



Mineralization Technology for Carbon Capture, Utilization, and Storage

Colin D. Hills^{1*}, Nimisha Tripathi¹ and Paula J. Carey^{1,2}

¹ Faculty of Engineering Science, University of Greenwich, Chatham, United Kingdom, ² Carbon8 Systems Ltd., Chatham, United Kingdom

OPEN ACCESS

Edited by:

Rafael Mattos Dos Santos,
University of Guelph, Canada

Reviewed by:

Hannu-Petteri Mattila,
Independent Researcher, Parainen,
Finland

Marius Bodor,
"Dunarea de Jos" University of Galati,
Romania

*Correspondence:

Colin D. Hills
c.d.hills@gre.ac.uk;
hc34@gre.ac.uk

Specialty section:

This article was submitted to
Carbon Capture, Storage,
and Utilization,
a section of the journal
Frontiers in Energy Research

Received: 17 April 2020

Accepted: 10 June 2020

Published: 14 July 2020

Citation:

Hills CD, Tripathi N and Carey PJ
(2020) Mineralization Technology
for Carbon Capture, Utilization,
and Storage.
Front. Energy Res. 8:142.
doi: 10.3389/fenrg.2020.00142

Carbon capture, utilization, and storage (CCUS) is a technology approach to the management of anthropogenic carbon dioxide gas emissions to the atmosphere. By injecting CO₂ into host rocks, or by employing an *ex situ* application step, geological formations can react with and store huge volumes of CO₂ as carbonate minerals. An alternative mineral feedstock material is the Gt of industrial process wastes that are often disposed to landfill. By applying an accelerated carbonation step to solid waste, there is potential to sequester meaningful quantities of CO₂ in carbonate-cemented products that have reuse potential. The manufacture of carbonated aggregates is commercially established in Europe, and recent advances in technology include a mobile plant that directly utilizes flue-gas derived CO₂ in the mineralisation process. The present work discusses the basis for mineralization in geologically derived minerals and industrial wastes, with a focus being on the manufacture of products with value. An assessment of mineralized construction aggregates suggests that carbon capture, utilization, and storage technology can manage significant quantities of this CO₂.

Keywords: carbon capture, utilization and storage, mineralization of carbon dioxide, calcium carbonate, flue gas, accelerated carbonation, accelerated weathering

INTRODUCTION

As carbon dioxide (CO₂) in the atmosphere increases due to anthropogenic activities, natural ecosystems are losing their ability to absorb this greenhouse gas (GHG). The major contributor of CO₂ to the atmosphere is generated from burning fossil fuels, biofuels, and wood. By way of context, a large thermal power station can easily emit more than 20 Mt of CO₂ each year (Armstrong et al., 2019).

The IPCC (2014) predicts that if GHG emissions continue, the global temperature will rise between 3.7 and 4.8°C by 2100. To reach the limit of 1.5°C, CO₂ emissions need to be reduced by 45% from 2010 levels by 2030 and net-zero emissions by 2050. The special IPCC (2018) report gives a revised target of <1.5°C temperature rise and emphasizes the need for further emission reductions¹ (IPCC, 2018).

¹<https://www.ipcc.ch/sr15>

The prospect of mitigating anthropogenic CO₂ emissions by carbon capture and storage (CCS) is attractive. By injecting CO₂, as a supercritical fluid, into porous rock strata in the geosphere, it is predicted that large amounts (Gt) of carbon can be managed this way. Injection into the deep ocean water column (>1,000 m), where CO₂ will remain dissolved for centuries, is also being suggested (UNESCO-IOC/SCOR, 2007). However, these processes are not yet commercially or technically viable, and alternative options for sequestering CO₂ are required. Discussed later is an approach based on nature.

REACTION OF CARBON DIOXIDE WITH MINERAL SYSTEMS

Mineralization is nature's way to sequester CO₂, but it is a slow process. The hydrolysis of CO₂ in moist air or water is a major driver of rock chemical weathering. The geological record indicates that tectonic forcing exposes large rock masses to the atmosphere and subsequent weathering occurs at a rate that can markedly reduce atmospheric CO₂ levels (Raymo and Ruddiman, 1992). Notwithstanding that natural weathering can remove some 30 Gt of CO₂ from the atmosphere each century, it is this natural process that has the potential to be accelerated and, therefore, industrialized. In this way, anthropogenic CO₂ can combine with a reactive substrate to form a mineralized product—carbonate salts, for ultimate disposal or conversion into valorized products. Mineralized products have the potential to be beneficially utilized in quantities that can ultimately reduce the effects of global warming. The common rock types and their reaction with CO₂ gas are discussed by Penner (2004) and Zevenhoven (2004).

CARBON CAPTURE, UTILIZATION, AND STORAGE

The concept of managed mineralization offers an environmentally sustainable opportunity that can work in synergy with carbon capture and storage. By incorporating a “utilization” option within a “storage” concept, captured CO₂ can be used as a feedstock for making products, products in which CO₂ gas is sequestered permanently. This unison is known as carbon capture, utilization, and storage (CCUS).

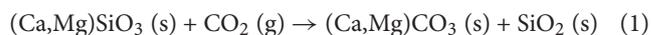
It is not widely appreciated that CO₂ gas has been used for centuries as a feedstock in industrial processes. Uses extend from carbonating beverages (18th Century) to the application of supercritical CO₂ in the manufacture of instant coffee, pharmaceuticals (20th Century), or construction materials (21st Century).

CO₂ gas-enhanced curing chambers have been utilized for conditioning and accelerating the hardening of cementitious materials since the 19th century (e.g., Rowland, 1870), as alkaline materials tend to be very CO₂ reactive. The tendency of minerals to react with CO₂ under the right conditions can be harnessed for CCUS, using both *in situ* and *ex situ* processing methods. By way of example of the former, Kelemen et al. (2019) report the

application of CO₂-rich fluid rather than natural water, in the *in situ* treatment of peridotite. In this approach, the dissolution of peridotite (and hence its carbonate-ability) can be increased by five orders of magnitude.

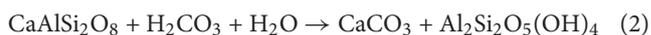
Rock masses found in the geosphere that are *materially* suitable for managed carbonation are ubiquitous, and the carbonated reaction products are stable in the geosphere. The National Academies of Sciences Engineering Medicine [NASEM] (2019) states that significant health and environmental benefits can arise from carbon mineralization, as the formation of carbonate is the safest carbon “storage” mechanism (Zhang and DePaolo, 2017).

The application of *in situ* mineralization technology has been described as mineral trapping or managed weathering and is simply an accelerated weathering step. Silicate rocks are the most suitable host rock formations for mineralized carbon, with the rate of carbonate mineral production being kinetically controlled. Huijgen and Comans (2003) and Zevenhoven and Fagerlund (2010) have reviewed the mineralization of CO₂. As mineral carbonation is an analog of natural weathering, the reaction between CO₂ and suitable silicate rocks can be summarized as (Equation 1):

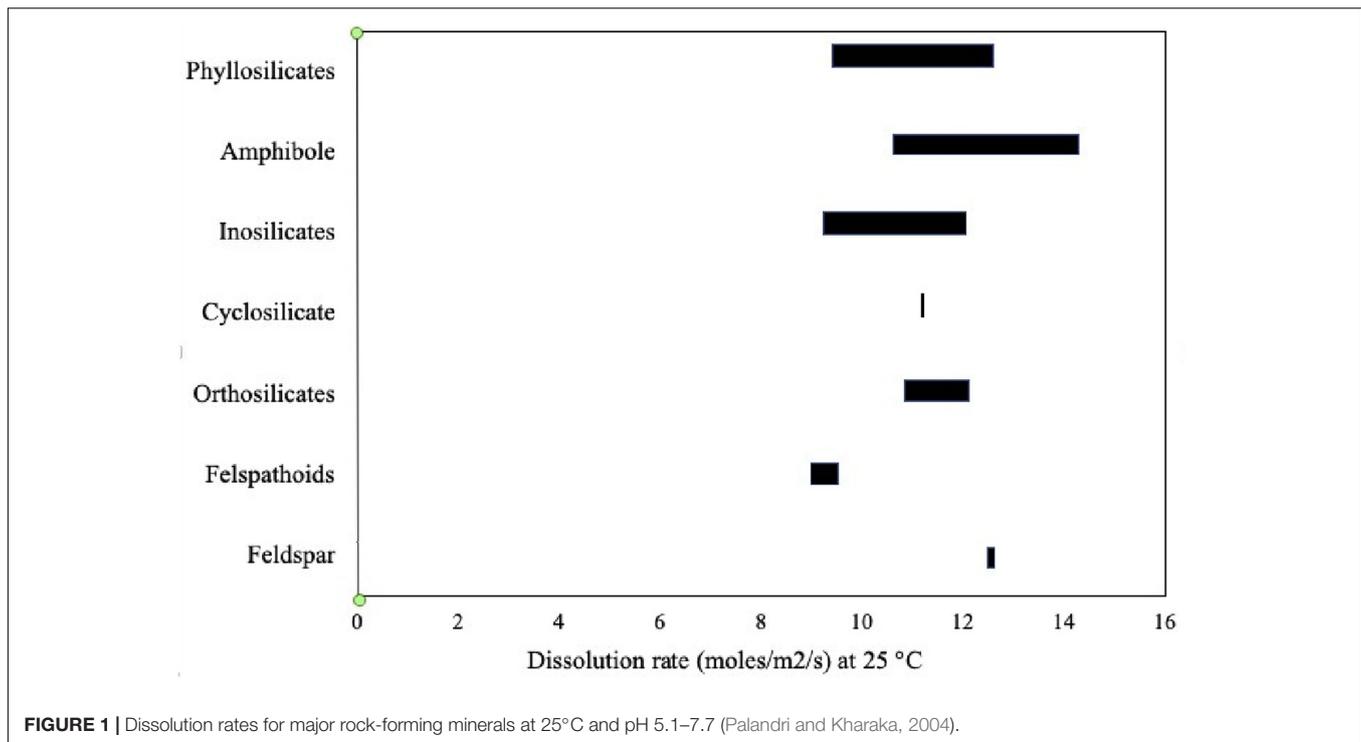


If we consider the course of natural weathering of olivine (Mg₂SiO₄) or serpentine [Mg₃Si₂O₅(OH)₄], it is the dissolution and ionization of CO₂ in the rain or groundwater that produces carbonic acid. This, in turn, dissociates into H⁺ and HCO₃⁻ or bicarbonate. As bicarbonate is conjugate acidic, it can chemically degrade susceptible minerals, liberating calcium and magnesium, which then bind with the bicarbonate to form solid carbonates. Beaulieu et al. (2012) estimate that 300 Mt of CO₂ gas is removed from the atmosphere via weathering and carbonate formation each year.

Potential geological storage “reservoirs” for mineralized CO₂ involve igneous (flood basalts, pillow lavas, and ultramafic rocks, such as peridotite), metamorphic (serpentinites and ophiolites), and perhaps, surprisingly, sedimentary rock formations, such as glauconitic and hematite-cemented sandstones. In respect of the latter, hematite has the potential to form siderite (FeCO₃) or ankerite (CaCO₃, MgCO₃, and FeCO₃), when Fe³⁺ is reduced to Fe²⁺. Arkosic sandstones, containing feldspar, are also a potential target host rock formation. Plagioclase (anorthite) found in igneous rocks and arkose may be a mineral suitable for carbonation; its reaction with CO₂ gas is summarized below (Equation 2):



Of particular interest are flood basalts with high porosity and a linked vesicular structure. These potential host rocks extend to thousands of cubic kilometers but can be geographically isolated, as are pillow lavas. Where basalt formations are saline, stable carbonates, including calcite, magnesite, and siderite, may be expected to form on contact with H₂CO₃⁻. Groundwater permeation through ultramafic rocks, such as dunite, is



accompanied by the dissolution of alkali and precipitation of carbonate (Canovas et al., 2017; Vankeuren et al., 2019).

The reaction rate of rock-forming minerals varies considerably, with wollastonite being one of the “fastest” and, hence, most reactive to CO₂. The dissolution of some common minerals at 25 and 180°C is discussed by National Academies of Sciences Engineering Medicine [NASEM] (2019). It should be noted that at higher temperatures, the rate of dissolution of minerals increases. By way of example, albite dissolution increases from an approximate log dissolution rate of between –11.5 and –12.5 at 25°C to –7.6 and –8.5 at 180°C. For olivine, a similar trend is observed, as the log dissolution rate increases from –8.2 and –10.1 to –4.2 and –5.7, at 25 and 180°C, respectively. The effect of small changes in pH can also be significant as exemplified for anorthite at 25°C, when the log dissolution of this phase decreases from –11.11 to –10.82, between pH of 8.1 and 8.4, respectively. **Figure 1** gives the solubility of different rock-forming mineral groups at 25°C between pH 5.1 and 7.7 (Palandri and Kharaka, 2004). It should be noted that not all the mineral groups shown are suitable for processing by carbonation.

The injection of CO₂ into the vicinity of active spreading ridges, where new oceanic crust is being formed, has been suggested as suitable host rocks, as they are characterized by saline hydraulic gradients (O’Connor et al., 2005). Seawater flows into pelagic sediments towards the spreading ridge itself, and injection of CO₂ would promote the formation of Mg and Ca carbonates. O’Connor et al. (2005) estimate that there is a capacity to store 8–40,000 Gt of CO₂ worldwide in the vicinity of spreading ridges. Olajire (2013) discusses several *in situ*

mineralization projects in basic and ultrabasic rock formations, including into basalts in Iceland (Carbfix, 2020).

The role of heterogeneities in potential host rock formations is of importance. Peuble et al. (2018) discussed the role of discontinuities in ultramafic rocks and their effect on chemical gradients, along preferential fluid pathways, with carbonate formation orientated normal to the hydraulic gradient.

Wherever suitable rock formations exist, their physical and chemical characteristics, including interlinked void space, pore solution chemistry, and temperature are key considerations for suitability for large-scale mineralization. There are several relevant reviews and reports on CO₂ mineralization, including IPCC (2005), Renforth et al. (2011), Romanov et al. (2015), and Kelemen et al. (2019). A further potential significant consideration is the role of iron-oxidizing bacteria on the degradation of silicate rocks, which is described by Daval (2018). This study reports the biologically induced formation of a passivating Fe³⁺-Si-rich layer on the surface of silicate minerals found in basic and ultrabasic rocks can, for example, reduce the dissolution rate of olivine by two orders of magnitude.

As such, one of the issues to be faced is the reaction yield, that is, how much CO₂ is mineralized per unit of mass of target mineral. Rarely is a target mineral reacted with CO₂ as predicted on stoichiometry grounds, meaning that in *ex situ* applications, high temperatures and pressures and the use of chemical reagents are required to achieve an acceptable yield in a short time. This will inevitably have an impact on cost, as the amount of target mineral required to trap 1 t of CO₂ (the R_{CO₂}) already varies widely; for example, the feedstock requirement for olivine, wollastonite, basalt, and magnetite is 1.6, 2.6, 4.9, and 5.3 t, respectively (Penner, 2004; Zevenhoven,

TABLE 1 | Common minerals and their reactivity with carbon dioxide.

Mineral	Formula	Reaction pathway	Potential CO ₂ uptake (% w/w total weight)
Olivine (Fosterite)	Mg ₂ SiO ₄	Mg ₂ SiO ₄ + 2H ₂ CO ₃ → 2MgCO ₃ + H ₄ SiO ₄	63
Serpentine polytype	Mg ₃ Si ₂ O ₅ (OH) ₄	Mg ₃ Si ₂ O ₅ (OH) ₄ + 3H ₂ CO ₃ → 3MgCO ₃ + 2H ₄ SiO ₄ + H ₂ O	48
Portlandite	Ca(OH) ₂	Ca(OH) ₂ + H ₂ CO ₃ → CaCO ₃ + 2H ₂ O	59
Brucite	Mg(OH) ₂	Mg(OH) ₂ + H ₂ CO ₃ → MgCO ₃ + 2H ₂ O	76
Larnite	Ca ₂ SiO ₄	Ca ₂ SiO ₄ + 2H ₂ CO ₃ → 2CaCO ₃ + H ₄ SiO ₄	67
Anorthite	CaAl ₂ Si ₂ O ₈	CaAl ₂ Si ₂ O ₈ + H ₂ CO ₃ + H ₂ O → CaCO ₃ + Al ₂ Si ₂ O ₅ (OH) ₄	16
Jennite	Ca ₉ Si ₆ O ₁₆ (OH) ₂ ·4H ₂ O	Ca _{1.67} SiO _{1.57} (OH) _{4.2} + 1.67H ₂ CO ₃ → 1.67CaCO ₃ + H ₄ SiO ₄ + 1.77H ₂ O	47
Rankinite	Ca ₃ Si ₂ O ₇	Ca ₃ Si ₂ O ₇ + 3H ₂ CO ₃ + H ₂ O → 3CaCO ₃ + 2H ₄ SiO ₄	38
Akermanite	Ca ₂ MgSi ₂ O ₇	Ca ₂ MgSi ₂ O ₇ + 3H ₂ CO ₃ + H ₂ O → 2CaCO ₃ + MgCO ₃ + 2H ₄ SiO ₄	48
Wollastonite	CaSiO ₃	CaSiO ₃ + H ₂ CO ₃ + H ₂ O → CaCO ₃ + H ₄ SiO ₄	38
Tobermorite	Ca ₉ Si ₆ O ₁₆ (OH) ₂ ·4H ₂ O	Ca _{0.83} SiO _{1.53} (OH) _{2.6} + 0.83H ₂ CO ₃ → 0.83CaCO ₃ + H ₄ SiO ₄ + 0.13H ₂ O	39
Pyroxene (Diopside)	CaMgSi ₂ O ₆	CaMgSi ₂ O ₆ + 2H ₂ CO ₃ + 2H ₂ O → CaCO ₃ + MgCO ₃ + 2H ₄ SiO ₄	41
Tremolite	Ca ₂ Mg ₅ Si ₈ O ₂₂ (OH) ₂	Ca ₂ Mg ₅ Si ₈ O ₂₂ (OH) ₂ + 7H ₂ CO ₃ + 8H ₂ O → 2CaCO ₃ + 5MgCO ₃ + 8H ₄ SiO ₄	38
Enstatite	MgSiO ₃	MgSiO ₃ + H ₂ CO ₃ + H ₂ O → MgCO ₃ + H ₄ SiO ₄	44
Laumonite	CaAl ₂ Si ₄ O ₁₂ ·4H ₂ O	CaAl ₂ Si ₄ O ₁₂ ·4H ₂ O + H ₂ CO ₃ + H ₂ O → CaCO ₃ + Al ₂ Si ₂ O ₅ (OH) ₄ + 2H ₄ SiO ₄	9

Where necessary, the reaction pathway has been normalized. Figures for % CO₂ uptake (w/w) assume the mineral reaction has been fully completed.

2004; Sanna et al., 2014). Olajire (2013) reports that rocks comprising olivine and serpentinite normally have an R_{CO_2} ranging between 1.97 and 2.51.

The reaction of CO₂ with olivine and serpentine in the presence of water is given in **Table 1**, together with other common minerals of interest (O'Connor et al., 2000; Renforth et al., 2011; Alexander and Maroto-Valer, 2018). The amount of CO₂ that is reacted (with the yield), on a percentage w/w basis, is calculated from the equation given. As can be seen, this varies widely and is dependent on the chemistry of the mineral in question and is further impacted by reaction kinetics, purity of the mineral system, and other considerations. Thus, a 100% reaction condition is rarely achieved.

As the rate of reaction between rock and bicarbonate is slow, a managed mineralization step requires a high-energy environment and complex reaction conditions to overcome kinetic constraints (Balucan et al., 2013; Eikeland et al., 2015; Ebrahimi et al., 2018). Thus, until mineralization technology advances, the *in situ* injection of CO₂ into a rock for storage (as opposed to the utilization of sedimentary formations found in depleted oil wells) is unlikely to be widely adopted. Wang J. et al., 2018; Wang F. et al., 2018 discuss some of the economic considerations involved, whereas Verduyn et al. (2011) review the options for CO₂ mineralization in rock. Majumdar and Deutch (2018) provide a context for mineralization and other CCUS options that may be deployed at scale. Kelemen et al. (2019) have most recently overviewed the status and challenges of mineralization in the geosphere.

ACCELERATED CARBONATION OF MINERAL SYSTEMS

The mining and fine grinding of basaltic or other rocks have been investigated for ameliorating soil to enhance mineral

weathering and produce bicarbonate (Kohler et al., 2010; Wilson et al., 2014; Renforth et al., 2015; Sackler Forum, 2017). Notwithstanding, the impacts on the chemistry of the oceans, the logistics involved in mining, processing, transporting, and widely applying this technology are likely to be prohibitive. Studies on accelerating mineralization by, for example, raised temperatures and pressures, controlling moisture content, and the partial pressure of CO₂ include those of Gerdemann et al. (2007), Dri et al. (2014), and Gadikota and Park (2015).

It is often assumed that the use of *ex situ* treatments may overcome many of the issues associated with the costs and technical complexity of the *in situ* mineralization of CO₂ in host rocks. An advantage of *ex situ* processing is the ease of auditing and confirming the permanence of carbon stored in the mineralized product(s). The drawbacks in respect of the energy requirement associated with processing and subsequent carbonation are augmented by one issue not receiving much attention to date, and that is the disposal of the mineralized media back into the geosphere. As mineralized products occupy significantly more volume than the originally excavated rock as it now contains carbonate, the environmental impacts associated with “landfilling” more rock that was excavated must be fully costed, as environmental harm will undoubtedly arise.

Geologically derived feedstock materials for mineralization, such as olivine and serpentine, are abundant, and mining technologies are mature enough to handle appropriate quantities of processed mineral media for carbonation. The high costs of intensive reaction conditions can, to a certain extent, be mitigated if commercially attractive mineral products, such as silica or magnesia, can be made available for sale. Teir et al. (2007) and Zevenhoven et al. (2017) have described processing of serpentinite by dissolution followed by carbonation and the recovery of mineral products.

For the industry to apply mineralization technology, changes in commercial and regulatory operating environments are

needed. This is particularly important if the mineralization processes used manufacturing products that are suitable for reuse or sale into the market. Unless facilitating tax regimes are in place to support processing and the disposal of mineralized products to land, the cost involved can only be mitigated by the manufacture of products for sale, such as silica or finely divided calcium carbonate.

That said, there are commercial opportunities involving the mineralization of wastes to divert them from landfill into products with value. Residues are subject to waste management regulations in many countries and have a significant negative value. Furthermore, because of the waste hierarchy, there are options to apply “end of waste” via the manufacture of products that are fit for purpose, providing the wastes are risk-managed. In Europe, the Waste Framework Directive provides a pathway to “end of waste” and product status (European Commission, 2019). Products can be a substitute for virgin stone, a mineral filler, or another commercially attractive material/product. Wastes suitable for processing are often found close to market, relatively consistent, alkaline and have a high surface area, properties that facilitate further processing by mineralization.

Within the last decade, the use of CO₂ to treat industrial waste via an accelerated carbonation step to make carbonate-cemented products has become established in Europe. There are several estimates of the global quantities of suitable industrial wastes for mineral carbonation (e.g., Gomes et al., 2016), and suitable arisings are likely to be ≥2 Gt each year. As such, there is mounting interest in potential carbonate-able wastes to sequester mineralized CO₂ but also to valorize the waste and reduce the risks associated with disposal to land.

There are newly commercially available and emerging CCUS technologies involving mineralization, and these have been summarized in Hills et al. (2019). Although it is not yet possible to be certain of the potential total global amount of CO₂ that can be mineralized in waste, estimates in the 1–5 Gt range are available. The GCI (2016) estimates that with the right commercial incentives/government support, 3.6 Gt/year of CO₂ could be mineralized in construction aggregates by 2030. As the annual world demand for aggregate is currently in the order of 50 Gt and rising, a market for manufactured carbonated aggregates is well established.

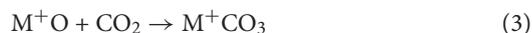
It should be noted that treatment of wastes can involve high-water containing “wet” systems, where waste particles are suspended in or dissolved in aqueous solution. The addition of CO₂ enables finely divided carbonated products to be produced, including precipitated calcium carbonate (PCC). PCC is used as a mineral filler in paper and plastic and has recently been recognized as a product in which CO₂ is permanently bound. As such, PCC meets the requirements of emissions trading (European Union [EU], 2018), a key development that enables the benefits of CO₂ sequestration to be recognized with value generation from carbon credits. Therefore, if CO₂ from an Emissions Trading Scheme (ETS) installation is ultimately not released into the atmosphere, either because it is transferred for geological storage or is mineralized, that amount should be subtracted from the emissions of the originating ETS installation. Recognition of this for PCC follows

the legal case initiated by Schaefer Kalk (C-460/15, EU:C:2017:29) (Curia.europa.eu, 2020).

Table 2 gives a general comparison of the strengths and weaknesses of *in situ* and *ex situ* mineralization technologies. It can be seen that there are benefits from both approaches; however, technology readiness and the investment involved are important impediments, especially where *in situ* technologies are concerned.

TREATMENT OF INDUSTRIAL PROCESS WASTE

The mineralization of CO₂ in waste has been variously demonstrated in the laboratory and commercially, including at ambient temperature and pressure conditions. The mineralization reaction primarily involves metal/mineral oxides, hydroxides, and silicates with CO₂ gas. Mineralization can be achieved (in minutes), but in reality, the reactions concerned are rarely fully completed. Calcium and magnesium carbonates, on account of the environmental impact and stability, are the favored reaction products and are simply represented as M⁺ (Equations 3 and 4):



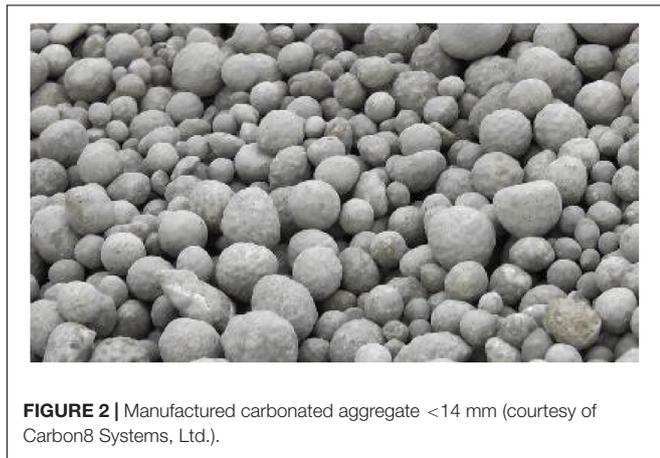
The reaction pathway involved may be more complicated than shown in Equations (3) and (4), as there are options to produce more than one valuable product from a mineralization step. For example, Xie et al. (2015) used CO₂ from flue gas to treat phosphogypsum waste in a reaction involving ammonia to produce ammonium carbonate, which then reacts with phosphogypsum waste in a reactor. The final products were calcium carbonate and ammonium sulfate, a source of nitrogen and sulfur for a slow release fertilizer.

A “world-first” long-term study on the sequestration of CO₂ in soil via field-scale carbonate-based solidification treatment of contaminated soil in South East England was carried out in 2000. This work demonstrated the possibility of treating “problem” soils to change their physical properties and leaching behavior (Antemir et al., 2010; Hills et al., 2019). Pure CO₂ was used to carbonate-solidify and granulate the soil concerned. Building on this work, flue gas extracted from a point source was used to treat locally sourced industrial solid wastes in a carbonation step. Wastes included air pollution control residues (APCr), which were mineralized with CO₂ derived from a landfill flare. The aggregated products were found to comply with construction material standards, and the approach was further developed (Gunning et al., 2011) and scaled up and commercialized (see **Figure 2**) in the United Kingdom (see Carbon8, 2020b).

The mineralization of CO₂ in waste is attractive, as it is a circular economic approach that can be achieved via direct use of available point-source CO₂. Potential carbonate-able wastes and CO₂ sources are generally co-located, and this provides an unique opportunity to strip the CO₂ gas directly from a flue to manage both solid- and gaseous-waste streams efficiently.

TABLE 2 | Example strengths and weaknesses of mineralization technologies.

Mineralization "type"	Strengths	Weaknesses
In situ (e.g., CO ₂ pumped into basalt or peridotite)	<ul style="list-style-type: none"> • Huge rock "reservoirs" available ($\times 10^6$ km³). • Reservoirs are found all over the world, e.g., ocean floor. • Potential to store CO₂ emissions for mega-annums. • Carbon capture and storage technology is under development. • Demonstration projects showing promise. • Large industrial projects (e.g., oil extraction) demonstrate potential to scale. • Government interest in large-scale solutions. • Mining/oil companies, e.g., have potential capacity to respond. 	<ul style="list-style-type: none"> • Infrastructure requirements are prohibitive, as "reservoirs" can be far from sources of waste and CO₂. • Technology is poorly developed. • Engineering effort required is "high." • Reaction kinetics are slow. • CO₂ needs to be dissolved or be supercritical. • Reservoir rock needs linked porosity/vesicules. • Mineralize basalt in the geosphere has no value. • Carbon credit status of <i>in situ</i> processing is uncertain. • Public perception of approach may not be favorable.
Ex situ (e.g., batch reaction with thermal alkaline residues)	<ul style="list-style-type: none"> • Thermal wastes and CO₂ are often emitted together. • Point-source CO₂ can be used directly, even at low partial pressure (e.g., 10% v/v). • Engineering effort required is "low." • Mineralized products have environmental and economic value. • Sources of solid waste and CO₂ can be close to market. • Infrastructure requirements are minimal. • Product transport costs can be low. • Reaction can proceed under ambient conditions. • Mineralized products proven to meet "end of waste." • Waste mineralization can reduce associated hazards (e.g., stabilize and solidify contaminants). • A Gt-size market exists for mineralized building products. • Meets the need of the circular economy. • Waste is diverted from landfill. • CO₂ is permanently stored. • Considerable carbon offsets can be realized. • Reduces pressure on virgin resources. 	<ul style="list-style-type: none"> • "Slow" reaction kinetics can be an issue. • Not all wastes are suitable for mineralization. • Some wastes need additional treatments, increasing cost. • Mineralization "yield" can be lowered by "blinding." • Accelerators or catalysts or more energetic reaction conditions may be required for some products. • Products compete in the market with virgin materials without subsidy (limits technology take-up). • No value (e.g., carbon credits) can be claimed for CO₂ volumes managed as yet. • Processes may fall under waste management regulations and be subject to restrictions. • Public perception may be adverse for waste containing products. • Industrial processes producing suitable voluminous wastes are being phased out in favor of low-carbon alternatives. • Market potential/competitive costs influence product acceptance.

**FIGURE 2** | Manufactured carbonated aggregate <14 mm (courtesy of Carbon8 Systems, Ltd.).

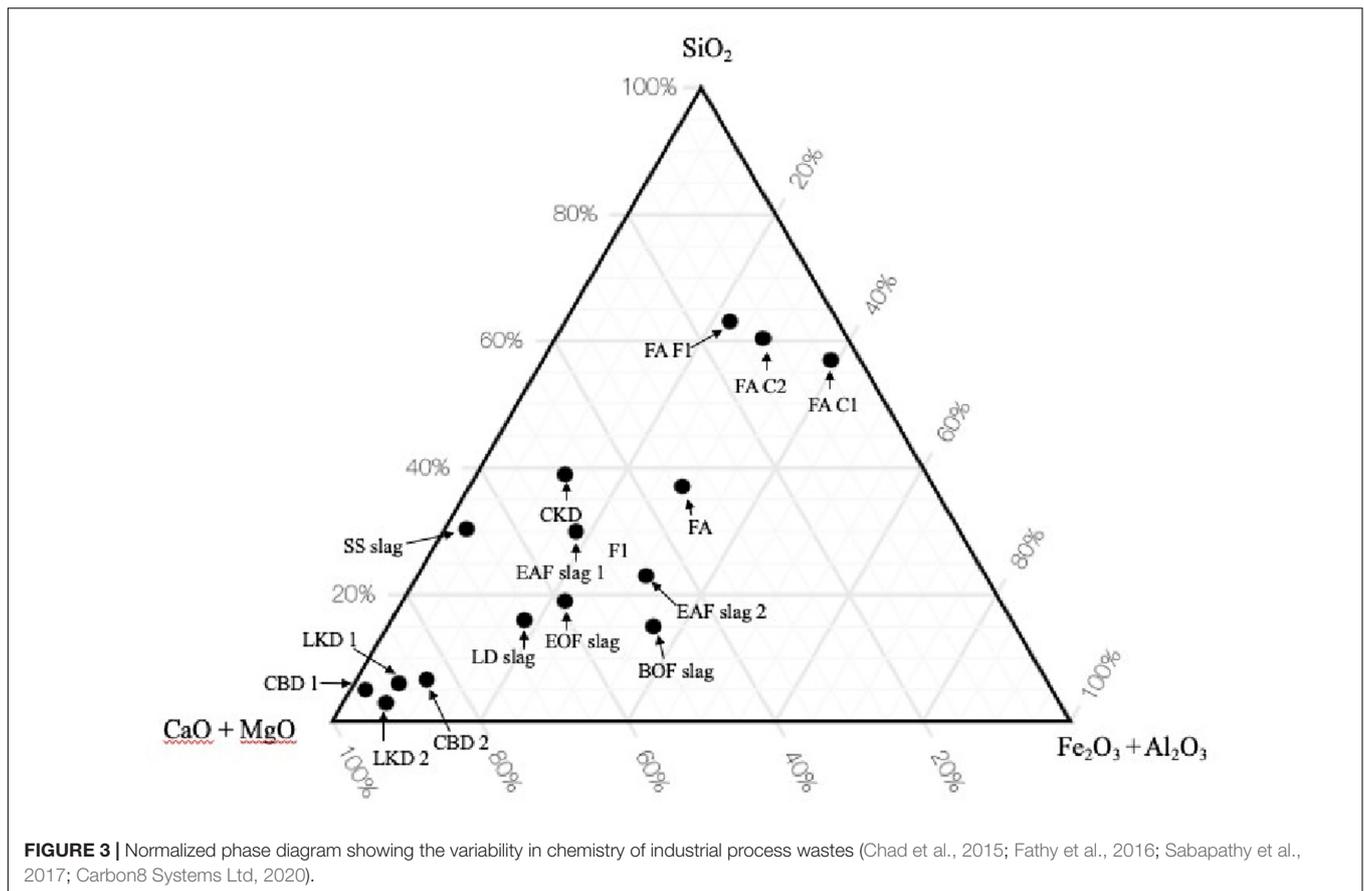
The formation of carbonates is an energetically downhill process, with the chemical reactions (e.g., as shown in Equations 3 and 4) being highly exothermic. The heat generated by a managed mineralization process can be recovered and reused. Important circular economic considerations related to mineralization include:

- Availability, location, and source of both solid waste and CO₂ gas;
- Optimal handling of feedstock solid and gaseous waste;
- Use of renewable energy in processing; and

- Facilitating standards for the use of the mineralized products.

Mapping sources and quantities can identify the availability of CO₂ as a feedstock. In Europe, it is mandatory to publish emission data if it exceeds 0.1 Mt/year, enabling the locations of CO₂ emissions to be mapped (Armstrong et al., 2019). Suitable CO₂ emissions that are easy to extract arise from the paper pulp industry (31.4 Mt/year), integrated (integrated coal gasification combined cycle) thermal coal plants (3.7 Mt/year), iron and steel manufacturing (151.3 Mt/year), and cement kilns (119.4 Mt/year) (Naims, 2016; Armstrong et al., 2019).

The waste materials that have the right chemistry, mineralogy, and physical properties can react with CO₂ under ambient temperature and pressure conditions (Fernandez-Bertos et al., 2004). The carbonate-cemented products can have the potential for reuse in engineering applications. However, as many industrial waste streams contain priority metals, there is a possibility of pre-processing to extract and recover the valuable metals before carbonation is carried out. By way of example, argon oxygen decarburization and basic oxygen furnace steel slags have been investigated by Gomes et al. (2016) and Ogden et al. (2017), whereas Quaghebeur et al. (2015) investigated the recovery of Cu, V, Zn, Ni, Mo, and Cr. A useful review of mineralization by Pan et al. (2012) discusses the various physico-chemical issues involved in waste mineralization. **Figure 3** shows the chemistry of example process wastes within the Ca(MgO)-SiO₂-Al₂O₃(Fe₂O₃) system and reflects the diversity



of wastes that have or may be presented for carbonation. As with geologically derived minerals and rock, the rate of calcium “leaching” from a waste (and, hence, its ability to combine with CO_2) is important. The ability of a waste to be mineralized is influenced by, for example, particle size, pH, solid-to-liquid ratio, temperature, pressure, mineralogy, etc. In reality, however, the formation of carbonate is subject to the balance between the kinetics of the reaction and the solubility of Ca or Mg in a waste feedstock (Pan et al., 2012).

There are several technical issues that potentially negatively impact the versatility of mineralization as a mechanism to carbonate wastes. One of those to avoid is the use of pure CO_2 , supplied as a process gas as the costs involved in compressing and transportation being relatively high. Thus, by capturing CO_2 directly from a flue gas without purification and liquefaction, costs are kept to a minimum as, for example, thermal residues can be reacted readily with the CO_2 released during their combustion without the need for transportation to a mineralization plant for onward processing. However, the amount of CO_2 in a flue gas can be low, and 10% v/v is not untypical. A point source with a low pCO_2 will necessitate extended reaction times and higher processing costs.

One approach to overcoming the kinetic constraints of flue gas with a low pCO_2 is the use of accelerators, and several homogeneous catalysts are available. These include inorganic

oxyanions (e.g., hypochlorite or sulfite), organic solutes (e.g., sugars and polyhydric alcohols), and amines and alkanolamines (which produce carbamates with CO_2) for the solvation and hydration of CO_2 in water that are available (Maries and Hills, 2013). It is important, however, to ensure that the presence of residual catalysts in the mineralized product does not affect reuse as might happen with enhanced sulfate or chloride leaching.

Another approach has been the biological catalyst, carbonic anhydrase, which is an enzyme better suited for use in higher water containing carbonation reaction environment. Power et al. (2016) reported that bovine-derived carbonic anhydrase increased the carbonation of a brucite slurry by 240%. The use of biomimetic metal–organic framework catalysts has been shown to enhance wollastonite dissolution (Lorenzo et al., 2018). Other work using a yeast-based catalyst, *Saccharomyces cerevisiae*, applied to coal fly ash, gave an increase in carbonation efficiency of 10% (Barbero et al., 2014).

LEGACY WASTES

Legacy residues, such as mine tailing, have the potential to be passively carbonated or mined as a feedstock for CCUS. This approach is sometimes called surficial carbonation, and tailings arising from mafic and ultramafic rock sources are candidate

wastes (Kelemen et al., 2019). National Academies of Sciences Engineering Medicine [NASEM] (2019) suggests that 10× Mt/year CO₂ can be readily sequestered in mine tailings this way. Hamilton et al. (2018) investigated chrysotile-processing residues/tailings from ultrabasic host rocks, whereas Pan et al. (2017) and Ebrahimi et al. (2018) investigated Ca-rich alkaline waste/tailings, including steel slag and electric arc furnace slag. Nickel-processing tailings, yielding serpentinite, were reported to be carbonated to a high degree by Teir et al. (2009). A review by Power et al. (2013) investigates the carbonation of alkali earth silicate and hydroxide-containing minerals. Xie et al. (2015) reviewed the mineralization of portlandite in various alkaline residues, including carbide, steel slag, paper mill waste, cement kiln dust, and coal fly ashes using sodium chloride to produce calcium bicarbonate. Treatment of both production and legacy wastes by *ex situ* processing has the potential to sequester Gt quantities of CO₂ each year (Renforth et al., 2011; Sanna et al., 2014). The relative costs involved in *in situ* and *ex situ* carbonation are discussed by Kelemen et al. (2019).

CARBON DIOXIDE MINERALIZATION IN THE CONSTRUCTION INDUSTRY

As mentioned, CO₂ has been long used to accelerate the curing of concrete and mortar. However, this application has rarely, if at all, been used to achieve a high degree of carbonation (i.e., where carbonate phases are responsible for hardening, rather than “normal” hydrated phases).

In the 1970–1980s, Berger and others investigated the reaction of CO₂ gas with cements, including individual calcium silicate phases (e.g., Klemm and Berger, 1972; Young et al., 1974). It was established that a rapid hardening of cement occurs at low water–solid contents via the decalcification of silicate phases leading to the precipitation of solid calcium carbonate. In largely unpublished work, Maries and Hills (1983) established a process for the rapid carbonate hardening of roof tiles, thereby omitting the need for steam curing. Shi et al. (2012) investigated

the kinetics of concrete carbonation and demonstrated that strength and dimensional stability are comparable with steam-cured articles.

The carbonate hardening of wastes arising from a mineralization step produces materials that can be used in engineering applications. Gunning et al. (2009) reported that lightweight carbonated aggregates with a bulk density of <1,000 kg/m³ and compressive strength >0.10 MPa could be manufactured from alkaline residues. Further developed by Gunning et al. (2011) for use with APCr, the carbonation process for manufacturing construction aggregates is now commercially established. Similar work was reported by Morone et al. (2014), with bonded aggregates made from basic oxygen furnace steel slags, which capture <10% CO₂ w/w. Salman et al. (2014) investigated monolithic products made from argon oxygen decarburization slag with strengths of 34 MPa after 3 weeks curing in 5% CO₂ and 60 MPa at 8 bar CO₂ and 80°C for 15 min. Similar results for stainless steel slag were reported by Quaghebeur et al. (2010) and Nielsen et al. (2017). Example CCUS processes delivering construction materials that are under development or commercially available are given in Table 3, together with their reported technical readiness level.

A notable recent development is a mobile carbonation plant producing construction aggregates, which is a flexible alternative to fixed plants using liquid CO₂ supplied by a road tanker. The mobile plant has a capacity of approximately 12 kt/year of manufactured carbonated aggregates using CO₂ directly stripped from a point source. This enables both gaseous and solid-waste streams to be economically captured and combined at relatively small emission locations or industrial plants with limited access. Figure 4 shows the “CO₂ntainer” (Carbon8, 2020a,b), which has been deployed at two cement plants, one in the United Kingdom and another in Ontario, the latter as a demonstration project funded under the Ontario Centres of Excellence Solutions 2030 initiative.

The capacity of the plant is designed to match the quantity of reactive waste residues generated by an industrial plant (e.g., 6–7,000 t for cement plants and 10–12,000 t for energy from

TABLE 3 | Selected mineralization processes yielding construction products.

Company	Technology	TRL	Product	References
Alcoa	Treatment of bauxite waste with CO ₂ (from an ammonia plant)	6	Construction fill, soil amendment	Global CCS Institute, 2011
Carbcrete	Carbonation activation of steel slag	6–7	Carbonated “concrete”	Savage, 2017; Carbcrete, 2020
Carbon8 Systems	Accelerated Carbonation Technology	9	Aggregates/fill, e.g., for blocks/concrete/screed	Carbon8, 2020b
Carbstone Innovation	Carbonation of steel slag	9	Construction materials including roofing tiles	Vito, 2020
Blue Planet	Carbonate coating over an alkaline substrate	6–7	Aggregate	Blue Planet, 2020
Carboclave	Nano-CaCO ₃ crystals producing a densification effect	7	Concrete blocks	Carboclave, 2020
Green minerals	Carbonation of olivine	3	Building materials	On-Site, 2020

TRL, technology readiness level.



FIGURE 4 | Mobile plant for manufacture of carbonated aggregates (courtesy of Carbon8 Systems, Ltd).

wastes), removing the need to transport the waste residues for treatment at a central site and also removing the need to purify the CO₂ for transport for use elsewhere. The containerized plant can be rapidly deployed and directly connected into the flue stack to extract the CO₂, with the remaining flue gas being returned to the stack.

LIFE CYCLE AND TECHNO-ECONOMIC ASSESSMENTS OF CARBON DIOXIDE MINERALIZED PRODUCTS

As we move to a circular economy as part of our wider sustainability efforts, the potential of mineralized CO₂-based products entering the marketplace mean the accounting of carbon is required. The key considerations concern the net carbon emissions of the process, materials processing/handling and transport, and energy requirements. At a system/sectoral level, the mineralization of CO₂ in wastes allows for the application of CCUS across a range of different industrial settings within an economy, making sure the varied inputs into the process are considered.

New technologies for CCUS are emerging, and their economic and environmental viability, and economic and environmental trade-offs have to be carefully analyzed. This involves a combination of life cycle assessment (LCA) and techno-economic assessment (TEA) and is key to informed decision-making and a standardized assessment. Several regulatory bodies of the European Commission (e.g., the Scientific Advice Mechanism, EIT Climate-KIC) have recognized the need of a common assessment guidelines involving LCA and TEA to enhance reliability, transparency, and compatibility of CCUS technologies and their technology readiness levels (Buchner et al., 2018).

An LCA can be used to assess the environmental impacts of products or services, being not limited to climate change only but also the other impacts, such as resource depletion (Bui

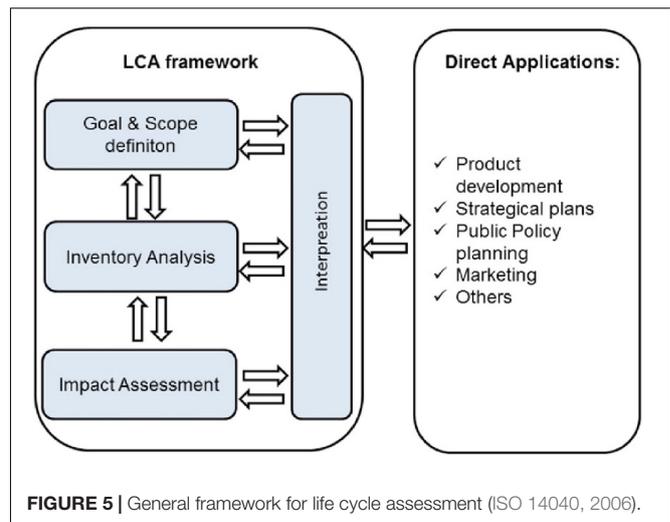


FIGURE 5 | General framework for life cycle assessment (ISO 14040, 2006).

et al., 2018). According to the International Organization for Standardization (ISO 14040, 2006), an LCA consists of the four interdependent phases, as shown in **Figure 5** (Armstrong et al., 2019). Artz et al. (2018) state that LCA studies for identical CCU technologies can also vary, so standardization of an LCA assessment is important under a general framework structured by ISO. Comprehensive guidance on LCAs has been produced for CCUS by the Global CO₂ Initiative (Deepblue, 2020) and the National Energy Technology Laboratory [NETL] (2019)². Both follow ISO 14040 (2006) (*Environmental Management–Life Cycle Assessment–Principles and Framework*) and ISO 14044 (2006) (*Environmental Management–Life Cycle Assessment–Requirements and Guidelines*) and provide additional guidance specific to CCUS projects. Further comprehensive guidance (arising from the same work) lists the needs for a standard methodology for LCA (Müller et al., 2020), as the choices made can differ widely and can significantly impact decision-making. Predefined assumptions on feedstock materials and utilities coupled with guidance on reporting enable standardization and a comparison between different CCUS technologies to be achieved.

It is worth noting that LCA has been applied to phosphogypsum recovery, both from wastewater and simulated solid-waste processing (Amann et al., 2018; Pell et al., 2019, respectively), showing that trade-offs between emissions and energy demand are required, further strengthening the need for a consistent approach, like that described by Müller et al. (2020). An LCA-based quantification of emissions re-use in the manufacture of different mineralized products is given by Zevenhoven (2020).

The TEA is a methodological framework that analyses the technical and economic performance of a process, product, or service (Armstrong et al., 2019). The major components of this framework involve the economic impact of research, development, demonstration, and implementation/deployment of technologies (Zimmermann et al., 2020). This is all

²www.netl.doe.gov/LCA/CO2U

incorporated in quantifying the cost of manufacturing and market opportunities with a particular technology.

The TEA is a widely used tool that considers several perspectives (e.g., research and development, corporate, and market) and varying guidelines according to application, technology development, and stakeholder needs (Zimmermann et al., 2020).

In the case of an LCA for CO₂ mineralization, the calculation mainly incorporates the emissions and turnover time of captured carbon (i.e., the duration of carbon storage in products). In a TEA for CO₂ mineralization, the CO₂ avoidance for product applications can be exemplified by lowering CO₂ emissions of another process (e.g., cement or steel) by waste treatment for industrial ashes. In market segments, CO₂ avoidance can be explained via an example of large-scale steel plants by making low-quality aggregates for low-cost concrete (Zimmermann et al., 2020).

Zimmermann et al. (2020) describe LCA and TEA of CCUS technologies and suggest that as a substitute of natural aggregates/concrete, 1 t of CO₂-mineralized concrete can be produced in a 50-kt/year plant with an output of over 20 years. The use and disposal of these products are likely to be the same as for benchmark (natural) products, and a gate-to-gate approach can be applied. However, to follow the cradle-to-grave approach, the integrated life cycle costing and LCA along with a TEA are suggested (Miah et al., 2017; Dong et al., 2018). The scope of TEA and LCA in the product life cycle is given by von der Assen (2016) and modified by Zimmermann et al. (2020), as given in **Table 4**.

The McCord et al., 2018 interpretation of TEA for CCUS-based production of secondary aggregates from APCr concludes that the production cost is high for a fixed plant producing 4,500 metric tons per year. However, the plant is profitable due to payment of gate fees for APCr treatment. The process inputs of this cradle-to-gate process are APCr, sand, cement, water, and CO₂ (flue gas). The process outputs are secondary aggregates, water, and flue gas (that is returned to stack), and the technology readiness level is 9.

In another interpretation of a cradle-to-grave approach incorporating CO₂-mineralized APCr blocks composed of carbonated material (lighter blocks than conventional), McCord et al., 2018 depicts that the blocks using the Carbon8

process (using substitution to remove the energy from waste impacts from the system boundary) result in a reduction of 22–34% of GHG emissions. This is evaluated considering the impacts arising from the APCr treatment and block production process.

FUTURE DEVELOPMENTS FOR CARBON DIOXIDE MINERALIZATION

Within the United Kingdom, the right commercial environment exists for the production of manufactured accelerated carbonated aggregates that are cost-competitive to natural stone and are market accepted. Acknowledging that the United Kingdom situation may be fortunate, in which aggregate sources are dwindling, landfilling of waste is subject to rising prices, and the market is amenable to new products.

However, as it currently stands, the general development of mineralization processes is largely held back by a mix of technical and economic reasons. As discussed in Chapter 7 of the IPCC special report on CCS (IPCC, 2005), although there has been some progress in the past 15 years, significant progress remains to be made (Khesghi et al., 2012). Current hurdles to overcome and their likely solutions are given in **Table 5**.

The mineralization of CO₂ in geologically derived and solid process wastes is attractive, not least that the quantities of mineral

TABLE 4 | The scope of LCA and TEA in CCU-based products.

Approach	Scope
Gate-to-gate TEA	<ul style="list-style-type: none"> For R&D/corporate perspective (preliminary studies). For substitute (chemically and structurally similar to benchmark material) products.
Cradle-to-gate LCA	<ul style="list-style-type: none"> For preliminary studies. For substitute products.
Cradle-to-grave LCA/TEA	<ul style="list-style-type: none"> For market-perspective studies. For non-substitute (chemically and structurally different benchmark material) products.

LCA, life cycle assessment; TEA, techno-economic assessment; R&D, research and development.

TABLE 5 | Problems and solutions for the development of mineralization technologies (Khesghi et al., 2012).

Problem	Solution	Timescale
Carbonation is kinetically controlled	Catalysts to increase efficiency of mineralization processes are required	Medium
Higher energy processing to increase “yield” is costly	Ensure renewable energy systems employed with catalysis	Medium
The cost of accessing and securing CO ₂ is too high	Develop processes or sorbents that directly strip CO ₂ from flue gas	Short
Regulation is immature and lacking for capture of CO ₂ in waste	Revise waste regulations to allow combining of gaseous and solid emissions	Short
Investment is hard to obtain/too costly, so business-related risk is not low enough	Government to underwrite and reduce risk profile for CCUS technologies	Medium
Materials standards do not recognize mineralized products	Ensure materials standards are cross-cutting and do not rely on virgin feedstock	Medium-long
Value for mineralized products has to be created	Government to provide incentives (e.g., tax breaks) for CCUS-derived products	Short-medium
Public acceptance needs to be improved	Implement a public awareness campaign of benefits of the circular economy	Medium
Government incentives are not available to grow a “mineralization” industry	Ensure climate change mitigation and CCUS is central to fiscal policy	Medium
Infrastructure remains lacking	Ensure <i>trans</i> -boundary infrastructural systems is built and available for use	Medium-long

feedstock available are suitable for sequestering Gt of carbon each year. As the technology and infrastructure required develop, the associated costs will decrease. How this will be paid for and by whom is a matter of current debate. In the meantime, efficiency measures, renewable energy sources, and other measures will go some way to limit emissions to the atmosphere.

The appeal of wastes as a feedstock for mineralization is promising, as the technology for manufacturing mineralized products is already commercially established and is being further developed (e.g., mobile carbonation plant directly using CO₂ from point-source emissions). In addition, the following advantages may also apply:

- Solid waste and point-source CO₂ are commonly co-located,
- Many solid process wastes are alkaline in nature, and
- Being often located close to the market is an added advantage, as proximity to the market is critical if valorized products are to be cost-competitive with virgin products.

An important implication of mineralization of CO₂ in waste for the production of construction materials is the environmental and economic benefits accrued via direct and indirect CO₂ emissions offset. The direct offset is achieved via permanently sequestering CO₂ in waste-based products, whereas indirect offsets can be realized by:

- Direct replacement of cement with carbonated materials made from waste,
- Replacement of hydraulic cement by carbonate-able binders,
- Reduction of transport involved in landfilling of waste,
- Replacement of quarried virgin stone,
- Reduced transport/materials handling, where solid waste and point-source CO₂ are co-located close to the market.

As the emphasis moves further towards low-carbon construction and wider certification of products becomes firmly established, the carbon offsets achieved by the manufacture and use of mineralized construction products will become more important.

The requirements for entering the “market” with a mineralized waste-based product involve complying with relevant regulatory instruments. In the European Union, this is governed by the Waste Framework Directive (European Commission, 2019). However, the laws of a particular territory sometimes preclude compliance, as the framework cannot be enacted. Furthermore, the needs and perceptions of the market and the supply chain in place may be an issue. In the United Kingdom, the Environment Agency is receptive to landfill avoidance strategies through the “end of waste” process and provides objective guidance. Thus, there are several technical and non-technical challenges involved, not least securing long-term contracts for waste feedstock and product offtake and the impact these “hurdles” have on investment confidence.

A consideration not widely reported is the public perception of using wastes in products, including those for construction. Industrial by-products have been widely used in construction, such as coal fly ash (pulverized fuel ash) and steel slag (ground-granulated blast-furnace slag). These materials are wastes but

have legal status as by-products. With many alkaline wastes falling under waste management regulations and remaining as wastes until they are sold to the market, a contradiction based on labeling emerges. As such, there is a need to address this either by legally changing the “label” or by educating stakeholders that mineralized products meeting “end of waste” are indeed products and not wastes by another name.

There are many considerations involved, and one might be to focus on:

- the embodied carbon within the mineralized product,
- its fitness for purpose including meeting internationally accepted material standards, and
- an accentuation of the sustainability gains, including protection of virgin resources.

Also, the move by the construction industry to carbon neutrality was an important consideration to the successful entry of manufactured carbonated aggregates into the United Kingdom market. With competitive pricing and technical advantages, such as lighter weight, augmented by carbon negativity, the United Kingdom construction block industry has been receptive. Furthermore, these benefits have also been somewhat recognized by the wider stakeholder community. Thus, inward investment supporting innovation and rising industry buy-in of the sustainability gains add strength to this CCUS approach.

Currently, in Europe, there is pressure for mineralization technologies to be included in the European Union ETS along with the generation of e-fuels, to avoid the release of fossil carbon. The mechanism being proposed is via the Emissions Monitoring and Reporting Regulation (European Union [EU], 2018) and relates to the ability of industrial flue gas to be transformed into useful materials, including mineralized building products. Changes to emission trading regulations, as proposed, will be a significant stimulus to help meet innovation challenges for the circular economy, the avoidance of CO₂, protection of natural resources, and the creation of wealth.

The world market for construction aggregates is in the order of 50 Gt/year and rising, so it is well placed to receive manufactured carbonated aggregate products. The total value of aggregate sales is projected to be US\$547 Bn by 2025 (Research and Markets, 2020), highlighting that value-added mineralized products can benefit from a growing global market. With the right incentives within emerging circular economies, such as those anticipated in Europe and Asia, sustainable mineralized products can play an increasingly important role in the building materials supply chain.

AUTHOR CONTRIBUTIONS

This work was conceived and primarily written by CH and NT. PC provided practical advice, guidance, and text, primarily to the commercially related aspects described in the manuscript, including the recent development of mobile carbonation plant and also elsewhere. All authors played a substantive role in the production and completion of the present work.

- Hills, C. D., Tripathi, N., Lake, C., Carey, P. J., Heap, D., and Hills, A. T. (2019). "General Mineralisation of CO₂ in solid waste," in *Carbon Dioxide Utilisation: Fundamentals*, Vol. 1, eds M. North and P. Styring (Berlin: De Gruyter Publications), 165–187.
- Huijgen, W. J. J., and Comans, R. N. J. (2003). *Carbon Dioxide Sequestration by Mineral Carbonation. Report Number ECN-C-03-016*. Petten: Energy Research Centre of the Netherlands (ECN).
- IPCC (2005). "IPCC special report on carbon dioxide capture and storage," in *Prepared by Working Group III of the Intergovernmental Panel on Climate Change*, eds B. Metz, O. Davidson, H. C. de Coninck, M. Loos, and L. A. Meyer (Cambridge: Cambridge University Press), 442.
- IPCC (2014). *Climate Change 2014 Synthesis Report Summary for Policymakers*. Available online at: <http://www.ipcc.ch/2018/02> (accessed April, 2020).
- IPCC (2018). *Summary for Policymakers of IPCC Special Report on Global Warming of 1.5° C Approved by Governments*. Available online at: <https://www.ipcc.ch/sr15> (accessed April, 2020).
- ISO 14040 (2006). *Environmental Management – Life Cycle Assessment – Principles and Framework*, Vol. 3. (Geneva: International Organisation Standardisation), 20.
- ISO 14044 (2006). *Environmental Management – Life Cycle Assessment – Requirements and Guidelines*. Int. J. Life Cycle Assessment, 652–668.
- Kelemen, P., Benson, S. M., Pilorgé, H., Psarras, P., and Wilcox, J. (2019). An overview of the status and challenges of CO₂ storage in minerals and geological formations. *Front. Clim.* 1:9. doi: 10.3389/fclim.2019.00009
- Kheshgi, H., de Coninck, H., and Kessels, J. (2012). Carbon dioxide capture and storage: seven years after the IPCC special report. *Mitig. Adapt. Strateg. Glob. Change* 17, 563–567. doi: 10.1007/s11027-012-9391-5
- Klemm, W. A., and Berger, R. L. (1972). Accelerated Curing of Cementitious Systems by carbon Dioxide: Part 1, Portland cement. *Cem. Concr. Res.* 2, 567–576. doi: 10.1016/0008-8846(72)90111-1
- Kohler, P., Hartmann, J., and Wolf-Gladrow, D. A. (2010). Geoenvironmental Potential of Artificially Enhanced Silicate Weathering of Olivine. *Proc. Natl. Acad. Sci. U.S.A.* 107, 20228–20233. doi: 10.1073/pnas.1000545107
- Lorenzo, F. D., Ruiz-Agudo, C., Ibañez-Velasco, A., Millán, R. G., Navarro, J. A. R., Ruiz-Agudo, E., et al. (2018). The carbonation of wollastonite: a model reaction to test natural and biomimetic catalysts for enhanced CO₂ sequestration. *Minerals* 8:209. doi: 10.3390/min8050209
- Majumdar, A., and Deutch, J. (2018). Research opportunities for CO₂ utilisation and negative emissions at the gigatonne scale. *Joule* 2, 801–809. doi: 10.1016/j.joule.2018.04.018
- Maries, A., and Hills, C. (2013). "Homogeneous catalysis of the accelerated carbonation of Portland cement," in *Proceedings of the 4th International Conference on Accelerated Carbonation for Environmental and Materials Engineering* (Leuven: ACEME).
- Maries, A., and Hills, C. D. (1983). Improvement in concrete articles. U.K. Patent No. 2,192,392.
- Miah, J. H., Koh, S. C. L., and Stone, D. (2017). A hybridised framework combining integrated methods for environmental Life Cycle Assessment and Life Cycle Costing. *J. Clean. Prod.* 168, 846–866. doi: 10.1016/j.jclepro.2017.08.187
- Morone, M., Costa, G., Polettini, A., Pomi, R., and Baciocchi, R. (2014). Valorization of steel slag by a combined carbonation and granulation treatment. *Minerals Eng.* 59, 82–90. doi: 10.1016/j.mineng.2013.08.009
- Müller, L. J., Kätelhön, A., Bachmann, M., Zimmermann, A., Sternberg, A., and Bardow, A. (2020). A guideline for life cycle assessment of carbon capture and utilization. *Front. Energy Res.* 8:15. doi: 10.3389/fenrg.2020.00015
- Naims, H. (2016). Economics of carbon dioxide capture and utilization- a supply and demand perspective. *Environ. Sci. Pollut. Res.* 23, 22226–22241. doi: 10.1007/s11356-016-6810-2
- Nation Energy Technology Laboratory [NETL] (2019). Available online at: www.netl.doe.gov/LCA/CO2U (accessed March, 2020).
- National Academies of Sciences Engineering Medicine [NASSEM] (2019). *Negative Emissions Technologies and Reliable Sequestration: A Research Agenda*. Washington, DC: The National Academies Press.
- National Energy Technology Laboratory [NETL] (2019). *Timothy J. S. P. E., Mutchek, M., Krynock, M., Cooney, G., Pegallapati, A., Rai, S., et al. Carbon Dioxide Utilisation : Life Cycle Analysis Guidance for the U.S. DOE Office of Fossil Energy. DOE/NETL-2019/2069*, eds Available online at: <https://www.netl.doe.gov/energy-analysis/details?id=3737> (accessed April, 2020).
- Nielsen, P., Baciocchi, R., Costa, G., Quaghebeur, M., and Snellingsa, R. (2017). Carbonate-bonded construction materials from alkaline residues. *RILEM Tech. Lett.* 2, 53–58. doi: 10.21809/rilemtechlett.2017.50
- O'Connor, W. K., Dahlin, D. C., Nilsen, D. N., Walters, R. P., and Turner, P. C. (2000). "Carbon Dioxide Sequestration by Direct Mineral Carbonation with Carbonic Acid," in *Proceedings of the 25th International Technical Conf. On Coal Utilization & Fuel Systems*, Coal Technology Assoc. (Clear Water, FL: U.S. Department of Energy).
- O'Connor, W. K., Dahlin, D. C., Rush, G. E., Gerdemann, S. J., Penner, L. R., and Nilsen, D. N. (2005). *Aqueous mineral Carbonation: Mineral Availability, Pretreatment, Reaction Parametrics and Process Studies. DOE/ARC-TR-04-002*. Albany, OR: Albany Research Centre.
- Ogden, M. D., Moon, E. M., Wilson, A., and Pepper, S. E. (2017). Application of chelating weak base resin Dowex M4195 to the recovery of uranium from mixed sulfate/chloride media. *Chem. Eng. J.* 317, 80–89. doi: 10.1016/j.cej.2017.02.041
- Olajire, A. A. (2013). A review of mineral carbonation technology in sequestration of CO₂. *J. Petrol. Sci. Eng.* 109, 364–392. doi: 10.1016/j.petrol.2013.03.013
- On-Site (2020). Available online at: www.on-sitemag.com/features/1003957345 (accessed April, 2020).
- Palandri, J. L., and Kharaka, Y. K. (2004). *A Compilation of Rate Parameters of Water-Mineral Interaction Kinetics for Application to Geochemical Modeling. US Geological Survey Open File Report 2004-1068*. Available online at: <http://www.dtic.mil/cgi-bin/GetTRDoc?Location=U2&doc=GetTRDoc.pdf&AD=ADA440035> (accessed April, 2020).
- Pan, S., Chang, E., and Chiang, P. (2012). CO₂ capture by accelerated carbonation of alkaline wastes: a review on its principles and applications. *Aerosol and Air Qual. Res.* 12, 770–791.
- Pan, S. Y., Chung, T. C., Ho, C. C., Hou, C. J., Chen, Y. H., and Chiang, P. C. (2017). CO₂ mineralization and utilization using steel slag for establishing a waste-to-resource supply chain. *Sci Rep.* 7:17227. doi: 10.1038/s41598-017-17648-9
- Pell, R., Wall, F., Yan, X., Li, J., and Zeng, X. (2019). Mineral processing simulation based-environmental life cycle assessment for rare earth project development: a case study on the Songwe Hill project. *J. Environ. Manage* 249:109353. doi: 10.1016/j.jenvman.2019.109353
- Penner, L. O. W. (2004). "Mineral carbonation: energy costs of pre-treatment options and insights gained from flow loop reaction studies," in *Proceedings of the 3rd Annual Conference on Carbon Capture and Sequestration*, Dublin.
- Peuble, S., Andreani, M., Gouze, P., Pollet-Villard, M., Reynard, B., and Van de Moortele, B. (2018). Multi-scale characterisation of the incipient carbonation of peridotite. *Chem. Geol.* 476, 150–160. doi: 10.1016/j.chemgeo.2017.11.013
- Power, I. M., Harrison, A. L., Dipple, D. M., Wilson, S. A., Kelemen, P. B., Hitch, M., et al. (2013). Carbon mineralization: from natural analogues to engineered systems. *Rev. Mineral. Geochem.* 77, 305–360. doi: 10.2138/rmg.2013.77.9
- Power, I. M., Harrison, A. L., and Dipple, G. M. (2016). Accelerating mineral carbonation using carbonic anhydrase. *Environ. Sci. Technol.* 50, 2610–2618. doi: 10.1021/acs.est.5b04779
- Quaghebeur, M., Nielsen, P., Horckmans, L., and Mechelen, D. V. (2015). Accelerated carbonation of steel slag compacts: development of high strength construction materials. *Front. Energy Res.* 3:52. doi: 10.3389/fenrg.2015.00052
- Quaghebeur, M., Nielsen, P., Laenen, B., Nguyen, E., and Van Mechelen, D. (2010). Carbstone : sustainable valorisation technology for fine grained steel slags and CO₂. *Refract. World Forum* 2, 75–79.
- Raymo, M. E., and Ruddiman, W. F. (1992). Tectonic forcing of late cenozoic climate. *Nature* 359, 117–122. doi: 10.1038/359117a0
- Renforth, P., Pogg von Strandmann, P. A. E., and Henderson, G. M. (2015). The dissolution of olivine added to soil: implications for enhanced weathering. *Appl. Geochem.* 61, 109–118. doi: 10.1016/j.apgeochem.2015.05.016
- Renforth, P., Washbourne, C.-L., Taylder, J., and Manning, D. (2011). Silicate production and availability for mineral carbonation. *Environ. Sci. Technol.* 45, 2035–2041. doi: 10.1021/es103241w
- Research and Markets (2020). Available online at: www.researchandmarkets.com (accessed April, 2020).
- Romanov, V. N., Soong, Y., Carney, C., and Rush, G. E. (2015). Mineralisation of carbon dioxide: a literature review. *ChemBioEng Rev.* 2, 231–256. doi: 10.1002/cben.201500002

- Rowland, J. L. (1870). Improvement in the manufacture of artificial stone. U.S. Patent No. 109669. Washington, DC: U.S. Patent and Trademark Office.
- Sabapathy, Y. K., Balasubramanian, V. B., Shankari, N., Kumar, A., and Ravichandar, D. (2017). Experimental investigation of surface modified EOF slag as coarse aggregate in concrete. *J. King Saud. Univ. Eng. Sci.* 29, 388–393. doi: 10.1616/j-jksues.2016.07.002
- Sackler Forum (2017). *Dealing with Carbon Dioxide at Scale*. Available online at: www.royalsociety.org/sackler-forum (accessed April, 2020).
- Salman, M., Cizer, Ö., Pontikes, Y., Santos, R. M., Snellings, R., Vandewalle, L., et al. (2014). Effect of accelerated carbonation on AOD stainless steel slag for its valorisation as a CO₂-sequestering construction material. *Chem. Eng. J.* 246, 39–52. doi: 10.1016/j.cej.2014.02.051
- Sanna, A., Uinbu, M., Caramanna, G., Kuusik, R., and Maroto-Valer, M. M. (2014). A review of mineral carbonation technologies to sequester CO₂. *Chem. Soc. Rev.* 43, 8049–8080. doi: 10.1039/c4cs00035h
- Savage, N. (2017). Carbicrete: Steel in, carbon out. *Nature* 545:S17.
- Shi, C. J., Liu, M., He, P. P., and Ou, Z. H. (2012). Factors affecting the kinetics of CO₂ curing of concrete. *J. Sustain. Cement Based Mater.* 24–25.
- Teir, S., Eloneva, S., Fogelholm, C. J., and Zavenhoven, R. (2009). Fixation of carbon dioxide by producing hydromagnesite from serpentinite. *Appl. Energy* 86, 214–218. doi: 10.1016/j.apenergy.2008.03.013
- Teir, S., Kuusik, R., Fogelholm, C., and Zevenhoven, R. (2007). Production of magnesium carbonates from serpentinite for long-term storage of CO₂. *Int. J. Mineral Process.* 85, 1–15. doi: 10.1016/j.minpro.2007.08.007
- UNESCO-IOC/SCOR (2007). *Ocean Carbon Sequestration: A Watching Brief of the Intergovernmental Oceanographic Commission of UNESCO and the Scientific Committee on Ocean Research. Version 2*. Paris: UNESCO.
- Vankeuren, A. N. P., Matter, J. M., Stute, M., and Kelemen, P. B. (2019). Multitracer determination of apparent groundwater ages in peridotite aquifers within the Samail ophiolite, Sultanate of Oman. *Earth Planet. Sci. Lett.* 516, 37–48. doi: 10.1016/j.epsl.2019.03.007
- Verduyn, M., Geerlings, H., van Mossel, G., and Jijayakumari, S. (2011). Review of the various CO₂ mineralization product forms. *Energy Procedia* 4, 2885–2892. doi: 10.1016/j.egypro.2011.02.195
- Vito (2020). Available online at: www.vito.be/en/carbstone (accessed April, 2020).
- von der Assen, N. (2016). *From Life-Cycle Assessment Towards Life Cycle Design of Carbon Dioxide Capture and Utilisation*. Available online at: <http://publications.rwth.aachen.de/record/570980> (accessed March, 2020).
- Wang, F., Dreisinger, D. B., Jarvis, M., and Hitchens, T. (2018). The technology of CO₂ sequestration by mineral carbonation: current status and future prospects. *Can. Metall. Q.* 57, 46–58. doi: 10.1080/00084433.2017.1375221
- Wang, J., Wu, H., Duan, H., Zillante, G., Zuo, J., and Yuan, H. (2018). Combining life cycle assessment and building information modelling to account for carbon emission of building demolition waste: a case study. *J. Clean. Prod.* 172, 3154–3166. doi: 10.1016/j.jclepro.2017.11.087
- Wilson, S. A., Harrison, A. L., Dipple, G. M., Power, I. M., Barker, S. L. L., Mayer, K. U., et al. (2014). Offsetting of CO₂ emissions by air capture in mine tailings at the Mount Keith Nickel Mine, Western Australia: rates, controls and prospects for carbon neutral mining. *Int. J. Greenhouse Gas Control* 25, 121–140. doi: 10.1016/j.ijggc.2014.04.002
- Xie, H., Yue, H., Zhu, J., Li, C., Wang, Y., Xie, L., et al. (2015). Scientific and engineering progress in CO₂ mineralization using industrial waste and natural minerals. *Engineering* 1, 150–157. doi: 10.15302/j-eng-2015017
- Young, J. F., Berger, P. L., and Breese, J. (1974). Accelerated curing of compacted calcium silicate mortars on exposure to CO₂. *J. Am. Ceram. Soc.* 57, 394–397. doi: 10.1111/j.1151-2916.1974.tb11420.x
- Zevenhoven, R. (2004). Mineral carbonation for long-term CO₂ storage: an exergy analysis. *Int. J. Appl. Thermodyn.* 23, 23–31.
- Zevenhoven, R. (2020). Metals Production, Mineralisation and LCA. *Metals* 10:342. doi: 10.3390/met10030342
- Zevenhoven, R., and Fagerlund, J. (2010). “Mineralisation of CO₂,” in *Developments and Innovation in CCS Technology*, ed. M. Maroto-Valer (Cambridge: Woodhead Publishing Ltd), 433–462. doi: 10.1533/9781845699581.4.433
- Zevenhoven, R., Slotte, M., Koivisto, E., and Erlund, R. (2017). Serpentinite carbonation process routes using ammonium sulfate and integration in industry. *Energy Technol.* 5, 945–954. doi: 10.1002/ente.201600702
- Zhang, S., and DePaolo, D. J. (2017). Rates of CO₂ mineralisation in geological carbon storage. *Acc. Chem. Res.* 50, 2075–2084. doi: 10.1021/acs.accounts.7b00334
- Zimmermann, A. W., Wunderlich, J., Müller, L., Buchner, G. A., Marxen, A., Michailos, S., et al. (2020). Techno-economic assessment guidelines for CO₂ utilisation. *Front. Energy Res.* 8:5. doi: 10.3389/fenrg.2020.00005

Conflict of Interest: The work reported is a review of technological advances involved in the mineralization of CO₂. Research is undertaken at the University of Greenwich, which has, in part, been successfully commercialized by its spin-out company, Carbon8 Systems Ltd. (C8S). It is explicitly recognized that PC is an executive director of C8S and CH is a full-time professor at the University of Greenwich but retains a non-executive technical role, in fulfillment of the Universities responsibility to its licensee spin-out company.

The remaining author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Copyright © 2020 Hills, Tripathi and Carey. This is an open-access article distributed under the terms of the Creative Commons Attribution License (CC BY). The use, distribution or reproduction in other forums is permitted, provided the original author(s) and the copyright owner(s) are credited and that the original publication in this journal is cited, in accordance with accepted academic practice. No use, distribution or reproduction is permitted which does not comply with these terms.