

Utilizing Iron and Steel Slag's in Environmental Applications-An Overview

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Introduction

In his 2001 Howe Memorial Lecture, Peter J Koros stressed the importance of utilizing to the fullest extent all resources, materials, energy and people within the boundaries set by the technologies, and economics of the iron and steel making industry. One of the areas highlighted in his address at this lecture to the Iron and Steel Society, was improved utilization of slag from steelmaking and ladle furnaces (Koros, 2003). Iron and steel slags are formed through the addition of fluxing agents such as but not limited to limestone or dolomite to blast furnaces and steel furnaces to strip the impurities from iron ore, steel scrap, and other ferrous feeds. The slag forms as a silicate melt that floats on top of the molten crude iron or steel and is tapped from the furnace separately from the liquid metal. After cooling to a solid form, the slag is processed and may then be sold and/or returned to the furnace. Apparent sales of iron and steel slags in the United States in 2006 totaled about 20.3 million metric tons (Mt), with an overall value of approximately \$406 million. (USGS, 2006)

Strictly speaking the utilization of iron or steel slag in any application can be viewed as an environmental application since the reuse or recovery of this material provides environmentally related benefits. For example the substitution of iron or steel slag for natural aggregate in asphalt not only saves on the energy that may be required to mine naturally occurring aggregate, but also eliminates the negative impacts associated with mining such as blight on the land. A more concrete example is in the reduction of CO₂ emissions as a result of fuel savings during cement manufacturing where iron and steel making slags are used in place of raw limestone and dolomite (Koros, 2003). Other uses of steel slag include ready-mixed concrete, concrete products, road bases and surfaces, fill, cementitious materials, gabions and rip rap, railroad ballast, landfill daily cover material, roofing granules, landscape aggregate, mineral wool (home and appliance insulation), soil remineralization and conditioning, etc. (USGS, 2006; Proctor et al., 2000). Table 1 adapted from the USGS 2006 Minerals Yearbook presents the ferrous slag sales by use in the US. The focus of this paper however is on those applications that directly affect environmental issues including remediation, and mitigation of activities that negatively impact the environment. It is within this framework that the following discussion will focus on the role iron and steel making slag has played in environmental and in a broader sense energy related issues.

Table 1. Percentage of Ferrous Slag Sales in the United states (USGS, 2006)

Use	2005			2006		
	Blast furnace slag		Steel slag	Blast furnace		Steel slag
	Air -cooled	Granulated		Air -cooled	Granulated	
Ready mixed concrete	17.8			15.7		
Concrete products	5.4			4.0		
Asphaltic concrete	13.4		15.6	13.3		12.1
Road bases and surfaces	32.5		53.0	41.3		51.0
Fills	12.3		10.5	14.6		17.9
Cementitious material		90.3			94.1	
Clinker raw material	4.9		6.9	1.5		6.9
Miscellaneous	9.6	0.9	2.3	7.6	0.6	0.8
Other or unspecified	4.2	8.8	11.7	2.0	5.3	11.3

Phosphate Removal

The impact of wastewater discharges on receiving waters depends on the watershed, in-stream quality, and other discharges and withdrawals. Frequent discharges of untreated wastewater with high quantities of phosphorous and nitrogen contribute to accelerated rates of algal activity and eutrophication. A basic method of algal control involves reducing the amount of nutrients available for algal growth and metabolism by controlling their inflow to the body of water (Raman, 1985). Nutrient control can best be accomplished through a comprehensive materials monitoring program in which nutrient inputs are identified for reduction or elimination (AWWA, 1987). Means of reducing nutrient inflow include amongst other practices wastewater treatment facilities designed to remove phosphorus and nitrogen (Reinert and Hroncich, 1990). Most of the chemical and advanced biological treatment technologies used for phosphorus removal in sewage treatment plants are expensive to install, difficult to operate efficiently, and produce considerable volumes of phosphorus-rich sludge requiring further disposal (Lee, 1998). Utilizing slag as a treatment media presents a low cost efficient alternative to phosphate removal.

Laboratory studies by Baket et al., (1998) have shown that basic oxygen furnace slag can be utilized to remove phosphorous from an onsite waste water disposal system. The researchers postulated that the slag could be used in single pass, self-contained treatment modules in alternative treatment systems. Their studies showed that the slag removed greater than 99% of PO_4 from a 10 mg/L PO_4 -P solution within 1 h of contact. Long-term attenuation capacities of the mixtures were assessed by continually loading bench-scale columns with a 3.3 mg/L PO_4 -P solution, at representative groundwater flow rates. A column containing 50 wt % silica sand, 45 wt % limestone, and 5 wt % iron/calcium oxide averaged >90% reduction in phosphate over 4 years (1450 pore volumes). X-ray and SEM microprobe analyses of the reacted solids showed phosphorus accumulations on the slag surfaces and discrete precipitates of microcrystalline hydroxyapatite [$Ca_5(PO_4)^{3-}OH$]. (Baker et al., 1998)

A waste water treatment plant (WWTP) for the township of Waiuku incorporates iron slag in its treatment process as an aggregate source. The plant was installed in 1993 and handles up to 3000m³ a day of treated effluent from the settling ponds. The WWTP is comprised of large aggregate filter beds with a hydraulic residence time of two days, the slag depth in the beds being about half a metre. There are 10 filter beds in all, utilizing some 15,000 m³ of slag aggregate (Bruce Wallace Partners 2004). The plant was initially designed to remove suspended solids and algae, and although phosphorous reduction was not part of the original brief, subsequent monitoring showed that it was removing a very high level of the incoming phosphorous – around 80% on average for the first five years - and most of the suspended solids and algae (NSA, 2008).

A Canadian project was implemented and administered by the Ontario provincial and municipal governments to remove phosphorus from septic system effluent. Since 1999, septic effluent from a single-family residence near North Bay, Ontario, has been treated in an aerobic sand filter, followed by a BOF slag chamber (Smyth et al., 2002). Phosphorus concentrations of approximately 5 mg/L PO₄-P in the septic system effluent were lowered to less than 0.02 mg/L PO₄-P in discharge from the BOF slag treatment chamber. The chamber also effectively removed E-coli from the wastewater. Neutralization of the pH occurred in groundwater adjacent to the subsurface discharge gallery. No maintenance of the influent and effluent lines of the BOF chamber was required for the first four years of operation. The lines were flushed hydraulically once to remove accumulated sludge (Smyth et al., 2004).

A concrete tank containing BOF slag was installed at the Massachusetts Alternative Septic System Test Center in October 2001. Aerobically treated communal wastewater with low biological oxygen demand (BOD) was introduced to the chamber via upward flow through the BOF slag mixture over a period of two years. The chamber received approximately 330 U.S. gallons (1,240 L) of communal wastewater that had been aerobically pre-treated daily. The wastewater had a residence time of approximately a day in the chamber. The wastewater had a BOD₅ between 3 and 7 mg/L, pH of 7 and phosphate (as PO₄) of 3.75 mg/L. A removal of total phosphorus and ortho-phosphate P to concentrations of approximately 0.05 mg/L or less was routinely achieved (Smyth et al., 2002, 2004).

Reduction of Hexavalent Chromium to Trivalent Chromium

Hexavalent chromium Cr (VI) is toxic to living cells and is believed to be a carcinogen. Unfortunately as an anion Cr (VI) is quite soluble over a very large pH range and readily penetrates cell membranes. Trivalent chromium Cr (III) is generally nontoxic and is believed to be essential in glucose metabolism in mammals (McBride, 1994). The National Academy of Science actually estimates a safe adequate intake of 0.05 to 0.20 mg/day (Tate and Arnold, 1990). Smith et al., (1998) demonstrated that steel slag removed hexavalent chromium from groundwater. Based on the characterization of steel slag, Ochola and Moo-Young (2004), noted that besides iron, steel slag is comprised of other minerals with the potential of adsorbing Cr (VI) and effectively reducing its concentration in solution. Bartlett and Kimble (1976) proposed that orthophosphate

(HPO_4^{2-}) competes with chromate for adsorption sites. They observed that in the presence of HPO_4^{2-} chromate adsorption was severely retarded, and there were also similarities between the adsorption and desorption behavior of chromate and phosphate in Spodosol horizons as pH was increased with CaCO_3 . Observations by Bartlett and James (1988) led them to the conclusion that there may be similarities on the adsorption of chromate (CrO_4^{2-}), sulfate (SO_4^{2-}), and phosphate (HPO_4^{2-}) on colloids with positively charged surfaces. Based on these observations, tests were conducted using solutions containing mixtures of CrO_4^{2-} with SO_4^{2-} , CrO_4^{2-} with HPO_4^{2-} , and CrO_4^{2-} with SO_4^{2-} , and HPO_4^{2-} batched with steel slag in order to determine whether adsorption, defined here as the mechanisms of electrostatic attraction and complexation reactions, was the mechanism responsible for the reduction of hexavalent chromium or whether reduction was the dominant Cr (VI) removal process (Ochola and Moo-Young, 2004). Simultaneous measurements of the redox potential during testing helped establish that redox reactions were primarily responsible for the reduction of hexavalent chromium Cr (VI) to trivalent chromium from water passed through steel slag. Furthermore they postulated that the mechanism responsible for the reduction occurred via a one electron transfer process within an inner sphere complex that occurs between steel slag and chromium. The process is repeated until the stable form of chromium Cr^{3+} is attained and due to the high pH of the solution results in the stable chromium ion being precipitated out (Ochola and Moo-Young, 2004).

Besides the treatment of contaminated groundwater as postulated by Smith et al., (1998), wastewaters from industrial applications that utilize hexavalent chromium may benefit significantly from utilizing steel slag as a primary treatment media for the removal of the spent hexavalent chromium from their effluent.

Remediating Acid Mine Drainage (AMD)

Acid mine drainage (AMD), can be described as the outflow of acidic water from abandoned metal mines, coal mines, or even large construction activities, usually within rocks containing an abundance of sulfide minerals. This discharge is characterized by elevated levels of potentially toxic metals, like, nickel, copper, lead, arsenic, aluminum, manganese etc. Treatment alternatives for AMD can be categorized into active and passive treatment systems. The conventional mine drainage systems involve neutralization by the addition of alkaline chemicals (such as limestone, lime, sodium hydroxide, sodium carbonate, or magnesia) to water. Active treatment systems generally require the installation of a plant with agitated reactors, precipitators, clarifiers, and thickeners. As a result of these requirements, active treatment systems carry a high installation and maintenance cost. Various researchers including Ziemkiewicz and Skousen (1998), Simmons et al., (2001), Ochola and Moo-Young (2005) have identified steel slag as a suitable candidate to remediate waters contaminated by acid mine drainage (AMD), since it has been shown to have a significant acid neutralizing potential that can be exploited to precipitate out a majority of dissolved metals by increasing solution pH. The Acid Neutralization Potential of steel slag was determined to be approximately 83% as calcium carbonate (CaCO_3) (Ochola and Moo-Young, 2005). Batch testing with steel

slag and solutions of various heavy metals including iron, zinc, aluminum, copper, and lead showed over 80% removal of aluminum, and over 90% removal of the other metals (Ochola and Moo-Young, 2005). Following is a brief discussion of two projects where steel slag has been successfully utilized to mitigate the effects of AMD.

McCarty Highwall

In October 2000 an abandoned surface mine located about 10 miles Southeast of Bruceton Mills in Preston County, West Virginia was the chosen site for an AMD remediation project. The site known as McCarty Highwall had water seeping out along an old spoil pile that was flowing into a channel and mixing with a second spoil seep approximately 500 ft downstream into Beaver Creek and eventually ending up in Cheat River. Earlier investigation had shown that the first seep was moderately acidic with a pH of 4.1 and a net acidity of 27.5 mg/L, while the second seep was similar with a pH of 3.9 and 24.5 mg/L acidity (Simmons et al., 2002). Remedial efforts involved the installation of a series of open limestone channels (OLCs) and steel slag leach beds were downstream of seeps 1 and 2. The leach beds consisted of a settling basin and steel slag check dam. Both check dams were formed from approximately 150 tons of steel slag and rip rapped along the downstream side with 6-8" limestone rocks. A 200 ft open limestone channel (OLC #1) was constructed from the upper spoil seep to the edge of the first settling basin. A secondary OLC (OLC #1b) was constructed to the left of OLC #1 to carry AMD from an intermittent spoil seep to the first basin. Water leaches from the basin through the center of a steel slag check dam and enters a 300 ft open limestone channel (OLC #2). OLC #2 exits into the second settlement basin, along with water from a 100 ft open limestone channel (OLC #3) constructed at the seep 2 site (Simmons et al., 2002). Water enters into settling basin #2 from OLCs #2 and #3 and exits the system through a second steel slag check dam and forms the headwaters of an unnamed tributary of Beaver Creek. Initial water samples collected from the leach beds contained extremely high concentrations of alkalinity (1479-1513 mg/L) and high pH values (11.6). However, by the fourth month, alkalinity had fallen to about 30 mg/L (pH 9.0) (Simmons et al., 2002)

Buckeye Furnace Project

The Buckeye Furnace Project includes extensive reclamation and treatment efforts along Buffer Run, a tributary to Little Raccoon Creek in Milton Township, Jackson County. The post-mining condition of this 1.9 acre basin includes numerous abandoned underground mines and an abandoned strip mine with 65-acres of barren coal refuse. High rates of erosion off the strip mine had literally buried the floodplain of Buffer Run in sediment and eliminated any semblance of a normal stream channel. Abandoned mine drainage (AMD) was continually discharging from numerous underground mine entrances and within the strip mine after heavy rain and snowstorms. The Little Raccoon Creek Abandoned Mine Drainage Abatement and Treatment (AMDAT) Plan identified Buffer Run as one of the top AMD producers in the entire Little Raccoon Creek basin. It was not uncommon for Buffer Run to be generating 25% of the total acid load in Little Raccoon Creek. Pre-reclamation water quality can be characterized as having low pH (2.5 - 3.5), high acidity load (900 - 4000 lbs/day), and abundant heavy metals (aluminum 23 - 320 lbs/day). The mean acidity load was on the order of 373 tons/year.

Reclamation and treatment involved a combination of source control measures (reshaping and recontouring of the coal refuse, capping the refuse with clay and soil, and placement of BYPRO (by product of Mead/WesVaco paper company) to reestablish vegetation) and passive treatment systems (successive alkalinity-producing system (SAPS), anoxic limestone drain (ALD), and steel slag leach beds). The SAPS and ALD systems are being used to treat drainage from the underground mines, while the steel slag leach beds are used generate additional alkalinity for the entire subwatershed. The post-reclamation water quality has a higher pH (2.7 - 6.1), lower acidity loads (130 - 290 lbs/day), and lower heavy metals (aluminum 8 - 45 lbs/day). The mean acidity load has dropped to 50 tons/year, which translates to an 87% reduction in acidity. (Ohio EPA, 2008)

Storm Water Treatment

Infiltration of rain water into the subsurface is retarded by conditions that include saturated grounds and impervious surfaces like parking lots, roads, buildings, and compacted soil. In most developed modern urban areas, drainage systems which collect runoff from impervious surfaces ensure that water is efficiently conveyed to waterways through pipes and networks. Anthropogenic activities result in deposition of pollutants on roads, lawns, roofs, farm fields, etc. that ends up in surface waters following a storm event. Many of these contaminants are potentially toxic and can have detrimental effects on the receiving environment by accumulating in sediments, plants and filter feeding organisms, causing health issues and habitat degradation (Ellis & Revitt 1991; ARC, 2003; ASA, 2006). In particular, 80% of all metals in particulate form remain potentially bioavailable and a source for further pollution (Colandini et al. 1995; Mikkelsen et al. 1996, ASA, 2006).

The Australasian (iron & steel) Slag Association (ASA) investigated six different types of iron and steel slag to determine their effectiveness in removing contaminants from artificially polluted storm water runoff. The slags utilized were identified as a SteelServ melter slag, a SteelServ electric arc furnace slag, a Whyalla blast furnace slag, a Whyalla basic oxygen steel slag, a Port Kembla blast furnace slag, and a Port Kembla basic oxygen steel slag. The contaminants investigated included 17 heavy metals listed as follows: aluminum, arsenic, antimony, boron, barium, beryllium, cadmium, chromium, copper, lead, manganese, mercury, molybdenum, nickel, selenium, tin, and zinc.

The study showed that all the six slags tested had the potential to be utilized as stormwater filter media as they reduced the concentrations of arsenic cadmium, copper, lead, nickel, zinc, phosphorus and nitrogen in the artificial stormwater. Some, but not all the slags also reduced the concentrations of aluminum, chromium, manganese and molybdenum. Only 3 slags reduced the concentrations of boron while the other 3 released boron. All but one slag released concentrations of barium (ASA, 2006).

Based on the results from this study, it is reasonable to conclude that in order for steel slag to be utilized for storm water treatment, field trials are necessary to determine the optimal treatment design based on slag type and runoff characterization. Additionally

treatment should be incorporated into a treatment train to ensure and not as a stand alone treatment unit. The ASA study showed that the slags tested had high permeabilities with saturated hydraulic conductivities $K_{\text{sat}} > 100,000 \text{ mm h}^{-1}$ ($K_{\text{sat}} > 2.8 \text{ cm s}^{-1}$) and although clogging may not be expected, additional study is warranted to verify this assumption. The general recommendations pertaining to utilizing slag for stormwater treatment are listed as follow (ASA, 2006):

1. Laboratory studies provide an approximation of field conditions, and further testing should be done in a controlled field study since depending on the type of construction, slag may retain or provide a suitable surface to degrade other contaminants not tested here, e.g., oils often found in car parks (Bond et al. 1999). The removal of hydrocarbons should be studied. Additionally the complex interaction between environmental, physical and chemical factors is only accurately testable in a controlled field situation.
2. Cost/benefit should be identified by economic and lifecycle analysis of stormwater treatment systems.
3. Locations should be identified that are suitable for the installation of slag based stormwater treatment systems. The general features of such suitable locations are large paved areas, heavy vehicular traffic, and sites that use or store contaminants (Pitt, 2002).

Sand Capping

Overview

Sand-capping is a technique used in the marine environment to suppress the elution of nutrient salts that cause eutrophication of seawater, and hydrogen sulfide that cause blue tides. In this technique organic sea-bottom sediments such as sludge layers are capped with sand in order to mitigate the effects of organic decomposition that lowers the amount of oxygen dissolved in the water. An added benefit of this technique is an improvement of grain size in seabed sand which helps create a suitable habitat for marine organisms. Current practice utilizes natural sand excavated from other areas which not only degrades the environment, but is a limited resource (Takahashi and Yabuta, 2002).

Analysis

The NKK Corporation of Japan has analyzed the viability of using blast furnace slag as a capping alternative to natural sand. In their analysis a comparison of the hydrogen sulfide generation suppression properties was made between slag, natural sand and uncapped bottom sediment. The results of the analysis presented in Figure 1 show that the hydrogen sulfide concentration in the interstitial water within the slag sand layer remained at levels markedly lower than that within the untreated bottom sediment or beach sand layers over a two-year period (Takahashi and Yabuta, 2002). Their analysis also showed that the seawater in the area where BF slag sand was laid had higher silicate concentrations than that in the area where bare bottom sediment was exposed. Silicate concentrations of more than 0.28 ppm are required for the growth of diatoms, the primary food source for various marine organisms, and diatoms are said to compete with dinoflagellates that cause red tides. Utilizing BF slag sand therefore has the potential to

prevent the occurrence of red tides. More telling of the environmental benefits associated with utilizing BF slag as capping sand in marine environments is its apparent effect in cultivating marine organisms. The total number and weight of benthic species living in the sea-bottom sampling areas, each covered by BF slag, beach sand, and untreated bottom sediment, was determined and is presented in Figure 2. The results showed that significantly more organisms lived in the slag-covered sea bottom area than in the sediment-covered area (Takahashi and Yabuta, 2002). Both the number of species and number of individual organisms are affected by sand grain size. It is reported that more benthonic organisms live in the sea bottom composed of sand of a larger grain size (Takahashi and Yabuta, 2002). BF slag sand has intermediate grain sizes of 1.0 to 1.5 mm, which are larger than those of typical natural sand (0.3 to 1.0 mm). NKK's BF slag sand-capping technology has been adopted in the Naka-umi Sea Purification and Sand capping Project carried out by the Ministry of Land, Infrastructure and Transportation in Shimane Prefecture, West Japan. Naka-umi is an enclosed area of sea connected to the Sea of Japan by a narrow channel. Takahashi and Yabuta (2002) concluded that BF slag sand has the potential to create a sea-bottom environment beneficial to a more diverse range of marine organisms.

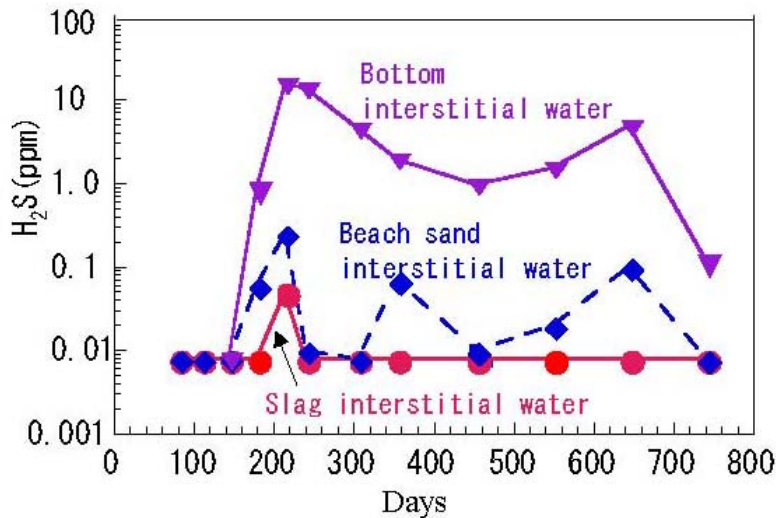


Figure 1. Interstitial Hydrogen Sulfide H₂S Concentrations in Slag, Beach Sand, and Bottom Sediment (Takahashi and Yabuta, 2002).

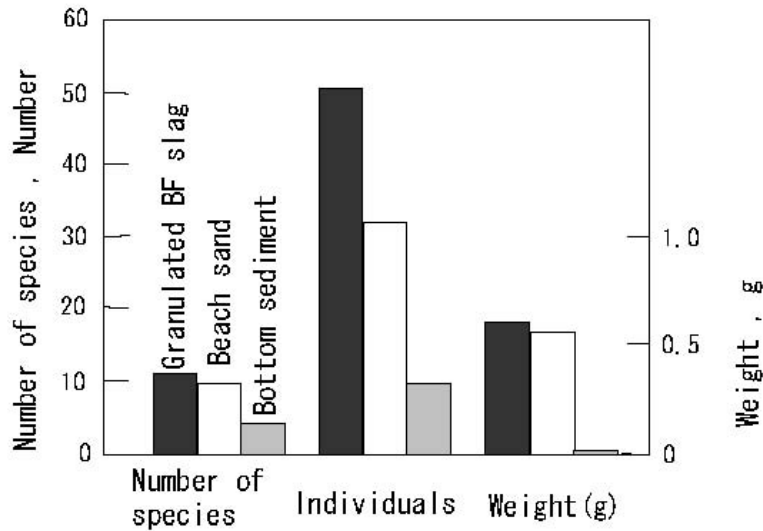


Figure 2. Observational Results of Benthic Organisms in Granulated BF Slag, Beach Sand, and Bottom Sediment (Takahashi and Yabuta, 2002).

Carbon Sequestration

One of the more interesting and innovative potential applications for utilizing slag is in carbon sequestration. To adequately understand what is involved in carbon sequestration, a brief description of carbon capture and storage is warranted. Carbon capture and storage (CCS) is an approach to mitigating global warming based on capturing carbon dioxide (CO₂) from large point sources such as fossil fuel power plants, biomass energy facilities, industries with major CO₂ emissions, natural gas processing, synthetic fuel plants, and fossil fuel-based hydrogen production plants and storing it instead of releasing it into the atmosphere. CCS applied to a modern conventional power plant could reduce CO₂ emissions to the atmosphere by approximately 80-90% compared to a plant without CCS (IPCC, 2005). Capturing and compressing CO₂ requires much energy and would increase the fuel needs of a coal-fired plant with CCS by about 25% (IPCC, 2005). These and other system costs are estimated to increase the cost of energy from a new power plant with CCS by 21-91% (IPCC, 2005). These estimates apply to purpose-built plants near a storage location: applying the technology to preexisting plants or plants far from a storage location will be more expensive.

Capture

Currently there exist three different types of technologies for capturing CO₂, post combustion, pre-combustion, and oxyfuel combustion. During *post-combustion*, carbon dioxide is captured from flue gases. This is a well understood technology currently used in various industrial applications. The other scenario *pre-combustion* involves the partial oxidation of the fossil fuel which results in the production of CO and H₂. When the CO reacts with water a reaction known as a water gas shift (WGS) results in the production of CO₂ and additional H₂. The resulting CO₂ can now be captured from a relatively pure exhaust stream while the H₂ can now be used as fuel. This technology is widely applied

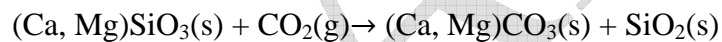
in fertilizer, chemical, gaseous fuel (H₂, CH₄), and power production (US DOE, 2008). The third technique *Oxy-fuel combustion* (Sweet, 2008) involves fuel burned in oxygen instead of air, with the cooled flue gas being recirculated and injected into the combustion chamber. The flue gas consists of mainly carbon dioxide and water vapor, the latter of which is condensed through cooling. The result is an almost pure carbon dioxide stream that can be transported to the sequestration site and stored.

Mineral storage

Reactions between either magnesium Mg or calcium Ca containing minerals and carbon dioxide CO₂ to form carbonates are advantageous in the following ways (Goldberg et al., 1998):

1. The reaction is thermodynamically favorable and occurs naturally since carbonates have a lower energy state than CO₂.
2. The raw materials such as magnesium based minerals are abundant.
3. The produced carbonates are unarguably stable and thus re-release of CO₂ into the atmosphere is not an issue.

The only drawback to storage via conventional carbonation pathways is that it occurs very slowly under ambient temperatures and pressures as exemplified in the weathering of rock over geologic time periods and described by the following equation.



The significant challenge therefore is to identify an industrially and environmentally viable carbonation route that will allow mineral sequestration to be implemented with acceptable economics (Goldberg et al., 1998):

Slag utilization

Huijgen, et al., (2005) investigated utilizing steel slag for CO₂ sequestration. In their study ground steel slag was carbonated in aqueous suspensions to study its reaction mechanisms. The researchers determined that based on the total Ca content of their slag sample the maximal CO₂ sequestration capacity was 0.25 kg of CO₂/kg of steel slag. Particle size, temperature, carbon dioxide pressure, and reaction time, were systematically varied, and their influence on the carbonation rate was investigated. Their results showed that using a particle size of < 38 μm, 19 bar CO₂ pressure at 100 °C, a carbonation degree equivalent to 74 % of the Ca content could be achieved in 30 minutes (Huijgen, et al., 2005). The carbonation reaction was found to occur in two steps; the first step involved leaching of calcium from the steel slag particles into the solution; followed by a second step whereby calcite precipitates on the surface of these particles. The first step particularly, the diffusion of calcium through the solid matrix toward the surface appeared to be the rate-determining reaction step (Huijgen, et al., 2005).

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