



Using CaO- and MgO-rich industrial waste streams for carbon sequestration

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Abstract

To prevent rapid climate change, it will be necessary to reduce net anthropogenic CO₂ emissions drastically. This likely will require imposition of a tax or tradable permit scheme that creates a subsidy for negative emissions. Here, we examine possible niche markets in the cement and steel industries where it is possible to generate a limited supply of negative emissions (carbon storage or sequestration) cost-effectively.

Ca(OH)₂ and CaO from steel slag or concrete waste can be dissolved in water and reacted with CO₂ in ambient air to capture and store carbon safely and permanently in the form of stable carbonate minerals (CaCO₃). The kinetics of Ca dissolution for various particle size fractions of ground steel slag and concrete were measured in batch experiments. The majority of available Ca was found to dissolve on a time scale of hours, which was taken to be sufficiently fast for use in an industrial process.

An overview of the management options for steel slag and concrete waste is presented, which indicates how their use for carbon sequestration might be integrated into existing industrial processes. Use of the materials in a carbon sequestration scheme does not preclude subsequent use and is likely to add value by removing the undesirable qualities of water absorption and expansion from the products.

Finally, an example scheme is presented which could be built and operated with current technology to sequester CO₂ with steel slag or concrete waste. Numerical models and simple calculations are used to establish the feasibility and estimate the operating parameters of the scheme. The operating cost is estimated to be US\$8/t-CO₂ sequestered. The scheme would be important as an early application of technology for capturing CO₂ directly from ambient air.

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1. Introduction

Climate change due to anthropogenic carbon dioxide (CO₂) emissions is a mounting concern. In order to prevent rapid climate change, it will be necessary to stabilize CO₂ concentrations, which, in turn, requires that net anthropogenic emissions of greenhouse gases be reduced to a small fraction of their current value. Emissions reductions are required under the framework convention on climate change and various subsidiary agreements such as the Kyoto protocol. Cost-effective control of CO₂ emissions in developed economies will likely require the imposition of a monetary cost on carbon emissions through such mechanisms as a tax or tradable permit scheme. A price on carbon emissions implies an equivalent subsidy for negative emissions, that is for industrial processes which remove CO₂ from the atmosphere.

Here, we examine possible niche markets in existing industrial systems, particularly the cement and steel industries, where it might be possible to generate a limited supply of negative emissions (carbon storage or sequestration) at relatively small cost. We examine relevant chemical and economic considerations for concrete waste from construction and demolition projects and steel slag, a byproduct of steel manufacture. These industrial waste streams have high alkalinity and calcium content, allowing them to sequester carbon in the form of stable carbonate minerals. Carbon in this form is virtually permanently removed from the atmosphere. Also, this type of carbon sequestration is suited for removing CO₂ directly from ambient air. The example scheme described in Section 6 can be considered an early, low-cost implementation of the technology, which can later be applied to larger-scale systems for capturing CO₂ from ambient air.

The waste streams we consider represent an opportunity to reclaim some of the carbon dioxide emitted during the manufacturing process. With concrete, for instance, energy was expended during the calcination process of cement manufacturing to separate carbon dioxide from calcium. This process accounts for about 50% of carbon dioxide emissions from cement manufacturing, which, in turn, accounts for 1.3% of United States (US) CO₂ emissions [12]. By carbonating the cement embedded in waste concrete, we can reclaim some of the CO₂ emitted in initial production.

Concrete has a wide variation in composition but is generally 10–15% cement, with the remaining mass comprised of inert aggregate (e.g. gravel and crushed rock) and water. The cement portion contains calcium oxides (CaO) and hydroxides (Ca(OH)₂), which can be reacted with carbon dioxide dissolved in water to produce calcite (CaCO₃) or other carbonate phases.

Steel slag is a consolidated mix of many compounds, principally of calcium, iron, magnesium and manganese. The proportions vary with the conditions and the feedstocks for the particular furnace where the slag is generated. Calcium compounds are the largest constituents, comprising 40–52% by mass as CaO [17]. The particular compounds formed depend on the rate of cooling during production. In the samples used in our experiments, the dominant phase was found by X-ray diffraction to be Portlandite (Ca(OH)₂). Calcium from the dissolution of the Portlandite in steel slag can be combined with carbon dioxide as for cement. Indeed, the spontaneous leaching of Ca from slag and subsequent formation of CaCO₃ is readily observed in industrial applications of slag as fill [17].

Table 1
Estimated annual sequestration potential of concrete and ferrous slag in the United States

	t-CO ₂ per ton material	Flow of material [Mt/year]	Sequestration potential [Mt-CO ₂ /year]	Percent of the industry's emissions
Steel and blast furnace slag	0.27 ^a	16.9 ^c	4.6	8 ^e
Waste concrete	0.06 ^b	68 ^d	4 ^d	6 ^{d,f}

^a From [11] and assuming 75% of Ca contained in the material binds to CO₂.

^b From [12] and [16] and assuming 75% of Ca contained in the material binds to CO₂.

^c From [11].

^d The estimate for the flow of waste concrete is highly uncertain since no formal reporting system captures the majority of construction and demolition waste. This is derived from [3]. The true figure is probably much larger.

^e Total steel and iron manufacturing emissions were 59 Mt-CO₂ in 2001 [6], which excludes emissions from primary energy generation for the process.

^f Total industry emissions of 67 Mt-CO₂/year are based on emissions from cement manufacturing [12] and exclude transport and processing of the aggregate material, which is comparatively small.

Rough estimates of the carbon-binding potential of steel slag in concrete are shown in Table 1. The experiments and analysis in the remaining sections are designed to assess the feasibility of making this conversion (i.e. the conversion of slag or concrete-derived Ca to CaCO₃) on an industrial scale. Sections 2 and 3 aim to demonstrate that this conversion is chemically and physically possible to perform on an industrial time scale. Sections 4 and 5 explain how this conversion might fit into the existing industrial flows of these materials and what the effects, other than CO₂ sequestration, might be. Finally, Section 6 presents a specific example of a system which could be built and operated to perform this conversion along with rough estimates of the capital and materials requirements for the system.

2. Methods

To assess the feasibility of extracting free calcium (Ca) from steel slag and concrete waste, a series of experiments measuring the dissolution of these materials in aqueous solution was performed.

A 500 ml cylindrical glass reaction chamber was filled with de-ionized water and a pH buffer. Then, dissolved CO₂ was eliminated with the introduction of pure N₂ bubbles. The chamber was stirred via a magnetic stir bar at 450 rpm. A quantity of slag or concrete was added such that the solubility of Ca(OH)₂ would not be exceeded in the pH range tested, i.e. a total expected Ca concentration of 0.01 mol/l was used. The chamber was either sealed or kept at positive pressure of N₂ during the experiment to prevent intrusion of atmospheric CO₂ and subsequent formation of CaCO₃.

Slag and concrete samples were dry-sieved into size fractions of particle diameter (D_p) between 45 and 74 μm , 74 and 300 μm , and 300 and 600 μm . X-ray diffraction analysis suggests that the fine and course size fractions of slag are not chemically different, having essentially the same peak patterns when ground to comparable sizes. In both cases, the identifiable Ca-containing phases were Portlandite and calcite.

Solutions were prepared with buffers known to have negligible binding capacity with Ca and Mg. CAPS (3-(Cyclohexylamino)-1-propanesulfonic acid) was used to hold a pH between 10.5 and 10.6, TES (2-[2-Hydroxy-1,1-bis (hydroxymethyl)ethylamino]ethanesulfonic acid) held a pH between 7.4 and 7.6, and unbuffered solutions ranged in pH from 10.8 to 12 over the course of the experiment.

Samples of 3–5 ml were taken periodically and immediately filtered through 0.45 μm syringe filters to remove suspended solids, leaving only the dissolved fraction of Ca. Additional 0.02 μm filtration and post filtration acid-digestion on some samples were both found to have no effect on measured Ca concentration, indicating the presence of colloids in the filtered samples to be negligible. Total dissolved Ca was measured by titration with EDTA (Standard Method 3500-Ca-D) and with Ion Chromatography (Dionex DX-500).

Steel slag was run for several size fractions and several pH values. Concrete was run for several size fractions in an unbuffered solution.

3. Results

Measurements of the dissolution of Ca from steel slag for several pH's and size ranges are presented in Fig. 1. The initial rate and extent of dissolution of calcium is higher for smaller particle sizes and for lower pH, as expected. The data for slag particles with diameter between 45 and 74 μm at pH 7.5 are shown at linear scale in Fig. 2. The dissolution curve has a hockey stick shape, with rapid initial kinetics in the first hour or so, and then settling to a slower, log-linear rate. Approximately half of the Ca observed to dissolve does so within the first minute. By 160 h, 230 mg of Ca per gram of slag has dissolved, which corresponds to 84% of the total Ca in the slag based on elemental analysis performed by the manufacturer.

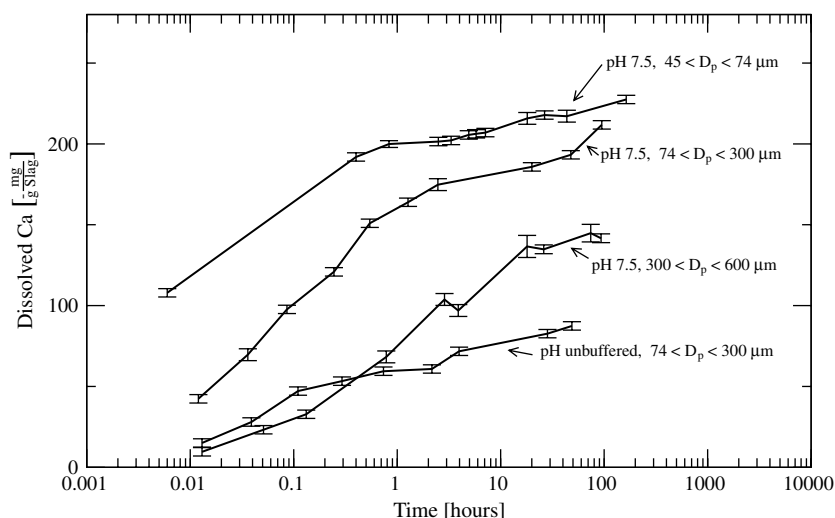


Fig. 1. Dissolution of calcium from steel slag. (Error bars represent 95% confidence intervals on the analytical measurements.)

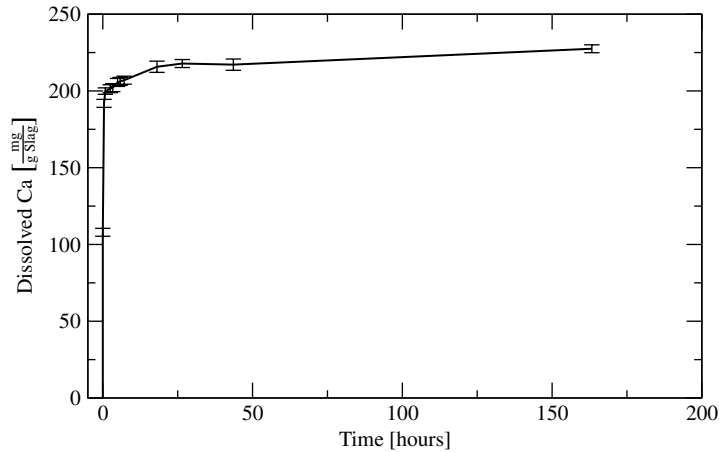


Fig. 2. Dissolution of calcium from steel slag: pH 7.5, $45 < D_p < 74 \mu\text{m}$. (Error bars represent 95% confidence intervals on the analytical measurements.)

Both the extent and rate of dissolution become notably smaller for particles in the larger size ranges, however, the dissolution rate is not as sensitive to particle size as simple homogeneous-particle theory would predict. For instance, the ratio of dissolution rates for the 45–74 and 74–300 μm size fractions, averaged over the first hour, is 1.25. However, we would expect the dissolution rate to be proportional to the specific surface area, holding pH and other variables constant. Using a hard sphere approximation, the ratio of surface areas for these fractions is 2.4, fitting the slag particles to a log-linear particle mass distribution. This discrepancy may be due to the heterogeneous composition or high porosity of slag particles, or to incomplete fractionation in the sieving process.

Dissolution rate is also strongly influenced by pH. This is consistent with the theoretical dependence of dissolution rate on saturation state. The saturation state of $\text{Ca}(\text{OH})_2$, the dominant phase of Ca in slag, decreases (the solubility increases) as pH decreases.

The data for concrete are shown in Fig. 3. The general shape of the curve is the same, with initial rate of dissolution similar to, if not faster than, steel slag. Here, the extent of Ca dissolution is not a monotonic function of particle size. It appears that the middle size range, $74 > D_p > 300 \mu\text{m}$, has the highest concentration of calcium. This is unsurprising since the concentration of cement in ground concrete is known to vary with particle size.

3.1. Summary

The results of the experiments above are intended to give a rough characterization of the dissolution of $\text{Ca}(\text{OH})_2$ from steel slag and waste concrete. They indicate that the rate and extent of dissolution are large enough to utilize on an industrial timescale, i.e. it seems possible to dissolve a substantial fraction of Ca with solids residence times on the order of hours. In light of this, it is reasonable to propose a scheme such as that outlined in Section 6.

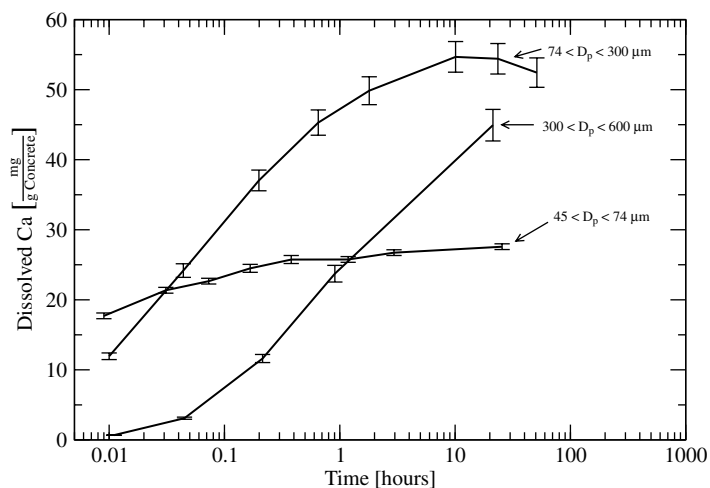


Fig. 3. Dissolution of calcium from concrete in unbuffered solution. (Error bars represent 95% confidence intervals on the analytical measurements.)

4. Options for steel and iron slag

Virtually all of the slag produced in the United States is currently sold and used for various applications, predominantly for asphalt and concrete aggregate and road base. The average market value for iron and steel slag at the point of production was \$8 per metric ton in 2001 [11], equivalent to \$25 per ton of CO₂. Given that likely CO₂ credits are of this order, this cost is probably prohibitive for a scheme where one buys slag solely for purposes of CO₂ sequestration and then disposes of it.

Use of the slag for sequestration, however, does not necessarily exclude subsequent uses. A possible scheme for carbonation of slag is outlined in the next section that is non-destructive. In this scheme, a substantial portion of the Ca and Mg oxides are dissolved from the slag. The Ca and Mg carbonates formed are deposited back into the slag as very fine particles. Thus, there is both a chemical and physical change in the slag properties, but both of these changes can be beneficial for further use. Furthermore, there are two reasons why slag intended for other applications is amenable to intermediate use in a sequestration scheme.

Most applications for slag require that it be crushed and sieved into relatively small particles. Some applications, such as use as a cement additive, require extremely fine grinding. Since the sequestration process requires small particles, using slag crushed for these applications can avoid additional energy and cost of grinding specifically for sequestration.

Secondly, the dissolution of Ca and Mg oxides from the slag may add value to the slag for use in the most common applications. Slag tends to have high water absorption and expansion properties due to the hydration of Ca and Mg oxides over time [11]. This is an undesirable property for concrete and asphalt aggregate and for base and fill material [17]. This property would be eliminated in any kind of carbonation scheme. In fact, washing or weathering is often already required for slag used as road base [17]. A modification of the process to enable capture of carbon dioxide may be all that is required to harness the sequestration potential of slag.

5. Options for concrete

Reinforced concrete waste is the single largest component of demolition and construction waste. It is generally removed from demolished buildings, roadways and other structures mixed with other sorts of rubble. While tipping fees at landfills are relatively low for concrete waste (which is considered “clean” waste), the extremely large quantities of it can make transportation and disposal expensive. About half of concrete waste is currently sent to landfills, while the rest is recycled in a variety of ways [3]. The decision on whether to recycle and how to recycle concrete waste rests on the balance of tipping fees and transportation costs versus the cost of sorting and grinding and (possibly) transportation of the resulting material minus the value of the recycled material. These costs vary regionally and are all of the order of dollars or a few tens of dollars per ton [15].

In large projects, much of the concrete waste is reused on site as fill and sub-base. It can also be used as aggregate in new concrete, obviating the need to import new aggregate. A limitation to this is that concrete made with recycled aggregate tends to absorb more water, expand more, and have slightly impeded performance characteristics compared with concrete made from new aggregate [14]. This is due to hydration of the original cement particles.

For all major recycling applications, steel and other major contaminants are removed, and the concrete is ground to various degrees of fineness, depending on the application. If recycled concrete, after grinding and before final application, could be put through a carbonation scheme like the one described below, many of its undesirable characteristics would be eliminated, adding value to the product. Additionally, the carbon capture scheme could be run without incurring the cost of additional grinding or interrupting major material flows. One can imagine a carbonation step being added to the recycling process at construction waste recycling facilities and large construction sites.

There may also be niches within the concrete industry that are particularly suited to sequestration schemes. For instance, after grinding, the concrete is often separated into coarse and fine fractions with, typically, the fine fraction passing a No. 4 sieve ($D_p < 4.75$ mm) [18]. The fine fraction has a disproportionate share of cement—about 40% [15]—making it particularly undesirable for most applications, but the small particle size and high-cement content make it ideal for carbonation. Once washed in a scheme like the example given, the fines fraction should have a better market potential.

6. An example carbonation scheme

We will assess the economics and energetics of a simple scheme for sequestering carbon using steel slag or concrete waste. A diagram of the scheme is shown in Fig. 4. An aqueous solution is cycled continuously through the system. The solution is sprayed from 10 m above on a bed of ground slag or concrete. The solution trickles by gravity through the bed, and the runoff is collected in a wet well. The solution, now saturated in $\text{Ca}(\text{OH})_2$, is then pumped back through the sprayers, collecting CO_2 from the air as it falls. The solution deposits solid CaCO_3 in the bed and dissolves more $\text{Ca}(\text{OH})_2$ as it passes through.

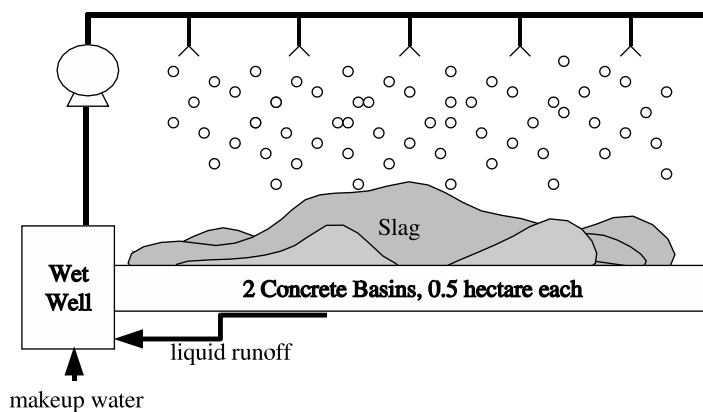


Fig. 4. An example carbonation scheme.

Two beds are operated in parallel, allowing one to be drained and unloaded while the other continues to operate. Materials can be collected and moved with standard front loaders. We assume 5% down time for maintenance and material exchange. Capital equipment and maintenance are priced for a 30 year lifetime.

For the purposes of this calculation, we assume that the flow rate and bed geometry have been chosen such that the solution exiting the bed is saturated with respect to calcium, i.e. 900 mg/l, or 0.023 mol/l, using the value measured for our sample of steel slag in water at equilibrium.

6.1. Spray characteristics and pumping energy

Other research has established the general physical feasibility of extracting CO_2 from the air [5] and of doing so with a caustic solution [8,9]. For our particular application, a mathematical model of a falling droplet was used to assess the feasibility of capturing CO_2 with a spray of $\text{Ca}(\text{OH})_2$ solution and to estimate the size of droplets required for efficient operation.

Drops were assumed to fall by gravity through 10 m of air with, on average, half of atmospheric CO_2 concentration. Mass transfer of CO_2 into the drop was modelled with boundary layer theory. Resistance in the liquid layer was considered to be negligible since internal circulation in drops of the size range considered is relatively fast [13]. Mass transfer through the air boundary layer was estimated with an empirical relation developed for raindrops [2] and integrated over the fall of the drop.

The resulting CO_2 absorption as a function of droplet diameter is shown in Fig. 5. The working solution effectively saturates with respect to CO_2 at 0.023 mol/l, so according to this model, we need drops smaller than about 0.8 mm. Spray nozzles that generate droplets in this range are common. A hollow-cone nozzle, for instance, is a relatively low-energy-cost option [7]. Having chosen an appropriate nozzle, 0.3–0.8 mm drops can be readily generated with a 10 psi pressure drop [1].

Integrating the results in Fig. 5 across a typical drop size distribution for a hollow-cone spray nozzle (adapted from [7]) gives an average CO_2 absorption of 0.019 mol/l, utilizing 84% of the Ca in solution.

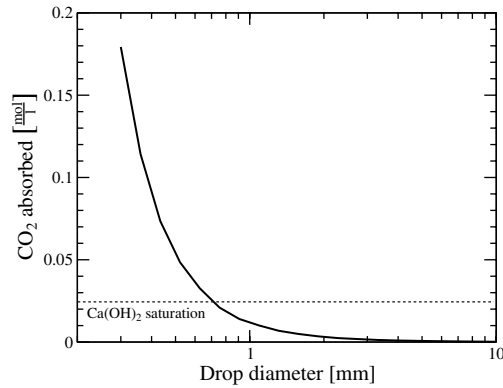


Fig. 5. CO₂ absorption by falling drops.

6.2. Water use

While the water for the working solution is recycled through the system, water must be added to make up for losses by two mechanisms: evaporation and entrainment.

To estimate the quantity of water lost by evaporation, a mathematical model was built, based on boundary layer theory, which estimates the quantity of water evaporated as a function of CO₂ absorbed. Numerical integration of the mass-transfer equations yields a ratio of water evaporated to CO₂ absorbed. The results are shown in Fig. 6. The solution depends strongly on initial conditions, i.e. the ambient temperature and relative humidity, and is found for the average conditions at several locations in the United States: Phoenix, AZ ($T = 21\text{ }^{\circ}\text{C}$, $\text{RH} = 37\%$), Miami, FL ($T = 23\text{ }^{\circ}\text{C}$, $\text{RH} = 76\%$) and Anchorage, AK ($T = 2\text{ }^{\circ}\text{C}$, $\text{RH} = 38\%$). The solution also depends on the efficiency of CO₂ capture from the air. Higher capture efficiencies require disproportionately more air-liquid contacting, leading to more evaporation. The values range from 2–22 m³/t-CO₂. For the example scheme, a capture efficiency of 50% and a moderate climate is assumed (5 m³-water/t-CO₂).

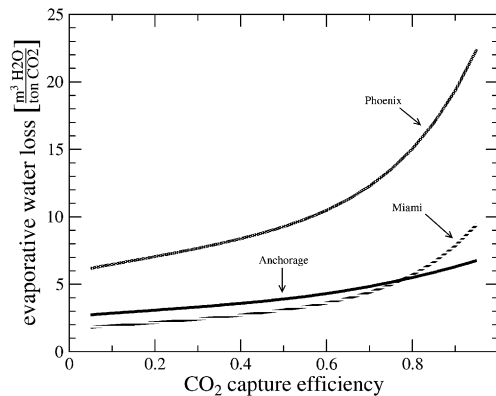


Fig. 6. Evaporative water loss for average conditions at three locations.

As the system requires a fine spray, some fraction of the droplets will be small enough to be carried by wind such that they fall outside of the collection basin. If we assume droplets with settling velocity smaller than 1 m/s are entrained in this way, using the drop-size distribution discussed in Section 6.1, 4% of the solution is lost. With careful choice of spray system, however, and some engineering optimization, a much better collection efficiency of water could probably be achieved.

In our example, the water loss to evaporation is 400 m³/d and to entrainment is 4000 m³/d. Thus, the makeup water requirement for the system is about 4500 m³/d, or 1.2 million gallons per day, which is considerable. The bulk of the water requirements, however, are due to the large fraction of droplets assumed to be entrained. If entrainment can be controlled either with control of the spray drop-size distribution, or with some kind of particle trapping mechanism, the water requirements would be quite manageable.

Water requirements of this magnitude will limit the placement of this sort of system to locations where large amounts of water are available. However, the input water has few quality requirements, and natural or secondary industrial flows can conceivably be used. In fact, it is particularly advantageous to use acid waste streams, which would be neutralized by the slag and would speed the kinetics of dissolution.

Much as local circumstances are important for determining the availability and cost and transport characteristics of concrete or slag, local circumstances with respect to water availability and cost are crucial to the success of this scheme. For the purposes of the cost calculation in Section 6.4, a typical western agricultural water price of \$30 per acre-ft is assumed.

6.3. Materials flow

The flow of materials is based on the CO₂ flux through the system, which has a square, hectare sized footprint and a 10 m high capture area, at an average wind velocity of 3 m/s. An assumed average capture efficiency for CO₂ entering the system of 50% fixes the rate of CO₂ sequestration. The resulting flows are summarized in Table 2. It may be possible to capture CO₂ at higher efficiencies than 50%, however, there is a diminishing return in terms of energy expended per quantity of CO₂ captured as efficiency is increased. 50% seems a reasonable trade-off and technically achievable based on our calculations.

The amount of slag or concrete required depends on the extent of dissolution of the available Ca(OH)₂. For the calculation, 70% of the total was assumed to dissolve. The results in Section 3 indicate that with reasonable particle sizes and with a residence time of the slag or concrete of hours or more, this should be achievable.

With these assumptions this scheme can sequester 32 kt of CO₂ per year, generating 73 kt of CaCO₃ per year. It requires a minimum of 140 kt of slag, 680 kt of concrete, or 200 kt of high-cement concrete fines per year. Potentially hundreds of such facilities could be supported by current US production levels of these materials. For the purposes of this example, procurement and transportation of these materials is assumed to be revenue-neutral. The cost of transportation and the value added to the material by this process are both on the order of dollars per ton and depend strongly on local circumstances [11,15]. It is assumed that this facility will be built where the value added for subsequent use will at least break even with the materials transportation and handling costs.

Table 2
Estimated cost and materials flows for example carbonation scheme

<i>Annual capital and operating costs [USD/year]^a</i>	
1 ha concrete basins	22,000 ^b
100 hp pumps, 4 in all	22,000 ^b
Piping	11,000 ^b
Wet well	3600 ^b
100 gpm sprayers, 300 in all	45,000 ^b
Pump operation and maintenance	8000
Front-loader operation, 18 days/year	13,000
Operator	60,000
Electricity	31,000
Water	40,000
Total annual cost	\$250,000
<i>Annual materials flows</i>	
Steel slag	32 kt ^c
Waste concrete	680 kt ^c
Concrete fines	200 kt ^c
Water	1.7 × 10 ⁶ m ³
Electricity	500 MW h ^d
CO ₂ sequestered	32 kt
Average cost of CO ₂ sequestration	\$8/t-CO ₂ ^e

^a These numbers are rough estimates based on discussions with industry personnel.

^b Capital costs are annualized at 6% interest for a 30 year plant life.

^c Any of the three materials—steel slag, concrete, or concrete fines—or a combination, can be used. These numbers represent the quantity required if the material is used alone.

^d With the US average electricity mix [4], this produces about 300 t-CO₂/year ≈ 1% of the CO₂ sequestered.

^e This is meant only to capture the cost of operating the scheme. It excludes some potentially important costs such as materials transportation to and from the site.

6.4. Cost estimates

Table 2 shows an estimated cost breakdown for the facility described, including capital costs and operation and maintenance costs. Dividing by the quantity of CO₂ captured gives an average cost of sequestration of about \$8/t-CO₂. This is small compared with the value of carbon credit necessary to meet Kyoto protocol targets, for which the Intergovernmental Panel on Climate Change gives \$55 as a lower bound estimate [10], and with economic model predictions of global trading prices, which range from \$18 to \$450 per ton of CO₂. On this comparison, it seems feasible to run such a facility profitably. It should be noted, however, that this is a very rough analysis. There are still many uncertainties on which the revenue-cost balance depends strongly. The cost of procurement and transportation of both water and raw materials will depend on local circumstances and have potential to dominate the economics. The efficiency of CO₂ capture from air passing through the system and the extent of available calcium dissolved from slag or concrete will dictate the magnitude of the water and materials flows. We consider these to be optimization issues and beyond the scope of this paper. However, from our rough analysis it appears possible that this sort of system can be operated profitably.

7. Conclusions

While many unanswered questions remain with respect to the use of slag and concrete for sequestering carbon, it is clear that the idea is worthy of further inquiry. With current information, it seems reasonable that a profitable scheme of this kind can be constructed, but it also appears that no large scale, uniform system will be effective in all circumstances. Effective application of carbon sequestration technology will probably become an exercise in industrial ecology, as it depends crucially on the matching of inputs and outputs from different industries, using existing flows and infrastructure, and clever rethinking of current practices. The potential value of carbon credit available from the flows in Table 1 is on the order of \$170 million annually, so this effort ought to be worthwhile economically. Socially, such endeavors are valuable as early implementations of carbon sequestration. Much of the technology and engineering required is applicable to other processes which capture carbon from ambient air. The niche facilities described here will provide valuable technological experience, making the way for larger-scale operations.

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