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Eggshell and Seashells Biomaterials Sorbent for Carbon Dioxide Capture

Abarasi Hart and Helen Onyeaka

Abstract

This review aims to explore the application of natural and renewable bioceramics such as eggshell and seashells in carbon dioxide (CO₂) capture from power plant flue gas. CO₂ capture, utilisation and storage (CCUS) is considered a means to deliver low carbon energy, decarbonising industries, power plants and facilitates the net removal of CO₂ from the atmosphere. The stages involved include CO₂ capture, transport of the captured CO₂, utilisation and secure storage of the captured CO₂. This chapter reports the use of eggshell and seashells biomaterials as an adsorbent to separate CO₂ from other gases generated by power plants and industrial processes. The capture of carbon dioxide by adsorption is based on the ability of a material to preferentially adsorb or carbonate CO₂ over other gases. In light of this, calcined eggshell and seashells biomaterial rich in calcium carbonate from which calcium oxide (94%) can be obtained have demonstrated a strong affinity for CO₂. These biomaterials are abundant and low-cost alternative to zeolite, activated carbon and molecular sieve carbon. The mechanism of CO₂ capture by eggshell and seashells derived CaO adsorbent comprises of a series of carbonation-calcination reactions (CCR): calcium oxide (CaO) reacts with CO₂ resulting in calcium carbonate (CaCO₃), which releases pure CO₂ stream upon calcinations for sequestration or utilisation, and as a consequence, the biomaterial is regenerated. Findings reveal that these biomaterials can hold up to eight times its own weight of CO₂ from flue gas stream. It was also found that the combination of 2 M acetic acid and water pretreatment improved the reactivity and capture capacity of the biomaterial for successive regeneration over four cycle's usage. Unlike activated carbon, these biomaterials are considered stable for high-temperature adsorption through carbonation.

Keywords: carbon dioxide capture, eggshell, seashells, adsorption, carbonation, calcination

1. Introduction

The combustion of fossil fuels such as coal, oil and natural gas for energy generate a large amount of carbon dioxide (CO₂) emission, causing global warming and climate change. Presently, legislation such as the Paris Agreement of 2015, provided a framework on dealing with greenhouse-gas-emissions (GHG) mitigation, and it is anticipated across the industrialised world to cut down the amount of CO₂

emissions and limit global warming to less than 2°C [1]. Additionally, the demand for energy is expected to increase by 50% in 2030, and also oil and gas are considered the principal feedstock of about 90% of chemicals produced worldwide, and it is forecasted that petrochemical industries will become the largest driver for global oil consumption by 2050 [2]. In this light, it is therefore important to mitigate the environmental impact of burning carbon-based fuels, in which potential progress has already been made in CO₂ capture, utilisation and storage (CCUS) technologies [3]. The CCUS is considered a means to deliver low carbon energy, decarbonising industries, and facilitates the net removal of CO₂ from the atmosphere. The stages involved include CO₂ capture, transport of the captured CO₂, utilisation and secure storage of the captured CO₂.

Carbon dioxide capture will play a significant role as fossil fuel will continue to meet world energy needs during this transition to sustainable low-carbon energy system [4]. It has also been reported that this transition phase will linger for a long time, providing sufficient time for the development and commercialisation of renewable energy systems. The transportation sector especially logistics operations majorly depend on fossil fuels, resulting in large carbon footprint on the environment. Based on World Bank data, the shift into low-carbon energy such as renewable energy in logistics operations prove to minimise carbon emission and other greenhouse gases, create sustainable environment as well as improve economic performance [5, 6]. In 2018, the global CO₂ emissions increased to 37.1 Gt which is forecasted to rise by about 10% in 2040, majorly due to the combustion of fossil fuels from industrial processes and transportation sector [7]. Hence, the impact of carbon emissions from logistics operations on the environment, global warming, climate change and health can be reduced remarkably by adopting renewable energy and green vehicles [6]. Therefore, government policy and legislations such as the Paris Agreement of 2015 are necessary to drive research and development into low-carbon energy and environmental sustainability. As a result of these policies, renewable energy and carbon capture technologies are being developed, and their implementation is expected to improve environmental quality and sustainability [5, 8, 9]. Unlike fossil fuels, renewable energies promote eco-friendly environment. Hence, CCUS technologies will enable the use of fossil fuels in a cleaner way when integrated with power plants to mitigate global warming and climate change effects. CO₂ has found utilisation in the following areas mineralisation, biological utilisation, food and beverages, energy storage media, chemicals, enhanced oil recovery, coal bed methane and hydraulic fracturing processes [7]. However, public awareness and acceptance of CCUS is still low in spite of the attention shown by the scientific communities, industries and governments. Findings by Tcvetkov et al. [10] show that most studies on CCUS are dedicated to carbon dioxide storage in geological formation with less attention on capture and transportation. Hence, this study focuses on carbon dioxide capture using natural and renewable biomaterials such as eggshells and seashells.

The essence of carbon capture is to separate carbon dioxide from other gases produced as a result of the combustion of fossil fuels for power generation and industrial processes. **Figure 1** shows the three main approaches to accomplish this, which are pre-combustion capture, post-combustion capture and oxy-fuel combustion methods.

Before now, the capture of carbon dioxide is commonly achieved in the industry through absorption using liquid solvents such as selexol, rectisol, and mono-ethanol-amine, MEA [11]. The absorption process involves the use of two columns, namely the absorber and the stripper. This makes the process cost intensive in addition to corrosion issues. Consequently, a large amount of energy is needed to absorb CO₂ [12]. On the other hand, physical adsorption via solid adsorption

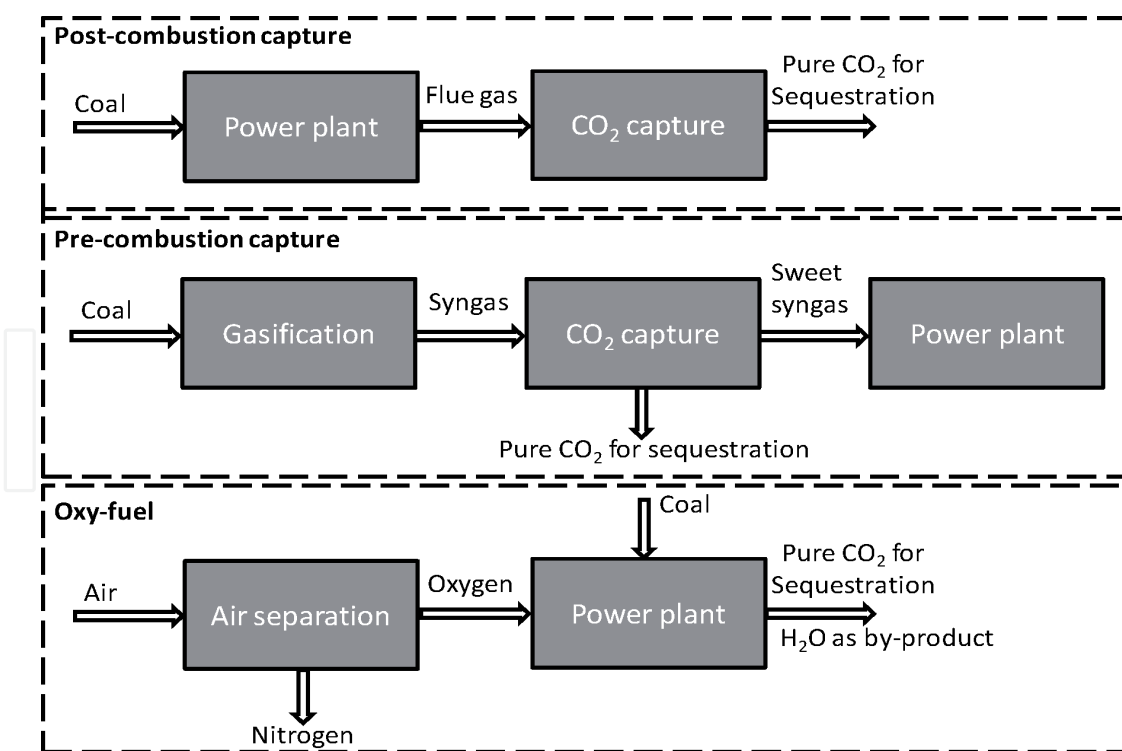


Figure 1.
 Three common carbon dioxide capture approaches for coal fired power plant.

processes can selectively separate carbon dioxide from flue gas mixture. The advantages of adsorption include high selectivity, operation simplicity, low-cost, ease of regeneration, and low corrosiveness of adsorbent compared with solvent processes [11, 12].

The carbon dioxide adsorption approaches rely on the ability of the adsorbing material to preferentially adsorb CO₂ over other gases. This is achieved through a packed bed system of the adsorbent materials. The adsorbent materials will continue to absorb CO₂ until it is saturated, which is its adsorptive capacity. At this point, the packed bed undergoes desorption either through pressure swing adsorption (PSA) or temperature swing adsorption (TSA), which causes the release of the adsorbed CO₂ to the point where the adsorbent material is at equilibrium [12, 13]. The commonly used adsorbent materials include zeolites, activated carbon, microporous/mesoporous silica, carbonates, carbon molecular sieves and metal organic frameworks. These materials possess adequate surface area and pore network structures that are highly microporous to accommodate and capture CO₂ [12, 13]. The adsorbent materials are evaluated on the basis of adsorption capacity, preferential adsorption affinity for carbon dioxide over gases from flue gas stream, adsorption and desorption kinetics, low-cost, tolerance of impurities, mechanical strength, multicycle durability and regeneration of stability [13]. Additionally, the porous structure of the adsorbent material is engineered to improve mass transport by reducing diffusional resistance, and the microstructure and morphological texture must demonstrate the capacity to hold captured CO₂ during multi cycling between the absorption and regeneration steps [13, 14]. However, since the process is based on gas-solid interaction, operational conditions such as gas flow rate, temperature and vibration could cause disintegration of adsorbent material due to crushing and abrasion, and consequently collapse pore network structures. It is also rare to find a single adsorbent material that maximises all the above highlighted attributes. Therefore, this review explores the use of other materials such as eggshell and seashell rich in calcium carbonate through reactive adsorption, which involves carbonation – calcination of CaO/CaCO₃ for carbon dioxide capture.

Alkaline earth metal oxides have demonstrated a strong affinity for acidic gas such as carbon dioxide and sulphur oxides. These metal oxides, particularly calcium oxide (CaO), are effective for the removal of CO₂ via carbonation at moderate temperatures of less than 700°C [11]. Hence, calcium oxide has proven a good sorbent material for carbon dioxide capture. With regards to availability and cost, an excellent source of CaO is calcium carbonate (CaCO₃). The most widely natural source of CaCO₃ includes dolomite and limestone. However, these natural resources are non-renewable, energy intensive to exploit, their mining cause damage to the environment as well as landscape. More also, CaO sorbent derived from natural limestone decreases in its reactivity over a number of cycles of reaction with CO₂ [15]. As a result of this, attention has been shifted to renewable sources such as eggshells, seashells and snail shells. These waste biomaterials provide sustainable source of calcium carbonate (CaCO₃) in the range of 90–96% [16]. Calcined egg-shell and seashells such as oyster shell are rich in lime (CaO) and can be combined with post-combustion and pre-combustion systems to separate CO₂ through cyclic carbonation of CaO (calcined eggshell/seashell) to CaCO₃, and subsequently the calcination of CaCO₃ to release pure CO₂ and regenerate back to CaO, as shown in **Figure 2** [15, 17–19]. This reversible reaction between CaO and CO₂ is a promising approach of removing CO₂ from flue gas from power plants, producing a pure stream of CO₂ ready for geological sequestration [15, 19]. To achieve this objective, the material should exhibit sufficient reactivity and thermal stability. Eggshell and seashell are a low-cost and abundant alternative to synthetic calcium carbonate and lime sorbents.

The poultry and seafood industries generate millions of tonnes of waste shells annually, which are disposed of in landfills. These biomaterials are rich in calcium carbonate, and subsequently, a large source of calcium oxide. The discarded eggshells and seashells after consumption of their food content, the heap waste shell is a habitat for microbes which causes environmental and air pollution due to emission of intensive odour especially during microbial decomposition [16]. These waste shell biomaterials can be recycled and used as a source of calcium oxide material for carbon dioxide capture purposes. Remarkable costs can be saved when these waste shells biomaterials are re-used, with emphasises on economic and sustainable

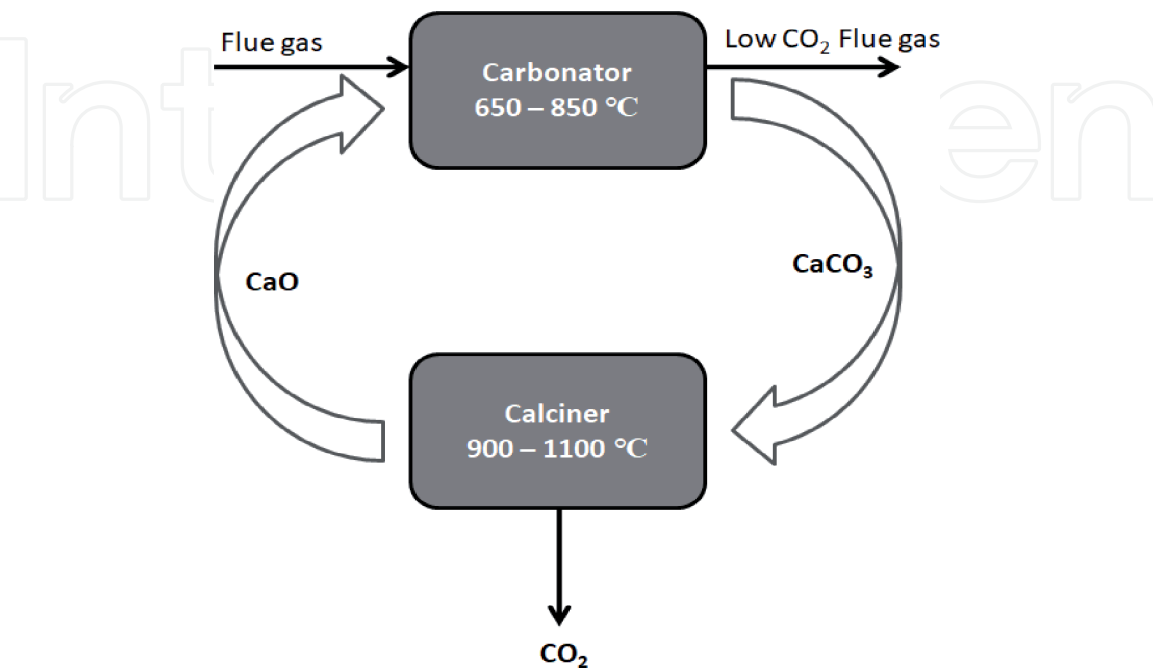


Figure 2.
Carbonation – calcination process in calcium looping cycle application for carbon capture.

environmental benefits of recycling instead of disposing. However, the carbon dioxide capture capacity of synthesised calcium oxide sorbents from eggshell and seashells decreases, as cycles of carbonation and calcination increases because of sintering over time [17]. To remedy this, it is important to generate more porous surface structure in the biomaterials through pre-treatment and regeneration processes.

2. Physicochemical properties of eggshell and seashells biomaterials

The major solid mineral component of eggshells and seashells is calcium carbonate in the range of 92–96% and minor trace elements such as silica, alumina, phosphorous, magnesium, sodium, potassium, zinc, manganese, iron, and copper. A detail composition of eggshell and seashells has been reported elsewhere [16]. The physical properties of some calcined eggshells and seashells biomaterials such as surface area, pore volume and pore diameter are shown in **Table 1**. These waste shells biomaterials exhibit the type-IV isotherm which an attribute of mesoporous texture morphology characterised with a network of micropores. The pore size re-affirms their microstructure characteristics to accommodate captured CO₂. During calcination, the specific surface area and pore volume of the crushed eggshells and seashells biomaterials increases, as the calcination temperature increases. This is because of the evolution of porosity within the material as a result of the release of CO₂ from CaCO₃, leading to the formation of CaO [16, 20]. However, at a temperature greater than 900°C, the surface area and pore volume decreased due to prolonged thermal effect, resulting in sintering [16, 20].

Figure 3 shows the X-Ray Diffraction (XRD) patterns of uncalcined (natural) and calcined (thermally treated) eggshell (quail) and seashell (oyster shell). The major component visible on the XRD pattern of the natural crushed shells is CaCO₃

Parameter	Mussel shell	Oyster shell	Chicken eggshell	Ostrich eggshell
Surface area (m ² /g)	89.91	24.00	54.60	71.00
Pore volume (cm ³ /g)	0.130	0.04	0.015	0.022
Pore size (nm)	3.5	6.6	0.54	0.61
Reference	[21]		[22]	

Table 1.
Surface area, pore size and volume of calcined seashells and eggshells biomaterials.

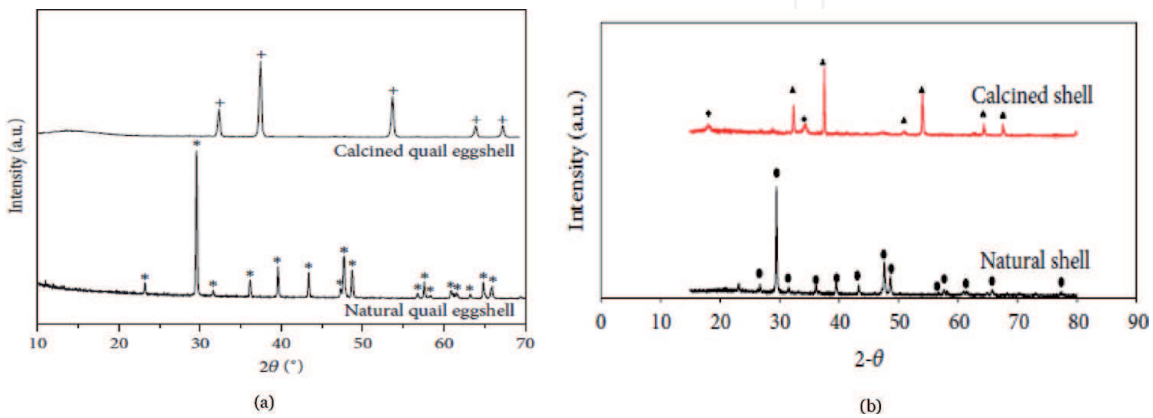


Figure 3.
XRD pattern of shells natural and calcined: (a) quail eggshell (*CaCO₃, natural + CaO, calcined at 900°C) [23] and (b) oyster shell (symbols: ●CaCO₃, ▲CaO, and ◆Ca(OH)₂) [21].

and a small amount of $\text{Ca}(\text{OH})_2$. Both the quail eggshell and the oyster shell share identical diffraction patterns for both the natural and calcined forms.

This suggests a similar mineralogical identity. After calcination (thermal treatment process), the diffraction lines attributed to rhombohedral phase for CaCO_3 disappeared, with new diffraction patterns arising around $2\theta = 32.3^\circ, 37.4^\circ, 53.7^\circ, 63.9^\circ$, and 67.3° assigned to cubic phase for lime (CaO) appeared (**Figure 3**). It is worthy to note that the quail eggshell exhibited a crystallite size of 315 nm (CaCO_3), while its calcined counterpart showed a size of 240 nm, CaO [23]. This crystallite size decrease can be ascribed to the exothermic natures of the calcination process. However, the lower intensity peaks for calcined eggshell and oyster shell could be related to the reduction in the crystallite size [21, 23]. Hence, the changes in the XRD pattern as a result of calcination are because of the release of carbon dioxide from the decomposition of CaCO_3 into CaO .

3. Methods of sorbent preparation

The associated complexity and high cost for the production of carbon dioxide capture adsorbent materials such as activated carbon or zeolite has shifted attention to exploiting and developing cheap and renewable materials such as eggshells and seashells biomaterials. **Figure 4** shows the procedure involved in the preparation of sorbent material from eggshells and seashells. The waste eggshells and seashells first undergo pre-treatment, which begins with acetic acid treatment with a concentration in the range of 1–10 molar to remove dirt, membrane layer, fibrous matters, proteins and other impurities as well as improve pore structure of the biomaterial [24]. Exposing the waste shells to acetic acid promotes the detachment of protein-collagen membrane depending on the extent, concentration and duration. At the end of this process, the sample is filtered and rinsed with distilled or deionised water. The separated eggshell or seashell is dried at $100\text{--}200^\circ\text{C}$ for 5 h [16]. The dried biomaterials are crushed and then sieved into different particle size ranges depending on the application. The particles are calcined; the calcination process involves heat treatment to decompose the major component CaCO_3 into CaO . The temperature of calcination could range from 500 to 1000°C depending on the application. It has been reported that at 900°C , the CaCO_3 undergoes complete conversion into CaO [21]. The material produced after calcination is the sorbent material, which is placed in a desiccator to curtail the chances of coming in contact with humidity and carbon dioxide in the air.

In the pre-treatment phase, the reaction of acetic acid with CaCO_3 results in the formation of calcium acetate, which has a larger molar volume than CaCO_3 and CaO [25]. The acetic acid treatment helps to expand and improve particle pore structure. As a result of the expanded and enhanced pore network structure, improve performance is achieved over multiple carbonation-calcination reaction

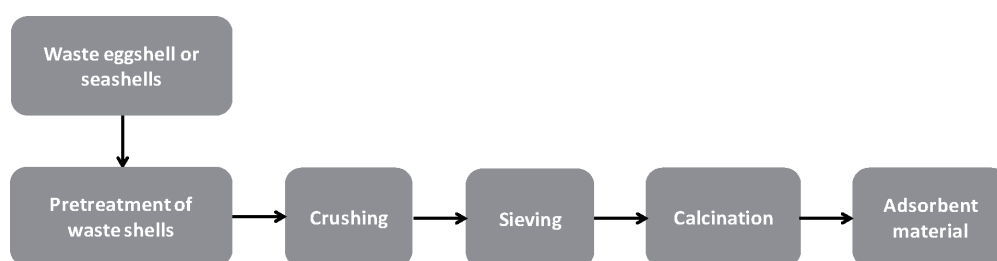


Figure 4. Adsorbent material preparation procedure from eggshell and seashells.

(CCR) cycles [24, 26, 27]. Hence, the increased porosity within the microstructure of the synthesised CaO sorbent biomaterial from eggshell or seashells leads to increased reactivity over time.

4. Adsorbent performance

The continuing reliance on fossil fuels such as coal, natural gas and crude oil emits greenhouse gas (GHG) especially carbon dioxide (CO₂), a major contributor to global warming. The application of physical and chemical absorption using solvents such as selexol, rectisol, and mono-ethanol-amine (MEA) to remove carbon dioxide from flue gas streams is limited by low-temperature, cost and energy-intensive to regenerate [11]. Produced CaO sorbent material from eggshells or seashells through the method outlined in **Figure 4**, has proven a good candidate for carbon dioxide capture from flue gas stream of power plants. This is owing to their affinity to carbonate in the presence of CO₂; resulting in the formation of CaCO₃ which is regenerated back to CaO via calcinations while pure CO₂ is released for sequestration in the process as shown in **Figure 5**.

Unlike the adsorption process for CO₂ capture using activated carbon or zeolite adsorbent materials, eggshells and seashells biomaterials are low-cost and offer exclusive environmental and economic benefits. Additionally, eggshell or seashell-derived CaO sorbent are abundant, renewable, simple to prepare and also possesses excellent thermal stability. The mechanism of CO₂ capture by these biomaterials comprises of a series of carbonation-calcination reactions (CCR): calcium oxide (CaO) derived from eggshell or seashell reacts with CO₂ in the flue gas stream, leading to calcium carbonate (CaCO₃), which then undergoes calcination resulting in the release of a pure CO₂ stream for sequestration, and at the same time is regenerated into CaO as shown in **Figure 5** [24]. The pilot-scale demonstration of the concept has been reported for eggshell and oyster shell in the literature [24, 26–29]. The reactions are summarised as follows: carbonation ($\text{CaO} + \text{CO}_2 \rightarrow \text{CaCO}_3$) of the eggshell-derived CaO through reaction with CO₂ forms calcium carbonate (CaCO₃), while the calcination process ($\text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2$), regenerates the CaO bio-composite material, and liberate pure stream of CO₂ for sequestration. Sacia et al. [27] investigated CaO sorbents derived from chicken eggshell for CO₂ from coal-fired power plants. In the work, they discovered that the pre-treatment of the eggshell with acetic acid enhanced

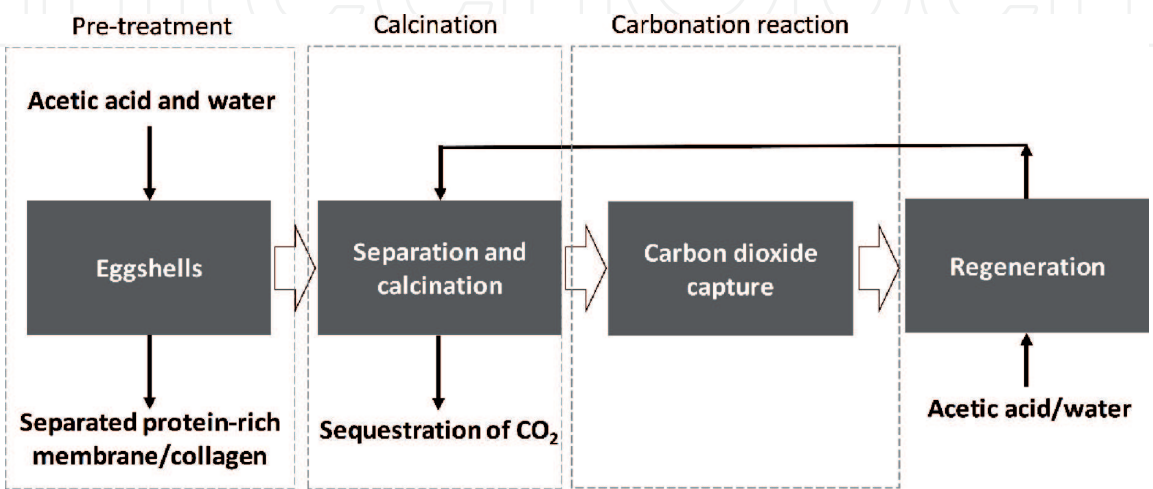


Figure 5.
Schematic of the eggshell or seashell carbonation – calcination processes for carbon dioxide capture.

and expanded the derived-sorbent material pore structure and surface area, which favoured CO₂ diffusion as mass transport is improved.

Figure 6 shows the effect of acetic acid concentration and treatment time on CO₂ capture over multiple cycles. It is clear that the acetic acid treated eggshell outperformed the untreated counterpart. On the other hand, derived CaO from eggshell treated with a low concentration of acetic acid exhibited better reactivity and CO₂ capture capacity than that treated with higher concentration. This can be attributed to the improved reactivity and porous surface structure within the bio-materials when treated with an optimised concentration of acetic acid [24, 26, 27].

Figure 6 also demonstrates that subjecting the eggshell or seashell to a higher strength acetic acid solution or for a longer treatment time could affect the pore structure, strength and stability of the derived CaO sorbent biomaterial. This is consistent with the result of the investigation reported by Sacia [17], on the use of eggshell for CO₂ capture. Hence, the observed decrease in the reactivity and CO₂ capture capacity under this condition. More also, the data shows that the derived sorbent from eggshell or seashell cannot be continuously regenerated over multiple cycles, as a result, fresh sorbent would be added as make-up during the process to sustain capture capacity (**Figure 6**). Depending on the acetic pre-treatment time, it has been reported that the CO₂ capture ranges from 70 to 80% in the first cycle, and gradually drop to about 40% in the fifth cycle [27].

Figure 7 shows simulated thermogravimetric analyser (TGA) results to prove CO₂ capture capacity of eggshell-derived sorbent using a typical flue gas stream (10% CO₂ for 60 min cycles at 700°C). The weight of the sample indicates reactivity, while the weight increase signifies carbonation due to CO₂ capture; the decrease represents the calcination process because of CO₂ liberation. It is clear that the CO₂ capture performance and reactivity gradually diminishes for multicycles over time.

The reactivity and CO₂ capture capacity of the eggshell or seashell derived CaO sorbent decline over time, so regeneration of sorbents in-situ is pivotal to maintaining CO₂ capture. The regeneration can be carried out using deionised water and acetic acid solutions [27]. The effect of regeneration of the eggshell derived CaO sorbent on CO₂ capture is shown in **Figure 8**. It is clear that regeneration with acetic acid is more effective than with water. Sacia [17] ascribed this observation to two factors. First, the use of acetic acid resulted in calcium acetate, which exhibited a higher molar volume than only Ca(OH)₂ formed when water is used. Also, the combination of water and acetic acid allows for a surface structure rearrangement due to the solubility of calcium acetate in water. It has been found that the use of

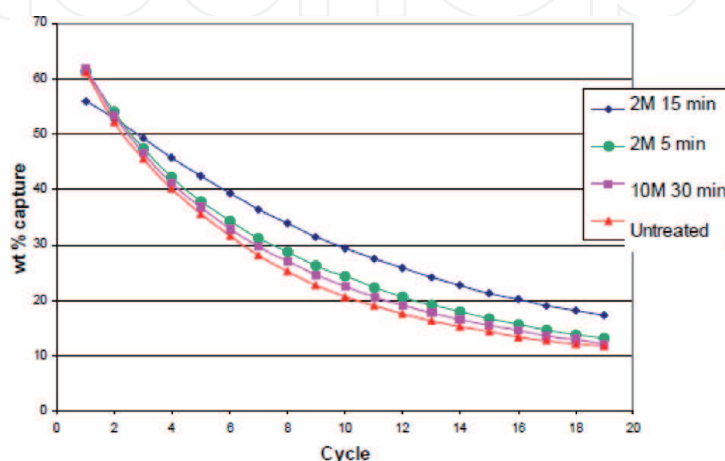


Figure 6. Effect of acetic acid and treatment time on weight per cent CO₂ capture using chicken eggshell [24].

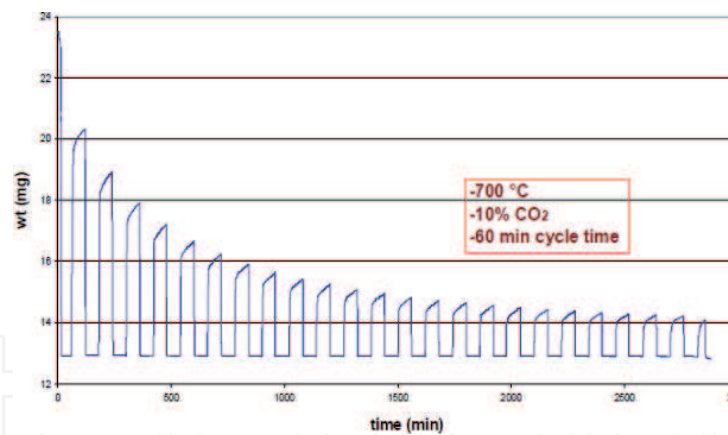


Figure 7.
Weight vs. time of eggshell-derived adsorbent for CO₂ capture using TGA [24].

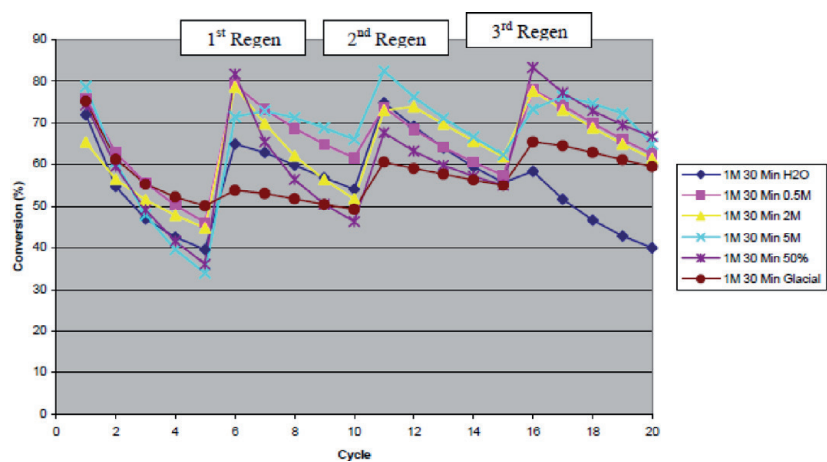


Figure 8.
Conversion vs. regenerations of eggshell derived CaO sorbent treated with a 1 M acetic acid for 30 min [17].

2 M acetic acid offers the best performance after multiple cycle regeneration in terms of reactivity and CO₂ capture [17, 24]. It can be observed that over three regenerations, all of the sorbent showed similar results trend.

In the investigation of Sacia et al. [27], it was found that regeneration restored the reactivity of the eggshell-derived CaO sorbent, and subsequently, CO₂ capture capacity in the range of 70–80% was achieved. The CO₂ capture capacity increased on average after successive regeneration, as can be seen in **Figure 8**. This suggests that periodic regeneration can effectively increase the reactivity of the spent eggshell or seashell-derived CaO sorbent. In another study by Banerjee et al. [30], it was reported that after four successive regenerations over multi-cycles usage, the carbon dioxide capture capacity of the eggshell-derived sorbent material decreased from 6824 mg CO₂/g to 1608 mg CO₂/g an average compared to the fresh material. This indicates that the eggshell-derived CaO sorbent biomaterial could hold about eight times its own weight of CO₂ from flue gas. Furthermore, Ma and Teng [31] investigated and reported the carbonation – calcination loop of CaO/CaCO₃ process for CO₂ capture using CaO derived sorbent from oyster shells. Though compared to reagent grade CaO from CaCO₃, the oyster shell derived CaO possess bigger crystallite size and lower specific surface area. It was reported that at 740°C carbonation temperature, the oyster shell-derived CaO sorbent in cyclic carbonation exhibited superior performance to the reagent-grade CaO obtained from CaCO₃. Therefore, utilising this waste biomaterial in CO₂ capture encourages the reuse of materials in the industries, which will reduce the risk, cost and energy associated with mining

limestone and dolomite for CaCO_3 and CaO , and subsequently offers economic and environmental benefits. However, these benefits will be significant if the system is scaled-up to industrial standards.

5. Conclusion


There are large tonnes of eggshells and seashells discarded in landfill annually from poultry and food industries. Most of the seashells and eggshells are piled up on the seashore and thus would cause risks to water resources and public health. The applications of these biomaterials in construction such as concrete and cement production, catalyst manufacture, adsorbent for wastewater treatment, source of calcium in animal feed, manufacture of hydroxyapatite biomaterial, and additive in plastic manufacture has been explored extensively in the literature. These biomaterials contain about 96% calcium carbonate mineralogical component from which calcium oxide can be produced through thermal treatment. The carbonation – calcination loop of CaO/CaCO_3 process has been investigated for CO_2 capture potentials. Herein, the application of eggshell and seashell derived- CaO sorbent in the capture of carbon dioxide from flue gas is reviewed. The utilisation of this waste shell offers economic as well as environmental benefits because they are abundant, renewable and cheap. The CaO sorbent derived from eggshell and seashell has demonstrated the potential for carbon dioxide capture. It was also found that pre-treatment and regeneration provide means of restoring reactivity and CO_2 capture capacity over multicyclic usage. Although this ensured sustainability and sorbent recyclability, the performance decreases ten cycles after regeneration. The future outlook will be to improve the carbon dioxide capture capacity and thermal stability of these biomaterials over multicycles operations.

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