CARBON CAPTURE USING WASTES: A REVIEW

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Abstract

Potential key strategies for the management of anthropogenic CO_2 emissions include mineral carbonation and storage in oil wells and in the oceans. In Europe, a large-scale demonstration of carbon capture and storage (CCS) has recently been given the go-ahead, and the application of mineral carbonation technology (MCT) to serpentine and olive-type minerals. Although less controversial in its approach, MCT involves intensive pre-treatment of the mineral feedstock, and a consequent high sequestration cost USD100-120/tonne CO_2 treated.

Mineralisation by carbonation is reliant upon the long-term storage of CO_2 in thermodynamically stable and environmentally benign carbonate-based reaction products that are persistent over geological-timescales. The use of solid industrial process wastes for storing carbon (via waste carbonation technology, WCT) may provide a shorter-term gain, as the industrialisation of CO_2 mitigation technologies takes place.

With WCT, CO_2 is reacted with alkaline waste residues, to both risk-manage a high pH, and utilise waste CO_2 gas, can be used as a pre-treatment prior to landfilling, facilitate valorisation and production of new materials.

The present work examines the current status of waste carbonation and investigates the utilisation of seven 'common' alkaline industrial residues showing that they have potential to sequestrate 1Gtonne of CO_2 worldwide. The projected average cost of USD38-95/tonne of CO_2, is competitive with landfill and projected carbon taxes. If WCT is more widely commercially developed an option for the management of significant amounts of carbon could become more quickly established.

Keywords: CO_2 emissions; Carbon utilisation; Mineral sequestration; Waste carbonation; Accelerated carbonation

Introduction

Strategies to control anthropogenic CO_2 emissions involve meeting national-term targets, improvement in energy efficiency, alternative fuels, 'greener' industrial processes and the adoption of targeted tax schemes and integrated environmental policies, such as ICCP [1-3].

By way of example of tax-based initiatives, Alberta, Canada, has a carbon offset tax where large emitters are charged CD15 (€14) per tonne of CO_2 with proposals to increase this to CD40 (€37) in order to generate more revenue [4]; on the other hand in British Columbia, the carbon tax system is neutral.

The progress towards a more eco-friendly society is slow, mainly due to government inertia, industrial resistance, the investment required for energy efficiency, coupled with the slow integration of renewables into the energy supply mix.

The potential of carbon capture and storage (CCS) for managing anthropogenic CO_2 rests on the yield that can be achieved at a reasonable cost. The routes to CCS involve the capture, transportation and subsequent storage of CO_2 in appropriate geological and ocean sites, or the storage as carbonates/bicarbonates via mineral carbonation [1].

However, it is not widely recognised that high volume alkaline waste streams can sequester CO_2. The chemical similarity of wastes to natural silicates is often overlooked and if waste carbonation can be widely achieved, then there is potential to manage significant amounts of CO_2 by this method.

The current work reviews mineral carbonation technology (MCT) and compares its potential with the other sequestration methods. In addition, the potential for waste carbonation technology (WCT) annual production and maximum sequestration capacity of seven alkaline wastes is estimated. Finally, an appraisal of companies applying mineral and waste carbonation is made via a comparison of their process routes.

Carbon Capture

Carbon capture and storage is projected to consume up to 60% of overall anthropogenic carbon emissions [6]. The process of CCS involves three steps:

1) the removal and capture of CO_2 from specific locations,
2) transportation to proper storage sites and
3) CO_2 long term storage [4].

Large quantities of carbon are emitted by coal and gas plants, where three capture technologies are used: Post-combustion capture, Pre-combustion capture and Oxy-combustion capture [5]. After transportation, the captured CO_2 can be stored in appropriate geological and ocean sites [6]. In Europe, a large-scale demonstration known as the 'White Rose' CCS project centred upon the Drax coal-fired power station in the UK has been given the go-ahead. This CCS project will see CO_2 transported via a pipeline for permanent storage in the North Sea in depleted oil wells [7].

Alternatively, mineral carbonation involves the production and disposal of environmentally benign minerals that are stable over geological timescales. Potential feedstocks for MCT comprise minerals comprising calcium and magnesium ions, including wollastonite, basalt, olivine and serpentine. The potential storage capacity exceeds 10,000Gtonnes at an average cost of USD100-120/tonne CO_2 [8-10].

Table 1 overviews the available carbon capture and storage technologies, their advantages/disadvantages, estimated capacity and likely costs[1].

Table 1: Overview of CCS technologies

Technology	Advantages	Disadvantages	Capacity[a] (Gt)	Cost[b] (USD)
Geological storage [11,12]	• Established technique • Low storage cost • Positive perception	• High risk of leakage • Requires continuous monitoring • Lack of appropriate sites	1800	5
Ocean storage [13]	• High capacity • Global availability • No monitoring requirements	• High environmental risk • Higher cost than geo-storage • Negative/hostile perception	10000	18
Mineral carbonation [8-10]	• Environmentally benign • Exothermal reactions • Wide availability of feedstocks	• High costs • Pre-treatment • Unsecure future potential	>10000	120

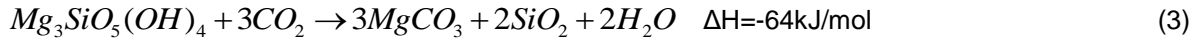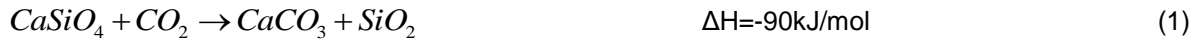*a: maximum estimated capacity, b: average sequestration cost*

Mineral Carbonation Technology (MCT)

In countries with large reserves of suitable rock resources i.e. Finland, Australia, USA and Canada, the potential for future carbon mitigation depends upon future research advancement.

Mineral carbonation mechanisms

Mineral carbonation involves the formation of stable carbonates and bicarbonates which are chemically equivalent to calcite, magnesite and dolomite [9]. Silicates naturally abundant in rocks and mining ores can be used as feed stocks e.g. peridotite, serpentine, olivine, wollastonite, gabbro and basalt. After appropriate treatment, carbonated products may be used as paper fillers and coating materials [14,15]. The following equations show the principal reactions occurring for calcium and magnesium silicates; wollastonite (1), olivine (2) and serpentine (3) [16].

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[1] Costs expressed in US Dollars. At current exchanges rates x0.77 for Euros, x0.61 for British Pounds Sterling

$$CaSiO_4 + CO_2 \rightarrow CaCO_3 + SiO_2 \qquad\qquad \Delta H = -90kJ/mol \qquad\qquad (1)$$

$$Mg_2SiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2 \qquad\qquad \Delta H = -89kJ/mol \qquad\qquad (2)$$

$$Mg_3SiO_5(OH)_4 + 3CO_2 \rightarrow 3MgCO_3 + 2SiO_2 + 2H_2O \quad \Delta H = -64kJ/mol \qquad\qquad (3)$$

All the reactions are exothermic and no energy is added to the system. However, slow kinetics, low yield under mild conditions, heat pre-treatment of silicates adversely impact on the cost of the processing [17]. The cost of transportation accounts for 77-94% of the total costs involved in MCT [18].

During the mining of silicate ores, the cost of grinding, crushing and milling must also be considered. It is estimated that, in order to be applied at industrial scale, the maximum cost of mineral carbonation should range between USD20 and USD30 per tonne of CO_2 [10]. Indicative costs of wollastonite and serpentine were estimated at USD200 and USD126-185 respectively [10,19].

Carbonation of mineral residues

Mineral sequestration can be achieved close to emitting plants by transporting the raw materials to site (ex-situ MCT), or via direct diffusion of CO_2 into rocks containing calcium and magnesium minerals (in-situ MCT).

The ex-situ carbonation of feed-stock occurs in three stages [20]:
1. Thermal and mechanical pre-treatment of the mineral
2. Reaction of CO_2 with ions of Ca^{2+} and Mg^{2+} accompanied by energy release
3. Disposal/reuse of the final carbonated products

One of the biggest challenges of ex-situ processing is the distance that often exists between CO_2 emitters and the location of mineral sources, as invariably they do not coincide. The need for handling large quantities of CO_2 and mineral silicates impacts negatively on the costs of material transportation.

Therefore, the location of bespoke carbonation plants should be as close to the natural mineral resources as possible. Picot *et al.* [21] investigated candidate locations around the world, which combine coal-fired plants emitting more than 1Mtonne of CO_2, with available minerals within a distance of 300km. The identified regions are located at Botswana, China, South Africa, Russia, Kazakhstan, and in some isolated places in Northern Europe, USA and Australia. Unfortunately, there is lack of mineral availability in industrial regions with vast carbon emissions.

Although ex-situ processing is a preferred option, the slow kinetics (unless finely ground feedstock is used), the pre-treatment of feedstock, and the elevated temperature and pressures required, have focussed attention towards in-situ treatments [22].

CarbFix is an ex-situ MCT research project, using CO_2 emitted from a geothermal plant in Iceland. The mineral 'host' is basalt, a silicate rich in calcium, magnesium and ferrite ions [23]. The installation consists of a CO_2 separation plant and injection facility, with a 3km pipeline for the carbon transportation and monitoring systems. The current overall capacity of CO_2 injection is estimated at 2.2ktonnes CO_2/per year [24] and it can reach 60ktonnes at overall cost of USD66/tonne of CO_2 sequestrated [25].

The Samail ophiolite in Oman is composed of peridotite, which naturally reacts with an estimated 0.1Mtonnes of CO_2 per year. The potential for reaction with carbon dioxide may reach 1Gtonne, if accelerated carbonation were to be applied at elevated temperature and pressure [26]. Other relevant research has been made at Mount Keith Nickel Mine in Canada, Linnajavri in Norway and various locations in Italy [27-29].

Routes for mineral carbonation

Carbonation routes (for mineral carbonation) are classified into two main categories: direct and indirect. During the direct route, silicates react with gaseous or aqueous CO_2 in a single step, without the extraction of calcium or magnesium ions [30]. Via the indirect route involves more than one step, including ion extraction. Table 2 summarises the available carbonation routes [31].

Table 2: Routes for direct and indirect carbonation

Route	Description	Advantages	Disadvantages	Cost US$/t
Gas-solid [30-32].	Particulate metal oxides react directly with CO_2,	• Simplicity • Production of steam and electricity • Mining integration potential	• Slow reaction • Thermodynamic constraints • Industrially unfeasible	
Aqueous [16, 30,33-40].	Mixing (minerals) with a liquid medium such as bicarbonate/salt before the reaction with CO_2.	• Conversion rate	• High energy requirements • Requires extra chemicals • No recycling • High costs	50-100
HCl extraction [9,16, 41].	Hydrochloric acid used to extract ions of Ca+ and Mg+ from a silicate matrix	• Recoverable reactants	• High energy requirements • Expensive	>188
Acetic acid extraction [22, 47-49].	Similarly to HCl extraction, the use of acetic acid is used	• Less energy intensive • Economical	• Higher recoverability needs to be achieved	78
Molten Salt [40].	Molten salt as a less energy intensive sorbent than HCl.	• Less energy intensive than HCl	• Extremely corrosive • Undesirable by-products	
Ammonia extraction [41, 42]	Ammonium salts used to promote the CO_2-silicate reaction	• High purity carbonates • Good kinetics • Recoverable reactants	• High current costs • More research needs to be done	

Waste Carbonation Technology (WCT)

As an alternative to natural minerals, solid wastes can be reacted with carbon dioxide [43]. This process is known as waste carbonation technology (WCT) and the most significant advantage, compared to MCT, is that pre-treatment is rarely necessary and the general proximity of these residues to point sources of anthropogenic CO_2 [16].

Wastes such as those from bauxite processing, cement manufacture, coal combustion, iron and steel manufacture and the incineration of municipal waste, are composed of a significant proportion of calcium and magnesium. These wastes are widespread around the world. A number of the high volume waste streams with potential for WCT are identified in Table 3:

Table 3: Characterisation of alkaline wastes according to European Directive 2000/532/EC [66]

Code	Main Category	Code	Sub-category	Waste
01	Wastes resulting from exploration, mining, dressing and further treatment of minerals and quarry	01 03	Wastes from further physical and chemical processing of metalliferous minerals	Bauxite residues
10	Inorganic wastes from thermal processes	10 01	Wastes from power stations and other combustion plants	Coal fly ash
		10 02	Wastes from the iron and steel industry	Steel slags
		10 13	Wastes from manufacture of cement, lime and plaster and articles and products made from them	Cement kiln dust
19	Wastes from waste treatment facilities, off-site waste water treatment plants and the water industry	19 01	Wastes from incineration or pyrolysis of waste	MSWI bottom ash
				MSWI fly ash

Bauxite residue (Red Mud)

'Red mud' is the solid waste produced by the Bayer process when alumina is extracted from bauxite ores. The transport, re use and disposal of red mud is extremely difficult due to its high alkalinity (pH>13) and elevated sodium concentration [44].

The carbonation of bauxite residues can be achieved by neutralisation involving the reaction of aqueous solutions of red mud with carbon dioxide [45-46]. The carbonation capacity of the process is estimated at 53kg of CO_2/tonne of red mud [47].

Cement kiln dusts

Cement kiln dust (CKD) is formed during the manufacture of cement clinker at high temperature (around 1400°C) and it is a mixture of fine particles, unburned and part-burned raw materials and contaminants. Many cement manufacturing processes involve CKD recycling. However, the degree of recycling depends on various parameters including dust composition, particularly alkali content, and the standards that need to be met [48-50].

Historically, large amounts of CKD have been landfilled and could be a potential resource for CO_2 sequestration. Depending on the type of cement kiln, 250-300kg of cement kiln dust is formed per tonne of cement.

Pulverised fuel ash

Pulverised fuel ash (PFA) is produced from the burning of coal. PFA is classified as either Class C or Class F, differentiating between those with high and low calcium contents, respectively [51,52]. Where high CaO containing PFA is available, there is great potential to capture CO_2 with minimal handling and transport costs [53].

MSWI bottom ash and APCr

The incineration of municipal solid wastes produces bottom (grate) ash and fly ash. Depending upon the composition, the two residues may or may not be combined. Air pollution control residues (APCr) are also produced as a result of flue gas treatment. APCr is sometimes combined with the fly ash.

Bottom ash former comprises 80% of the total incineration residue and is composed of ash powder along with glass and metal fragments. Bottom ashes are invariably used in road pavement construction, glass and ceramics, and in agriculture and waste water treatment [54, 55]. The presence of heavy metals is a concern, and an accelerated carbonation treatment-step has been shown to be a promising management option [56-59].

Fly ash is a fine grained airborne material which is removed via electrostatic precipitators. Typically, the heavy metal loading is higher than bottom ashes, which is often the determining factor whether it can be combined with the bottom ash.

APCr is typically a mixture of lime and activated carbon, that is injected into the flue gas to remove volatile heavy metals and neutralise acidic gasses [60].

Steelmaking slags

Steel manufacture generates slag, comprised of calcium, iron, silicon, aluminium, magnesium and manganese oxides. These residues are highly alkaline (pH 12) and when they react with carbon dioxide, stable products comprising carbonates are produced [41, 61]. Moreover, slag production is around 400Mtonnes worldwide [62], with 45Mtonnes in the EU [63] and 5.2Mtonnes in the UK [64]. Their carbon-uptake potential is promising whilst transportation costs are negligible, assuming the CO_2 can be captured.

Carbon Sequestration Potential

Based upon available data (between 2007 and 2011), it is possible to sequester up to 1Gtonne of CO_2 by carbonating 6 alkaline residues found widespread around the world (Table 4).

WCT ensures the utilisation of both CO_2 and solid waste residues (arising from the same plant) without additional transportation and monitoring costs. The average cost of WCT is estimated at USD67/tonne of CO_2, which is considerably lower than MCT. Furthermore, process operating conditions including temperature and pressure are generally lower. However, the complexity of waste systems and the frequent presence of heavy metals and/or radioactive elements in some residues necessitate pre-treatment, which may increase the cost of handling and processing these wastes. Finally, in many countries the regulation of waste does not lend itself to treatment and valorisation at this time, and this is an impediment to utilising waste for the sequestration of CO_2 gas.

Table 4: Alkaline residues suitable for waste carbonation

Waste	Annual world production in million tonnes	Sequestration capacity in kg CO_2/tonne of waste	Max potential CO_2 utilised[c] (MTonnes)	Min potential CO_2 utilised (MTonnes)
Bauxite [44-47, 65-68]	120	53	6.36	6.36
CKD[a] [50, 70-73]	990	15-115	113.85	14.85
PFA [51-53, 74-75]	600	8-264	158.4	4.8
MSWI bottom ash[b,c] [54-60, 76-80]	80	247-475	38	19.76
MSWI fly ash[b] [57-59, 76-78, 80-81]	20	30-120	2.4	0.6
Steel slags [61-64, 81-84, 85-88]	400	227-300	120	90.8

a: estimated based upon 0.25-0.30 tonnes of kiln dust per tonne of cement. b: annual production is estimated at 100Mt. Assumed 80% as bottom ash and 20% as fly ash. c: estimated using the highest and lowest sequestration capacities identified

Commercial Applications

Several companies are already using mineral and waste carbonation to produce valuable products, including aggregates and secondary building materials. Table 5 presents data on these commercial/near commercial companies and their processes, using data/ metrics (CO_2 utilised, products value, energy penalty) taken from two reports from Carbon Sequestration Leadership Forum [89-90], companies web-sites and other information sources.

Summary

There is great potential to manage carbon emissions to protect the environment. Technologies capable of doing this are in development, and cost reduction remains a priority.

The application of Carbon Capture and Storage (CCS) and Mineral Carbonation Technology (MCT), have the greatest potential to mitigate the environmental impacts of large volumes of CO_2. However, in the shorter-term the use of waste may provide significant gains as the often waste and CO_2 are emitted by the same process, are generated near to industrial centres, or close to where the carbonated products might be used, or disposed of.

Many industrial residues have the potential to act as capture media, including wastes from bauxite extraction, cement manufacture, coal combustion, municipal waste incineration, and steelmaking. Applying Waste Carbon Technology (WCT) to these six residues alone has the potential to sequester up to 1Gtonne of CO_2 each year.

Several companies have been established tor are developing WCT processes. Currently, there are two, Carbon8 and Recmix, who are operating commercially to produce construction materials from wastes treated using WCT. Several others are in the process of bringing new technologies to the market.

Table 5: Summary of ventures applying mineral and waste carbonation

Name	Activities and Technology	Raw Materials	Key Process Steps	Output materials
Commercially Active				
Carbon8	Carbon8 produces carbon negative construction aggregates from hazardous wastes including MSWI ashes, CKD, and steel slags. [91-92]	1. Alkaline wastes 2. CO_2 3. Reagents	1. Pre-treatment of waste 2. Blending reagents 3. Pelletising 4. Aggregate production	1. Aggregates
Recmix	Stainless steel-slag is carbonated to produce aggregates, fillers for concrete and asphalt and blocks. [93]	1. Stainless steel slag 2. CO_2	1. Granulation 2. Mixing 3. Carbonation 4. Shaping and curing	1. Aggregates 2. Concrete/asphalt fillers 3. Artificial aggregates
Commercialising				
Solidia	Production of low-energy cement from blended calcium silicates, and hardening using CO_2. [96]	1. Solidia cement 2. CO_2	1. Kiln treatment 2. Mixing with water 3. Curing with CO_2	1. Solidia concrete
Pilot Scale				
Calix	Enhanced calcination technology offering the potential for efficient mineral sequestration of carbon. Sorbent is regenerated and recycled [97]	1. Limestone/Dolomite/ Magnesite/Kaolin/Gypsym/ Diatomite/Bauxite	1. Minerals Grinding 2. Minerals pre-treatment 3. Calcination 4. Separation of products	1. Steam 2. Magnesium Oxide 3. Semidolime 4. Metakaolin 5. Organic Phosphate
Carbon Engineering	Direct air capture (DEC) and production of high quality carbon dioxide for enhanced oil recovery, algae growth in industrial-scale ponds, and synthesis of liquid hydrocarbons. [98-99]	1. Atmospheric air 2. Energy 3. Chemicals	1. Air capture 2. Separation of CO_2 3. Regeneration cycle	1. Pure CO_2
CO_2 Solutions	Utilises the enzyme catalyst carbonic anhydrase for removing carbon from flue gases at emitting plants. [100-102]	1. Flue gas 2. Low energy solvents	1. CO_2 Absorption 2. Enzyme catalysis 3. Thermal heating	1. Pure CO_2
ICS	The novelty of ICS approach is that integrates the flue gasses capture step with the carbonation of silicate minerals. [103-104]	1. Flue gas 2. Mineral silicates	1. Capture of flue gas 2. Mineral preparation 3. Carbonation 4. Storage	1. Mineral Carbonates
Laboratory Scale				
CCC	Mineralisation of magnesium and calcium silicates to carbonated products, along with generation of zero carbon electricity [105-106]	1. Silicate minerals/wastes 2. Flue gas	1a. Digestion of the process fluids 1b. Carbon capture & power generation 2. Carbonation 3. Fluid regeneration	1. Silica by-products 2. Carbon free electricity 3. Carbonate materials

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