. In this way, less consumption of natural resources for new concrete production will be ensured, as well as a reduction in the amount of CDW stored in landfills, which has serious effects on the environment. It is envisaged that with the measures to be taken in this direction, efficiency can also be achieved in the European economy (European Commission, 2011).

According to the projection given in the road map published by GCCA, the contributions of the measures that can be taken towards the carbon zero target are given in Figure 2.7 according to their importance percentages. As can be seen from the figure, the main precaution to be taken is the integration of CCUS techniques into PC production processes.

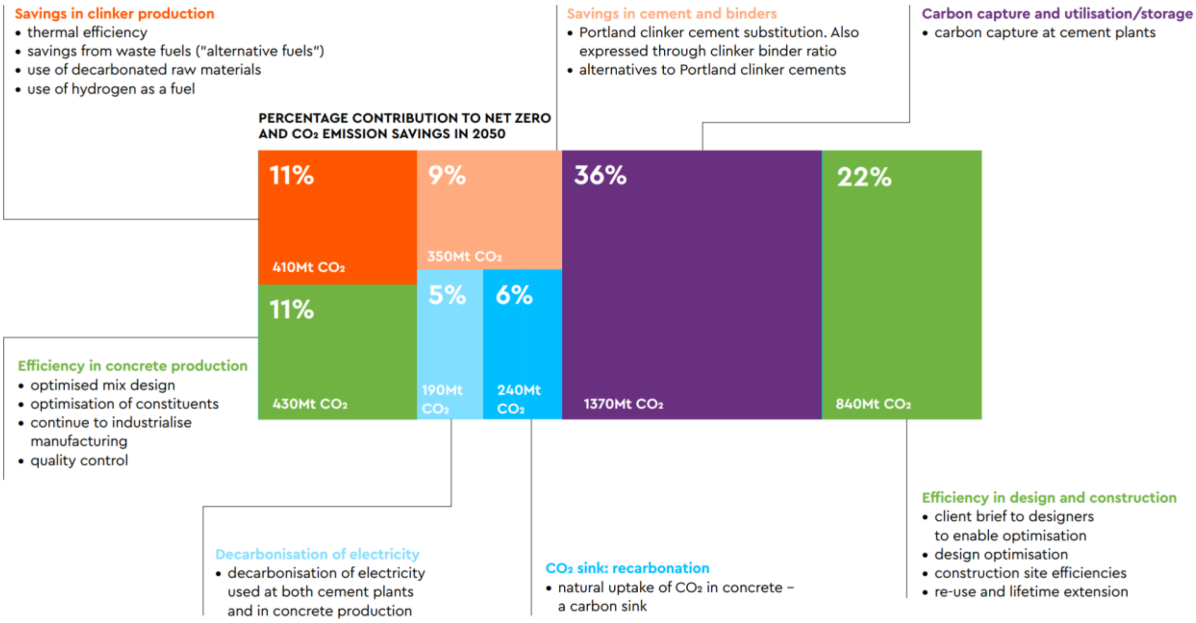


Figure 2.7 Envisaged contribution potential of the measures that can be taken according to the “2050 Net Zero Road Map” (GCCA, 2021)

CCUS methods are promising in terms of preventing the release of CO_2 gas into the atmosphere and reducing carbon emissions, without preventing the use of fossil fuels used in industries with high energy needs. CO_2 can be captured with CCUS methods in the PC industry and can be used to carbonate various materials used in concrete.

Due to its nature, concrete can form chemical bonds with free CO_2 in the air. During its life and after its destruction, $Ca(OH)_2$ and CO_2, which are found in the porous structures in concrete, can react in humid environmental conditions. In this way, buildings can actually compensate for a certain part of the CO_2 they cause during their lifetime and in the waste state after they are demolished. However, this absorption occurs with a slow reaction that lasts for years and spreads over the long term. This process occurs faster in CDW. Because wastes are ground into small particles, they provide a higher surface area for carbonation than mass concrete which continues its lifetime. In particular, processing industrial RCA in large-scale designed carbonation reactors before its use in new building construction has the potential to contribute to reducing the carbon footprint of concrete in life cycle analyses. So much so that, in a study, it was stated that by carbonating RCA before its secondary life, it is possible to absorb approximately 40% of the CO_2 resulting from the production of the PC to be used with it (Collins, 2010). For this reason, even if a structure made of concrete reaches the end of its life, it should be included in recycling processes, and CO_2 should be absorbed back through appropriate processes in these stages. When the situation where a concrete structure has a primary and secondary life is taken into account, the carbon footprint of concrete will be significantly lower. This double positive effect was also among the motivations of this thesis study. Starting from this point, RCA obtained from the demolition waste of end-of-life structures was carbonated with an industrial CO_2 source representing the CO_2 released from the flue gas of cement factories. The ultimate goal here is to prove the feasibility of this process, which has serious potential in reducing the carbon footprint of concrete.

2.3. Construction Demolition Wastes

Concrete, the most commonly used material in line with housing and infrastructure needs, is designed to have certain mechanical properties and to maintain its strength over a certain period of time. When many standards are examined, concrete structures are generally expected to be designed to have a lifespan of at least 60-70 years. However, due to environmental and mechanical effects such as earthquakes, it is estimated that many of these types of structures begin to deteriorate after 20-30 years, so the buildings built in the 1960-1990s are about to complete their service life. In structures that are nearing the end of their lifespan or have some structural defects, efforts are being made to extend their lifespan by using strengthening techniques. Ultimately, all of the serious amounts of various wastes that arise in processes such as demolition, strengthening, repair of structures that have completed their service life or the construction of new structures are called “Construction and Demolition Waste”.

It is stated that approximately 1/3 of the amount of solid waste produced worldwide is CDW (Du et al.,2020). The annual amount of 3 billion tons of CDW reported in 2012 is estimated to be around 10 billion tons today (Bose, 2022). It was reported that a total of 3 billion tons of solid waste was produced in the European Union (EU) member countries in 2011, of which approximately 30% consisted of CDW (European Commission, 2011). It has been reported that 23% of the total solid waste generated in Hong Kong is CDW resulting from construction activities (Lu and Yuan, 2011). It was emphasized that, according to data obtained in 1999, the annual amount of CDW in the USA reached around 136 million tons, and this figure corresponds to 464 kg/person (Franklin Associates, 1998).

Considering the world average, it is seen that the ratio of CDW in total solid waste is around 30-40%, and approximately 60% of the CDW content consists of concrete waste. For this reason, in this thesis study, we focus on the evaluation of concrete waste as RCA, which is the most occurring CDW when we look at the world in general. Although wastes such as glass and bricks have also been examined as recycled aggregate sources, RCA has been the focus since they are in large quantities.

Since a large part of Turkey's surface area is in the first and second degree earthquake zones, a large part of Turkey's building stock is faced with the threat of earthquake. When the existing building stock is examined, it is seen that there is a large proportion of buildings that are about to complete their life. As a result of earthquakes and urban transformation works initiated in the country, serious amounts of CDW are released. As a result of the devastating earthquakes in the Marmara Region in 1999, 13 million tons of debris emerged, almost all of which was moved to empty storage areas, and some even poured into the sea (Coşgun and Salgın, 2012). In addition, as a result of the devastating earthquakes that took place in Kahramanmaraş, Turkey on February 6, 2023, thousands of buildings were destroyed and a significant amount of CDW was released. It is also estimated that millions of buildings will be affected by the urban renewal law in Turkey.

2.3.1. CDW Prevention, Reuse, Recycling and Recovery

When studies in the literature are examined, it is seen that reuse or recycling of CDWs is possible with many different methods. However, although many studies have shown that reuse or recycling is possible, CDWs are generally stored on clean soil in empty lands, which poses serious environmental problems.

The production of materials used in new building construction processes creates many harmful effects on the environment. The contribution of fossil fuels consumed in these processes to greenhouse gas emissions, natural resources consumed, and other harmful wastes released during production can be eliminated by reusing or recycling CDW. The use of CDW in new buildings through recycling rises to 30% on average in Europe, and up to 90% in the most developed member countries (Deloitte, 2017). These high recycling rates are a source of hope and inspiration for other countries in terms of protecting natural resources and reducing environmental pollution.

In addition to using the waste resulting from demolition in new construction activities, it is also recommended to minimize or prevent waste before it reaches the final stage in the waste hierarchy. With the waste prevention process, raw material consumption can be reduced and the burden on waste management processes can be reduced. Prevention activities recommended by many organizations are essentially associated with durable and long-lasting building designs.

It is possible to use CDW waste in new buildings. However, in many cases, using these wastes as they are, can cause serious uncertainties and weaknesses in the new structure. For this reason, CDW that is planned to be used in new buildings must be thoroughly analyzed physically and chemically, and its properties must be improved if necessary. In particular, RCA obtained from concrete waste can cause some mechanical and durability problems in new concrete due to their high water absorption capacity (Bravo-German et al., 2021).

In the study of Bose (2022), the reasons why the CDW recycling industry still cannot reach satisfactory levels were investigated and the following findings were put forward as the main reasons:

- Failure to plan CDW recycling facilities correctly,
- Poor communication between the recycled material producer and end users,
- States' lack of appetite for recycling,
- Insufficient awareness and appetite for on-site and selective collection of CDW,
- Collection or processing of CDW using inappropriate techniques,

- Monopoly global companies or organizations, etc., that do not want CDW waste to be included in the market as alternative raw materials after recycling.

There are also some obstacles in terms of occupational health and safety to the widespread use of recycling practices and the proliferation of recycling facilities on an industrial scale. The preferability of these facilities is decreasing due to the fact that the dust generated in these facilities poses a risk to the health of workers and the need to take many precautions to eliminate these risks. On the other hand, whether these wastes are of sufficient quality for recycling creates another trust problem (European Commission, 2016).

2.3.2. CDW Content

CDW, which contains large amounts of concrete and aggregate waste, also includes various different materials such as glass, brick, plastic, wood, plaster, insulation materials, asphalt, and excavation waste (Figure 2.8). When the ratio of the components in CDW is examined, it is seen that perhaps more than half of it consists of elements such as concrete and brick. Waste concrete, which is the main material handled within the scope of CDW, largely arises from construction demolition activities, but can also arise from wastage in factories producing prefabricated concrete elements or ready-mixed concrete factories. For this reason, waste concrete and waste bricks are considered the most important components in CDW (Patel et al., 2015). Which materials the CDW content consists of and in what proportions it contains these materials can vary greatly depending on the type of building being demolished, the region, city or country where the building is located. It has been determined that approximately 800 million tons of CDW is produced every year in Europe, and it has been stated that these arise from infrastructure construction and demolition activities such as highways and railways, new building construction and demolition activities of structures that have completed their lifespan (Deloitte, 2017).

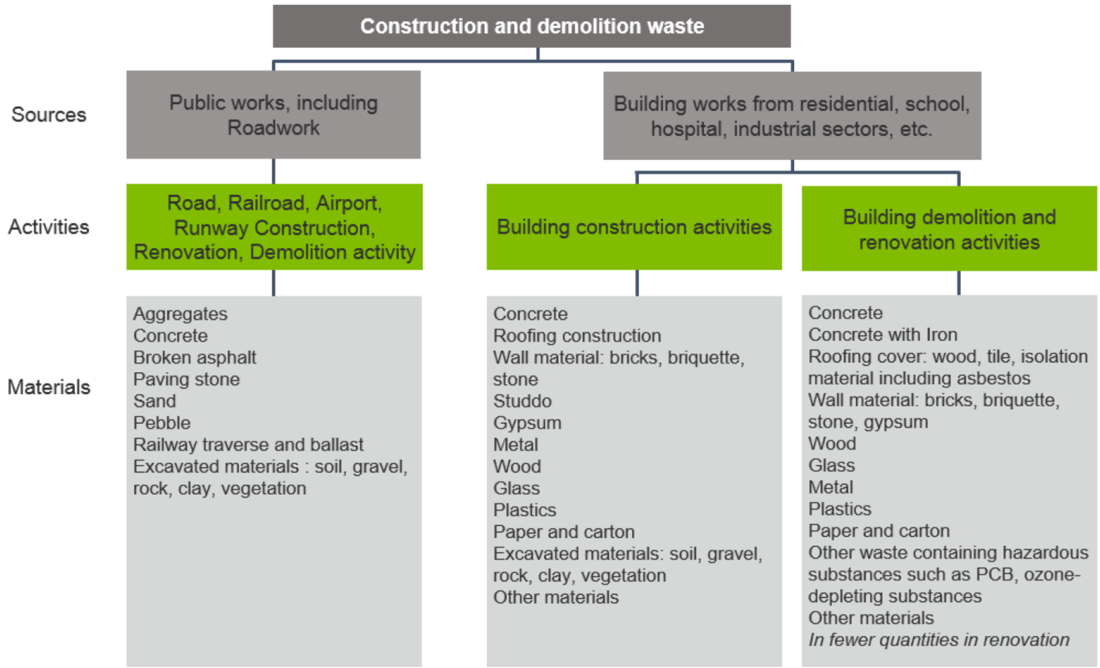


Figure 2.8 CDW content and resources (Deloitte, 2017)

Asphalt produced as the upper wear layer on highways is obtained with aggregate, bitumen and some additive materials. Asphalt, which can generally be completely recycled with appropriate processes, is removed from the road surface with appropriate machines, and the recycled materials in recycling facilities are used in new mixtures under appropriate conditions. It is guaranteed that asphalt waste will be 100% recyclable if it is collected with appropriate machines and under appropriate conditions and is uncontaminated (Bio Intelligence Service, 2011).

Another type of waste included in CDW is wood waste, which is generally recycled in the form of timber. However, collecting wood waste without deterioration or contamination during construction demolition processes is difficult due to occupational health and safety regulations and limited project timelines. It is usually mixed with other CDW waste. This limits the conversion of wood waste into high value-added recycled industrial products. This disadvantageous situation experienced in the demolition of residential buildings is not experienced in the waste collection of wooden electric poles or wooden sleepers used under railway during the maintenance and reconstruction works of infrastructure facilities (Australian Government, 2012). There are also studies where waste wood materials are used as fiber reinforcement in cementitious composites (Tuğluca et al., 2023). However, in all processes, these wastes must first be pre-treated.

Gypsum is a building material used for plastering purposes on interior wall surfaces in buildings or in manufacturing such as partition walls, which are relatively thinner than brick walls. It has been reported that gypsum waste, obtained in low amounts compared to other CDWs, was released as 10 million tons worldwide and 4 million tons in Europe in 2007 (Bio Intelligence Service, 2011). Gypsum-based materials, especially used in residences, are highly recyclable materials if collected under appropriate conditions. However, it is very difficult to distinguish this material from the CDW resulting from the demolition of a building, as it is mixed with other wastes in powder form.

Ceramic materials are used for different purposes in many building types. Ceramic materials are frequently used on roofs, interior and exterior coatings of buildings, interior walls of buildings, and infrastructure facilities for drinking water or wastewater transmission. Ceramic material produced with easily accessible clay raw material generally appears in the form of bricks and tiles. Compared to other CDWs, these materials are easier to selectively separate from structures before demolition, and it is even possible to reuse them in construction activities in other structures. The parts that cannot be reused can be used for different purposes in recycling processes. Tiles & Bricks Europe (2011) reported that ceramic waste can be recycled as display material in stabilized roads, subbase in special road construction, aggregate in building construction, ground material in tennis courts, and substrate for various plants.

The plastic material found in construction demolition waste generally manifests itself in the form of Polyvinyl chloride (PVC). Apart from this, there are very few cable coatings used in electrical installations. Since PVC, which is used as coating, door, pipe or window in buildings, is a thermoplastic material, it softens when heated, can be reshaped into the desired shape and can be cooled and used in its new form. This feature shows that PVC is a suitable material for recycling. However, like other materials in CDW, PVC's contamination with other pollutants makes its recycling difficult. Because it requires many processes to recover it with high cleanliness (Australian Government, 2012).

Although CDW contains many valuable materials that can be recycled, materials such as asbestos, phenols, and plumbic-based paints that have serious harm to the environment and human health can also be released. It is important that these materials are removed from structures and disposed of with special care, without mixing with other CDW content. According to the Hazardous Waste Directive 91/689/EEC3 regulation in force in Europe, it is mandatory to first identify and record harmful materials in the process of separating such materials from CDW and to take necessary precautions to prevent them from mixing with other wastes in CDW (European Commission, 2011).

Reinforced concrete structural elements generally consist of a high proportion of concrete and a very low proportion of reinforcing steel. So much so that the ratio of reinforcing steel in reinforced concrete elements is roughly 1-5% (Mehta and Monteiro, 2005). Although it is produced in relatively small amounts, the reinforcement steels obtained from structures that have completed their service life are completely recyclable (Bio Intelligence Service, 2011). This shows that structural steel is a good resource to be recycled and used in new buildings or other industrial productions.

2.3.3. CDW Separation Processes

Different methods are used to separate different materials in CDW. First of all, before demolition, materials such as pipes, glass, doors and bricks that can be used in the new building can be collected without damaging them. After the structure, which contains materials that cannot be reused as they are, is demolished, the rubble goes through a series of processes. The concrete and aggregate part can be crushed in crushers and prepared for RCA or for use in road sub-bases. Materials such as wood, plastic, metal and glass that cannot be separated from the building without being damaged can also be separated by different methods. Contaminated plastic, wood and other hazardous materials that are not in suitable conditions for recycling can be destroyed by burning them for energy production. Other wastes that are not suitable for these processes are sent to appropriate waste storage areas.

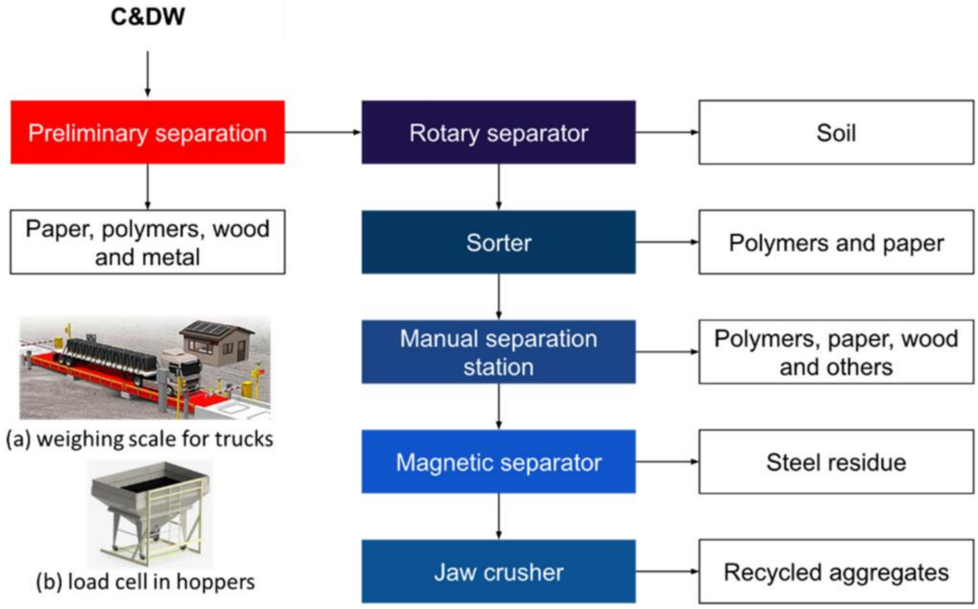


Figure 2.9 CDW separation processes (Maury-Ramirez et al., 2022)

On-site selective CDW separation is important in structures that have been or will be demolished. For these operations, portable material separator and crusher vehicles can be used at demolition sites. After CDW is formed, special equipment is available to separate relatively light products such as wood, plastic and insulation materials. Metal content such as reinforcing steel and steel pipes can be separated with the help of large magnet machines. The final CDW part, which contains a large proportion of concrete and aggregate, can be processed through crushers and evaluated as RCA (Figure 2.9). The finer part of the aggregate, which occurs in different particle sizes, is not preferred in new concrete production due to its high water absorption capacity values.

In a demolition activity, it is important that the company that will carry out the demolition carries out and reports the entire process within a certain waste planning. It is important to obtain as clean materials as possible for the recycling process before and after demolition. Proper separation of materials is directly related to the success of the recycling process to which they will subsequently be subjected. Materials such as doors, windows, installation components, valuable woods, metals and radiators should be collected before demolition in appropriate conditions and, if possible, redirected to reuse. All these measures allow the materials remaining in the waste stream to be purified and obtained as quality raw materials.

The Regulation on the Control of Excavation Soil, Construction and Demolition Wastes, which came into force in Turkey in 2004, specifies the principles regarding the collection, disposal, storage, transportation and recycling and evaluation of wastes generated during construction/maintenance activities, natural disasters and construction/demolition stages. According to the regulation, CDW should be reduced to a minimum amount at the source and reusable content should be collected at the source with a selective collection approach. It is stated that the persons or institutions responsible for the management of these wastes are obliged to take the necessary precautions in terms of environmental pollution and human health. The regulation specifically regulates the recycling or reuse of CDW and excavation wastes as low-quality infrastructure material; it also requires special care to be taken to avoid mixing CDW and excavation wastes with each other. The regulation recommends that the non-recyclable part of CDW be ground and used as cover material in solid waste landfills.

Well-organized waste management plans are needed for the success of all these processes. In these plans, steps such as identifying and reporting waste, collecting materials that can be selectively collected before demolition without being damaged or contaminated, where the waste will be transported after demolition and how it will be stored, and what the recycling processes will be like should be clearly stated. These plans are also expected to define processes on how occupational health and safety measures will be taken and how hazardous wastes will be evaluated or disposed of (European Commission, 2016).

2.3.4. CDW Recycling Methods

Industrial wastes such as fly ash and blast furnace slag can be turned into a binding structure similar to cement by the method called geopolymerization. This method is based on the activation of a suitable aluminosilicate source in powder form with an alkaline activator of the appropriate type and appropriate molarity (Wu et al., 2019). This method, which has been studied for many years, is also being tried for the disposal of CDW. Materials such as waste concrete, waste brick, waste ceramics can be pulverized and subjected to geopolymerization, thus having the potential to be used as an alternative binding material source to cement in new building construction (Kul et al., 2024).

In the geopolymerization method, solid-powder aluminosilicates react with alkaline hydroxide or silicate solutions under suitable conditions and form a new binding structure. Alkaline activators used in this process are generally sodium silicate and sodium hydroxide. Different materials such as blast furnace slag and fly ash can be used as aluminosilicate. Aluminosilicate materials in the amorphous phase are activated by alkaline solutions. This activation creates a structure similar to the calcium-silicate-hydrate gels obtained in conventional cement paste, and high mechanical properties similar to conventional concrete can be obtained (Balaguer et al., 2014). In some studies in the literature, construction demolition waste has also been tested as a source of aluminosilicate. Geopolymerization methods have also been applied to materials such as waste concrete, brick and glass powder and reasonable mechanical properties have been achieved (Ahmari et al., 2012; Kul et al., 2024; Yildirim et al., 2023; Ulugöl et al.,2021). On the other hand, there are also studies in which waste glass powder is used as an alkaline activator in geopolymerization applications (Torres-Carrasco et al., 2015). The presence of high amounts of alkali oxides and amorphous silica allows glass powder to be also used in this role.

Robayo-Salazar et al. (2020) followed the procedure in the Figure 2.10 in their study where they turned such materials obtained from CDW into binders using the alkaline activation method. In this study, CDW powders were brought into a binding structure similar to cement mortar with alkaline activators and mixing water. Additionally, RCA was used as aggregate. Experimental studies have shown that CDW can be completely recycled as both binder and aggregate and can be used in structural concrete production. Since the CDW powder used as a binder contains mixed wastes such as bricks, concrete and tiles, it has been shown that these wastes, which are difficult to separate, can be evaluated in the recycling process with the innovative method in this study.

Figure 2.10 The process of evaluating CDW as a binder using the alkali activation method (Robayo- Salazar et al., 2020)

Waste concrete and waste wall elements, which have a significant share in CDW content, are generally turned into RCA after simple crushing and screening processes, and are then used in construction activities such as road or pavement foundations where structural bearing properties are not required (Khatib, 2009; Arulrajah et al., 2013). The fact that RCA is not used in qualified construction activities may be associated with the low mechanical and chemical properties of RCA. The main factor here is the presence of a high-void structure caused by cement paste or mortar residues on the surface of the aggregate. For these reasons, widespread use of RCA remains limited.

Although fine dust formation in RCA production creates environmental and occupational health problems, similar effects are also encountered in NA production. In addition, NA is transported from its source to construction sites after it is produced, resulting in a serious carbon emission rate due to transportation-related fuel consumption. These negative effects can be eliminated by using RCA in production facilities or construction sites near the region where it is produced (Bio Intelligence Service, 2011).

In his study, Demir (2009) revealed the negative effects of NA production with a simple approach. According to the results of the research conducted in the USA, it has been stated that 2.7 billion tons of aggregate are used annually, and based on the information that NA has an average specific gravity of 2.7 tons/m^3, it has been concluded that approximately 1 billion m^3 of the natural environment is destroyed to obtain this amount of NA.

Considering that the most commonly used material in concrete production is aggregate, with a rate of nearly 80%, it becomes clearer how large a volume of aggregate is needed for concrete, which is one of the most produced products in the world. To meet this need, natural resources are destroyed in very high volumes and harmful pollutants are produced for the atmosphere. For this reason, it is important to produce RCA from construction wastes with high recovery and to make it safe to use in new buildings (Etxeberria et al., 2022).

In this thesis study, solutions were sought to the problems encountered in new concrete mixtures of RCA, which constitute a serious potential for recycling and a serious raw material supply in CDW content. In this direction, the negative properties of RCA were tried to be eliminated by the accelerated carbonation method, and the engineering properties of carbonated RCA, whose properties were improved, were revealed by using it in new mortar mixtures. The results obtained gave clues that it may be possible to use RCA in new structures.

2.4. Recycled Aggregates (RA)

Considering that approximately 3/4 of conventional concrete consists of aggregate and that more than 5 billion tons of concrete is produced annually in the world, it is easily understood that a large part of the environmental impacts resulting from concrete production are caused by aggregate (WBCSD, 2012).

Aggregates, as the most widely used material in concrete, are one of the components that affect the properties of concrete the most. Therefore, the quality of the aggregate is very important. The production of NA, which has suitable properties to exhibit the expected performance in concrete, has reached very serious levels due to the increasing need for construction. This causes the destruction of natural resources. On the other hand, the use of CDW, which accumulates in landfills and causes serious harm to nature, as aggregate in the production of new concrete, is a good opportunity for the disposal of these wastes. For this reason, reducing the use of NA and using CDW as aggregate is important for environmentally friendly concrete production.

In many sources, it is stated that NA production has reached very serious levels due to increasing demand, serious amounts of carbon and harmful gas emissions occur during the production phase, natural resources, which are aggregate sources, are gradually decreasing and the natural balance is disrupted, causing serious damage to the ecosystem. In order to obtain river sand, which is one of the natural resources from which aggregate is obtained, structures such as flow direction, flow bed, and shoreline of rivers can be disrupted, causing serious damage to the ecosystem (Pavlu, 2018; Nedeljkovic et al., 2021). For these reasons, it is important to use RA as an alternative to NA in structural productions as well as in low-quality productions.

The use of RA dates back to ancient times. After World War II, large amounts of debris from buildings, roads and various infrastructures emerged in the cities destroyed in Europe. Both the need to dispose of these wastes and the need for new construction after the war led to the recycling of CDW and its use in RA production (Buck, 1977).

There are various materials such as brick, glass and ceramic that can be used instead of NA in CDW. Materials such as glass, ceramics and brick in the waste can be selectively collected without being damaged before demolition, therefore, the majority of the material emerging after demolition consists of waste concrete pieces (Figure 2.11). Therefore, the main source of RA is waste concrete and RA coming from the waste concrete is called recycled concrete aggregate (RCA).

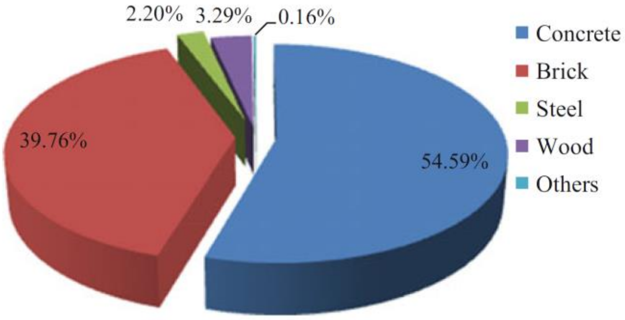


Figure 2.11 Content and proportions of Construction Demolition Waste (Xiao, 2018)

Recycling of waste concrete, which contains a large amount of CDW, generally occurs in low-quality production such as pavement and road sub-base, simple wall blocks, and precast structural elements. However, it is also possible to produce higher quality structural concrete. Fine RCA is considered as RCA with a size of 4.75 mm and below, as in NA. Compared to coarse RCA, their use is quite limited and is not recommended by many standards. For this reason, fine RCA is generally used in low-quality construction activities such as road and pavement foundations and fillings.

Increasing the replacement rate of RCA, where the replacement rate with NA is limited due to some physical and chemical negativities, by various methods to maximum levels appears as a very environmentally friendly approach, considering that aggregates are used in high volumes in concrete. The use of RCA instead of NA in newly produced concrete provides economic benefits as well as environmental benefits. RCA prices are quite low compared to NA unit prices. The unit price of RCA, which may vary depending on the region where it is obtained and the costs at the recycling facility, starts from 1 $/ton and can rise up to 18 $/ton (USGS, 2000). Based on this, the use of RCA in concrete can reduce the unit cost of concrete by up to 60% (ECCO, 2018).

The use of fine RCA, whose use is not recommended and restricted by some standards, instead of fine NA also provides significant environmental benefits. It has the potential to prevent carbon emissions that will occur in the production of fine NA and other harmful gases released into the atmosphere. Although some environmental damage may occur as a result of the industrial processes required to produce fine RCA, it is likely to have a very low level of environmental impact compared to fine NA production. In a study, it was observed that around 23-33 kg of CO_2 was emitted in the production of 1 ton of fine NA, while this value decreased to 12 kg in the production of fine RCA (Hossain et al., 2016).

According to different usage purposes, RCAs are processed in different types of crushers in the facilities and brought to small sizes. These include machines such as impact crusher, jaw crusher, cone crusher (ACI 555, 2001; Nedeljkovic et al., 2021). In NA production, while aggregates in the desired particle size distribution can be produced with optimum settings in the crushers, it is difficult to grind RCA to a certain standard due to its multiphase structure. In the process initiated to obtain an RCA of a certain size with crushers, there is a high probability of formation of particles in a wide range of sizes (Pedro et al., 2017).

2.4.1. Standards related to RA

Although there are studies in the scientific literature on the use of RA at high rates and without processing, there are standards and various reports that limit or regulate its use in concrete production (RILEM, 1994; ACI 555, 2001).

Since RA contains a higher amount of voids than NA, it has the ability to absorb a much higher amount of water. In new concrete mixture design, the water absorption capacity of the aggregate planned to be used is one of the most important parameters. Some regulations state that the aggregate to be used should not have more than 5% water absorption capacity. When the values in the literature are examined, it is seen that RA generally has a capacity above this rate. For this reason, the use of RA is restricted in many standards (Etxeberria et al., 2007).

Rilem (1994) does not find fine RA appropriate in the use of RA. It imposes some limits on coarse RA. In addition, it does not allow the use of RA, which contains other substances and contaminations that delay the setting time of the concrete more than 15% and damage its other properties. It divides RAs that can be used in buildings into 3 classes according to their types. Those obtained from waste wall elements are called Type I, those obtained from waste concrete are called Type II, and RAs mixed with NA in certain proportions are called Type III. Rilem (1994) also determined sub-criteria in these classes. The use rate of Type I aggregate in mixtures is limited to 10%, and the use rate of RA in Type III aggregates is limited to 20%. As in the Rilem example, other standards also recommend separating fine RA from the aggregate. This is because not enough studies have been conducted on fine RA yet and there are no comprehensive guidelines.

The use of RAs in new concrete mixtures is technically similar to the processes in traditional concrete production. However, care must be taken in material selection and use. ACI 555 (2001) states that in concrete where RA is used, it should be subjected to pre-wetting process due to the high water absorption feature of RA and the content of RA with grain size below 2 mm should be separated. It emphasizes that if a plasticizer admixture is not used, the free water demand in the system containing RA will be 5% higher.

2.4.2. Factors Affecting Recycled Concrete Aggregate (RCA) Features

Concretes produced with RCA may exhibit different behaviors depending on the properties of the waste concrete from which RCA is obtained. The amount of mortar residue on the RCA and the structure of this residue affect this behavior. Additionally, the properties of the old NA in the parent concrete also play a role in the quality of RCA.

Generally, as the RCA grain size decreases, the amount of mortar residue in the total volume increases. On the other hand, it is seen that the mortar residue is affected by the w/c ratio of the parent concrete and this factor directly affects the performance of RCA in the new mixture (Etxeberria et al., 2007).

CDW brought to recycling facilities is separated by various techniques and the part containing mostly waste concrete is finally ground. The crushing and sorting techniques used to convert waste concrete into RCA in recycling facilities can also affect the quality of RCA and the mortar residue rate (Katz, 2003). Additionally, CDWs from different building types can mix with each other in these facilities. CDW arriving at the facility may have been exposed to different environmental aggressive effects during its life cycle and depending on storage conditions after demolition. Therefore, it may not be possible to achieve a certain standard in the RCA produced.

In general terms, the factors affecting the quality and various physical and chemical properties of RCA are shown in Figure 2.12.

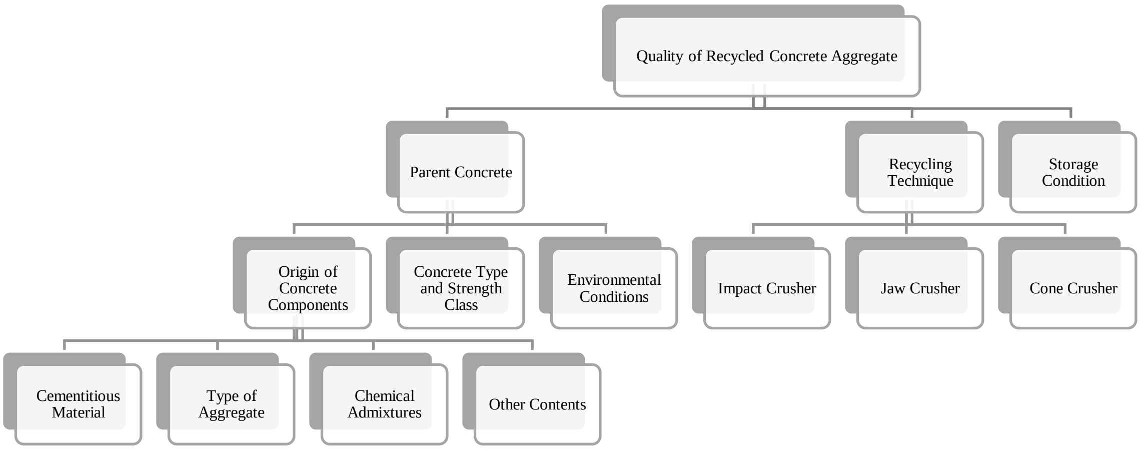


Figure 2.12 Factors affecting the quality of RCA

2.4.3. Chemical Properties of RCA

The chemical structure of RCA may vary depending on the content of waste concrete and the types of binders, aggregates and different additives used in old concrete. While studies in the literature show high levels of SiO_2 in some RCAs (Angulo et al., 2009), CaO is observed as the dominant compound in others (Bianchini et al., 2005). This shows that the aggregate used in the parent concrete consists of silicious materials or limestone type aggregates. As seen in studies examining the chemical structure of RCA, high loss of ignition (LOI) values have been observed, especially in sub-mm fine RCA. This is attributed to the predominance of waste concrete dust in fine fractions, the possibility of this waste dust being highly carbonated, and the calcareous sand grains it contains.

2.4.4. Engineering Performance of RCA

One of the most fragile areas in concrete or mortar made by RCA is the interfacial transition zone (ITZ) formed between the new cement paste and the old mortar residue on the RCA surface. As can be seen from Figure 2.13, when look at the microstructure, the old NA contained by RCA, the mortar residue adhered to the old NA, the old ITZ between this mortar residue and the old NA, and the new ITZ between the new cement paste and RCA and the old mortar residue can be observed.

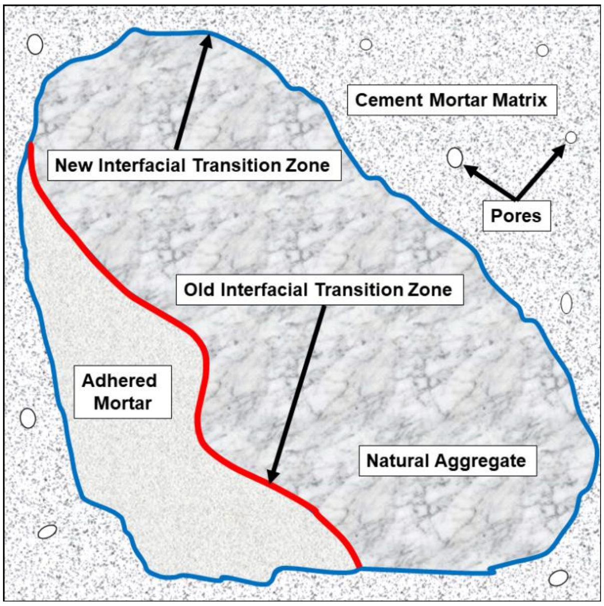


Figure 2.13 Representative RCA microstructure and the different phases it contains (Adams and Jayasuriya, 2019)

Cocnrete obtained with the use of RCA has negative properties compared to traditional concrete produced with NA. Commonly observed situations are low strength, low workability, drying shrinkage and other mechanical problems. This situation varies depending on the high water absorption of RCA, the properties of the source waste concrete from which RCA is obtained, the substitution rate in the mixture, w/c ratio, the amount of mortar residue on the RCA, and the amount of cement used in the mixture. For this reason, before the production of concrete for use in structural systems, it is recommended to pre-test issues such as the source from which the RCA is obtained, what level of water absorption it has, and with what w/c ratio appropriate workability and strength values can be achieved (McNail and Kang, 2013).

RCA has a permeable structure because the mortar residue on its surface is highly porous and has a highly cracked structure during its production. This makes RCA a low quality aggregate source (Tam et al., 2007). The high void ratio of the mortar residue on the RCA surface reduces the density of the RCA. The density of the mortar residue is therefore lower than the density of both the old NA embedded in the RCA and the newly produced NA. For this reason, some negative effects may occur in some mechanical properties of concrete (McNail and Kang, 2013). On the other hand, unhydrated cement areas can also be found in the mortar residue, and this part can contribute to the strength of new mixtures (Manzi et al., 2013).

NA, which generally has a smooth and angular grain shape, provides a suitable structure for the desired workability in concrete. However, RCA generally has a negative impact on workability due to its highly rough and uneven shape. The shape of RCA may vary depending on the variety and working principles of the crushing machines used in its production (Etxeberria et al., 2007).

Other obstacles to the widespread use of RCA are the possibility of the mortar residue on the RCA turning into additional dust content, the possibility of being contaminated with various sulfates or chlorides, and even in the best case scenario, the formation of additional ITZ zones at a higher rate than in the NA system (Singh et al., 2022).

Disadvantages experienced with the use of RCA can be minimized by changing the mixing ratios, trying different mixing techniques, using high proportions of cement, changing the substitution rate of NA with RCA, and using water-reducing admixtures.

Concrete may also exhibit poor behavior in terms of durability due to the properties of the RCA it contains. It has been reported that water absorption, capillary water permeability and chloride permeability in concrete samples in which RCA was replaced by 100% increased significantly compared to samples in which 100% NA was used (Evangelista and de Brito, 2010). The effects of RCA on the general durability performance of concrete were compiled in the study of Guo et al. (2018) and summarized in Table 2.1.

Table 2.1 Factors affacting durability properties of concrete due to RCA usage (Guo et al., 2018)

Factors	***Change***	***Effect***
Water/Cement ratio	increase	strong negative
RCA size	decrease	mild negative
Residual Mortar amount	increase	strong negative
RCA/NA ratio	increase	strong negative
Parent Concrete Strength	increase	strong positive
Mineral Admixture		positive
Enhancement of RCA		strong positive

The freeze-thaw feature, which generally depends on parameters such as the humidity and temperature conditions of the environment where the concrete is located, the aggregate it contains and the void amount of the mortar, is negatively affected by the use of RCA. The excess water absorbed by RCA due to mortar residue accelerates the damage of concrete due to additional stress during freezing and thawing. It is accepted that the RCA substitution rate and fine RCA/coarse RCA ratio are inversely proportional to the freeze-thaw performance of concrete. The properties of the waste concrete from which the RCA obtained may also affect the freeze-thaw performance of the concrete. For example, it is seen that RCA coming from an old and high-performance concrete in which air-entraining additives were used can provide a higher performance, assuming that the voids are more homogeneously distributed (Ajdukiewicz and Kliszczewicz, 2002).

Due to its low performance, RCA has limited potential for use in structural concrete production that is expected to be exposed to aggressive environmental effects (Gonçalves et al., 2020). At this point, fine RCA and coarse RCA ratios determined in the mixture design can also have a serious impact on concrete performance. The fact that fine RCA contains more mortar residue than coarse RCA causes fine RCA to create a more porous structure and cause more durability problems in aggressive environments (Evangelista and de Brito, 2014). The fact that coarse RCA has a lower porous structure than fine RCA is related to the lower surface area of coarse particles. Fine RCA, on the other hand, contains a larger surface area in this respect. Thus, the total mortar residue in coarse RCA is expected to be considerably lower than in fine RCA. The high-porous structure of fine

RCA affects the permeability of the cocnrete more than coarse RCA (Guo et al., 2018). The high void structure of fine RCA due to its high content of mortar residue also increases the chloride permeability of the concrete in which fine RCA is used. So much so that the negative effect of coarse RCA on chloride permeability is much lower than the effect of fine RCA (Evangelista and de Brito, 2010). The effect of the properties of fine RCA on the permeability becomes very evident. In a study examining the change in chloride permeability caused by the amount of mortar residue on the surface of the RCA, it was revealed that as the amount of mortar residue increased, the chloride permeability increased (Duan and Poon, 2014).

The fact that RCA has a more porous structure and higher water absorption capacity than NA may be advantageous in some cases. For example, the excess water absorbed by RCA may provide additional curing for the hydration process in concrete, thus reducing permeability. In addition, mineral additives that provide pozzolanic properties can be used to reduce permeability and the rate of porous and permeable structure caused by RCA can be reduced. On the other hand, it has been claimed that the porous structure resulting from mortar residue in mixtures using fine RCA allows the internal stresses that occur during freeze-thaw cycles to be absorbed (Nedeljkovic et al., 2021).

2.4.5. Effect of RCA Size on Properties of Concrete/Mortar

The use of RCA obtained from CDW with all grain sizes in new construction activities is a necessity in terms of an environmentally friendly and sustainable approach. However, fine RCA contains high amounts of mortar residue due to its high surface area, and therefore has a higher water absorption capacity, which negatively affects the workability properties of the mortar into which it is incorporated. It also causes weaknesses in terms of mechanics and durability. Especially due to its high permeable structure, it provides less resistance to many deteriorations in structural concrete located in chemically and mechanically aggressive environments (Nedeljkovic et al., 2021). These negative situations experienced in concrete using fine RCA are related to the amount of voids in the mortar residue, the size of the voids and the permeable structure formed due to the distribution of these voids.

Some standards and technical reports restrict or prohibit the use of fine RCA in the production of structural bearing concrete due to its high mortar residue content and high water absorption capacity (RILEM, 1994; ACI 555, 2001). The processes required to remove mortar residue from fine RCA, which is the reason for the negative properties of fine RCA, or to purify fine RCA contaminated with various CDW contents, may be more costly than the production of NA (Nedeljkovic et al., 2021). However, access to the fine river sand needed in concrete production is becoming more difficult day by day and natural resources are being irreversibly destroyed. For this reason, the number of studies on the use of fine RCA in structural concrete production as well as coarse RCA has been increasing in recent years (Evangelista and de Brito, 2010).

Due to the high water absorption feature of fine RCA, more water is used to achieve similar workability in concrete mixtures produced with NA, which changes the effective w/c ratio in the mixture. The water used more than in the mixture with NA concentrates in the new ITZ region formed on the fine RCA surface over time, and this causes the ITZ to take on a structure with higher porosity (Behera et al., 2019). The decreases in compressive strength caused by fine RCA are generally caused by this situation. In order to prevent this decrease, mineral additives with pozzolanic properties such as blast furnace slag, fly ash or metakaolin or chemical additives with different properties can be used. In this way, the effective water rate in the concrete mixture using fine RCA can be reduced and a structure with lower voids can be obtained.

The sub-mm portion of fine RCA, which generally consists of waste concrete dust and may contain different pollutants, poses risks for its use in new concrete in many studies. For this reason, it is stated that separation of content of 63 μm and below is necessary for the widespread use of fine RCA (Nedeljkovic et al., 2021). In addition, it can be predicted that fine RCA, which is generally stored outdoors, may vary depending on the external environmental conditions. In material stored in heaps, it is possible that fine RCA on the outer parts of the heap exposed to air may have carbonated. This shows that fine RCA on the outer surfaces and inside of the heap may have different chemical structures.

Although many sources state that fine RCA should be eliminated in concrete mixtures, there are also studies in which there is no loss in terms of mechanical performance with its use in a certain amount. Mardani et al. (2015) reported that the strength values obtained by substituting natural sand with fine RCA up to 60% were very close to those of the control mixtures, and stated that it was possible to use substitution rates up to 60%. It is also seen that sub-mm RCA can have a positive effect in some cases. It is possible to fill the voids, especially those consisting of larger sized aggregates, with fine fraction RCA and thus show a filler effect. In this way, the porous structure can decrease by a certain amount, the density of the concrete can increase and capillary permeability can decrease (Kwan et al., 2014).

In some studies conducted on mortar samples, it has been shown that, unusually, the use of fine RCA can give higher results than the strength of NA mortar samples (Topçu and Bilir, 2010). This situation is associated with the formation of a stronger interlocking between the cement paste and fine RCA due to the irregular and rough structure of the fine RCA grains. However, the mortar residue on the fine RCA may also contain unhydrated old cement. It is stated that when these parts are hydrated with new concrete mixing water, an increase in strength can be experienced as a result of the formation of an additional binding structure (Braga et al., 2012).

2.4.6. Mortar Residue Removal Methods

As frequently mentioned in the above sections, mortar residue on the RCA is one of the main reasons for the negativities encountered in the concrete including RCA. For this reason, different methods are being tried to get rid of mortar residue or improve its properties. Increasing the ratio of the binder material to be used in the mixture, saturating the RCA with water before mixing, covering the RCA with various cementitious mortars, subjecting it to freeze-thaw in chemical solutions, applying some mechanical abrasion processes are some of these (Singh et al., 2022). One of the most notable of these is study of Abbas et al. (2008) in which they developed a chemical freeze-thaw method. In this study, it is seen that both the mortar residue on the RCA can be removed and a suitable method is presented to determine the amount of mortar residue in the RCA samples. The technique discussed here appears as a revised method obtained by using the methods given in ASTM C666 and ASTM C88 together. RCA, which is kept in sodium sulfate solution and subjected to a certain number of freeze-thaw cycles, is sieved at regular intervals and the process is completed when the part remaining on the sieve reaches a constant weight. At the end of this process, the weight loss of the RCA is calculated and this result is accepted as the mortar residue in the RCA.

2.5. Improvement Techniques of RCA

Replacing NA, which is the most consumed material in concrete produced in high quantities, with RCA attracts attention as an environmentally friendly approach. In this regard, there are many studies on RCA. According to NA, RCA includes three different phases. These are the NA, the mortar residue on the NA surface, and the old ITZ between the NA and the mortar residue. This multiphase structure causes RCA to exhibit different behavior compared to NA. This different behavior usually manifests itself in the form of negative characteristics. The main reasons why RCA is disadvantageous are the ITZ between the old NA and the mortar residue, and the mortar residue in RCA has high porosity and permeability. For these reasons, RCA has a high water absorption capacity and causes the new ITZ formed between RCA and cement paste in new concrete mixtures to become a high-void structure (Memon et al., 2022).

Different approaches are being tried to eliminate these weaknesses of RCA. Some studies aim to physically remove RCA from mortar residue, while others aim to chemically improve the properties of mortar residue. Different methods are tried in these two approaches. Among these, methods include such as pre-wetting (Gonzalez-Fonteboa et al., 2011), acid dissolution (Ismail and Ramli, 2014), mechanical abrasion (Yanagibashi et al., 2005), thermal abrasion (Kuroda and Hashida, 2005), microwave heating (Choi et al., 2014; Bru et al., 2014), use of mineral additives (Zeng et al., 2020), coating with polymers (Kou and Poon, 2010; Santos et al., 2017), biological calcium carbonate precipitation with bacteria (Feng et al., 2021; Sonmez et al., 2021), accelerated carbonation (Zadeh et al., 2021; Dündar et al., 2023).

Unlike healing methods, different mixing techniques have been tried in the literature in order to prevent the disadvantageous situation caused by RCA that has not undergone any treatment. Studies can be found in which the problems caused by the excessive water absorption capacity of RCA can be eliminated by methods such as two or three stage mixing and equivalent mortar volume (Bahraq et al., 2022). It has been reported that some mechanical properties of mixtures prepared with these methods are superior to those of coarse RCA containing mixtures prepared with conventional mixing techniques.

Although a certain amount of improvement has been achieved in the methods exemplified above, it is obvious that some negative effects such as high energy requirements and the emergence of additional concrete dust are also observed. Moreover, their acceptance in construction is still limited due to being time- and energy-intensive, impracticability on large-scale and high cost. For this reason, it is necessary to improve with a more environmentally friendly, sustainable and more feasible method. For this purpose, in this thesis study, the method described as accelerated carbonation was used to improve RCA. Additionally, the effect of RCA improved by various methods on concrete durability is an issue that needs to be investigated (Bahraq et al., 2022). This is one of the main reasons why RCA is widely used and the treatment methods we encounter in the literature cannot be adapted to real life. Therefore, in this thesis study, the durability performance of RCA improved by accelerated carbonation technique was also examined.

2.5.1. Mechanical Abrasion

The process considered in this method generally involves processing RCA for a certain period of time in a mill operating at high speed and removing the mortar residue on the surface of RCA through the abrasive effect in the mill. RCA grains, which generally have a highly rough surface structure, acquire a smoother and angular surface structure as a result of this process. This minimizes the negative impact of RCA in terms of loss of workability. Apart from the energy requirement and production of additional powder material, the mechanical abrasion method is an easily applicable, simple and fast method. On the other hand, the intense energy and equipment maintenance costs used in this method increase the unit cost of the RCA to be obtained (Quattrone et al., 2014).

It has been reported that no change in the mechanical performance of the concrete was observed as a result of the replacement of 33% of the improved RCA with NA by the mechanical abrasion method applied with different mechanisms (Alqarni et al., 2021). In another study, it was stated that there was no loss in mechanical properties by replacing NA with 60% RCA, whose water absorption capacity decreased below 1% as a result of mechanical abrasion (Dilbaş and Cakir, 2021). On the other hand, a highly aggressive abrasion process may damage the aggregate removed from mortar residue and cause the formation of smaller grain size aggregates and cracks in the structure of these aggregates (Feng et al., 2022).

2.5.2. Thermal Process

The heating method, which is one of the methods developed according to the principle of removing the mortar residue that causes the RCA to have poor properties, is based on the degradation of hydration products in the mortar residue on the RCA and weakening its structure as a result of exposing the RCA to high temperatures. In the heating method designed to get rid of mortar residue from RCA, the process is generally carried out at temperatures exceeding 500^oC. When these temperatures are reached, CH and CSH, which are the hydration products in the mortar residue, decompose and CaO formation occurs. As a result of this decomposition, the mortar residue and the ITZ attached to the aggregate lose their strength and begin to crumble and fall off the RCA surface. This separation is also caused by the different expansion behavior of aggregate, mortar residue and ITZ at high temperatures. Mechanical abrasion method can also be used together with the heating method (Katz, 2004; Kumar et al., 2023). In fact, the heating process is a kind of pre-treatment that facilitates the mechanical abrasion method and increases the rate of mortar residue removal.

In this method, the mortar residue, which is heated and whose strength is weakened, can be removed from RCA at a high rate by mechanical abrasion. It is seen that more than 90% of the mortar residue can be removed by considering these two methods together (Feng et al., 2022). There are also studies where the mechanical abrasion step is not used by going to higher temperature levels. However, at temperatures above a certain level, not only the mortar residue but also the NA's properties begin to deteriorate. (Chauhan and Singh, 2023).

On the other hand, this process requires high energy, as in mechanical abrasion. In addition, since mechanical abrasion mechanisms are generally needed after heating, both the equipment supply and the high energy requirement make this method far from being an economical and environmentally friendly method.

2.5.3. Acid Dissolution

The acid dissolution method is generally based on dissolving the mortar residue on the aggregate surface by keeping it in acid solution and obtaining a high rate of mortar removal. Dissolution here occurs by reacting with acid and the hydration products in the mortar residue. Mortar residue and old ITZ decomposed as a result of the reaction decompose from the NA in RCA over time. In studies in the literature, abrasion experiments have been carried out with different acids such as H_3PO_4, HCl, H_2SO_4, CH_3COOH, Na_2SO_4 (Tam et al., 2007, Kim et al., 2017). Similar to the thermal abrasion method, there are also studies in which acid abrasion method and mechanical abrasion are used together (Kazmi et al., 2019).

In studies in the literature, the success of this method is correlated with the result of the removal of mortar residue after the process. It is stated that the success of this process is affected by factors such as the type of acid used, the molarity in the solution, and the retention time in the solution (Tam et al., 2021).

In the experiment conducted on fine RCA treated with H_2SO_4 and HCl solution, it is seen that lower strength is obtained in the mortar samples produced with fine RCA treated with H_2SO_4 (Kim et al., 2017). It was observed that the mortar residue on the RCA surface could be removed, but a gypsum structure formed on the aggregate surface. Therefore, it has been observed that ettringite forms in the hydration on the aggregate surface, which causes a decrease in strength. The authors therefore emphasize that RCA should be washed to get rid of the gypsum formed after treatment with H_2SO_4. Although washing the aggregate seems effective, a large amount of water will be consumed for this process. Washing water will later turn into acidic water and become a waste that needs to be treated (Wang et al., 2017). Such situations appear as handicaps of the acid dissolution method.

2.5.4. Mineral Admixture Solutions

Mineral admixtures are often used as substitute binding materials in new cement concrete mixtures. When used as a binder, CH formed in the mixture can be transformed into CSH through pozzolanic reactions and can contribute to the mechanical and durability properties of concrete. It can be observed in studies in the literature that this feature of mineral admixtures can also be used for RCA improvement. In terms of waste disposal, this method offers an environmentally friendly and economical solution by combining CDW waste, RCA, and industrial wastes, such as fly ash and blast furnace slag. The use of pozzolanic materials not only improves RCA properties, but also contributes to increasing the rate of CSH, the main hydration product sought in new concrete. When studies in which improvements in RCA properties can be achieved with mineral admixtures are examined, it is seen that fly ash, blast furnace slag, metakaolin and various nanomaterials are used (Bahraq et al., 2022).

The mortar residue of RCA and the old ITZ region contain high amounts of CH. With the use of mineral admixtures, CH on the RCA surface is converted to CSH and mortar residue and voids within the ITZ can be reduced (Shi et al., 2016). In the near-surface areas of RCAs, which are generally treated with solutions prepared using cement along with mineral admixtures, the voids are filled with CSH formed as a result of the hydration of the cement. At the same time, CH, both in the mortar residue and from cement hydration, reacts with mineral admixture, resulting in the formation of additional CSH (Dao et al., 2024). As a result, there is a serious decrease in water absorption of RCA. This method ensures that RCA is completely covered and its mechanical properties are improved, rather than improving the mortar residue of RCA. By adding RCA to the new concrete mixture after this process, the structure of the ITZ to be formed will be different from the concrete prepared with RCA without treatment. As a result of reducing water absorption capacity, changing the surface structure and decreasing the crushing index, RCA provides higher mechanical properties in the concrete in which it is used.

In their study, Katz et al. (2004) kept the RCA samples in an aqueous solution containing silica fume for 24 hours and then dried the RCA in an oven. After the procedure, a weight increase in RCA was observed compared to before the procedure. In the SEM imaging performed in the study, it was observed that the void areas in the mortar residue decreased and a denser structure was formed. By applying silica fume to RCA, the porous and cracked structure of RCA can be improved, the water absorption capacity of RCA can be reduced, and the new ITZ to be formed in the new concrete mixture can be strengthened.

In study of Dao et al. (2024), RCA samples were kept in cement + fly ash slurry for 24, 48 and 72 hours. As a result of the process, it was observed that the surface of the RCA became less rough and had fewer voids, and some mechanical properties improved. In study of Shi et al. (2018), in addition to silica fume, they also studied the effect of fly ash and nano SiO_2. RCA samples kept in solutions prepared with these materials and dried were examined, and it was found that fly ash showed the highest level of pozzolanic effect. However, it has been reported that a higher microhardness value was obtained with silica fume solution and a more intense ITZ was observed.

The effectiveness of mineral admixtures depends on their fineness. As the particle size of the additive decreases, its absorption from the capillaries on the RCA surface will increase. Apart from mineral admixtures, nanomaterials used for RCA treatment can be highly absorbed by RCA due to their small size. While the strength and durability properties of RCA are significantly improved by soaking RCA in nanomaterial solution, on the other hand, a significant disadvantage occurs in terms of cost. Therefore, researchers have tried different application methods (Li et al., 2021). The results of this study showed that the nano material applied to RCA by spraying did not increase the density value of RCA much, but there was a reasonable improvement in properties such as workability and strength in the concrete mixture in which it was used. It was observed that nano silica applied to the RCA surface by spraying method consumed the CH on the RCA surface, formed new CSH gels and improved the properties of the newly formed ITZ.

It is claimed that when RCA coated with cement + nano silica mixture is used in new concrete, some of the nano material on its surface disperses into the new ITZ region and exhibits nucleation properties for the hydration of the binder cement in the new concrete mixture. As a result, it has been reported that a denser ITZ can be obtained with nanomaterial (Zhang et al., 2016).

Mineral additives appear to be an effective method to improve the porous structure of RCA. However, it is stated that it lags behind the accelerated carbonation method in terms of property improvement (Memon et al., 2022).

2.5.5. Sodium Silicate Solution

The use of sodium silicate solution, which is a method similar to healing with mineral additives, is also among the RCA healing methods. In this method, the CH on the surface of the RCA treated with the prepared solutions bonds with the silicate in the solution and forms CSH. In this respect, mineral additives are similar to the method used. In this method, which is beneficial in reducing the void ratio of RCA, the high concentration of the prepared solution may have a harmful effect. RCA treated with high concentration solutions can affect the alkalinity of the new mortar mixture, making the produced concrete susceptible to alkali-silica reactions. In addition, it has been reported that there are decreases in compressive strength in new concrete mixtures with RCA processed with this method (Shaban et al., 2019).

2.5.6. Polymer Coating

In this method, which is applied by impregnating or spraying solutions prepared using different polymers onto the RCA, a highly water-impermeable layer of equal thickness is generally formed on the surface of the RCA. The polymer solution applied to the RCA surface dries quickly and makes the RCA surface waterproof. Thus, an RCA with low water absorption can be obtained and the workability of the mixture can be positively affected. Depending on the polymer type used and the concentration of the solution, the rate of decrease in the water absorption capacity of RCA may vary. For example, there are studies where the water absorption capacity can be reduced from 4.5% to 0.75% with the polymer impregnation method (Spaeth and Tegguer, 2013). However, the fact that the relationship between RCA and the new mortar mixture is significantly reduced may cause negative effects on the strength of the new concrete. This situation is associated with two reasons. First of all, preventing RCA from entering water into the mortar residue may cause the unhydrated cement content in the mortar residue to not be able to meet with water and additional CSH gels cannot be produced. The second reason is that the water-repellent structure created on the RCA surface has low strength of the connection between the new cement paste and RCA (Shi et al., 2016). On the other hand, there are comments that the polymeric coating on the RCA surface may be worn and damaged by friction during the new mortar mixing process (Feng et al., 2022). If this situation occurs, the effect provided by the polymer coating can be eliminated very quickly.

2.5.7. Bacterial Calcium Carbonate Precipitation

Another method that has been tried to improve RCA features is to create calcium carbonate (CC) precipitation using bacteria. In this method, CC precipitate forms in the RCA voids in a bacterial environment under appropriate conditions and RCA voids are reduced (Sonmez et al., 2021). This biomineralization method, which can significantly reduce the water absorption capacity of RCA, is one of the promising methods in improving the properties of RCA. This method, which appears as an environmentally friendly approach since the bacteria used in the application are naturally occurring bacteria and does not require any cleaning method, is still one of the subjects widely studied in the literature and seems far from being a practical method for real environment applications (Memon et al., 2022).

The metabolic behavior of bacteria ensures the formation of CO_3^{-2} in their environment. If Ca^{+2} is present in their environment, these two products combine to form CC precipitation. The CC structure obtained by this method usually appears as vaterite and calcite (Chen et al., 2023). The amount and crystal structure of the precipitation that occurs may vary depending on the type of bacteria used, the metabolic activity of this species, the alkalinity of the environment, the type of Ca source and the duration of application (Dhami et al., 2013). As the application time increases, CC accumulation increases and capillarity on the surface of the RCA decreases. Therefore, it is difficult to achieve gains in a short time in the biocalcification method. Therefore, the potential for use of this method in real engineering applications seems low.

2.6. Accelerated Carbonation

Carbonation processes with CO_2 curing under suitable conditions are seen as one of the most environmentally friendly and sustainable RCA improvement techniques (Shi et al., 2016; Li et al., 2017; Zhan et al., 2018). With the accelerated carbonation application, which is performed much faster than natural carbonation with atmospheric CO_2, RCA pores can be refined and lower water absorption capacity, higher density and higher mechanical properties can be obtained (Dündar et al., 2023).

Carbonation reactions occur very slowly and over long periods of time under normal environmental conditions. In special cabins where the environmental conditions can be changed, these times can be shortened considerably and an improvement technique for RCA can be obtained. This method, called accelerated carbonation, provides high amounts of CC formation and appears as an environmentally friendly approach that improves the properties of both old and new ITZ.

During carbonation, CH and CSH in the mortar residue react with CO_2 under suitable conditions, and CC, silica gel and water emerge as the final product (Zadeh et al., 2021) (Figure 2.14). Apart from this, different reactions may occur depending on the different minerals in the mortar residue. Reactions during carbonation generally occur as in the following equations:

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$$

$$C\text{-}S\text{-}H + CO_2 \rightarrow CaCO_3 + SiO_2.nH_2O$$

$$Ca/Mg\text{-silicate} + CO_2 \rightarrow (Ca/Mg)CO_3 + SiO_2$$

$$(Ca/Mg)^{+2} + CO_3^{-2} \rightarrow (Ca/Mg)CO_3$$

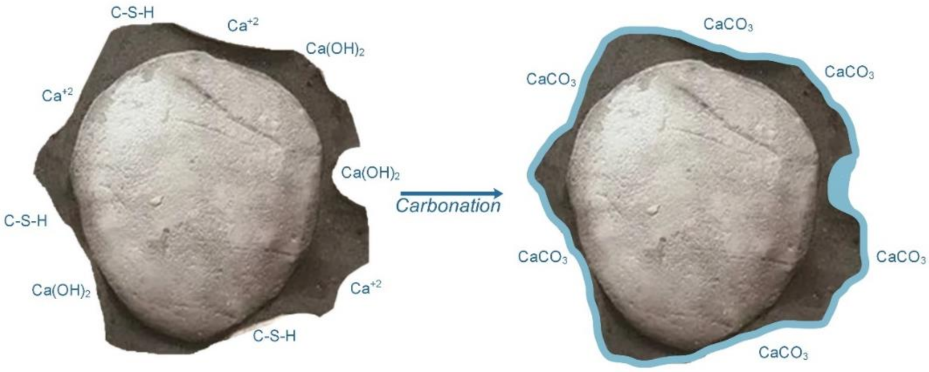


Figure 2.14 Schematic accelerated carbonation reaction taking place on the RCA surface

Mineralization of RCA via accelerated carbonation provides RCA with improved density, lower porosity, lower water absorption capacity, higher strength, and resistance to external detrimental agents. In the current literature, the efficiency of accelerated carbonation was enhanced by applying various arranged operational- and material-related parameters during the carbonation procedure. In this regard, the effects of humidity, pressure, temperature, carbonation duration, the particle size of RCA, CO_2 concentration, static/dynamic carbonation methodologies, and incorporation of carbonation accelerator agency on the efficiency of the accelerated carbonation process were some influential parameters tested in the literature (Dündar et al., 2023). However, the literature studies showed that there is no optimum accelerated carbonation condition valid for each type of RCA. Depending on the parameters such as sources and particle size distribution of RCA, temperature, humidity, pressure, carbonation medium, etc., the optimal condition for each parameter range may differ under different carbonation parameters (Pederneiras et al., 2022).

2.6.1. Effect of Relative Humidity

As is known, in completely water-saturated or high relative humidity (RH) environments, CO_2 ingress into samples slows down or may even stop completely. Therefore, the optimum condition in terms of humidity is a low/medium humidity condition. Zhan et al. (2014) stated in their study in this context that the 40-70% RH range is the most suitable conditions for accelerated carbonation. In some studies, it is seen that RCA is subjected to some pre-treatments such as soaking or wetting/drying. However, in Zhang et al. (2015) and Xuan et al. (2016) studies, it is seen that keeping RCA in a carbonation cure chamber at 20±2 ^{o}C temperature and 60±5% RH without any pre-treatment is very effective.

In a study investigating the effect of RH on the carbonation of $Ca(OH)_2$ nanoparticle (López-Arce et al., 2011), 75% and 90% RH environments caused calcite, vaterite and aragonite formation and compared to lower RH rates (33%, 54%), 75% and 90% RH caused CC precipitation in higher crystal structure and larger particle sizes.

Xu et al. (2022), who investigated the effect of RH on carbonation reactions in their review article, states that there is a consensus in the literature on the optimum RH, that carbonation reactions are limited in a low RH environment, and that the transport of reactants becomes difficult in high RH and therefore carbonation reactions are restricted.

Galan et al. (2013), who researched the carbonation kinetics of cement paste in long-term natural carbonation and accelerated carbonation scenarios, states that in very low RH, there will not be enough water to dissolve the reactants and to carry out the reactions, and in a high RH environment, the pores will be filled and CO_2 diffusion will be prevented. It means that in an environment of medium RH, the reactants can easily dissolve and there will be enough water for CO_2 diffusion and enough space for carbonation products. Since there is also cement paste residue on the RCA surface, the carbonation kinetics of this residue will also require a similar RH condition. In another study examining the carbonation of hardened cement paste (Doruet et al., 2019), it was emphasized that as the RH decreases, the formation of calcite in a stable structure decreases, and the calcite formed is in a metastable structure.

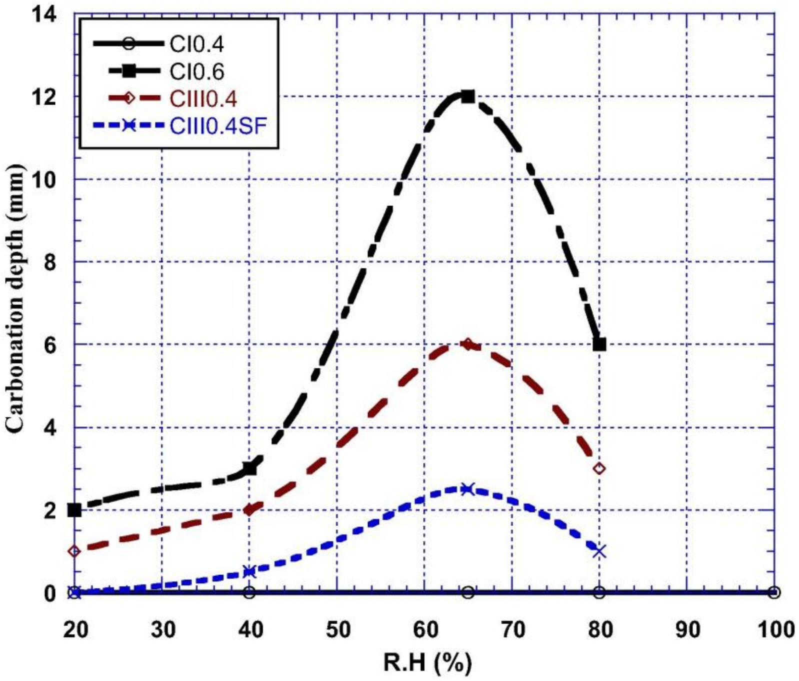


Figure 2.15 Variation of carbonation depth of concrete samples in different strength classes according to relative humidity (Leemann and Moro, 2017)

Leemann and Moro (2017), who examined the carbonation depth of concrete samples of different strength classes, examined the effect of RH on the carbonation depth. As seen in Figure 2.15, 60-70% RH increased the carbonation depth considerably compared to lower or higher RH rates.

RH affects both the amount and structure of the carbonation reaction products. It is stated that product formation accelerates in carbonation at optimum RH values and amorphous CC and monohydrocalcite formation is observed, while vaterite and portlandite are observed in a lower RH environment and this reaction occurs slowly (Liang et al., 2020).

2.6.2. Effect of Pressure

Ambient pressure is also considered as a parameter by researchers in accelerated carbonation processes, and has a serious effect on carbonation performance of RCA. (Xuan et al., 2016; Kashef-Haghighi et al., 2015; Fang et al., 2017). The usual situation is that the increase in pressure increases the rate of dissolution of CO_2 in the porous structure and the carbonation reaction accelerates (Carroll et al., 1991; Hyvert et al., 2010). However, using too high pressure in the process may cause negative results. High pressure can cause the degradation of mortar residue, especially on the low-quality RCA surface, the formation of cracked structure and the formation of undesirable fine-sized material (Liang et al., 2020; Xuan et al., 2016; Zhan et al., 2017). Some studies in the literature state that the ambient temperature may increase due to excessive pressure and microcracks may occur even in the carbonated structure on the RCA surface (Xuan et al., 2016; Liang et al., 2019; Fang et al., 2017; Zhan et al., 2016(b); Wu et al., 2022).

Xuan et al. (2016) examined the changing properties with an increase in pressure from 0.1 bar to 5 bar in the curing chamber. It is stated that with increasing pressure, an increase in values such as density, crushing value and carbonation percentage in RCA is observed. On the other hand, Vivian et al. (2016) stated that some brittle, soft RCA may break with high pressure and fine-grained materials of undesirable sizes may form. In this regard, it has been stated that significant healing performance can be achieved with low pressure and longer curing time.

2.6.3. Effect of Temperature and Carbonation Duration

Another effective parameter observed in the carbonation process of RCA is ambient temperature (Li and Wu, 2022; Wang et al., 2019). It is seen that the carbonation rate increases when the temperature is increased to a certain level (Fernández Bertos et al., 2004). Liu et al. (2001) states that the CO_2 capture rate will increase as the temperature increases to 60^{o}C, and the dissolution rate of CO_2 gas in water will decrease at higher temperatures. On the other hand, it is also stated that the heat generated due to the exothermic nature of the carbonation reaction enables the CC formed as a result of the reaction to form in a meta-stable structure and that carbonation must be carried out at lower temperatures to obtain calcite in a stable polymorphous structure (Asavapisit et al., 1997).

The temperature parameter also affects the crystal structure, polymorph structure, amount and particle size of the CC formed. Lu et al. (2022), who studies such details on the carbonation mechanism of cement paste, claims that with an increase in temperature, the transfer rate and kinetic energy of the particles in the environment will increase, thus accelerating the carbonation reactions (Figure 2.16). It was stated that while vaterit was observed in CC formation at low temperatures, a more stable polymorphic structure such as aragonite with larger particle sizes was observed at high temperatures (50^{o}C).

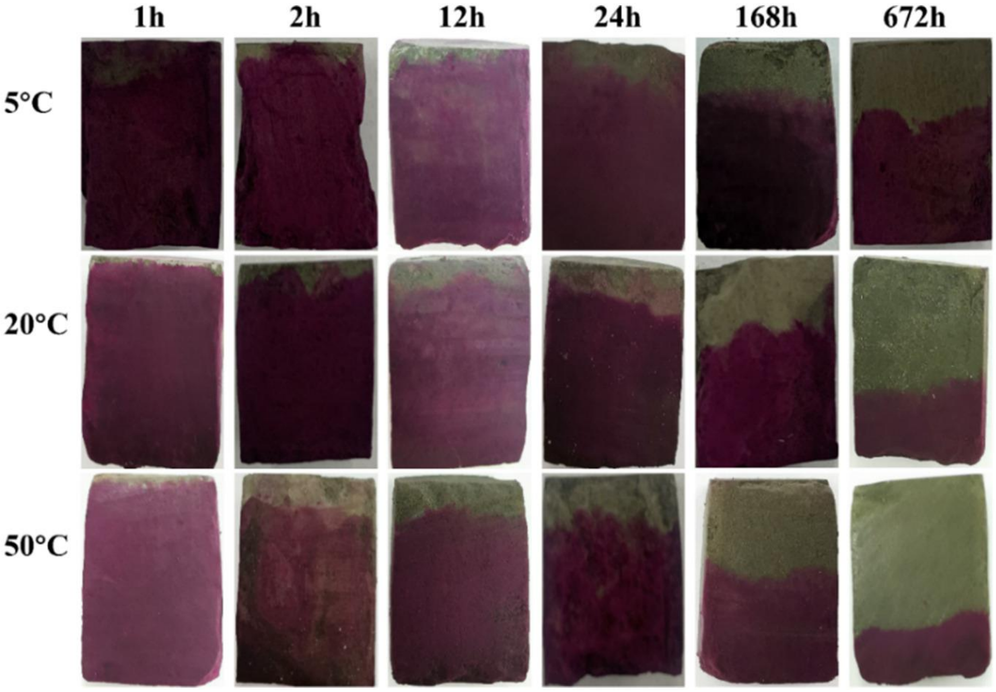


Figure 2.16 Carbonation depths of cement paste samples carbonated at different temperatures and durations (Lu et al., 2022)

In the study of Lu et al. (2022), it was stated that the carbonation rate increases as the temperature increases. On the other hand, there was a decrease in the carbonation rate with increasing temperature and increasing carbonation duration. A rapid formation of CC occurred at the beginning of the carbonation, and the porous structure was filled by the rapid coating of the uncarbonated structure in the inner parts with CC. As the carbonation time increases, CC formation continues, but the formation rate slows down considerably. This is associated with the pause in CO_2 diffusion due to the filled porous structure. Similar effects are observed in the carbonation of cement paste residue on the RCA surface. Zhan et al. (2014) reported the carbonation rates obtained at the end of 0.5, 1, 2, 3 and 4 hours of carbonation in their experiment with 0.1 bar CO_2 pressure in the carbonation chamber (Figure 2.15). As understood from these results, it was observed that there was a significant increase in the carbonation percentage with the curing application for up to 3 hours, but this increase slowed down after 3 hours. Additionally, another point emphasized in this study is that the increase in carbonation rate and the weight increase in RCA are experienced mostly at the beginning of the curing period.

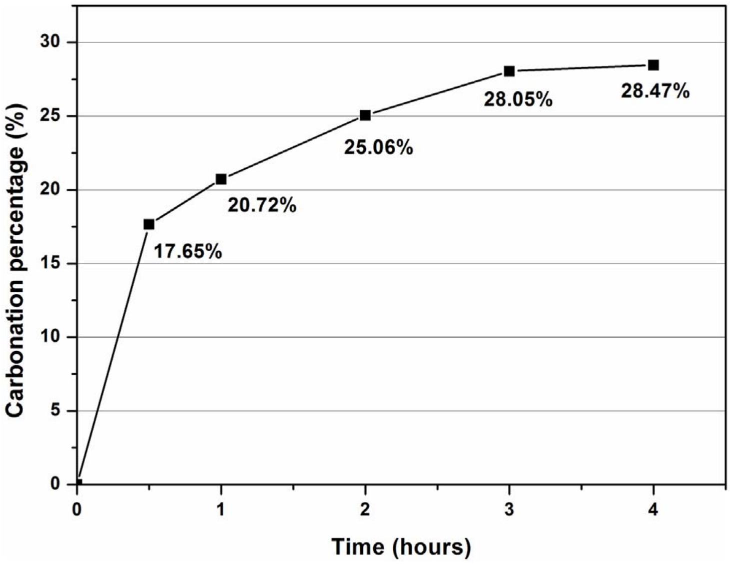


Figure 2.17 Relationship between CO_2 cure application time and carbonation rate (Zhan et al., 2014)

As can be seen in the author's article and the results section of this thesis, as the carbonation time increases, CC formation continues, but after a while it slows down. It is understood that the reason for this is that after CC formation on the cement-based mortar residue on the RCA surface, a passivation film is formed by covering the surface of the RCA and CO_2 permeability decreases (Zhan et al., 2016; Kurda et al., 2019; Shi et al., 2012; Kou et al., 2013, Silva and Andrade, 2017; Ahmed et al., 2018). It is thought that this passivation film also prevents the release of uncarbonated Ca^+ ions in the inner parts.

As a result, the common conclusion drawn from the author's study and other examples in the literature is that the increase in temperature accelerates the formation of CC in the early times of the carbonation process, and the formation of CC slows down with the increase of the carbonation time, regardless of temperature.

2.6.4. Effect of Particle Size

The particle size of RCA is one of the main parameter affecting the RCAs’ CO_2 uptake rate during the accelerated carbonation process. It is seen from the literature that this parameter is generally related to the total surface area provided by the RCA sample for carbonation, according to its particle size distribution.

Li et al. (2019) who examined the fracture strength properties of RCA particles of different sizes (20-30, 30-40 and >40 mm) after accelerated carbonation process, found that RCA with small grain size captured more CO_2. They stated that concrete samples containing finer carbonated RCAs showed lower water absorption rate, higher microhardness and higher crushing strength.

Kikuchi and Kuroda (2011) also found a similar result. They showed that RCA with finer particles captured more CO_2 when carbonation was carried out with the wetting-drying cycle.

Thiery et al. (2013), who parametrically examined the effect of the w/c ratio of the mortar residue on the RCA surface and the particle size of the RCA on the CO_2 capture performance in the carbonation process, similarly emphasized that the degree of carbonation is higher in RCAs with partile sizes of 2 mm and below.

Sereng et al. (2021), in his study on a similar subject, where he examined many parameters in the accelerated carbonation process of different types of RCAs, stated that RCAs in the 1-4 mm size range of each different type capture much more CO_2 than those in the 12-20 mm size range. They found that as the grain size decreases, the time required for CO_2 diffusion also decreases.

Zhan et al. (2014) also observed in their study that CO_2 absorption increased as RCA particle size decreased. This is associated with the availability of surface area that provides higher contact with CO_2 in small particle sizes. In the study, it was observed that

56% of the CO_2 applied in the accelerated carbonation process performed on RCA samples with particle sizes between 5-10 mm was captured by the RCA and there was a total weight increase of 3.88% in the RCA. It has been reported that this effect is considerably greater than the effects observed in samples in the 10-14 and 14-20 mm particle size range. In the published article of the author (Dündar et al., 2023), it was reported that in the carbonation process carried on different particle size ranges, the size range that captures the highest amount of carbon is 2-4.75 mm.

2.7. Valorization of RCA in Cement-based System Through Carbonation

Studies have shown that RCA has many disadvantages compared to NA. These include negative features such as high water absorption capacity, permeability, and high void ratio. These problems prevent the use of RCA in new concrete as it is, and cause weaknesses in terms of mechanics and durability in the new concrete mixture in which it is used (Shi et al., 2016; Tam et al., 2018; Ai et al., 2020). To overcome this obstacle, various improvement practices mentioned above have been tried. However, many of these methods have been shown to be infeasible in terms of time, cost and energy. As an alternative to these methods, carbonation with a CO_2 curing chamber stands out as a cost-effective method (Dündar et al., 2023). For this reason, the accelerated carbonation technique was discussed in this thesis study, and a number of mechanical and durability tests were carried out with the improved RCA obtained by this method.

In the author's previous study (Dündar et al., 2023) and other studies in the literature on a similar subject, parameters such as curing time, cabin pressure, temperature, humidity rate, CO_2 concentration, RCA particle size, static/dynamic application methods were examined in order to achieve optimum carbonation. In this study, where many parameters such as temperature, pressure, CO_2 concentration, curing time, particle size were examined, the highest CO_2 capture rate was achieved with a particle size of 2.00-4.75 mm. It has been reported that the optimum carbonation conditions are 50°C temperature, 70% relative humidity, 1 bar ambient pressure, and 48 hours of curing time. In the thesis study, CRCA with these optimum values were used in new cementitious mortar mixtures. Some mechanical and durability tests were carried out on the produced mortar samples.

Some of the studies in the literature dealing with mechanical and a small number of durability tests on samples produced with CRCA are summarized below.

2.7.1. Mechanical Performance Provided by CRCA

It is seen from the studies in the literature that with CO_2 cure, the density of RCA increases and their water absorption capacity and crushing values are reduced. In these studies, performance tests were also conducted on cementitious mortars produced with cured RCA. It has been observed that the compressive strengths of mortar samples produced with CO_2-cured RCA are close to the compressive strengths of mortar samples produced with natural sand and give higher values than those produced with untreated RCA. It is stated that the underlying reason for these results is the improvement in the surface of RCA and thus the formation of a denser and stronger ITZ (Figure 2) (Zhang et al., 2015).

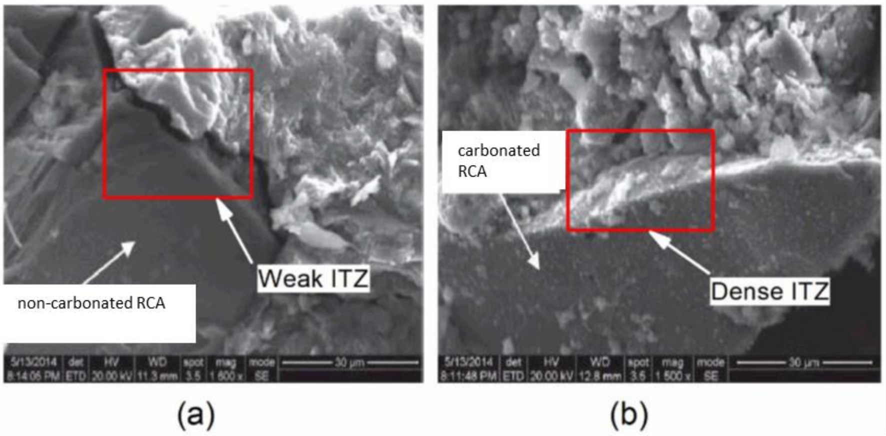


Figure 2.18 Interfacial transition zone of mortar produced with (a) untreated and (b) CO_2 cured RCA (Zhang et al., 2015)

However, it has also been reported that a decrease in drying shrinkage and an increase in resistance to chloride diffusion were observed in mortar samples produced with carbonated RCA. In another study (Vivian et al., 2016), it was also emphasized that other mechanical properties of samples using 30% carbonated RCA were very similar to the properties of samples using completely NA. Xuan et al. (2016), confirming this situation, stated that with the use of up to 60% carbonated RCA, there will be no serious loss in mechanical properties compared to the use of NA.

Tam et al. (2020) found that the flow performance of CRCA is much better than RCA. Pu et al. (2021) based CRCA's better slump value on the reduction of voids with the formation of CC and therefore the reduction of water absorption capacity. It has been stated that the change in slump values with NA, CRCA and RCA is also due to the difference in particle shapes (Kim et al., 2018). In another study, while a flow value of 220 mm was observed in the mortar produced with 100% CRCA, it was reported that a flow of 140 mm was observed in the mortar produced with 30% CRCA + 70% RCA. It has been claimed that this situation is evidence that there is a decrease in porosity in CRCAs and that water ingress into the aggregate is prevented (Tam et al., 2016).

Xuan et al. (2016-b) concluded that the micro-hardness value of ITZ in the CRCA system was higher than the ITZ of the RCA system. This development in ITZ has also increased the bending and splitting tensile strength. The improvement in the ITZ was attributed to the relatively lower w/c ratio around the CRCA as a result of its low water absorption (Waseem, 2022; Xuan et al., 2016-b). In the study of Wang et al. (2021), a 17.54% increase in bending strength was observed with the use of CRCA compared to RCA. In fact, it has been observed that there is almost no loss in bending strength when CRCA is replaced with NA up to 60% (Xuan et al., 2016-b).

As it is known, CRCA generally has lower water absorption capacity than RCA. Since less water is used in the production of CRCA concrete than in the production of RCA concrete in order to reach the desired slump value in the prepared mortar or concrete mixture, it is expected that the effective w/c ratio in the mixture will remain low, and therefore the compressive strength will be higher (Wang et al., 2020). Waseem (2022) stated that 11% more strength was achieved with CRCA compared to RCA, and only 5.9% lower strength was observed compared to NA. This increase is attributed to the strengthening of the ITZ due to the increase in its density as a result of carbonation causing the formation of CC in the pores of the RCA.

More voids and cracked structures were found in the ITZ observed in an SEM analysis performed on the concrete sample containing untreated RCA. This structure, which causes low strength, was observed much less in concrete using CRCA. The reason why

CRCA gave a structure with low voids and cracks was that the formed CC and silica gel filled the void structure and CC could react with the aluminate phase (Lu et al., 2019). The formation of structures with high voids and column-like ettrengite is frequently observed in the untreated RCA system. This causes expansion and additional stress in the internal structure. In the CRCA system, fiber and cotton-like calcium and silica gel products were formed, which contributed to a decrease in the void ratio and an increase in strength (Shi et al., 2018).

It is seen that the degree of water absorption can be significantly reduced in concrete samples produced with CRCA. As a result of 28 days of curing, it was determined that concrete including CRCA absorbed 21.12% less water than that including RCA (Waseem, 2022). This situation is associated with the formation of CC, lower void ratio and a denser matrix formation. Apart from carbonation, there are also studies that reduce the amount of water absorption with different surface improvement methods. Zhu et al. (2013) reported that the mixture using RCA processed with water-repellent materials increased its resistance to capillary water absorption, carbonation and chloride ion permeability in terms of durability.

It is seen that carbonation also improves sorptivity and the difference between RCA and NA can be compensated to a certain extent. In the study of Waseem (2022), it was observed that sorptivity gave the highest value in samples including RCA, followed by CRCA and NA, respectively. It has been interpreted that this situation is parallel to the trend in water absorption properties. Similarly, Gomez et al. (2021) obtained a much lower capillary coefficient with fine CRCA for 5 hours than with uncarbonated fine RCA.

2.7.2. Durability Performance Provided by CRCA

The high water absorption capacity and high degree of water saturation in RCA reduces the freeze-thaw resistance of the concrete in which it is used (Zaharieva et al., 2004). The excess water absorbed by RCA used in mortars can pass into the cement paste phase during freeze-thaw and reduce the freeze-thaw resistance. For this reason, the resistance of concretes where RCA is used directly in aggressive thermal environments is generally low and therefore the direct use of RCA is not recommended (Guo et al., 2018). On the other hand, studies where CRCA is used in new mortar or concrete samples, showed that CRCA greatly increases the freezing and thawing resistance of concrete according to that of concrete made up with RCA (Li et al., 2017; Kazmi et al., 2020).

Surfaces with old mortar residues with high CH cause very harmful void structures. This porous structure makes it easier for water and chloride ions to leak into the concrete. Since this void structure decreases and improves with the formation of CC, it is expected to observe much lower chloride permeability in concrete produced with CRCA. For this reason, the chloride diffusion coefficient decreases with the use of CRCA. In a study, it was reported that concrete produced with CRCA had approximately 41-45% lower chloride permeability than those containing RCA (Shi et al., 2018). It has been discovered that RCA provides an easier environment for the passage of chloride ions, while CRCA significantly reduces ion permeability due to the increase in density as a result of the decrease in porous structures due to the formation of CC (Waseem, 2022). In another study, it was observed that the permeability coefficient decreased by 44, 64.5 and 79.9% with the use of CRCA with residual mortar w/c ratios of 0.35, 0.45 and 0.55, respectively. From this result, it is understood that carbonation has a more serious effect in low quality RCA. It has been evaluated that the carbonation percentage may be higher than that of high quality RCA, since low-quality RCA has a higher percentage of voids on its surface that will allow carbonation (Liang et al., 2019).

There are statements that the risk of reinforcement corrosion may actually increase in CRCA concretes due to the decreasing alkalinity degree as a result of the carbonation of RCA (Tang et al., 2018). It is claimed that carbonation can reduce free alkali metal and hydroxyl ions, thus the pH of the environment may decrease and, as a result, the passivation in the reinforcement may be weakened (Weerdt et al., 2019). On the other hand, since permeability is significantly reduced by carbonation, the entry of harmful ions into the concrete is prevented and the risk of corrosion initiation can be indirectly reduced (Xuan et al., 2017).

In this thesis study, an answer was sought whether the disadvantages caused by RCA in terms of mechanics and durability could be eliminated by improving RCA with the accelerated carbonation method. Studies in the literature that comprehensively address multiple mechanical and durability effects are very limited. In one of them, it was found that there were very few studies examining the durability effects on samples produced with coarse CRCA and fine CRCA; In fact, it has been stated that there are no studies that combine these two (Liang et al., 2019). It has been emphasized that there are very limited studies in the literature with different results in terms of durability, especially gas and chloride ion permeability, and therefore detailed and comprehensive studies should be carried out in terms of durability (Li et al., 2022). On the other hand, it is understood that the effect of carbonation has more impact on durability properties rather than mechanical properties (Zhan et al., 2014). For these reasons, this study focuses on effects in terms of durability along with mechanical effects.

In some studies, it has been mentioned that studies examining the negative effects caused by fine RCA are quite limited and comprehensive studies in terms of durability cannot be found (Kirthika and Singh, 2020). It has been stated that fine RCA affects the impermeability of concrete more than coarse RCA, and that this effect is associated with the higher capillary voids and cracked/permeable structure in fine RCA (Bravo et al., 2015; Martinez-Lage et al., 2012). It has been stated that fine RCA affects chloride permeability more than coarse RCA because it contains more mortar residue or clay content (Bravo et al., 2015; Evangelista and Brito, 2010). As an example, in the study of Duan (2014), it was reported that higher ion permeation resistance was achieved with coarse RCA with removed mortar residue. As a result, the scope of the study was limited to the fine RCA particle size range of 0.85-4.75 mm due to the reasons mentioned above, such as the fact that fine RCA affects the mechanical and durability properties of concrete more than coarse RCA, the fact that there are few comprehensive studies examining the durability properties of fine CRCA, and especially fine RCA captures more CO_2 than coarse RCA.

3. EXPERIMENTAL STUDIES

3.1. Studies on Aggregates

In the first stage of the thesis study, low quality RCA was obtained from rubbles originated from CDW and was made ready for the accelerated carbonation process. In this first stage, the carbonation performance of RCA in the light of all parameters was examined with various analysis methods.

3.1.1. RCA Preparation and Carbonation Techniques

Low-quality end-of-life concrete rubbles were acquired from an Ankara urban transformation area in the form of various-sized pieces of rough fragments (Figure 3.1). RCA was obtained by crushing via jaw crusher and sieving by following the ASTM C136. Obtained RCA was then classified according to particle size ranges of <0.85 mm, 0.85-2.00 mm, 2.00-4.75 mm, 4.75-9.50 mm, and 9.50-14.00 mm. The physical appearances of the obtained RCAs with various particle sizes were presented in Figure 3.1. As the demolished buildings were at the end of service life and constructed with expired specifications providing relatively low mechanical performance (with average compressive strength values of 8.7 MPa measured from the demolished buildings by using obtained core samples), the used RCAs can be regarded as low-quality.

Figure 3.1 Views of concrete rubbles and RCAs in different particle sizes

To improve the properties of RCA, to increase the speed of carbonation reactions of RCA and to provide an effective process for permanently removing CO_2 from the atmosphere, a dynamic carbonation technique was followed in this study. In this regard, a lab-scale carbonation reactor designed by the research team was used to provide an accelerated carbonation process for low-quality RCA. The reactor was designed to provide a reliable, controlled environment by arranging the relative humidity, pressure, temperature, and CO_2 concentration for the carbonation reaction. The details of the developed carbonation reactor are demonstrated in Figure 3.2. In this reactor, the temperature was driven by the internal heaters and sensors, and temperature settings range from 20±3 ^{o}C to 110±3 ^{o}C. The relative humidity of the medium was controlled by a humidifier and can be operatable in a range of 30±5% and 95±5%. The CO_2 concentration of the reactor which has been chosen considering the CO_2 concentration of the emitted gas through the cement plants, run from 0 to 15±2% via a concentrated CO_2 tank (Torrenti et al., 2022). The pressure of the reaction medium is arranged with an air tank and can be set from 1 to 6 bar. The reactor has a cylindrical steel chamber with a capacity of approximately 25 l. However, the filling ratio of the reactor for effective process control is up to ~10%. In order to keep the CO_2 contact at a high level and to provide homogeneous carbonation, a mixer providing rotational mixing was designed for the carbonation reactor. The rotation speed of the mixer can be arranged in a range of 0 to 1 rpm. There is a control panel that stabilizes the interior of the chamber through sensors and adjusts the interior conditions.

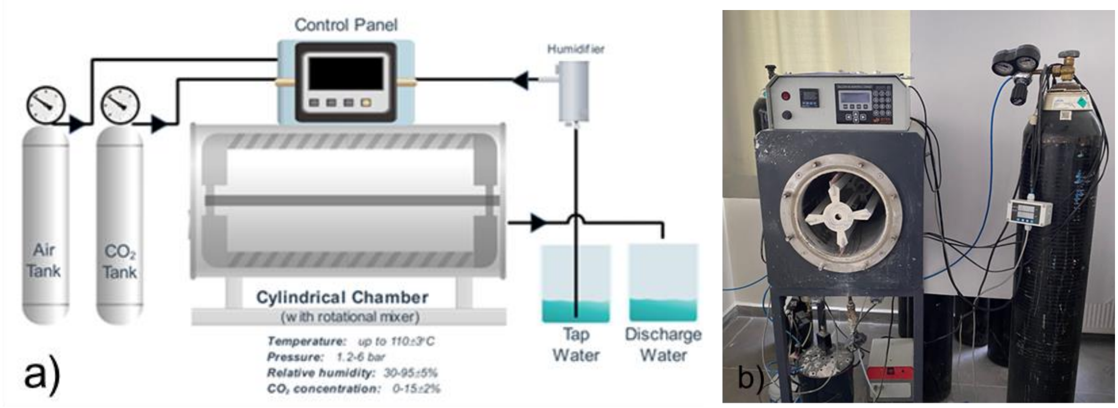
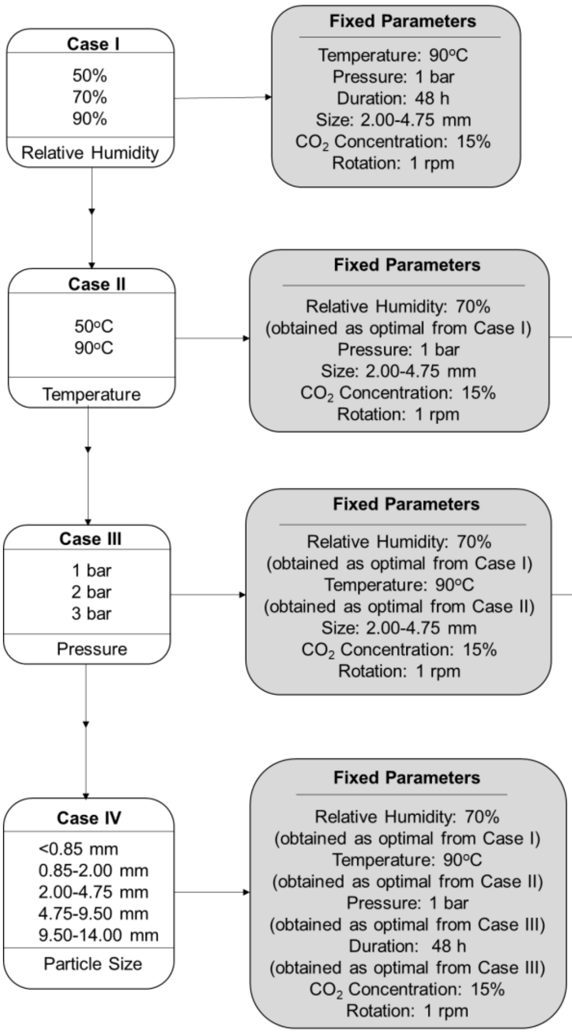


Figure 3.2 a) Schematic view of carbonation chamber setup, b) the view of the carbonation chamber

In this study, to optimize the carbonation efficiency of RCA, a series of carbonation processes were implemented on the low-quality RCA under various operational conditions arranged by the pre-set parameters summarized in Table 3.1. Process parameters were designed comprehensively considering the industrial implementation and emitted flue gasses (Torrenti et al., 2022). In total, four different cases were designed to investigate the effects of relative humidity, temperature, carbonation duration, pressure, and particle size of RCA on the carbonation efficiency of the low-quality RCA. When designing these four cases, it was aimed to prevent a sharp increase in the effort to figure out the optimum carbonation process with the rise of the number of independent variables. In this regard, in favor of simplifying the experimental program, the main effective parameters were treated as independent variables. The relevant parameter was investigated by fixing some other parameters that will be investigated in the next cases.

Table 3.1. Flowchart of parameters selected for different carbonation cases



3.1.2. Testing Methods on Aggregates

To investigate the effects of various chemical carbonation processes implemented on the low-quality RCA, several microstructural analyses were performed on CRCA and RCA, including thermogravimetry (TG/DTG) analysis, scanning electron microscopy (SEM) analysis, and X-ray diffraction (XRD) analysis. Water absorption capacity test was also carried out to assess the surface modification level of RCA after the carbonation process. This experiment was conducted by following the procedure given in ASTM C128.

3.1.2.1. Thermogravimetry (TG/DTG) Analysis

TG/DTG analysis was performed to assess the chemical composition of CRCA and RCA by focusing on the consumption of CH and the formation of different forms of CC. For the analysis, the RCA was first crushed to obtain powder specimens and then dried in a laboratory-type oven at 40 ^{o}C. The RCA specimen with a weight of 10–30 mg was used for testing. In each analysis, at least three separate specimens were used. Analysis was carried out on the specimens by heating up to 1000 °C from 40 °C at the rate of 10 °C/min. by using Shimadzu DTG-60H. Heat flow-temperature and weight-temperature data graphs were obtained and used to assess the efficiency of carbonation process parameters. The fluctuations observed in these graphs were used to determine the amounts of CH and CC in the sample. The weight loss observed in the temperature range of 375-450 °C (Δ_m 375-450) indicates the decomposition of CH. In addition, CC is characterized by weight loss in the temperature range of 600–890 °C(Δ_m 600-890). In this temperature range, CO_2 in the CC structure dissolves and decomposes (Alarcon-Ruiz et al., 2004; Zhang et al., 2015). To calculate the CO_2 uptake rate of RCA after the accelerated carbonation process, the differences between CC amount of CRCA and RCA were considered (Morandeau et al., 2014).

3.1.2.2. Scanning Electron Microscopy (SEM) Analysis

SEM was used to examine morphological changes because of the carbonation process. The SEM examination was performed under vacuum conditions with a working distance of 10 mm. RCA samples were dried in a 40 $^{\circ}$C oven before analysis. The morphologies of carbonation reaction products were observed in dried samples.

3.1.2.3. X-Ray Diffraction (XRD) Analysis

The crystalline structures of CRCA and RCA were assessed by XRD analysis by focusing on the changes in crystalline peaks. Before analysis, materials were dried at 40 $^{\circ}$C in a laboratory-type oven. The dried samples were then scanned between angles of 10-55° with an increase of 0.02°.

3.1.2.4. Determination of Water Absorption Capacity

One of the most visible differences in the aggregate structure in the carbonation technique is the water absorption capacity. As the cracks on the RCA surface are filled and closed, it is expected that the water ingress into the particle will decrease and the amount of water required for the surface to become water saturated (SSD) will decrease. In this regard, water absorption capacity measurements, one of the important parameters, were carried out in different particle size ranges by following the steps specified in ASTM C127 and C128 standards. Among the aggregate samples prepared in different sizes, those over 4.75 mm were measured according to the ASTM C127 standard, and those below were measured according to the ASTM C128 standard. The basic principle in both of these standards is based on finding the difference between the weights of the samples in their SSD state and their oven-dry weights. At the same time, specific gravity values can be calculated after determining the weight of the samples in water.

According to ASTM C127, coarse aggregate samples are kept in water for 24 hours after being dried and cooled in the oven. The sample removed from the water is made SSD by means of an absorbent towel and weighed. The sample is then placed in the water tank via a wire basket (Figure 3.3) and its weight in the water is determined. After the measurements are completed, the sample is dried in the oven at 110^{o}C until it reaches a constant weight and the final weight is recorded.

Figure 3.3 Water tank, wire basket and weight measuring device used to measure the weight of the aggregate in water

According to ASTM C128, fine aggregate samples are kept in water for 24 hours after being dried and cooled in the oven. At the end of this period, the excess water in which the sample was kept is carefully drained. In a suitable tray, the sample is dried using a hot air source (e.g. hair dryer) to bring it to SSD state. The sample, which is checked to become SSD with the cone test specified in the standard, is taken into the pycnometer (Figure 3.4). Water is filled to a certain level. Up to this level, pycnometer filled with only water and pycnometer filled with water + aggregate$_{(SSD)}$ are weighed. After the measurements are completed, the sample is dried in the oven at 110^{o}C until it reaches a constant weight and the final weight is recorded. Water absorption capacities and specific gravity values of the samples are calculated with the suggested formulas.

Figure 3.4 Cone/hammer used to control the SSD state of fine aggregate and pycnometer used for weight measurements

3.2. Studies on Mortar Phase

The efficiency of the accelerated carbonation technique on low quality RCA was investigated in the study covering above section (Dündar et al., 2023). In this study, in which many parameters such as temperature, pressure, CO_2 concentration, curing time, particle size were examined. The optimum conditions determined in the study were preferred for the carbonation process. In this regard, interior conditions were set as relative humidity 70%, temperature 50^{o}C, pressure 1 bar, and fine RCA for the mortar samples was subjected to accelerated carbonation for 48 hours under these conditions.

3.2.1. Materials and Sample Preparation

In this study, it was aimed to work with aggregates in the range of 0.85-4.75 mm in order to conduct controlled experiments on mortar samples and considering that fine aggregates exhibit higher CO_2 capture performance. Xuan et al. (2016) stated that aggregates smaller than 5 mm capture 2.15% CO_2, while aggregates 5-10 mm in size capture 0.81% CO_2. In addition, other studies have found that aggregates in the range of 0.16-2.5 mm exhibit much higher capture performance than larger sizes (Zhang et al., 2015; Dündar et al., 2023). On the other hand, it has been stated that the use of fine RCA smaller than 0.63 mm negatively affects the mechanical properties of cementitious mortars due to high water absorption rates (Zhao et al., 2015). In fact, although powder RCA has relatively positive effects in older ages, they create uncertainties in their use in mortars due to their complex content. It has been reported that when powder RCA is replaced by 30% in natural sand, there is a 20% loss in compressive strength (Duan et al., 2020). Therefore, in the study, the minimum aggregate size was determined as 0.85 mm. Concrete rubble (Figure 3.6) crushed with a crusher was sieved according to ASTM C136, and fine RCA was obtained in accordance with the particle size distribution (PSD) in Figure 3.5.

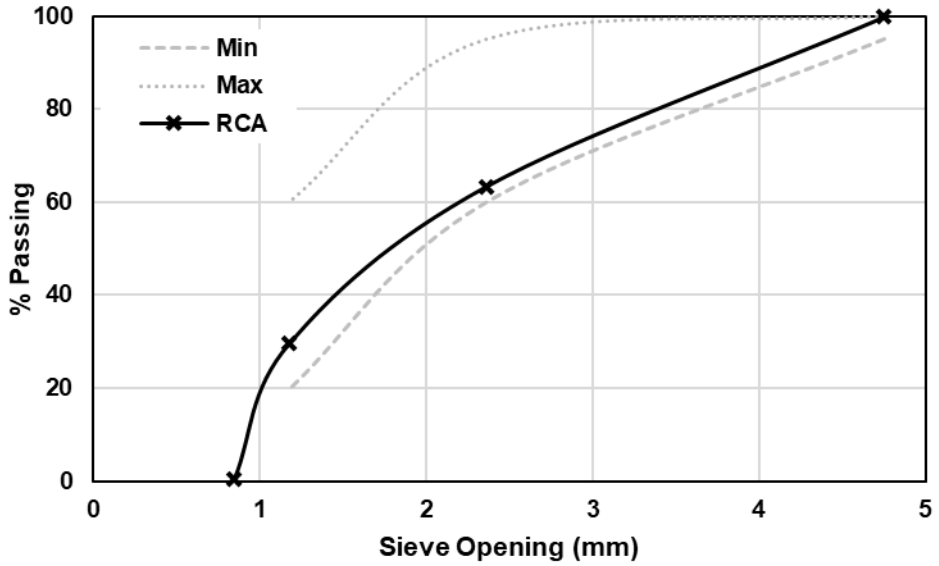


Figure 3.5 Particle size distribution of RCA prepared for mortar samples

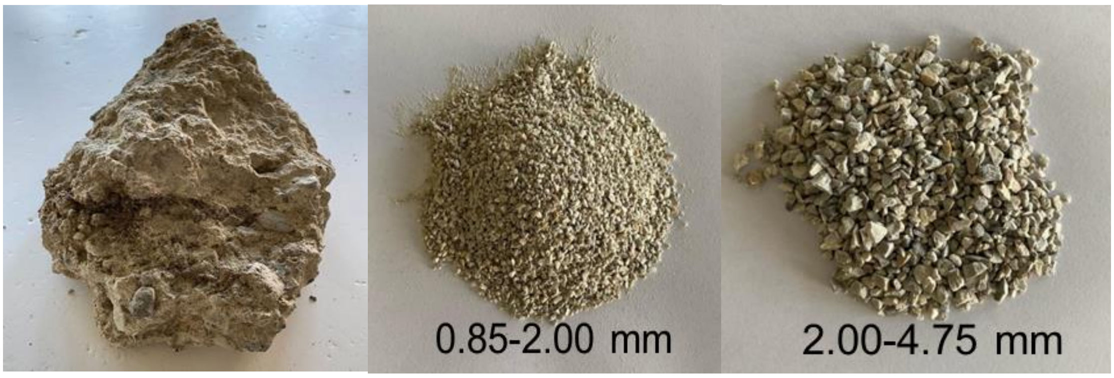


Figure 3.6 Views of concrete rubbles and RCA in two different particle sizes

In some studies, it has been mentioned that studies examining the negative effects caused by fine RCA are quite limited and comprehensive studies in terms of durability cannot be found (Kirthika and Singh, 2020). It has been stated that fine RCA affects the impermeability of concrete more than coarse RCA, and that this effect is associated with the higher capillary voids and cracked/permeable structure in fine RCA (Bravo et al., 2015; Martinez-Lage et al., 2012). It has been also stated that fine RCA affects chloride permeability more than coarse RCA because it contains more mortar residue or clay content (Bravo et al., 2015; Evangelista and Brito, 2010). As an example, in the study of Duan (2014), it was reported that higher ion permeability resistance was achieved with coarse RCA with removed mortar residue. As a result, the scope of the study is limited to 0.85-4.75 mm size RCA due to the fact that fine RCA affects the mechanical and durability properties of concrete more than coarse RCA, that there are few comprehensive studies examining the durability properties of fine CRCA and that fine RCA captures more CO_2 than coarse RCA.

Chemical analysis was also carried out with samples taken from NA, RCA and CRCA aggregates with PSD in the range mentioned above. The first thing that draws attention in the results seen in Table 3.2 is the high LOI rate. This shows that the natural aggregate used is calcareous (limestone) aggregate and the parent aggregate contained in RCA is also calcareous aggregate (Alexandridou et al., 2018). When the CaO ratios are examined, it is seen that NA contains slightly more CaO. On the other hand, it is seen that RCA and CRCA contain more SiO_2, Al_2O_3 and Fe_2O_3 than NA. This difference is due to the adhered mortar on the surface of RCA and CRCA.

Table 3.2 Chemical characterization of aggregates used in mortar samples

%	*NA*	*CRCA*	*RCA*
SiO_2	1,62	3,18	2,66
Al_2O_3	0,17	0,92	0,83
Fe_2O_3	0,01	0,49	0,50
CaO	53,96	50,83	51,19
MgO	0,71	1,25	1,75
SO_3	0,04	0,39	0,38
Na_2O	0,23	0,23	0,22
K_2O	0,32	0,33	0,33
TiO_2	-	0,06	0,05
Cr_2O_3	-	-	-
Mn_2O_3	-	0,01	0,01
P_2O_5	0,05	0,14	0,15
ZnO	-	0,01	0,01
SrO	0,02	0,03	0,03
LOI	**42,87**	**42,14**	**41,91**

Water absorption capacity test was carried out in accordance with ASTM C128 to examine the modification on the surface of RCA after the carbonation process and to see the difference between it and NA. NA used in the test was prepared similar to the PSD of RCA and CRCA. As a result of the test, while the water absorption capacity of NA is 1.03%, that of RCA is determined as 6.20%. Due to the carbonation effect, the water absorption capacity of CRCA decreased to 3.43% (Figure 3.7).

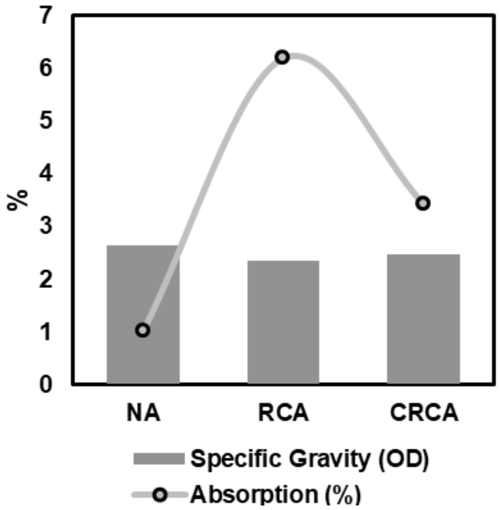


Figure 3.7 Specific gravity and water absorption capacities of RCA, CRCA and NA

Within the scope of the study, in the mortar mixtures prepared with NA, RCA and CRCA, saturated surface dry (SSD) states were taken into account. For this reason, pre-wetting was applied to RCA and CRCA in proportion to their differences in the water absorption capacity of NA (Table 3.3). It is possible to come across studies in which the aggregates are pre-wetted so that the surface becomes saturated before preparing mixtures where CRCA and RCA are replaced at a rate of 100% (Liang et al., 2019; Lu et al., 2019; Waseem, 2022). In the study of Pan et al. (2017), the amount of water was decided by targeting equal slump of 130±5 mm. When the studies in the literature were examined, it was seen that the pre-wetting process was applied at different time intervals. In one of them, RCA with a size of 0.15-4.75 mm was pre-wetted with water according to its water absorption capacity for 15 minutes before adding it to the mixture (Ma et al., 2022). In another study, saturating the surface of 0-4.75 mm RCA too early caused segregation, so water was added to the mixture a certain time ago. 15 minutes was found to be short for fine RCA to absorb water, while 1.5 hours was found to be sufficient (Behera et al., 2019). Within the scope of this study, it was decided to apply the pre-wetting process with the help of spray 1 hour before mixing.

Table 3.3 Mixture proportions of mortar samples prepared

	Agg. (g)	***Presoak (g)***	***Water (g)***	***Cement (g)***	***Apperent w/c***
NA	3.000	0	500	1.000	0,50
RCA	3.000	155	500	1.000	0,50
CRCA	3.000	72	500	1.000	0,50

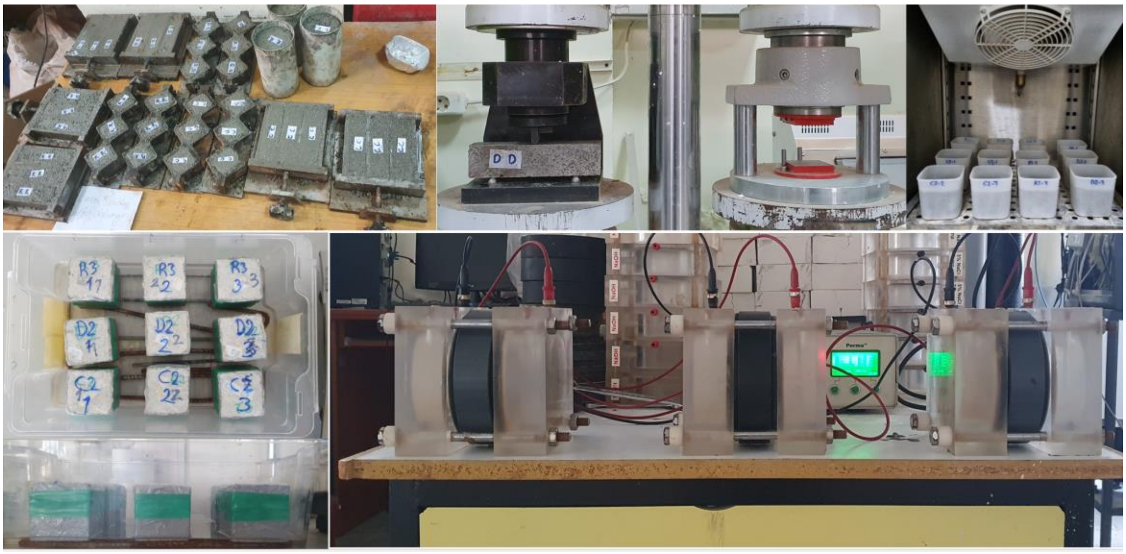


Figure 3.8 Views of samples prepared for various tests

Mortar mixtures were prepared with CEM I 42.5R Portland cement (PC), tap water and 3 different aggregates without the use of chemical additives (EN 196-1). The prepared 3 groups of mortar samples were cast into molds in sizes appropriate to the standards of the tests to be applied. The samples were removed from the molds after 24 hours and subjected to water cure until the test day.

3.2.2. Test Methods on Mortar Samples

3.2.2.1. Flow Properties

The flow table test was primarily carried out in accordance with the ASTM C1437 standard to assess the flowability of the produced mortar mixtures. The process started with filling half of a cone with fresh mortar and rodding it 20 times. The cone was then fully filled with mortar and rodded 20 times before excess mortar was removed, and the cone was lifted vertically. Then, the flow table was dropped 25 times, and the diameter of the spread was recorded in two different directions. The average of these values was taken as an indicator of the flowability.

3.2.2.2. Flexural and Compressive Strength

The flexural strength of prismatic samples in size of 40x40x160 mm was measured following the guidelines outlined in the ASTM C348 standard. To assess compressive strength, half of the prismatic samples that were broken during the flexural strength tests underwent testing in accordance with the ASTM C349 standard. Both compressive and flexural strength tests were conducted on a minimum of three replicates for each mixture at curing ages of 7 and 28 days. The mean of the test results obtained from these three replicates was reported as the strength values.

3.2.2.3. Water Absorption and Apparent Density

Absorbtion test, as per ASTM C642, was also used for assessing voids in hardened mortar samples. For this test, 50x50x50 mm cubic samples were subjected to a 1-day drying process in an oven at 100 ± 5°C. Upon removal from the oven, the samples were allowed to cool in dry air before being weighed (oven-dry mass). Subsequently, the samples were immersed in tap water, and their mass (saturated mass of the samples after immersion) was monitored at 24-hour intervals until the mass increase became negligible (less than 0.5% of the larger mass). The absorbtion of the samples after immersion was then calculated by dividing saturated mass of the sample after immersion by the oven-dry mass of the sample.

The apparent density of the cubic hardened mortar samples, measuring 50x50x50 mm, was also determined following ASTM C642 standard. In this experiment, the immersed apparent mass was determined by weigthing the sample suspended in water. The apparent density of the samples was calculated using the equation given below.

$$Apparent\ density = \ [m_o/(m_o - m_w)].\rho$$

where:

m_o = mass of oven-dried sample in air, g
m_w = apparent mass of sample in water after immersion and boiling, g
ρ = density of water, = 1 g/cm^3.

3.2.2.4. Rate of Absorption (Sorptivity)

Sorptivity (capillary water absorption) tests were conducted on 28-day-old cubic samples with a side length of 50 mm (three samples for each series of mortar) in accordance with the ASTM C1585 standard. During the test, only one surface of the sample was allowed to come into contact with water. To ensure one-directional flow through the sample, the sides were sealed with a silicone coating. Then, the samples were placed in water on steel rods and the depth of water was maintained at 2-3 mm. The increase in mass of the samples was monitored at specified time intervals while allowing it to absorb water through capillary suction. The rate of absorption-I (mm) was determined by dividing the change in sample mass (g) by the exposed area of the test sample (mm^2) and the density of water (g/mm^3). This data was then plotted against the square root of time ($s^{1/2}$) and both initial and secondary absorption values of the samples were determined from the resulted graphes.

3.2.2.5. Freezing and Thawing Test

The mortar samples were subjected to the rapid freeze and thawing test in accordance with the prescribed procedures outlined in the ASTM C666 standard. In this test, 28-day-old cubic samples with dimensions of 150 × 150 × 150 mm were exposed to 300 cycles of freezing and thawing. In the experiment performed up to 300 cycles in the cabin, each cycle was set to +4^{o}C for 4 hours and -18^{o}C for 4 hours. At the end of every 36 cycles, the weight loss in the samples was noted. At the conclusion of every 300 cycles, the strength loss of mortar samples were also computed.

3.2.2.6. Rapid Chloride Permeability Test

The 100 mm diameter samples prepared in cylindrical shape were cut into 3 pieces at 50 mm height and their chloride ion penetration resistance properties were determined. In the test carried out in 6 hours with the Perma brand device produced in accordance with the ASTM C1202 standard, the current (mA) passing through the samples was recorded at 30-minute intervals by applying a voltage of 60±0.1 V and the total charge passed (C) was calculated for each sample.

4. RESULTS AND DISCUSSION

4.1. Investigating the Operational/Materials-Oriented Parameters on the Carbonation Performance of RCA

First stage of the thesis focuses on the examination of the parameters affecting the carbonation performance of low-quality RCA obtained from end-of-life buildings and having low mechanical and chemical properties. The accelerated carbonation process was implemented by using a lab-scale carbonation reactor, providing a dynamic carbonation process. Thermogravimetry was the test method used to quantify the CO_2 uptake rate of low-quality RCAs. In addition, the water absorption capacity of RCAs was determined to gain insight into precipitated calcium carbonate. To evaluate the microstructure of carbonation products, scanning electron microscope (SEM) and X-ray diffraction (XRD) methods were used.

4.1.1. Parameters Affecting the Carbonation Performance

In the scope of this stage, as the main operational parameters, RH (50-70-90%), medium pressure (1-2-3 bar), and temperature (50-90^{o}C) were determined. Carbonation processes were implemented on the low-quality RCAs with 5 different particle size ranges, including <0.85, 0.85-2.00, 2.00-4.75, 4.75-9.50, and 9.50-14.00 mm. Effects of carbonation durations (2-4-6-12-24-48-72-96-120 hour) on the carbonation performance of RCAs were also investigated.

4.1.1.1. Effect of Relative Humidity

The carbonation efficiency of RCA significantly depends on the RH of the carbonation medium as water is a vital player in the chemical reactions of carbonation and diffusion of CO_2 into the inner parts of RCA (Zhan et al., 2014; López-Arce et al., 2011; Xu et al., 2022). In the carbonation chamber, the solubility of reactants (e.g., CO_2) decreases at the lower RH because of the presence of limited solvents in the medium (Maia Pederneiras et al., 2022; Galan et al., 2013; Doruet et al., 2019). However, the diffusion of CO_2 into RCA is restricted at the extremely high RH due to the formation of a liquid layer on the pore surface (pore blockage by water presence), preventing the diffusion of CO_2 (Fernández Bertos et al., 2004; Elsalamawy et al., 2019; Leemann and Moro, 2017). Therefore, lower, or higher RH has adverse effects on carbonation efficiency. Considering these, optimal RH determination is crucial for the efficient carbonization of RCA. Although in the literature, most of the studies suggest the optimal RH range of carbonation medium as between 40-70% (Gholizadeh-Vayghan et al., 2020; Liang et al., 2020; Li and Wu, 2022), depending on the other parameters such as sources and size of RCA, temperature, and pressure of reaction medium, etc., optimal RH range may differ under different carbonation parameters (Maia Pederneiras et al., 2022). To clarify the effects of the RH on the carbonation efficiency of low-quality RCA carbonated under 15% CO_2 concentration, 90 ^oC temperature, and 1 bar pressure conditions, the obtained CO_2 uptake rates are given in Figure 4.1. Results showed that the CO_2 uptake level increased with the increase in the RH up to 70%. This could be attributed to the increment of solvent materials in the reaction medium, enabling more carbonation reaction and CC precipitation (Zhan et al., 2014; Galan et al., 2013). The decrement in CO_2 uptake was observed with the increase in RH from 70% to 90%. The increment in humidity may cause lower permeation of CO_2 into the inner zones through cracks, leading to less precipitation of CC (Fernández Bertos et al., 2004; Elsalamawy et al., 2019; Leemann and Moro, 2017). Considering the uncarbonated RCA, the CO_2 uptakes were recorded as 3.72%, 7.40%, and 6.36% for the CRCA carbonated under the RH condition of 50%, 70%, and 90%, respectively. The water absorption capacity results of the uncarbonated RCA and CRCA were also presented in Figure 4.1. Similar trend was observed compared to TGA results for the varying RH level. Results showed that CC precipitation fills the cracks/pores of RCA, leading decrement in the water absorption capacity of CRCA (Sonmez et al., 2021). The decrement in the water absorption of CRCA considering the uncarbonated RCA was recorded as 17%, 32.7%, and 28.2% for the 50, 70, and 90% RH, respectively. According to the CO_2 uptake and water absorption results, the optimum RH level was obtained as the RH of 70% for the carbonation condition designed for this case, which is consistent with the literature studies (Elsalamawy et al., 2019; Chen et al., 2019). However, although it was expected to obtain more CO_2 uptake and less water absorption capacity from the CRCA under 50% RH compared to that under 90% RH, the opposite is the case for the low-quality RCA carbonated under the predefined condition of 15% CO_2 concentration, 90 ^oC temperature, and 1 bar pressure. The possible reason for obtaining such a result could be attributed to that high water content may benefit carbonation at elevated temperatures compared to the lower water content presence in the medium. Because in high temperatures, diffusion of CO_2 and the leaching of calcium ions accelerate, but the solubility of CO_2 and calcium ions in liquid decreases (Li and Wu, 2022). To overcome the decreased solubility because of elevated temperature, a high-water content in the medium can benefit the carbonation reactions. Therefore, RH level of 90% yielded better carbonation efficiency compared to the 50% RH, yet 70% RH provided the optimal water content that balances the solubility and diffusion process during the carbonation reactions.

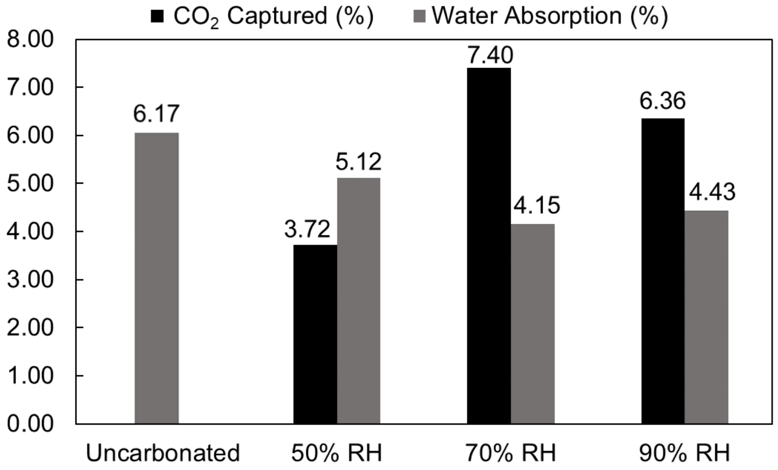


Figure 4.1 Comparison of relative humidity conditions in terms of captured CO_2 and water absorption capacities

4.1.1.2. Effect of Pressure

Gas pressure has a significant effect on the carbonation performance of RCA (Xuan et al., 2016; Kashef-Haghighi et al., 2015; Fang et al., 2017). In general, when the pressure is increased, more CO_2 is dissolved in the pore solution (Carroll et al., 1991), resulting in faster carbonation reactions (Moorehead, 1986; Hyvert et al., 2010). However, there is an optimum pressure for exposing the RCA to CO_2, and excessive pressure can degrade the RCA's characteristics. This could be due to excessive pressure causing new cracks to develop in the attached old RCA mortar, resulting in significant porosity and water absorption of the RCA (Liang et al., 2020; Xuan et al., 2016; Zhan et al., 2017). The formation of new cracks and new dust content with some pressure increase becomes even more effective for such low-quality RCA. Previous research has also shown that excessive gas pressure is unfavorable for the carbonation reaction of RCA because it generates significant heat, increasing the likelihood of microcracking (Xuan et al., 2016; Liang et al., 2019; Fang et al., 2017; Zhan et al., 2016; Wu et al., 2022; Bukowski and Berger, 1979). In this study, 1, 2, and 3 bar (0.10-0.20-0.30 MPa) CO_2 and dry-air mixed gas pressure were examined throughout 48 hours-duration. Figure 4.2 depicts how the pressure affects RCA carbonation performance and water absorption capacity.

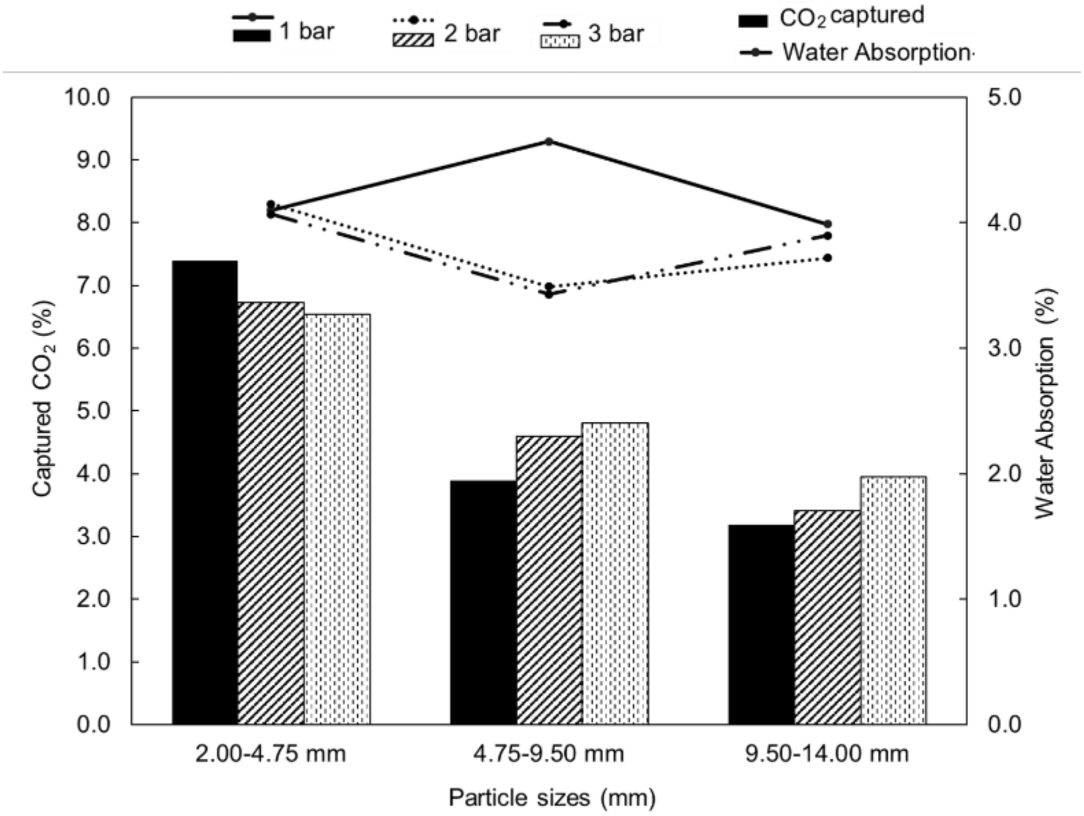


Figure 4.2 CO_2 capture rates (represented in columns) and water absorption values (presented in lines) of three different particles size under 1, 2 and 3 bar pressure for 48 hours

As illustrated in the graph, the connection between CO_2 uptake and water absorption is not always linear. This relationship may be affected by parameters such as particle size and source of RCA (Wang et al., 2019; Drouet et al., 2019; Liang et al., 2022). Also, Figure 4.2 shows that after 48 hours of the carbonation process, the maximum CO_2 uptake performance was obtained from RCA with the particle size range of 2.00-4.75 mm carbonated under 1 bar of pressure. On the other hand, the lowest CO_2 uptake was observed with the particle sizes range of 4.75-9.50 mm and 9.50-14.00 mm carbonated under 1 bar pressure. The CO_2 uptake rate reduced for the RCA having a particle size range of 2.00-4.75 mm when the pressure was increased from 1 bar to 2 bar. This decrease with pressure increase can be attributed to the fact that under high pressure, some vulnerable RCAs may be broken, and undesirable fine-grained materials may form and clog the fine pores, preventing CO_2 diffusion into deeper parts of RCA. It can be stated that the pores in this size are much smaller than the pore structure in coarser RCA particles, which causes this effect to be negative in the size of 2.00-4.75 mm. On the other hand, it can be concluded that undesirable damage and formation of fine-grained materials due to high pressure create additional surface area for CO_2 capture. However, the results confirm the conclusion that this surface already contains residual mortar whose carbonation is almost complete. Accordingly, with low pressure, remarkable CO_2 uptake performance can be expected in small-sized RCA (Tam et al., 2016; Zhan et al., 2016).

On the other hand, as the aggregate size increased (>4.75 mm), a slight increase in pressure resulted in both an increase in CO_2 capture rate and a decrease in water absorption capacity. For the RCA with the particle size ranges of 4.75-9.50 mm and 9.50-14.00 mm, when the pressure was changed from 1 bar to 2 bar, approximately an 18% and 7% increase in CO_2 uptake rate and 26% and 3% decrease in water absorption capacity was observed, respectively.

Furthermore, at all three pressures, a high carbonation rate occurred in the first 6 hours of carbonation, and the reduction in water absorption capacity of the samples exposed to 2 and 3 bar pressure was rapid in the first hours and slow in the subsequent hours (Figure 4.3). Carbonation/the amount of captured CO_2 significantly increased over time, but a slower rate of increment was observed in the following periods. Additionally, the decrease in water absorption capacity in the following hours may be related to the rapid filling of surface voids and the slower absorption of CO_2 in the first few hours. It has been observed that RCAs with higher CO_2 uptake percentages also have higher water absorption values (Figure 4.3). This is because the increased water absorption values of the RCA indicate a greater level of open porosity within the RCA that could allow CO_2 to penetrate the RCA. Even after most of the open porosity in the RCA is filled with free water, CO_2 still cannot freely enter the RCA (Zhan et al., 2014). In this case, CO_2 diffusion in the saturated RCA restricts the carbonation process of the interior of the RCA; this is also the non-linear relationship between the percentage of RCA carbonation and water absorption. Besides, the decrease in CO_2 uptake rate exhibited in Figure 4.2 and 4.3 with increasing pressure can be attributed to the breaking of some brittle soft RCAs, under high pressure, as well as the development of undesirable fine-grained compounds and clogging of the pores (Tam et al., 2016). While positive gains in water absorption and CO_2 uptake were noted with increasing pressure in the 4.75-9.50 mm and 9.50-14.00 mm grain sizes, it was observed that pressure had no noticeable effect on the carbonation process of fine RCA (<4.75 mm).

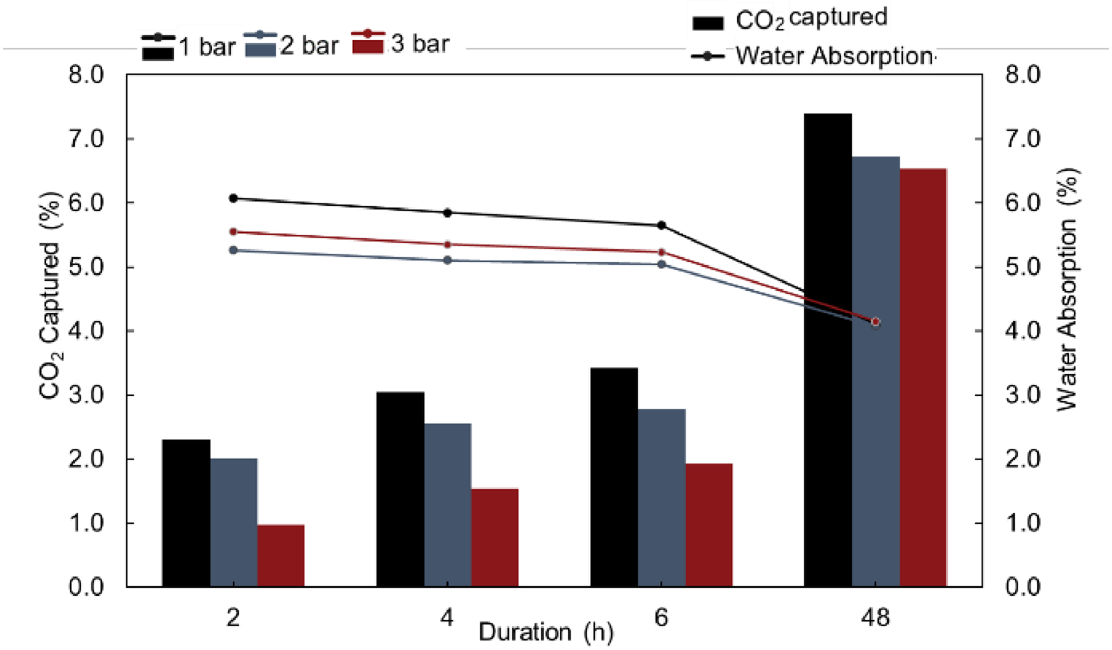


Figure 4.3. Results of 2.00-4.75 mm size RCA for water absorption (lines) and CO_2 captured rate (columns)

4.1.1.3. Effect of Temperature and Duration

The temperature is also one of the most critical factors affecting the carbonation efficiency of RCA (Li and Wu, 2022; Wang et al., 2019). Therefore, in this study, the carbonation potential of RCA was investigated considering the varied temperatures. In this regard, two different carbonation process temperatures, including 50 °C and 90 °C were performed. These carbonation processes were implemented with adjusted RCA particle size fraction (2.00-4.75 mm), CO_2 gas concentration (15%), pressure (1 bar), and relative humidity (70%) through different durations of carbonation treatment (2, 4, 6, 12, 24, 48, 72, 96 or 120 hours). The CO_2 uptake rates and water absorption capacities of both RCA and CRCA carbonated under different carbonation processes varied according to the temperature and carbonation treatment duration are shown in Figure 4.4. As shown in Figure 4.4, an increase in the carbonation process temperature from 50 ^oC to 90 ^oC favored the overall carbonation of RCA although the differences among them were not significant. Favoring effect of temperature was more evident for the specimens treated for up to 48 hours. This favorable effect of the increased temperature on the carbonation rate can be attributed to the acceleration of diffusion of CO_2 and the migration of the materials, leaching of calcium ions, and thereby acceleration of carbonation reactions (Fernández Bertos et al., 2004; Wang et al., 2019; Li and Wu, 2022; Kurda et al., 2019; Greve-Dierfeld et al., 2020; Lekakh et al., 2008; Lu et al., 2022). When the CO_2 uptake rates of RCA carbonated during different carbonation treatment durations are evaluated from Figure 4.4 it can be clearly seen that a longer carbonation process induced a higher CC precipitation rate, which means a higher CO_2 uptake level. This finding was also reported by several other researchers in the available literature (Xuan et al., 2016; Kurda et al., 2019; Limbachiya et al., 2012; Silva et al., 2015; Pu et al., 2021; Gholizadeh-Vayghan and Snellings, 2022) and can be attributed to the increase in the amount of carbonation reaction between CO_2 and relevant reagents available on the RCA surface owing to the increasing the specimens' exposure time to CO_2. The increase in the carbonation rate associated with the increased carbonation duration was more pronounced for up to the carbonation duration of 48 hours. Namely, the carbonation reactions continued progressively at the RCA surfaces attached to the mortar residues in the first 48 hours. After 48-hour of the carbonation process, the CO_2 uptake became relatively stable, irrespective of the carbonation process temperature. This result was most probably due to the formation of a passivating layer of carbonation products on the cement-based mortar residues affecting diffusion phenomena of CO_2 as the carbonation progressed over time (Zhan et al., 2016; Kurda et al., 2019; Shi et al., 2012; Kou et al., 2013, Silva and Andrade, 2017; Ahmed et al., 2018). Products occurring from the carbonation process resulted in progressive densification of the micro-structure of the cement-based zones on the RCA surfaces and coating of them. This caused delaying the leaching of calcium ions and decrements in the rate of CO_2 diffusion and dissolution and undermining the specimens' exposure to CO_2. Thus, the carbonation reactions slowed down as a function of time yielding stabilization of CO_2 uptake over time. Based on this result, it can be concluded that it is necessary to determine the optimum CO_2 exposure time providing maximum penetration, interaction, and CO_2 absorption by cement paste particles attached on the RCA.

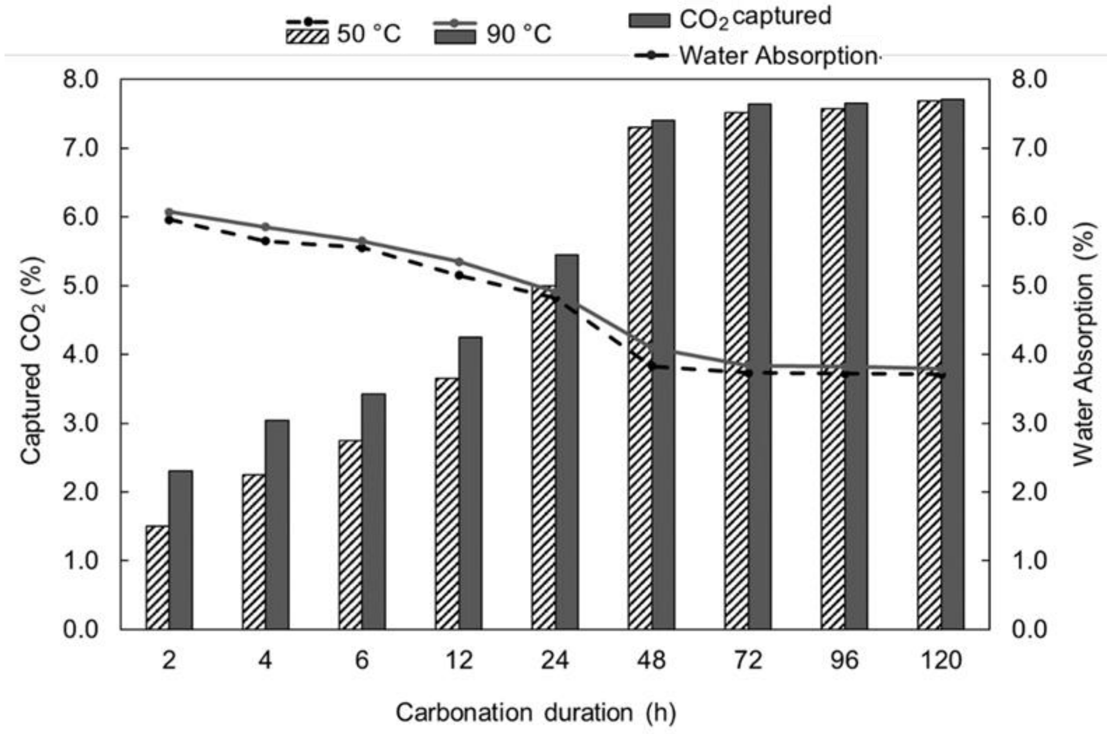


Figure 4.4 The CO_2 captured rate (columns) and water absorption capacities (lines) of RCAs treated under different carbonation processes varied according to the temperature and carbonation treatment duration

When Figure 4.4 are examined, it can be clearly seen that the overall pattern of the test results showing the water absorption capacities of carbonated RCAs was almost parallel to that of CO_2 uptake trend of them. As with the CO_2 uptake level, the water absorption capacities of RCAs were also fixed after a certain period of carbonation duration. Although this situation started from the 48th hour for the CO_2 uptake results, the stabilization in the water absorption capacities of RCAs started to occur from the 72^{nd} hour, regardless of the carbonation process temperature. On the other hand, although higher CC precipitation rates were obtained from the RCAs carbonated under relatively higher carbonation treatment temperature level (90 ^{o}C), the water absorption capacities of these specimens were slightly higher compared to those of RCAs carbonated under the temperature of 50°C. These results can be related to the form of the carbonation product occurring from the carbonation process. While more stable and denser polymorphs of the carbonation process is obtained at low temperatures, the formation of meta-stable forms of $CaCO_3$ can be induced under high temperatures (Fernández Bertos et al., 2004; Drouet et al., 2019; Lu et al., 2022; Tai and Chen, 1998; Chu et al., 2013; Liu et al., 2020). Therefore, the higher water absorption capacities of the RCA carbonated under the temperature of 90 °C can be related to their relatively higher content of meta-stable forms of $CaCO_3$.

4.1.1.4. Effect of Sizes of Aggregates

The particle size of RCA is another parameter affecting the RCAs' CO_2 uptake rate during the accelerated carbonation process. Experimental results of this study also presented that the particle size of RCA can influence the rate of calcium carbonate precipitation. According to CO_2 uptake results of RCAs with varying particle size ranges, a similar to normal distribution curve-shaped graph was observed (Figure 4.5), meaning that in the results there is a diffraction point where the trend was reversed. This may be connected to the fact that RCA formed via the crushing process can vary the amount of residual mortar available on the RCA due to different particle sizes. In this study, the RCA were crushed and classified into five size fractions of <0.85 mm, 0.85-2.00 mm, 2.00-4.75 mm, 4.75-9.50 mm, and 9.50-14.00 mm. Among the particle sizes, 2.00-4.75 mm sized RCA stood out by having about 55.4%, 46.3%, 67.9%, and 60.3% higher CO_2 absorption than <0.85 mm, 0.85-2.00 mm, 4.75-9.50 mm, and 9.50-14.00 mm, respectively. The CO_2 uptake rates of RCA exhibited an increase with the increase in the maximum particle size up to 2.00-4.75 mm, yet from 2.00-4.75 to 9.50-14.00 mm sized RCA showed a slight decrease in the carbonation performance (Li et al., 2019). The RCAs with particle size smaller than 2.00-4.75 led to more CO_2 uptake level compared to those with particle size higher than 2.00-4.75 mm (Kikuchi and Kuroda, 2011; Thiery et al., 2013).

As the particle size of RCA decreases, the residual mortar content may increase, meaning the availability of more reactants for carbonation reactions. However, the finer particles may also contain unhydrated cement particles even in low-quality RCA that can produce hydration products because of water interaction (Sereng et al., 2021). This hydration reaction may cause the clumping of RCAs to each other, and so that prevents the diffusion of CO_2 (Fernández Bertos et al., 2004; Leemann and Moro, 2017; Thiery et al., 2013). Besides that, the RCA with the finer size are prone to agglomerates in the presence of humidity, which is also supposed to be the main possible reason for the reduction in CO_2 uptake level from 2.00-4.75mm to <0.85mm. Compared to RCA having a particle size range of <0.85mm, a higher CO_2 uptake level was obtained from the RCA with a particle size range of 0.85-2.00 mm. This could be attributed to the agglomeration degree of finer RCA during the carbonation process, as seen in Figure 4.6. On the other hand, RCAs larger than 4.75 mm showed a sharp decline in CO_2 uptake performance. This can be associated with the RCA's smaller specific surface area and fewer mortar residuals because of larger particles, thereby, less interaction capacity (Kikuchi and Kuroda, 2011). To measure the amount of residual mortar presence on the RCA regarding the particle size, the proposed method, which is standardized guidelines provided in ASTM C88 and ASTM C666, was conducted by following the procedure given in a study (Abbas et al., 2008) by using sodium sulfate solution. As seen from Table 4.1, the amount of residual mortar obtained at the end of the chemical and physical degradation process based on the aforementioned standards and method available on the RCAs with smaller particle size was higher than that with coarser particle sizes, which proves the statement expressed above. The precipitation of calcium carbonates at the surface of coarser RCA may obstruct the CO_2 diffusion into inner zones through cracks after a certain degree of carbonation, which is also another reason for the low carbonation performance of coarse RCA compared the fine RCA which have a significantly higher surface area. Overall, the highest CO_2 uptake performance was recorded for the RCA having a particle size range of 2.00-4.75 mm, meaning that a balance between the given opposing effects on carbonation was obtained for this particle size range.

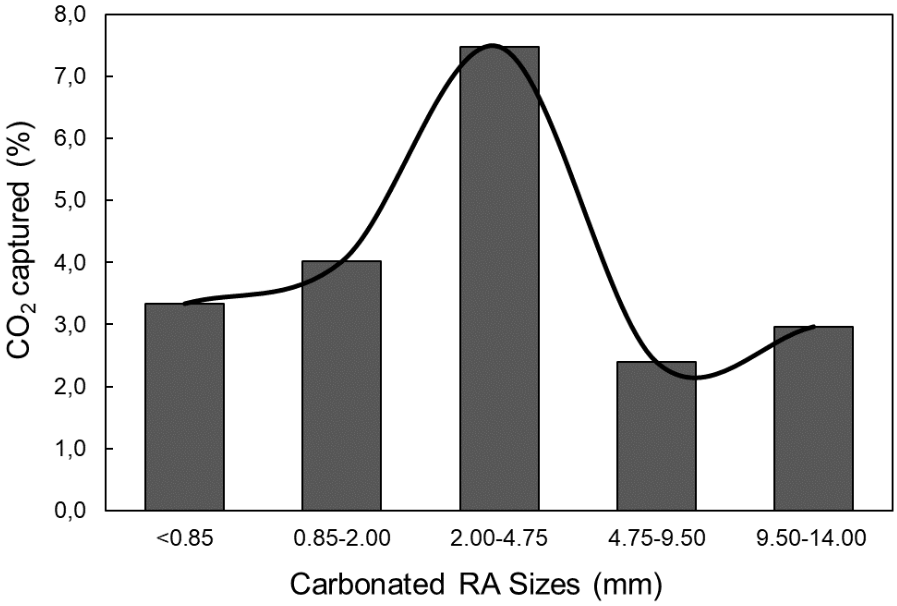


Figure 4.5 CO_2 uptake percentages of carbonated RCAs-under 90 ºC temperature, 70% RH and 1 bar pressure-with varying particle size

Table 4.1 Residual mortar masses obtained from 2.00-4.75 mm RCA and 4.75-9.50 mm sized RCA

Sieve Opening (mm)	*Retained Mass on Each Sieve*	
	4.75-9.50 mm	2.00-4.75 mm
4.75-9.50	159.6	--
2.00-4.75	39.1	123.7
0.85-2.00	28.2	41.0
<0.85 (residual paste)	23.1	85.3
Initial Cumulative Retained Mass (g)	250	250
Residual paste (%)	9.24	34.1

Figure 4.6. Images of RCA; a) uncarbonated <0.85 mm sized, b) carbonated and agglomerated <0.85 mm sized, c) uncarbonated 0.85-2.00 mm sized, d) carbonated and agglomerated 0.85-2.00 mm sized

4.1.2. Microstructural Properties of Carbonated RCA

4.1.2.1. X-ray Diffraction (XRD) Analysis

The XRD patterns of RCA and CRCA at temperatures of both 50 °C and 90 °C are presented in Figure 4.7. During analysis, peaks corresponding to portlandite, calcite, aragonite, vaterite, calcium oxide, and monocalcium aluminate were detected. Intensified peaks of calcium carbonate in the form of calcite were observed for all tested specimens regardless of the accelerated carbonation process. The presence of calcite peaks for the uncarbonated specimen can be attributed to the original aggregate content of obtained RCA. The natural carbonation process also can be another reason for the calcite peaks detected in XRD patterns of uncarbonated RCA, as the used low-quality RCA was obtained from the end-of-life structures served for years under the condition providing

CO_2-exposing. The observation of calcite peaks in all RCAs prevented the evaluation of the carbonation potential of the RCA in terms of calcite formation. The peaks representing crystal structures of portlandite, calcium oxide, and monocalcium aluminate recorded in the XRD pattern of uncarbonated RCA were not observed after the accelerated carbonation process regardless of temperature conditions because of the carbonation reactions (Sonmez et al., 2021; Kaliyavaradhan and Ling, 2017; Šavija and Luković, 2016; Fernández-Carrasco et al., 2008; Sugama et al., 2002). While crystalline peaks belonging to different forms of calcium carbonate of aragonite and vaterite were detected for the CRCA carbonated under the temperature of 90 ^oC, only aragonite peaks were recorded for the XRD pattern of CRCA carbonated under the temperature of 50 ^oC. In the previous study of the authors, vaterite peaks were also not recorded for the CRCA carbonated under a different carbonation condition (in terms of CO_2 concentration, relative humidity, duration, etc.) with a similar carbonation temperature of 50 ^oC (Sonmez et al., 2021). These findings are supportive of the formation of varied forms of calcium carbonate regarding the temperature, which is also stated under the section regarding the effects of temperature (Fernández Bertos et al., 2004; Drouet et al., 2019; Lu et al., 2022; Chu et al., 2013; Liu et al., 2020). Considering these results, it can be stated that high temperatures induce the formation of vaterite as a metastable form of calcium carbonates.

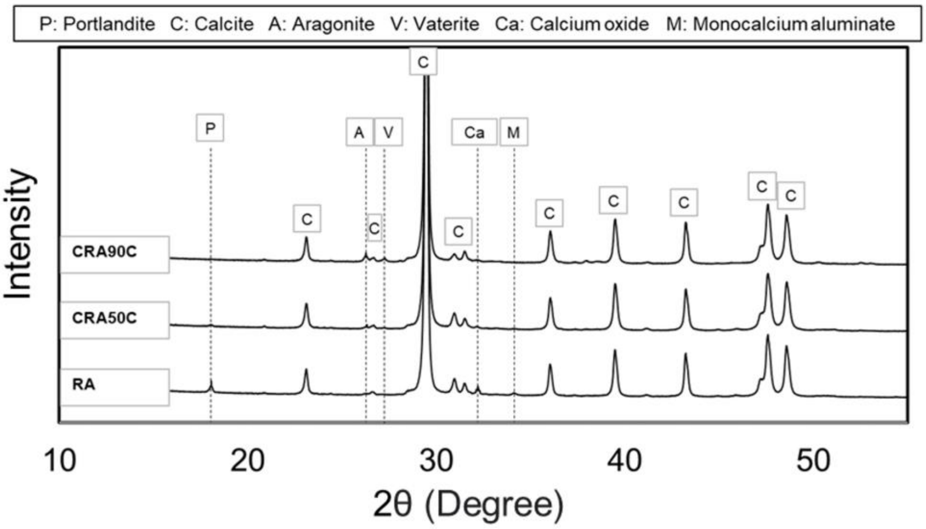


Figure 4.7 XRD patterns of RCA and CRCA under 1 bar, 70% RH, 50 and 90 °C

4.1.2.2. Scanning Electron Microscopy (SEM) Analysis

SEM micrographs of RCA and CRCA are shown in Figure 4.8 to verify the forming of carbonation products on the surface of both treated and untreated RCA. For microstructural observation, due to its efficient CO_2 uptake results, 2.00-4.75 mm size, and RCA carbonated under 1 bar, 50 ºC, 70% RH condition was selected. According to Figure 4.8, on the surface of RCA, products of chemical carbonation reactions could be clearly observable.

Calcium carbonate was the primary carbonation product. The most stable phase of calcium carbonate is called calcite, which is often granular in texture and rhombohedral or cubic in form (Park et al., 2020). According to findings from a study (Fernández Bertos et al., 2004), when RCA surfaces were treated with chemical carbonation, the surface of CRCA became less porous. It was covered with visually-inspected granular-textured and cubic-shaped calcium carbonate crystals. Due to the existence of old mortar remains, the surface of the RCA was rough and irregular, as can be seen in Figure 4.8a (Behera et al., 2014). However, the RA's EDX study findings also revealed that calcium carbonate was being produced on the surface, which may be related to either the chemical components of the RCA or calcite production as a result of CO_2 exposure over time (Wang et al., 2017). On the other hand, the CRCA surface's EDX results revealed higher calcite precipitation. The TG/DTG study reported in the previous section and the XRD analysis described in the previous section both supported the finding that this result was connected to the production of calcite on the CRCA surface (Figure 4.8b).

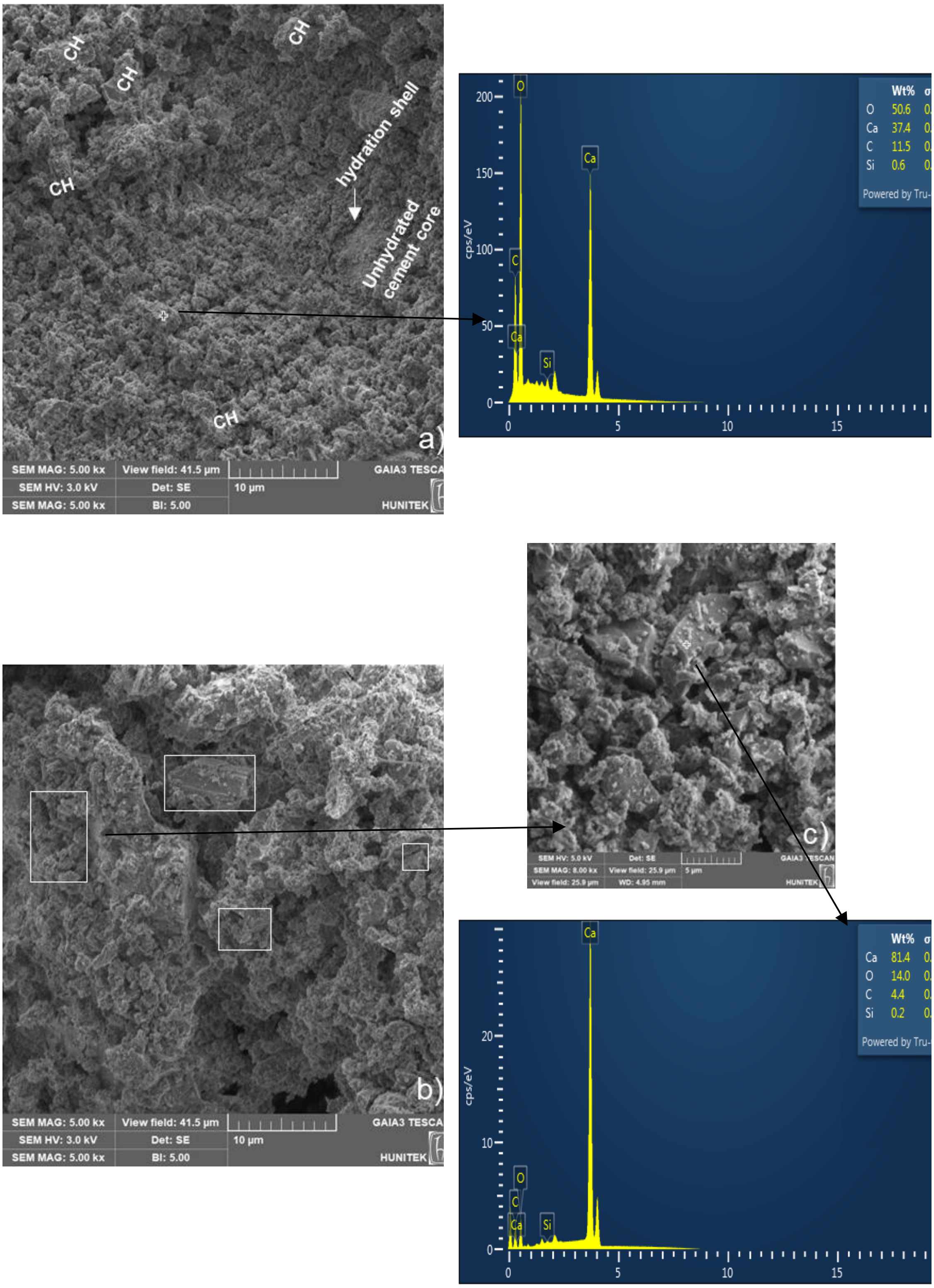


Figure 4.8 SEM micrographs of (a) RCA, (b) CRCA (squared area demonstrates extensive calcite precipitations)

4.2. Investigating the Effects of Carbonated RCA on Engineering Performance of Cement-based Systems

Second stage of the thesis focuses on the investigating the effect of fine fraction (0.85-4.75 mm) carbonated low-quality RCA, obtained from end-of-life buildings, on engineering properties of the prepared mortar. Firstly, for the carbonation, interior conditions of reactor were set as relative humidity 70%, temperature 50°C, pressure 1 bar, and fine RCA for the mortar samples was subjected to accelerated carbonation for 48 hours under these conditions determined in the first stage explained above. After that, series of mechanical and durability tests were conducted on prepared mortar samples to evaluate the performans of CRCA in the cementitious system. To examine the microstructure of mortar samples, scanning electron microscope (SEM) method was used.

4.2.1. Mechanical Properties of Cement-based Systems

4.2.1.1. Flow Properties

Fine NA, fine RCA and fine CRCA used in the mixtures, were used in surface saturated state. The mortar mixtures, which were cast in a common manner for all tests, were examined on the flow table and the flow value was measured before being poured into molds suitable for the targeted tests. Although all aggregates were added to the mixture in a surface saturated state, the flow value of the mixture with NA was measured as 160 mm, the mixture with RCA was 145 mm, and the mixture with CRCA was measured as 155 mm. Although similar results were obtained, differences in these results show that the flow value of the mortar mixture is not only related to the water absorption capacity of the aggregate used. In the literature, it has been stated that the change in slump values with NA, CRCA and RCA is also due to the difference in particle shapes (Kim et al., 2018). The results obtained from the experiments indicate that NA is the aggregate with the least roughness and RCA is the aggregate with the most. It can be assumed that the roughness of RCA decreases with the accelerated carbonation process. As a well-established principle in materials science, it is widely acknowledged that the angularity and rough surface characteristics of aggregates contribute significantly to heightened internal friction and increased resistance to flow. In the case of RCA, the recycling process itself can enhance angularity, while the presence of adhered residual pastes further contributes to a rougher aggregate surface. The close examination of NA and both treated and untreated RCAs in Figure 4.9 reveals the intricate details of their particle structures. As evident from the figure, it is apparent that NA exhibit a smoother surface texture, in contrast to untreated RCA characterized by a rougher surface and higher angularity. Additionally, it was observed that the carbonation process played a role in imparting a smooth surface to CRCA by effectively sealing the holes and cracks through the precipitation of CC.

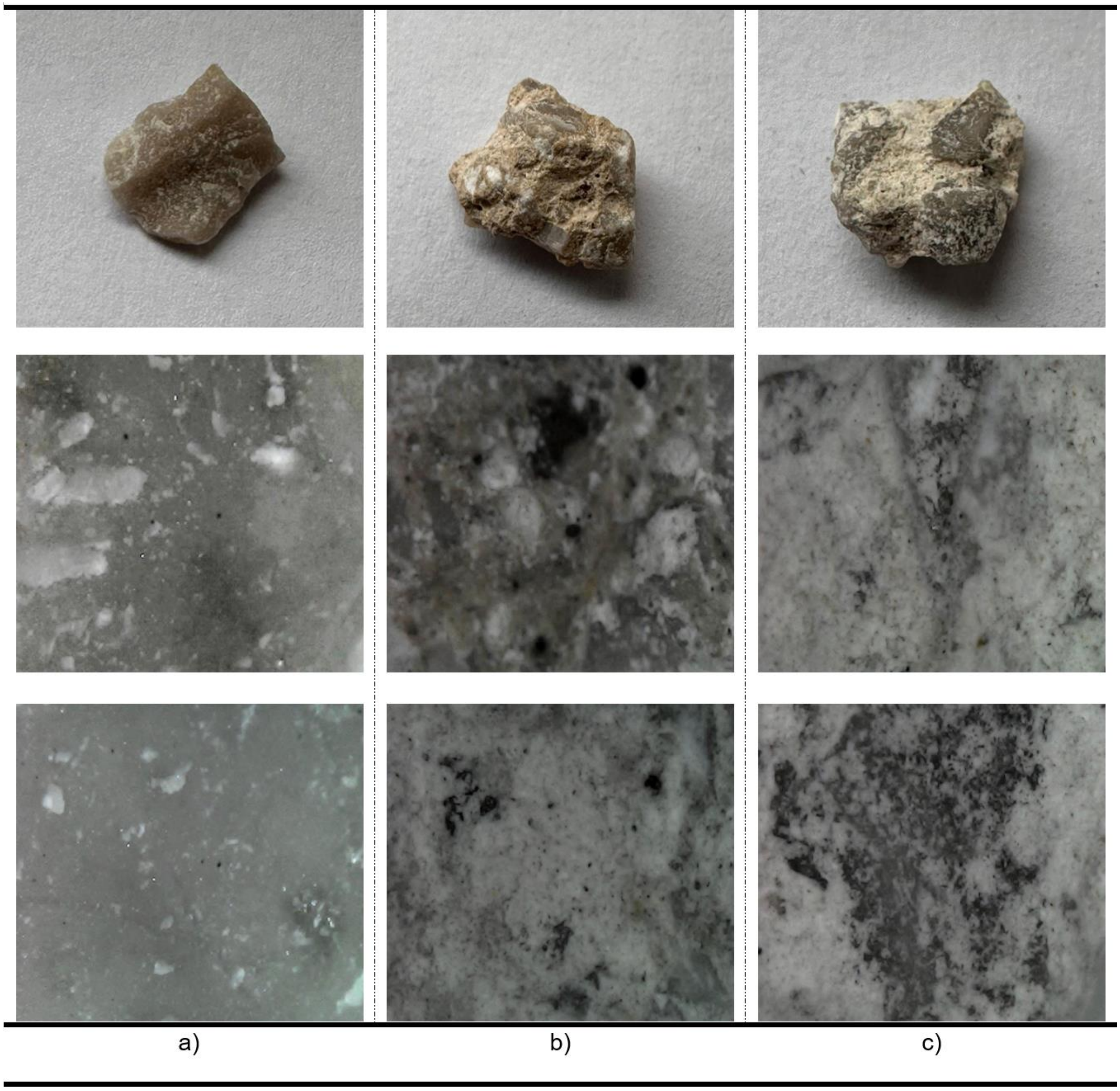


Figure 4.9 Digital image and close view of a) NA, b) RCA, c) CRCA

When the results were examined, it was understood that the negative effect of RCA on workability could be reduced by the accelerated carbonation process. Tam et al. (2020) found that the flow performance of CRCA is much better than RCA. Pu et al. (2021)

attributed the better slump value of CRCA to the reduction of voids with the formation of CC and therefore the reduction of water absorption capacity. In another study, while a flow value of 220 mm was observed in the mortar produced with 100% CRCA, it was reported that a flow of 140 mm was observed in the mortar produced with 30% CRCA + 70% RCA. It has been claimed that this situation is evidence that there is a decrease in porosity in CRCA and that water ingress into the aggregate is prevented (Tam et al., 2016). As seen in this study, although RCA and CRCA were used in same saturation state, it was observed that CRCA had a slightly better performance in terms of workability due to its structure with fewer voids and smoother surface.

4.2.1.2. Flexural and Compressive Strength

Xuan et al. (2016-b) stated that the micro-hardness value of ITZ in the CRCA incorporated system was higher than the ITZ of the RCA incorporated system. This development in ITZ has also increased the flexural and splitting tensile strength. Similar to this result in the literature, in this thesis study, the flexural strength of CRCA mortars was higher than RCA mortars and lower than NCA mortars (Figure 4.10). It can be clearly seen from this figure that due to ongoing hydration reactions, the extended aging/curing resulted in increments in the flexural strength test results, regardless of mixture design parameters. The highest flexural strength test results were recorded from sample containing NA for each curing age with the values reaching 6.9 MPa for 7-day-old sample and 7.6 MPa for 28-day-old sample. The flexural strength of mortar or concrete samples is widely acknowledged to be closely related to the structure of the ITZ situated between the aggregate and the hydrated cement paste (Singh et al., 2023; Liu et al., 2022; Gan et al., 2022). In standard concrete/mortar compositions, this zone is recognized as the weakest link in the system (Lyu et al., 2020). However, when RCA is incorporated into mortar mixtures instead of NA, the complexity and microstructure weakening effect of formed ITZs increases significantly. With RCA use, not only is there an interface between the RCA particles and the newly formed hydrated cement matrix, but there are also additional interfaces between the old aggregate contained within the RCA and the pre-existing hydrated cement paste adhering to these particles (Otsuki et al., 2003; Zhang et al., 2019; Zhao et al., 2017). In addition to this, crushing and grinding processes are required to prepare RCA and these processes lead to the formation of numerous cracks on the surfaces of the crushed material and seperation of ITZ locating between the aggregate and old hydrated cement paste adhering to these particles (Ozbakkaloglu et al., 2018; Kang et al., 2024). Furthermore, the ITZ between RCA and the new cement paste is characterized by the presence of numerous fine pores and cracks (Zhang et al., 2019). As a result, the increase in the number of interfaces and formation of weaker ITZ can significantly impact the mechanical properties of the RCA-incorporated systems. Therefore, all these phenemenons can be regarded as the possible reasons for the higher flexural strength results of the samples containing NA compared to those of samples containing RCA and CRCA. The incorporation of CRCA resulted in a notable enhancement in the flexural strength of mortar samples compared to those containing RCA. The similar findings were also found in previous studies (Xuan et al., 2016; Waseem, 2022). This outcome can be attributed to the pores becoming filled with calcium carbonate during the carbonation process of RCA. Consequently, the residual mortar adhering to RCA is strengthened, along with the existing ITZ between the aggregate and the pre-existing hydrated cement paste adhering to the aggregates. This reinforcement ultimately strengthens the CRCA, leading to an enhancement in the overall properties of the resulting mortar mixtures (Luo et al., 2022).

Table 4.2 Flexural strength values of mortar samples at the ages of 7 and 28 days

Sample	***Age***	***Mean (Mpa)***	***σ***	***CV (%)***
NA	7 days	6,85	0,13	1,84
RCA		5,11	0,36	7,01
CRCA		6,04	0,11	1,78
NA	28 days	7,78	0,30	3,83
RCA		5,66	0,09	1,67
CRCA		6,91	0,21	3,03

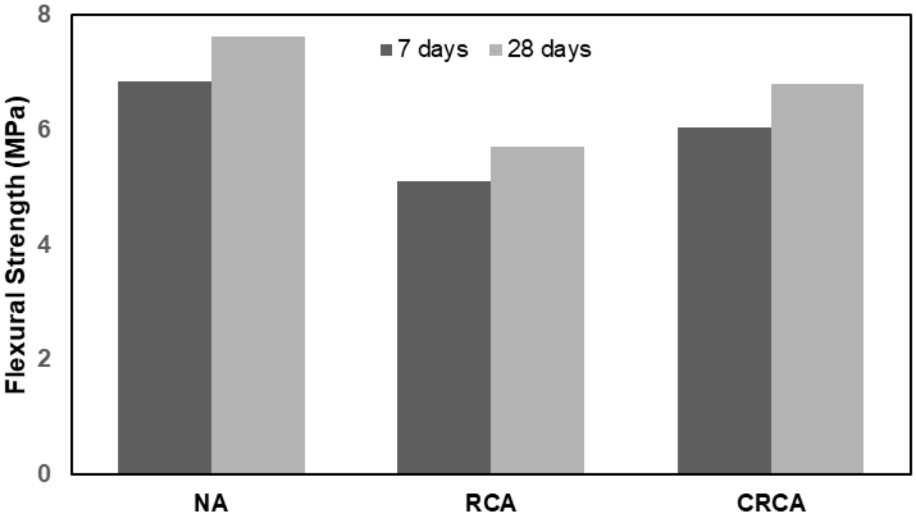


Figure 4.10 Flexural strength values of mortar samples at the ages of 7 and 28 days

The compressive strength test results were illustrated in Figure 4.11 to investigate the mechanical performance of cementitious systems incorporating RCA and CRCA in comparison to the use of NA. As indicated by the results, the compressive strength of the samples exhibited an increase with curing ages, regardless of aggregate types used. Upon examination of the results, mixtures containing NA and CRCA exhibit comparable mechanical performance, whereas the mixture with untreated RCA demonstrates lower performance, with compressive strength results of 44.23, 41.77, and 33.64 MPa, respectively, after 28 days. The decline in compressive strength associated with RCA can be ascribed to multiple factors:

i) the coexistence of two distinct ITZ within the matrix—representing both old and new interfaces,

ii) the presence of mortar residues on the RCA surface, characterized by low density and a highly porous ITZ (Waseem, 2022), potentially leading to reduced elastic modulus values and diminished strength in mortars incorporating RCAs,

iii) an elevated effective water-to-binder ratio linked to the use of RCA, resulting in the development of more porous structures (Behera et al., 2019),

iv) the existence of cracks, microcracks, holes, and fissures on the RCA surface attributable to the manufacturing process,

v) ettringite formation on RCA surfaces, causing volume expansion and additional internal stress in matrix.

The results conclusively demonstrate that the carbonation of RCA led to a significant improvement in mechanical performance, reaching levels comparable to those achieved with a cementitious matrix incorporating NA. This improvement can be attributed to the reduction of voids, cracked structures, and cavities within the RCA, especially in ITZ regions, resulting in a more stable and robust structure. Given that ITZ is commonly perceived as the weakest region responsible for crack propagation, the carbonation-induced modification of mortar residue on RCAs and the old ITZ leads to enhancement in mechanical performance. Additionally, the carbonation process led to a lowered effective water-to-binder ratio, contributing to a more compact microstructure.

Table 4.3 Compressive strength values of mortar samples at the ages of 7 and 28 days

Sample	***Age***	***Mean (MPa)***	***σ***	***CV (%)***
NA	7 days	34,17	1,60	4,69
RCA		24,33	0,64	2,64
CRCA		30,72	1,05	3,41
NA	28 days	44,23	1,69	3,83
RCA		33,64	0,52	1,54
CRCA		41,77	0,33	0,79

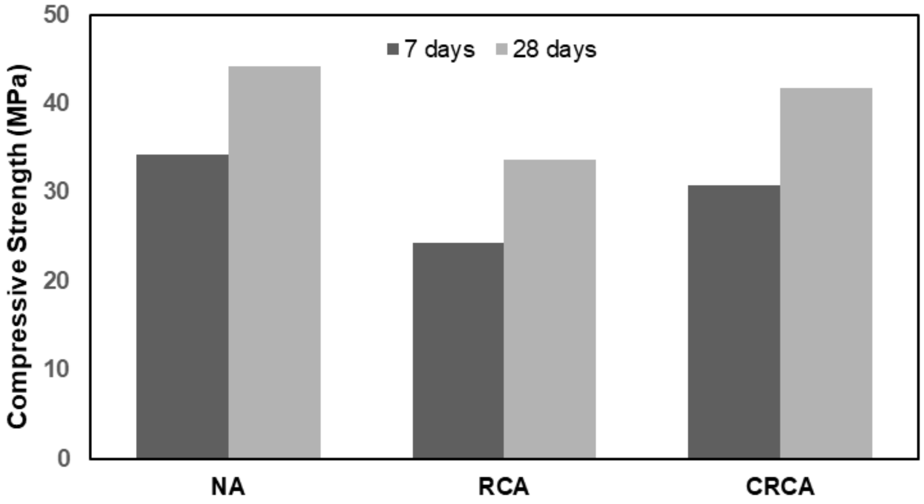


Figure 4.11 Compressive strength values of mortar samples at the ages of 7 and 28 days

Another study attracted attention, where serious decreases in strength were observed in mortars produced on the basis of equal flow with RCA in the range of 0-4 mm compared to NA. This situation is associated with the excessive void structure in the cement paste phase as a result of the excess water requirement due to RCA and the weak microstructure of the old mortar residue on the RCA surface. It has been stated that the high-void structure of RCA causes crack formation, propagation and eventually fracture as a result of stress concentration around the voids (Gonçalves et al., 2020). Depending on the source of RCA, the clay minerals in it contains also cause a decrease in strength (Alexandridou et al., 2018).

As it is known, CRCAs generally have lower water absorption capacity than RCAs. Since less water is used in the production of CRCA concrete than in the production of RCA concrete in order to reach the desired slump value in the prepared mortar or concrete mixture, it is expected that the effective w/c ratio in the mixture will remain low and therefore the compressive strength will be higher (Wang et al., 2020). The results obtained within the scope of the thesis also support this situation. As a result of the pre-wetting process to saturate the surface, the amount of water in the RCA mixture is higher than that in the CRCA and NA mixture. For this reason, it was evaluated that the local w/c ratio of ITZ on the surface of RCA particles may be higher than others. Similarly, the w/c ratio in the ITZ of the mixture with CRCA will be higher than that of the mixture with NA and lower than that of the mixture with RCA.

The only reason why CRCA offers a higher strength value is not that ITZ has become stronger. The cracked and void structure in the mortar residue on the RCA also causes low strength, and with carbonation, these voids and cracked structures are filled with CC and silica gel, thus increasing the strength. In a study, more voids and cracked structures were found in the ITZ observed in an SEM analysis performed on the concrete sample containing untreated RCA. This structure, which causes low strength, was observed much less in concrete using CRCA. The reason why CRCA gave a structure with low voids and cracks was that the CC and silica gel formed filled the void structure and CC could react with the aluminate phase (Lu et al., 2019). The formation of structures with high voids and column-like ettringite is frequently observed in the untreated RCA system. This causes expansion and additional stress in the internal structure. In the CRCA system, fiber and cotton-like calcium and silica gel products were formed, which contributed to the decrease in void ratio and increase in strength (Shi et al., 2018).

The disadvantageous situation in compressive strength is tried to be eliminated with RCAs processed with different techniques in the literature. It was observed that the compressive strength increased by approximately 14% in the samples produced with RCA pretreated with HCl and Na_2SO_4. This positive effect is explained by the elimination of the porous structure in the mortar residue by pretreatment and a denser interface can be obtained. (Kim et al., 2018). In Spaeth et al. (2014)'s study, it was reported that the water absorption of RCA pretreated with various polymers could be reduced, but there was a decrease in the compressive strength of the final product. As can be seen, some techniques that can improve the RCA surface and physical properties may not yield the expected results in compressive strength. CC formed on the RCA surface as a result of accelerated carbonation has a higher volume and microstrength than the CH present in high amounts before carbonation. Therefore, the CC formed contributes to the improvement in the physical properties of the mixture (Liang et al., 2019).

4.2.1.3. Water Absorption and Specific Gravity

Results in the literature show that RCA increases the water absorption of the mortar sample. This situation is generally attributed to the permeable structure of the new ITZ formed in the mortar sample and the old ITZ on the RCA surface allowing water ingress (Zhiming et al., 2022). In the study of Zhiming et al. (2022), it was found that the mortars prepared with RCA, in which the old mortar residue was removed, absorbed less water than the mortars prepared with RCA containing mortar residue on the surface. This shows that the water absorption rates of mortar samples are directly affected by the mortar residue on the RCA surface. For this reason, within the scope of this thesis study, it was examined whether the high water absorption, one of the negative effects caused by mortar residue on RCA, could be eliminated by the accelerated carbonation technique.

The presence and proportions of fine and coarse aggregate used in the mixture also affect the water absorption parameter. In the experiment conducted with concrete in which coarse and fine RCA were used with different replacement ratios, it was observed that the water absorption rate increased by 28% in the concrete using coarse RCA, but increased by 109% in the cocnrete using fine RCA. This is associated with fine RCA absorbing more water due to its higher specific surface area (Sasanipour and Aslani, 2020). As can be seen from this example, the part that most affects water absorption in concrete or mortar samples is the fine fraction of the aggregate used. It has been observed that the physical parameters of the fine aggregate will significantly change the water absorption value of the concrete or mortar samples.

In this thesis study, the results shown in Figure 4.12 were obtained in the water absorption test performed on mortar samples produced with NA, RCA and CRCA. It is seen that the degree of water absorption can be significantly reduced in samples produced with CRCA. As can be seen from Figure 4.12 at the end of 7 days of curing, 74.4% more water absorption was observed in RCA mortars and 22.8% more water absorption in CRCA mortars compared to NA mortars. At the end of 28 days of curing, these values were found to be 55% excess water absorption in RCA mortars and 13.5% in CRCA mortars. The first conclusion that can be drawn from these values is that the positive effects of the accelerated carbonation technique on RCA also provide significant benefits in terms of water absorption in the mortar/concrete in which RCA is used. The main reason for the decrease in water absorption is that the voids and capillaries contained in the mortar residue on the RCA surface are refined with CC. It can be inferred that ITZ, which is one of the regions where the permeability of mortars/concrete is most experienced, has a more dense and impermeable structure. In another study, as a result of 28 days of curing, it was determined that concrete including CRCA absorbed 21.12% less water than that including RCA (Waseem, 2022). This situation is associated with the formation of CC, lower void ratio and a denser matrix formation. According to the results in Figure 4.12, the RCA mortar absorbed 42% and 36.5% more water than the CRCA mortar, at the end of 7 and 28 days of curing, respectively. At the same time, the specific gravity value measured in the CRCA system was greater than that of the RCA system.

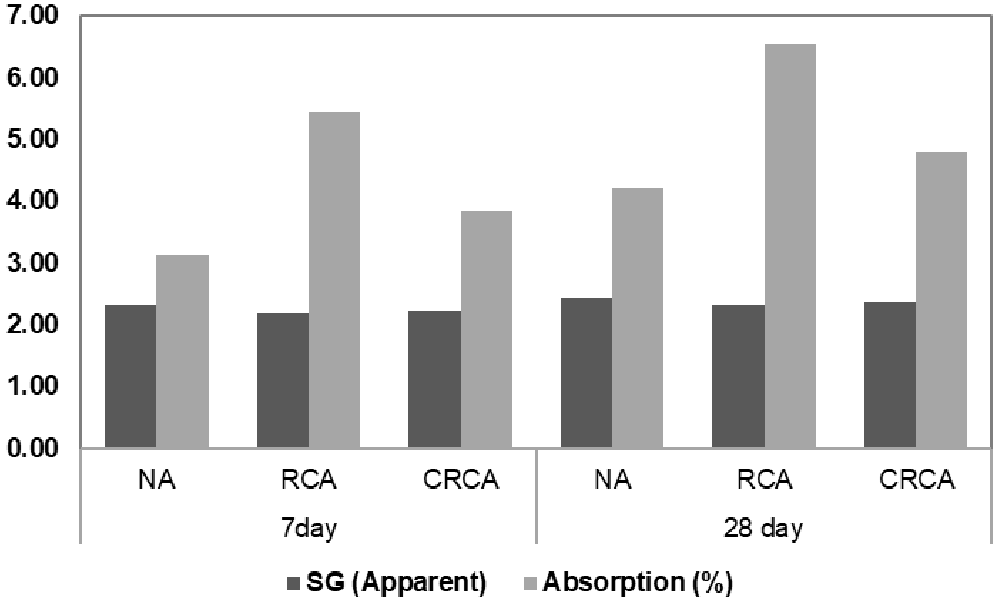


Figure 4.12 Water absorption (%) and specific gravity (apperent) values of mortar samples at the end of 7 and 28 days curing

Apart from carbonation, there are also studies that reduce the amount of water absorption with different surface improvement methods. Zhu et al. (2013) reported that the mixture using RCA processed with water-repellent materials increased its resistance to capillary water absorption, carbonation and chloride ion permeability in terms of durability.

4.2.2. Durability Performance of Cement-based Systems

4.2.2.1. Rate of Absorption (Sorptivity)

It is known that in addition to the water absorption capacity value in mortar samples, the rate of absorption (sorptivity) value is also an important source of information on the capillarity of the produced sample. In terms of durability, water leakage into the inner parts of the sample can cause serious problems. Therefore, in this study, rate of absorption testing was carried out on mortar samples produced with NA, RCA and CRCA in accordance with the ASTM C1585 standard. Mortar samples that were kept in water cure for 28 days were coated with epoxy, leaving only one surface open. The weight increase in the samples whose exposed surface was immersed in water was measured depending on time. The water absorbed by capillary effect rises to the upper parts of the sample and causes an increase in the weight of the sample. The weights of the samples, whose lower surface was immersed in water by 1-3 mm, were measured at certain periods for 9 days. The weight increase in the samples was proportional to the cross-sectional area of the surface in contact with water and the sorptivity (I) value in mm was calculated:

$$I = \frac{m_t}{a * d}$$

where:
I : the absorption
m_t : weight increase in the sample at time t, g
a : area of surface in contact with water, mm^2
d : density of water, g/mm^3

The absorption values measured for 9 days versus the square root of time (√sec) are plotted in graph. With linear regression analysis, the slope of the line graph was obtained and the initial and secondary rates of absorption (mm/√s) were obtained. The initial rate gives the absorption rate of the sample in the first 6 hours. The secondary rate of absorption value is obtained in measurements between 1 and 7 days. Both in initial and secondary rate measurements, the correlation coefficient in all measurements was above 0.98 and the measurements were considered valid.

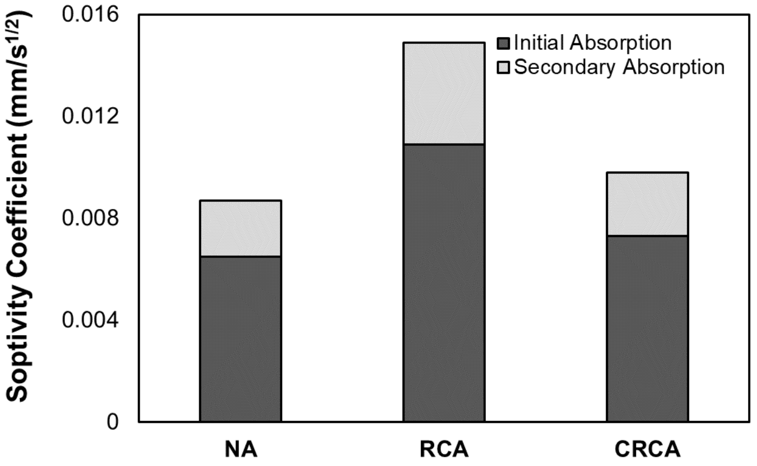


Figure 4.13 Initial and secondary rate of absorption of mortar samples

The slopes of the drawn line graphs gave the rate of absorption values of the samples. When these values in the Figure 4.13 are examined, it is seen that the initial rate of absorption of the RCA sample is 70.6% higher than that of NA. In a study in the literature, 25-50-75% RCA substitution resulted in a 12-14-52% increase in rate of absorption, respectively (Alexandridou et al., 2018). Gonçalves et al. (2020) obtained higher sorptivity values in mortars produced with 0-4 mm RCA than in mortars produced with natural sand. The high void structure resulting from the high water absorption capacity of RCA increased this value as expected. It is seen from the figure that the sample with CRCA is 10.3% higher than the sample with NA. According to the initial rate of absorption values, it was understood that 85.4% of the difference between NA and RCA could be eliminated by accelerated carbonation. The fact that the high sorptivity of RCA mortar due to high porous mortar residue could be reduced to this extent indicated that the pores and capillaries in the mortar residue were filled to a high degree by accelerated carbonation and the structure in this region was improved. Similarly, Gomez et al. (2021) obtained a much lower capillary coefficient with CRCA carbonated for 5 hours than with uncarbonated RCA.

Since the samples were in oven-dry condition at the beginning of the test, they absorbed water at a high rate in the first 6 hours of the test, and the water was quickly transported to the upper parts of the permeable channels in the sample by capillary effect. RCA samples allowed this transport more with their high permeable structure. When examined in Figure 4.13, it can be seen that secondary rates of absorption are lower than initial rates. The capillaries near the bottom surface of the sample become increasingly saturated during the first hours of the test. For this reason, there was a decrease in the rate of water transport upwards in the following hours and days of the test. When the sorptivity values between 1-7 days was examined, it was seen that the secondary sorptivity values of RCA and CRCA samples were 81.8% and 13.6% higher, respectively, than NA samples. Although the rates of absoprtion of all samples decreased in the later days of the test, this significant difference within them continued to show itself. As a result of the use of CRCA, it was observed that the serious sorptivity difference between RCA-NA was reduced by 83.4%. It is seen that carbonation improves sorptivity and the difference between RCA and NA can be compensated to a certain extent. Consistent with the literature, in the study of Waseem (2022), it was observed that rate of absorption gave the highest value in RCA concrete, followed by CRCA and NA, respectively. It has been interpreted that this situation is parallel to the trend in water absorption capacities of aggregate.

4.2.2.2. Freezing and Thawing

The high water absorption capacity and high degree of water saturation in RCA reduces the freeze-thaw resistance of the concrete in which it is used (Zaharieva et al., 2004). The excess water absorbed by RCA used in mortars can pass into the cement paste phase during freeze-thaw and reduce the freeze-thaw resistance. For this reason, the resistance of concrete where RCA is used directly in aggressive thermal environments is generally low and therefore the direct use of RCA is not recommended (Guo et al., 2018). In line with the aim of demonstrating that the use of RCA, discussed in the thesis study, can be made possible by accelerated carbonation, the produced mortar samples were subjected to a 300-cycle freeze-thaw test (ASTM C666) and the weight loss of the samples in this process was observed (Figure 4.14).

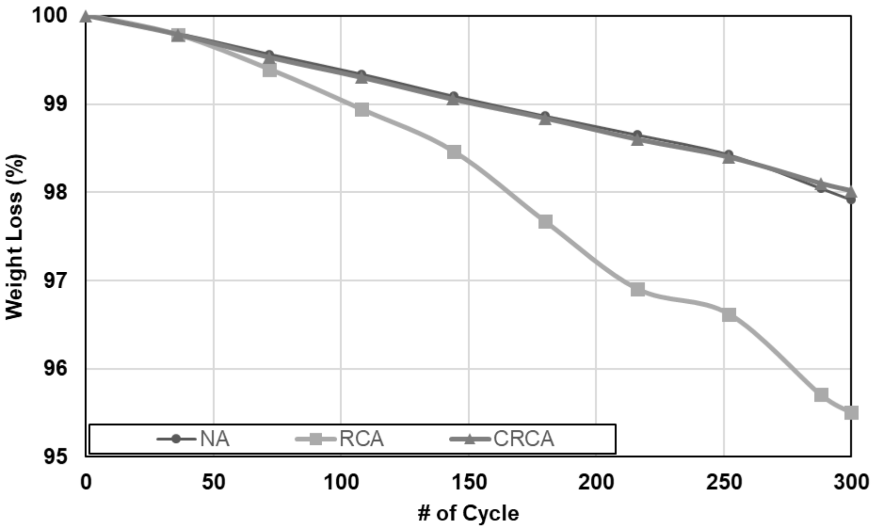


Figure 4.14 Graph of weight loss of samples (%) vs. number of cycles

It was understood that RCA showed a poor performance in the weight losses observed in Figure 4.14. At the end of 300 cycles, samples with RCA experienced a weight loss of 4.5%, while samples with NA and CRCA experienced a weight loss of 2%, very close to each other.

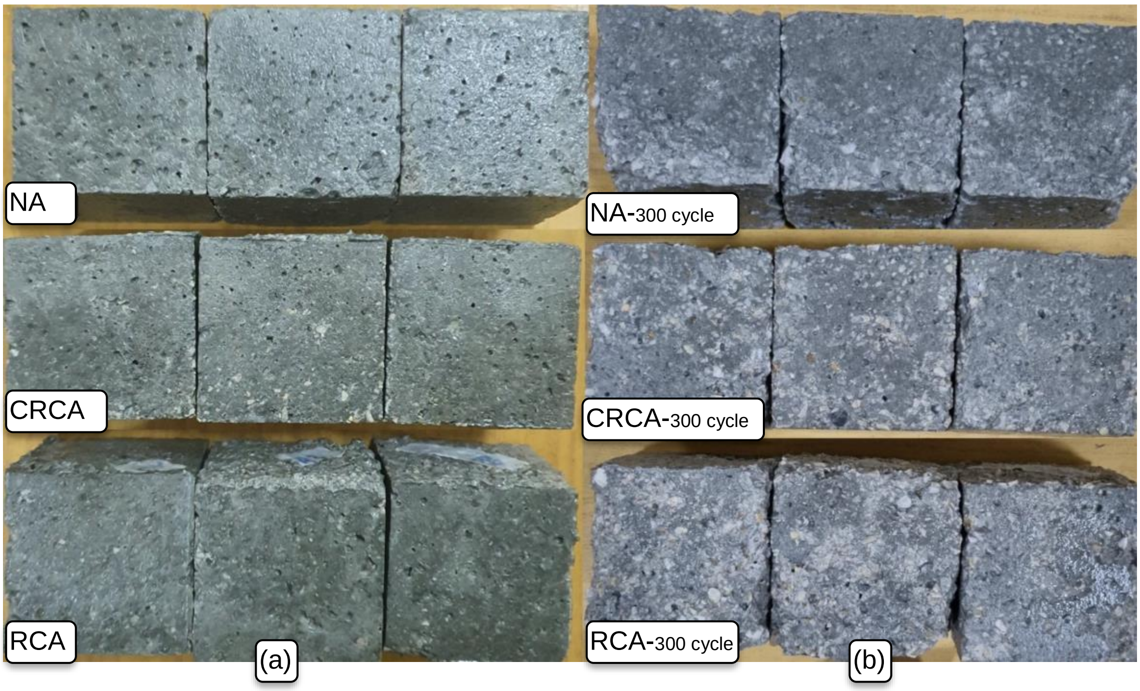


Figure 4.15 Image of samples (a) after 28 days of water cure and (b) after 300 freeze-thaw cycles

The difference between Figure 4.15 (a), which reflects the condition of the samples before the test, and Figure 4.15 (b), which shows the deterioration in the samples after 300 cycles, also confirms the weight loss values obtained. The positive effect of the accelerated carbonation improvement technique on RCA also showed itself in terms of frost resistance. It can be seen from these results how the porous structure, which plays a dominant role in freezing resistance, can be harmful when using RCA. On the other hand, it was revealed as a result of this test that RCA, whose use is avoided, can be made usable through the accelerated carbonation process.

The weight loss of the samples after completing 300 cycles is an important indicator. However, compressive strength also appears as an important indicator in order to see whether the carriying properties of samples, which are considered to remain in thermally aggressive environments, are lost after a certain number of freeze-thaw cycles. For this purpose, at the end of the test, the compressive strengths of the samples shown in Figure 4.15 (b) were measured. As can be seen from Figure 4.16, while there was a 19% loss of strength in NA samples after 300 cycles, there was a 27.7% loss in RCA samples and 19% loss in CRCA samples. After the test, the strength of the NA sample decreased to 35.5 MPa, while that of the RCA sample decreased to 25 MPa and that of the CRCA sample decreased to 33 MPa.

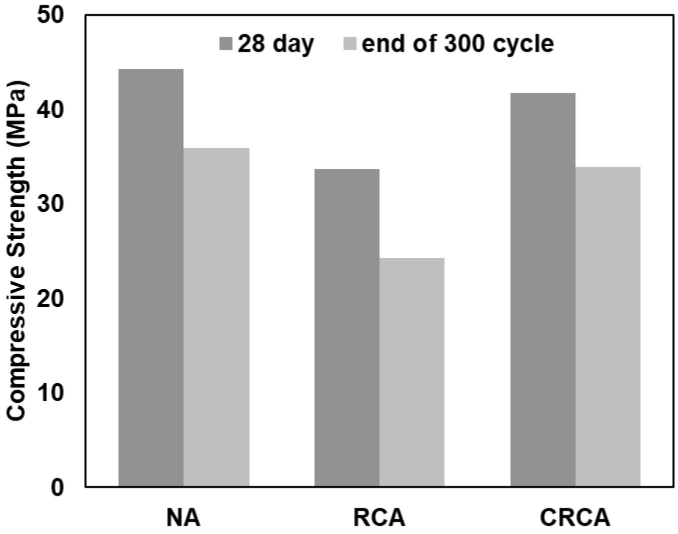


Figure 4.16 Compressive strength values of the samples after 28 days of water cure and end of 300 freeze-thaw cycles

As a result of this experiment, which was based on only the weight loss calculation and subsequent compressive strength measurement in the ASTM C666 standard, as it was carried out on mortar samples, it was determined how effective the accelerated carbonation technique was. The performance test in which this technique demonstrates itself most effectively is the freeze-thaw resistance test. With the use of CRCA, almost the same performance as NA samples was achieved in terms of weight loss and strength loss.

4.2.2.3. Rapid Chloride Permeability Test

The disadvantages brought by the use of RCA bring to mind the question to what extent the chloride ion permeability of the concrete produced with RCA may be affected. Determining this permeability is important in terms of predicting the risk of corrosion that the reinforced concrete structure using RCA may experience in the future. The corrosion chain begins with the destruction of the passivation layer on the surface of the steel reinforcement in reinforced concrete. Therefore, concrete must be designed in a structure that protects this passivation. This passivation may be negatively affected in situations such as natural carbonation, pH drop, cracked and permeable structure formation and Cl^- ion leakage that may occur in concrete. The most common situation in the literature about the use of RCA is that they create a highly porous and permeable structure in the concrete in which it is used. For this reason, it was decided to perform the chloride permeability test in this thesis study. It has been reported in the literature that chloride permeability increases significantly as the RCA substitution rate increases in mortars and concretes. It has been stated that due to high water demand, the porous structure increases and therefore ion transfer becomes easier. However, it has been mentioned that this negative effect can be eliminated by using different pozzolanic additives. Different techniques can be seen in the literature to overcome this disadvantageous situation regarding chloride ion transport. For example, the increase in permeability that occurs with the use of RCA is quite limited by the use of RCA with reduced mortar residue content (Ma et al., 2022).

Among various techniques, another technique that can reduce chloride ion permeability is the accelerated carbonation technique, which was discussed in the thesis. In a study, it was observed that the permeability coefficient decreased by 44, 64.5 and 79.9% with the use of CRCA which has residual mortar with w/c ratios (of parent concrete) of 0.35, 0.45 and 0.55, respectively. From this result, it is understood that carbonation has a more serious effect in low quality RCA. It has been evaluated that the carbonation percentage may be higher than high quality RCA since low quality RCA has a higher percentage of voids on its surface that will allow carbonation (Liang et al., 2019). Based on these results, it is thought that the accelerated carbonation technique can make a significant difference in the mortars produced with low quality RCA, which is the subject of this thesis study.

This test, which was discussed within the scope of the thesis study, was carried out inspired by the ASTM C1202 standard. Although this standard is recommended for concrete samples, it was thought that this standard could be used on mortar samples, since a qualitative comparison was aimed. Chloride ion permeation behavior, which can take years under real environmental conditions, can be demonstrated in a period of 6 hours with the help of this test. The electric current results on the samples for 6 hours by applying a 60V potential difference are shown in Figure 4.17. From the results in the graph in the figure, it is understood that the electric current in the sample with the use of RCA is significantly higher than that with NA, and the use of CRCA limits this increase. In the figure, maximum values of 207, 383, and 484 mA were reached in mortars containing NA, CRCA, and RCA, respectively. This test result showed that RCA creates a highly porous and permeable structure in mortar production. On the other hand, it has also been revealed to what extent the accelerated carbonation technique can compensate for this disadvantages. Waseem (2022) discovered that RCA provides an easier environment for the passage of chloride ions, while CRCA significantly reduces ion permeability due to the increase in density as a result of the decrease in void structures due to the formation of CC.

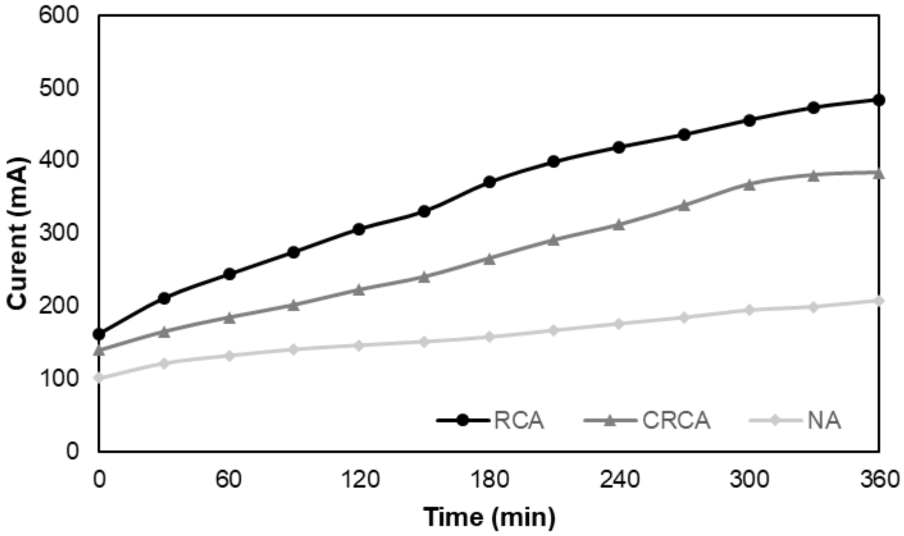


Figure 4.17 Electric current (mA) passing through samples according to time (min)

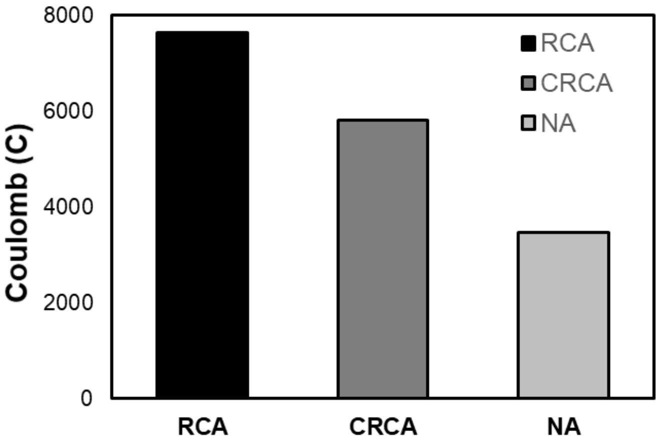


Figure 4.18 Total charge amounts passing through the samples at the end of the test

Figure 4.18 gives the total charge passing through the samples in coulombs at the end of the 6-hour test. These values are directly related to the chloride ion permeability of the samples and are perhaps one of the most important parameters in terms of durability. The total charge passing through the sample for 6 hours is an index that shows the electrical conductivity of the mortar sample. As seen from the graph here, the total charge passing through the RCA mortar sample is 2.2 times higher than the charge passing through the NA sample. This big difference clearly shows to what extent RCA mortars have a porous and permeable structure. Similar results are also found in a small number of studies in the literature. In one of these, it was observed that the permeability of mortars produced with

RCA increased due to the high void structure in the mortar residue on the aggregate surface, and that the total charge pass in mortars produced with RCA was up to 4.5 times compared to the control samples (Sasanipour and Aslani, 2020). In another study, chloride permeability increased by 15-40% with the use of 25-100% RCA (Thomas et al., 2018). In the literature, differences are observed in the permeability increase rates experienced with the use of RCA. It is thought that this situation is related to the different quality and chemical/mechanical properties of the demolition waste RCA produced from. On the other hand, another result was found that chloride permeability decreased by 21.25% as a result of using RCA with 30% replacement. However, in this study, with the use of RCA completely, the permeability increased twofold compared to the control sample. Similarly, weak and void ITZ due to free water and old mortar residue in the mixture were given as the reasons (Kirthika and Singh, 2020).

Surfaces with old mortar residues with high CH cause very harmful void structures. This porous structure makes it easier for water and chloride ions to leak into the concrete. Since this void structure decreases and improves with the formation of CC, it is expected to observe much lower chloride permeability in concrete produced with CRCA. For this reason, the chloride diffusion coefficient decreases with the use of CRCA. In a study, it was reported that concrete produced with CRCA had approximately 41-45% lower chloride permeability than those containing RCA (Shi et al., 2018). As can be seen from Figure 4.18, as a result of the test carried out within the scope of the thesis study, chloride permeability was reduced by 24% compared to RCA with the accelerated carbonation technique. This result shows that 44% of the permeability difference between NA and RCA can be compensated.

There are statements that the risk of reinforcement corrosion may actually increase in CRCA concretes due to the decreasing alkalinity degree as a result of carbonation of RCA (Tang et al., 2018). It is claimed that carbonation may reduce free alkali metal and hydroxyl ions, thus the pH of the environment may decrease and, as a result, the passivation of the reinforcement may be weakened (Weerdt et al., 2019). On the other hand, since permeability is significantly reduced by carbonation, the entry of harmful ions into the concrete is prevented and the risk of corrosion initiation can be indirectly reduced. (Xuan et al., 2017).

4.2.3. Microstructure of Cement-based Systems: SEM Analysis

The SEM micrographs showcased in Figure 4.19 depict significant microstructural differences among mortars incorporating NA, RCA, and CRCA. In Figure 4.19-a,b, NA mortar exhibited a dense and compatible cementitious matrix and aggregate, whereas RCA mortar displayed a rough surface characterized by ITZ cracks, ettringite formation, microcracks, and pores (Figure 4.19-c,d). The observable variations in microstructure between mortars containing NA and those utilizing RCA emphasize the potential problems linked to integrating recycled materials in a cementitious matrix. The effective approaches to these problems through chemical CC mineralization highlight the encouraging potential of treatments in improving the properties of recycled materials. CC emerged as the primary carbonation product, with calcite being the most stable phase, typically displaying a granular texture and assuming a rhombohedral or cubic form (Park et al., 2020). Previous observations in the literature noted that treating RCA surfaces with chemical carbonation led to a reduction in porosity and the formation of visually identifiable granular-textured and cubic-shaped calcium carbonate crystals on the surface of CRCA (Bertos et al., 2004). The rough and irregular surface of RCA mortar, attributed to the presence of old mortar remains, is evident in Figure 4.19-c (Behera et al., 2014). In contrast, CRCA mortar exhibited distinct precipitation of rhombohedral-shaped CC crystals (Figure 4.19-e,f). In addition, observation of needle-shaped ettringite is possible within the adherent mortar of RCA, yet no crystals are evident after the carbonation process, indicating their total disappearance. Furthermore, numerous micro-cracks, voids, and loosely bound hydrated products contribute to inadequate bonding between the attached mortar and RCA (Zhang et al., 2015).

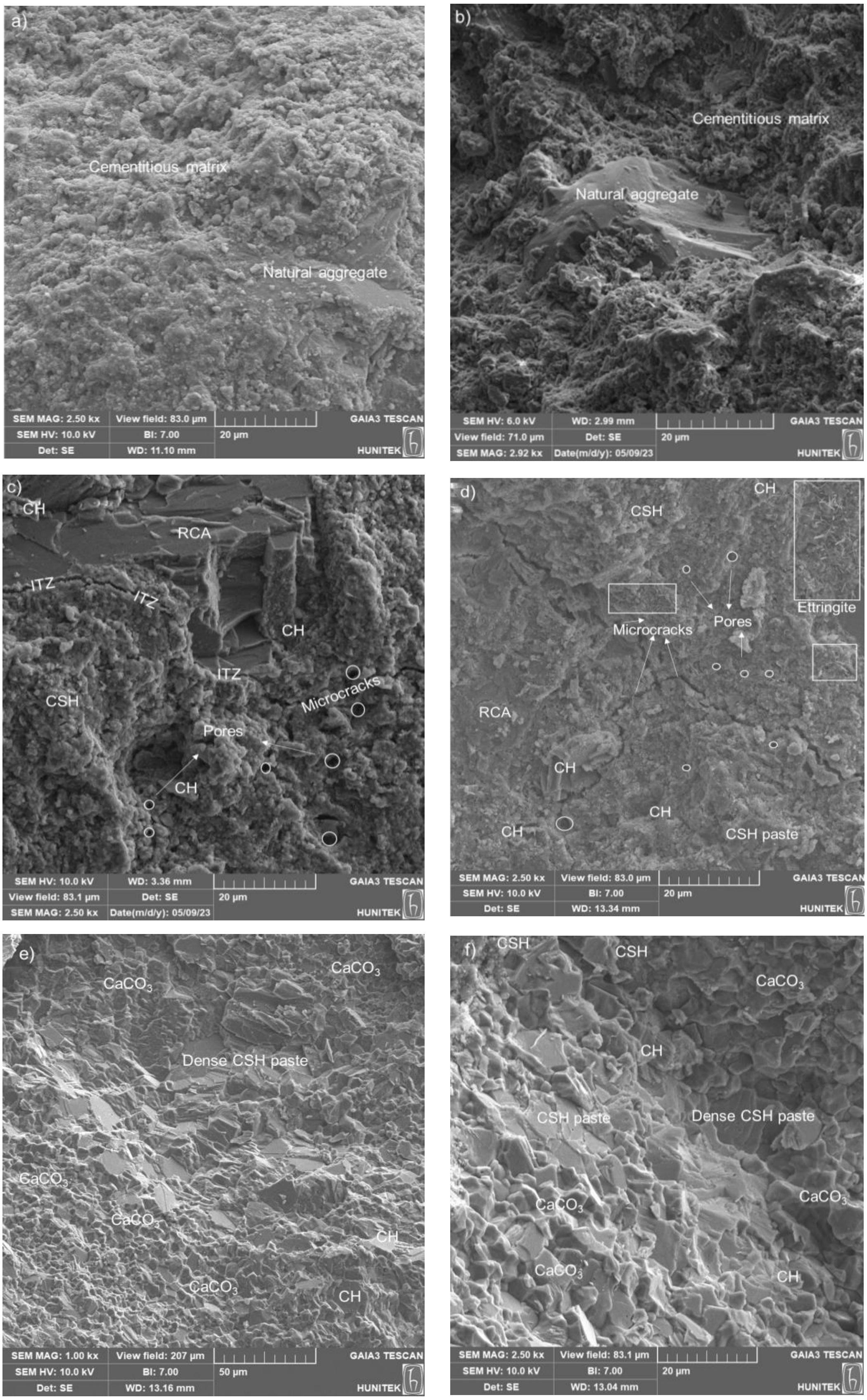


Figure 4.19 SEM micrographs of mortars including a-b) NA, c-d) RCA, e-f) CRCA.

*Squared areas in the figure show ettringite formation.

The diminished compressive and flexural strength observed in RCA mortar samples may also be associated with ITZ, microcracks, ettringite formation, and surface voids in RCAs. Moreover, the microstructure of CRCA mortar appeared denser, more uniform, and better covered with regular calcite crystals compared to RCA mortar. In contrast, in Figure 4.19-e,f, a densified ITZ with hydration products like CC is evident, indicating an enhancement in microstructural surfaces. Consequently, SEM analysis confirms the enhancement of ITZ in CRCA mortar (Li et al., 2019).

Consequently, chemical-induced CC mineralization effectively eradicated the porous and rough structures associated with RCA, potentially alleviating adverse effects on mortar microstructure. The presence of CC particles contributed to the densification of ITZs. Thus, the accelerated carbonation treatment markedly improved the quality of the attached mortar and the ITZs within the RCA. Overall, it can be indicated that the accelerated carbonation treatment enhances the original ITZ and the adherent mortar in the RCA (Zhang et al., 2015; Guo et al., 2018; Li et al., 2018).

5. CONCLUSION

First stage of the thesis study investigates the effects of different carbonation process-related parameters including relative humidity, pressure, temperature, duration, and particle size of RCA on the CO_2 uptake performance of low-quality RCAs obtained from end-of-life buildings. The following conclusions have been drawn from the experimental studies carried out within the scope of this stage:

- Among the RH of 50%, 70%, and 90%, considering the CO_2 uptake level and water absorption capacity results, the RH level of 90% resulted in better carbonation efficiency compared to the 50% RH; however, 70% RH provided the optimal condition for carbonation efficiency.
- As the RCA size increases (>4.75 mm), the pressure increase is beneficial in terms of both CO_2 capture and water absorption capacity, but it does not provide a benefit in fine RCA and even causes a decrease in CO_2 capture rate. In all sizes, there was a faster decrease in the water absorption capacity in the first hours of the curing period in 2 and 3 bar pressure environments, and this decrease slowed down in the following hours.
- An increase in the ambient temperature was found to be favorable for the overall carbonation performance of the RCA. While more stable and denser polymorphs of the carbonation process are obtained at low temperatures, the formation of meta-stable forms of CC is induced under high temperatures.
- The carbonation performance of RCA increased with the increase in carbonation duration. The carbonation reactions continued progressively in the first 48 hours but after 48-hour of the carbonation process, the CO_2 uptake rates became relatively stable.
- The findings revealed that RCA particle size influences the rate of CO_2 absorption during the carbonation process. Particles with a size of 2.00–4.75 mm distinguished among the others by absorbing CO_2 with 55.4%, 46.3%, 67.9%, and 60.3% higher than those with a size of <0.85, 0.85–2.00 mm, 4.75–9.50 mm and, 9.50-14.00 mm, respectively.

- SEM results indicated that the surface of RCA was rough due to the presence of old mortar residues. Still, RCA’s EDX findings showed that RCA's chemical components or the long-term effects of exposure to CO_2 on the synthesis of calcite or calcite production because of CO_2 exposure over time could be the cause. However, the CRCA surface's EDX data showed larger precipitation of calcite and vaterite.
- Calcite crystalline peaks were observed in the XRD patterns of both RCA and CRCA. As a result of the carbonation process, portlandite, calcium oxide, and monocalcium aluminate available on the RCA were transformed into carbonation products. Although calcite, aragonite, and vaterite were detected for the CRCA carbonated under 90^{o}C temperature, calcium carbonate in a form of vaterite was not observable in the RCA and CRCA carbonated under 50^{o}C temperature.

Overall, as a result of the first stage where optimum carbonation conditions of RCA are sought, by subjecting low-quality RCA to accelerated carbonation under the optimally selected and defined conditions of 70% RH, 50 °C, and 1 bar, approximately 8% CO_2 capture was achieved with 2-4.75 mm size range compared to untreated RCA.

In the second stage, where engineering properties were investigated, a series of tests were carried out on mortar samples. The following conclusions have been drawn from the experimental studies carried out within the scope of second stage:

- Although all aggregates were added to the mixture in a surface saturated state, the flow value of the mixture with NA was the highest, the mixture with RCA was the minimum. Change in slump values with NA, CRCA and RCA was due to the difference in particle shapes. NA is the aggregate with the least roughness and RCA is the aggregate with the most.
- The flexural strength of CRCA mortars was higher than RCA mortars and lower than NA mortars. The incorporation of CRCA resulted in a notable enhancement in the flexural strength of mortar samples compared to those containing RCA.
- Mortar samples containing NA and CRCA exhibit comparable mechanical performance, whereas the samples with RCA demonstrates lower performance,

with compressive strength results of 44.23, 41.77, and 33.64 MPa, respectively, after 28 days. Carbonation of RCA led to a significant improvement in mechanical performance, reaching levels comparable to those of mortar sample incorporating NA.

- The degree of water absorption of mortar samples can be significantly reduced with use of CRCA. At the end of 28 days of water curing, values were found to be 55% excess water absorption in RCA mortars and 13.5% in CRCA mortars according to the NA mortars. It can be inferred that ITZ in mortar sample including CRCA has a more dense and impermeable structure than ITZ in the mortar sample including RCA.
- The initial rate of absorption of the RCA sample is 70.6% higher than that of NA. The sample with CRCA showed 10.3% higher sorptivity than the sample with NA. When the secondary rate of absorption values were examined, it was seen that the sorptivity values of RCA and CRCA samples were 81.8% and 13.6% higher, respectively, than NA samples. It is seen that carbonation improves sorptivity and the difference between RCA and NA can be compensated to a certain extent.
- At the end of 300 cycles of freezing and thawing, samples with RCA experienced a weight loss of 4.5%, while samples with NA and CRCA experienced a weight loss of 2%, very close to each other. While there was a 19% loss of compressive strength in NA samples after 300 cycles, there was a 27.7% loss in RCA samples and 19% loss in CRCA samples. The performance test in which accelerated carbonation technique demonstrates itself most effectively was the freeze-thaw resistance test. With the use of CRCA, almost the same performance as NA samples was achieved in terms of weight loss and strength loss.
- According to the rapid chloride-ion permeability test results, the electric current in the mortar sample including RCA is significantly higher than that including NA, and the use of CRCA limits this increase. The total charge passing through the RCA mortar sample is 2.2 times higher than the charge passing through the NA sample. Chloride permeability was reduced by 24% compared to RCA with use of CRCA. It was understood that RCA provides an easier environment for the

passage of chloride ions, while CRCA reduces ion permeability due to the decrease in void structures with the formation of CC.

- The SEM analysis also showed notable enhancements in the ITZs in mortar including CRCA, displaying denser and more uniform microstructures compared to that including RCA, highlighting the effectiveness of carbonation treatment in improving both the initial ITZ and the surrounding mortar in RCA.

The low-quality RCA have shown improvement in microstructural, physical, mechanical, and durabilty aspects by studying the accelerated carbonation method comprehensively. Instead of being just a CDW waste, they can be actively used in the cementitious systems and their adverse environmental effects can be reduced. In addition, showed results in the thesis contributed to increasing the use of CRCA in newly produced concretes and thus to the sustainability of cement based systems.

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APPENDICES

Appendix 1- Publications Derived from Thesis

Dündar B., Sönmez Tuğluca M., İlcan H., Şahin O., Şahmaran M. The effects of various operational- and materials-oriented parameters on the carbonation performance of low-quality recycled concrete aggregate. Journal of Building Engineering, Volume 68 (2023), 106138, ISSN 2352-7102.