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Carbonation: An Efficient and Economical Process for CO_2 Sequestration

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ABSTRACT

Naturally occurring carbonation of cement-based materials is a slow phenomenon. Many factors control the rate of carbonation process in cement-based products. A comprehensive knowledge of the major factors controlling the rate of carbonation, and their effects on the properties of the materials subjected to it, is necessary for using this technology for CO_2 sequestration. Most of the studies related to the carbonation are limited to its effects on corrosion of reinforced steel, cover depth design, and service life prediction of reinforced concrete structures. The possibility of using carbonation process as a direct means for carbon dioxide sequestration is yet to be realized. This paper discusses the application of carbonation technique in cement-based products for CO_2 sequestration for reducing its concentration in the Earth's atmosphere. This technology would help cement, concrete, precast products, and other similar industries to reduce the contribution of CO_2 emission generated by them.

INTRODUCTION

It is now generally accepted that the global warming is caused by an increase in the concentration of greenhouse gases (GHGs) in the Earth's atmosphere from human activities. Carbon dioxide gas is the principal greenhouse gas. It exists in gaseous form in the Earth's atmosphere at a standard temperature and pressure. The major human activities which contribute to the emission of the CO_2 gas in the Earth's atmosphere include combustion of fossil fuels and deforestation. A natural emitter of large amount of CO_2 gas is volcanoes, hot springs, and geysers. However, the emissions of carbon dioxide gas by human activities are about 130 times greater than the quantity emitted by volcanoes; and, it is about 27 billion tonnes/year [USGS 2009]. The increasing concentration of carbon dioxide gas in the Earth's atmosphere has raised concerns about global warming, climate change, and their subsequent effects on its inhabitants. The global average atmospheric carbon dioxide concentration has risen from 280 ppm at the beginning of industrial revolution (about 150 years ago) to 380+ ppm today (2008). The annual growth rate of global average CO_2 concentration in the Earth's atmosphere since the beginning of continuous monitoring in 1959 is given in Figure

1. The maximum growth rate of atmospheric carbon dioxide was 1.9 ppm/year during 2000 – 2006 [Canadell et al., 2007]. Carbon dioxide is a toxic gas. Its effects on human body increase with an increase in its concentration. Some of these effects as reported by Davidson

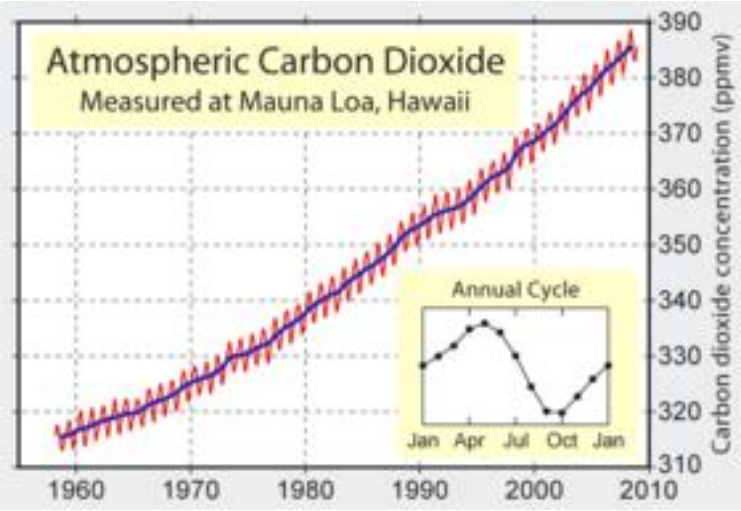

Fig. 1. CO_2 Concentrations Measured at Mauna Loa Observatory

[2003] are:

- 1% increase can cause drowsiness with prolonged exposure;
- 2% increase is mildly narcotic and causes increased blood pressure and pulse rate; and,
- At about 5% increase it causes stimulation of the respiratory centre, dizziness, confusion and difficulty in breathing

Therefore, scientists, engineers, researchers, environmentalist, geologists, and others, along with carbon dioxide contributing industries, are making tireless effort to develop efficient and viable technologies in their respective areas that could help in reducing carbon dioxide concentration in the atmosphere. From an environmental prospective, concrete construction industry is a very large consumer of natural resources such as stone, sand, and drinking water; and, at the same time, it is also one of the biggest generators of large amount of waste. Each of the primary ingredients of concrete i.e. cement, aggregate, and water has some adverse environmental impacts [Mehta, 2001 and 2002]. The production of cement, the main ingredient of concrete, is a highly energy intensive process and releases green-house gases (GHGs). Cement industry contributes approximately 6% of total anthropogenic CO_2 emission [Hendriks et al., 2004, Naik 2008]. Since global warming has emerged as the most serious issue of the recent time, and at the same time sustainability is becoming an important economical and political issue, there is an urgent need that the carbon dioxide contributing industries must develop technologies that could help in reducing carbon dioxide concentration in the atmosphere. The major environmental issue associated with the concrete construction industry is the CO_2 emissions from the production of portland cement.

CARBON DIOXIDE EMISSION

From 1850 to 2006 about 330 x 10^9 metric tons of CO_2 gas has been accumulated in the Earth's atmosphere from burning of fossil fuel and emission from cement industry [Canadell et al. 2007]. Fossil fuel and cement emissions increased from 7.0 PgC/y (1PgC = 1 petagram = 10^9 metric tons of C) in 2000 to 8.4 PgC/y in 2006, which is 35% above the emissions in 1990. The average growth rate of fossil fuel and cement emissions increased from 1.3% per year for 1990-1999 to 3.3% per year for 2000-2006. The annual atmospheric CO_2 concentrations measured at Mauna Loa Observatory and presented in the form of is presented in Figure 1 [NOAA, 2009].

Naik [2008] recommended increasing use of blends of portland cement. Such blended cements containing by-products materials such as fly ash and ground granulated blast furnace slag, are used in increasing quantities to replace the portland cement in concrete. However, the potential to reduce CO_2 emission by using blended cement varies from country to country depending on the availability of blending materials on the basis of coal combustion, pig iron production, and production of cement. Worrell [1995] estimated the potential for carbon emission reduction in 24 countries in the OECD, Eastern Europe, and Latin-America and reported a potential for CO_2 emission reduction between 0% and 29%. He further reported an average emission reduction of 22% for all countries accounted in his study. It was negligible for those countries already producing large share of blended cement or countries without iron production or coal fired thermal power plants. A large potential for blended cement is in countries without much production of blended cement (such as in the USA) and having coal fired thermal power plants and iron industries. Hendriks [2004] estimated that the global potential for CO_2 reduction through blended cement is at least 5% of total carbon dioxide emissions from cement making, but it may be up to 20%. Therefore, innovators and researchers working in the fields related to cement-based materials are exploring possibilities to develop economical, practical, and environmentally benign technologies for CO_2 sequestration in cement-based materials for reduction of carbon dioxide gas concentration already present in the Earth's atmosphere.

CARBONATION OF CEMENT-BASED MATERIALS

In concrete technology term, carbonation may be defined as a chemical process in which the pH of concrete is reduced from around 12.5 to below 9 through the absorption of carbon dioxide gas. Carbon dioxide mineralization in the hydrates of cement in cement-based material occurs either in natural way of carbonation or by some specific engineered way. Several factors such as water-to-cementitious materials ratio, portland cement and other cementitious materials type, quality of concrete, time, environmental conditions including relative humidity, level of CO_2 concentration, etc., affect rates of carbonation of cement-based materials. It is necessary to be aware of the factors that affect the rate of the process of carbonation and, therefore, CO_2 sequestration. Cement-based materials are inherently pervious; therefore, various substances (i.e., gases and liquids) penetrate the material through interconnected capillary pores originating from their surface. Carbonation of cement hydration products is a slow occurring natural phenomenon in the Earth's atmosphere. It is due to the interaction between atmospheric carbon dioxide gas and the alkaline environment of the cementitious materials matrix. Hydrated portland cement carbonates to form stable products, see Table 1. Natural carbonation is a slow process due to low CO_2 concentration in the air. Commonly used concrete in the construction of buildings carbonates due to the exposure to natural environment of the ambient air, at a rate of about one mm/year [Vasburd et al. 1997]. On the other hand, the presence of a higher concentration of carbon dioxide gas surroundings the cement-based materials such as concrete and other types of concrete products, cement plaster, controlled low strength material (CLSM), and other similar

construction materials causes accelerated carbonation at a much higher rate. Since the carbonation rate of cement-based materials exposed to the natural atmosphere is very slow, the carbonation testing and evaluation is usually performed by adopting accelerated processes in an environment of relatively higher carbon dioxide concentration [Fattuhi, 1986; Castellote

Table 1. Carbonation Products of Portland Cement Hydrates [Jana and Erlin, 2007]

Cement hydration products	Carbonation products
Calcium hydroxide	Calcite and water
Calcium silicate hydrate	Calcite, silica gel, and water
Calcium aluminate hydrate	Calcite, alumina gel, and water
Hydrated ferrite phases	Calcite, ferric oxide, alumina gel, and water
Ettringite and calcium monosulfoaluminate	Gypsum, alumina gel, and water

et al. 2008a; Sanjuan, et al., 2003; Castellote and Andrade, 2008; Chun et al., 2007]. Therefore, accelerated carbonation process is an important tool to compare carbonation rates of different types of cementitious materials including concretes.

ACCELERATED CARBONATION OF CEMENT-BASED MATERIALS

Accelerated carbonation is a method to speed up the rate of carbonation reaction in cement-based materials. Various techniques for accelerated carbonation of cement-based materials are in practice among which the use of higher concentrations of CO_2 in surrounding of cement-based materials is very common. The most widely adopted engineered way for the mineralization of carbon dioxide in cementitious materials is their early age carbonation curing. The early age carbonation curing coverts cement hydrates to stable calcium carbonate and silica gel. This provides a means to carbon dioxide sequestration in cement-based materials. This method involves use of bottled-CO_2 gas as a resource material. Several studies [Castellote et al. 2008a; Sanjuan, et al., 2003; Chun et al., 2007; Castellote et al. 2008b; Al-Kadhimi, 1996; Monkman et al. 2006; Shao et al. 2006] showed use of CO_2 concentrations, ranging from 0.15% to 100% for this method of accelerated carbonation. Sanjuan et al. [2003] conducted a comprehensive comparative study to evaluate the depth of carbonation in concretes exposed to natural and accelerated environments of carbonation. They created and used an artificial environment up to 100% CO_2 and noticed as much as 40 times greater carbonation rate for the accelerated process than the natural carbonation. The natural test consisted of exposure of the test specimens in the laboratory environment of 50 ± 5% RH and 22 ± 2 ºC, while the specimens subjected to accelerated environment had 60% RH and 100% CO_2. They further reported a five-fold increase in carbonation depth in the accelerated environment of 5% CO_2 concentration in comparison with an environment of 0.03% CO_2 concentration (found in typical ambient air). This indicated a profound effect of CO_2 concentration on the carbonation depth of concrete. They also observed a significant difference in the microstructure of the concrete carbonated at a higher CO_2 concentration verses naturally carbonated concrete. They concluded that CO_2 concentration up to 5% has significantly less effect on the microstructure compared to a higher CO_2 concentration. Castellote et al. [2008] studied chemical changes and phase analysis of ordinary portland cement (OPC) pastes carbonated at different CO_2 concentration (0.03% corresponding to natural, and 3, 10, and 100% for accelerated conditions) and found very similar microstructure for pastes carbonated in an environment up to 3% CO_2 concentration and naturally carbonated (i.e., 0.03% CO_2) pastes. However, they observed a significantly

different microstructure for paste carbonated in higher CO_2 concentration (between 10 and 100%) than that of microstructure obtained by the natural carbonation process.

Chun et al. [2007] investigated the effect of different curing environments on carbon dioxide sequestration in concrete containing ASTM C 618 Class C fly ash. In the study they used Class C fly ash at 0%, 18%, and 35% of total cementitious materials and three different curing environments (moist-curing (100% RH) and 0.15% of CO_2, drying room with 50% RH and 0.15% of CO_2, and a CO_2-chamber with 50% RH and 5% of CO_2 concentration) to investigate the carbon dioxide sequestration potential and subsequent effects on mechanical properties of concrete. Their study showed highest rate of carbonation in carbon dioxide chamber where the concentration of CO_2 was 5%. Furthermore, they observed mechanical properties of the concrete specimens kept in the carbon dioxide chamber to be similar to that for specimens cured in the moist-curing room. Al-Kadhimi et al. [1996] reported a rapid method for carbonation of concrete specimens. This method involved exposing concrete specimens up to 150 mm diameter, or 100 x 100 mm cross section, which had been dried to an internal relative humidity of 60% or less, prior being exposed to the atmosphere of carbon dioxide gas at 100% CO_2, at 1500 kPa (1.5 MPa) pressure. They claimed that such specimens could be fully carbonated in two weeks by adopting this method of accelerated curing. In their experiments, they used concrete mixture with cement content 300 kg/m^3 and a water-to-cement ratio of 0.63. A higher water-to-cement ratio was preferred to facilitate the diffusion of carbon dioxide when the specimens were exposed to 100% carbon dioxide environment. Numerous studies [Al-Kadhimi, 1996; Monkman, 2006; Shao et al 2006; Shao, and Monkman, 2006; De Schutter and Audenaert, 2004; Shi and Wu, 2008; Sims, 1994] have pointed out several advantages of the early age carbonation curing for concrete and other cement-based materials. Earlier age carbonation accelerates strength gain, leading to shortening of the time required for the production precast elements. Consequently it results in enhanced productivity for pre-cast concrete products. Durability of plain concrete is not impaired by carbonation; rather it is improved [Khunthongkeaw, et al. 2006].

Khunthongkeaw et al. [2006] performed a comparative study on carbonation of concrete with and without fly ash. They used two types of fly ashes with different CaO contents and compared the results with concrete made with cement only. They found that carbonation depth of concrete increases with the increase in the fly ash content. This finding was similar to the trend observed by other researchers [Ho and Lewis 1987; Papadakis et al 1992; Sulapha et al 2003; Shah 2005; Ramme 2008; Naik et al 2009]. Khunthongkeaw et al. [2006] further observed two to three times increase in the carbonation coefficient in concrete with fly ash content of 50% compared to that of concrete without fly ash. According to Khunthongkeaw et al. [2006] "this increase in carbonation coefficient is possibly because when a large amount of fly ash is used, the effect of reduction of CH (Ca(OH)2), by pozzolanic reaction of fly ash and by reduced cement content, dominates over the pore refinement. At the same water-to-binder ratio, fly ash delays the hydration and increases the porosity of concrete (at an early age)." They also observed higher carbonation depth for concrete incorporating low-lime fly ash (CaO = 8% or less) than high lime content (CaO = 15% or higher) at a similar fly ash content. The lower porosity of high-lime content concrete, as well as contribution of some amount of CH from this type of fly ash, were believed to be responsible for slowing down the further ingress of the carbonation front. They further reported that at an equal compressive strength at the 28-day age, the carbonation depth increases with the increase in the amount of fly ash. Jiang et al. [2000] also reported that at a given binder content, high-volume fly ash concrete showed a higher carbonation depth than ordinary portland cement concrete.

It is well known that carbonation of plain concrete is not harmful but it may be detrimental for reinforcing steel in concrete. Carbon dioxide decreases the pH of concrete. This decrease in pH destroys the passive coating that protects reinforcing steel from corrosion. Therefore, it increases the susceptibility of corrosion of the reinforcing steel in reinforced cement concrete construction. Concrete made for CO_2 sequestration requires special type of reinforcing steel or coating that could protect or reduce the chances of corrosion due to carbonation reaction before being used in reinforced structures. Study is being reported for development of such coating [Weiss et al, 2009].

CARBON DIOXIDE SEQUESTRATION IN CEMEN-BASED MATERIALS

In the past, most of the accelerated carbonation studies had been conducted to evaluate the effects of the aggressive environments on the corrosion of steel reinforcement in concrete and cover depth design. The possibilities for positive effects of carbonation in cement-based materials are the least documented studies. Some of such studies have been published based upon newer research activities [Shah 2005; Ramme, 2007; Chun et al 2007, Naik et al, 2009]. Some researchers such as Shao et al.[2006] used accelerated carbonation curing for the sequestration of CO_2 in cement-based products and claimed a consumption of carbon dioxide up to 9 to 16% by mass of cement in just two hours through their accelerated method. Accelerated carbonation in carbon dioxide rich environment makes CO_2 a resource material. Many companies claimed to have developed constructive use for carbon dioxide sequestration in cement-based materials. Carbon Sense Solutions [Inhabitat, 2008] of Canada has claimed to have developed a faster way to store more carbon dioxide in concrete through CO_2 by an accelerated curing of precast concrete elements. This procedure of the accelerated carbonation speeds up the rate of carbonation and gives an opportunity to utilize cement-based materials to mineralize carbon dioxide gas in to a stable calcium carbonate form. However, for such accelerated carbonation curing, special requirements such as a leak-proof carbonation chamber, pure carbon dioxide gas, monitoring devices for carbon dioxide concentration, etc. are required. Recently a practical and easy way of carbon dioxide sequestration in cement-based materials has been suggested by Naik et al. [2009]. Naik et al. [2009] conducted a research project for EPRI (Electric Power Research Institute) on carbon dioxide sequestration in cementitious products such as concrete and CLSM. ASTM C 618 Class C fly ash was used to replace cement in both the concrete and CLSM. Concrete mixtures were prepared with 0%, 30%, 40%, and 50% cement replaced with fly ash. The test specimens of concrete and CLSM were exposed to an environment favorable to carbonation reaction in cementitious materials (a relative humidity of 65 ± 25% and temperature of 20 ± 2 ^{o}C) until the time of testing. Most of the time, the relative humidity around the specimens was 55 to 65 %. The concrete and CLSM specimens were tested for the compressive strength and carbonation depth at different test ages (7, 28, 56, and 91 days). Typical depth of carbonations of CLSM specimens at 28, 56, and 91 days are shown in Figures 2 through 5. Figure 6 and Figure 7 show typical depths of carbonation in concrete specimens. Carbonation depth measured at different test ages for the concrete specimens are presented in Figure 8. Based on the maximum carbonation depth obtained for concrete and CLSM mixture at 91 days, Naik et al. [2009] estimated CO_2 sequestered per ton of cement used, Table 2. They reported that the maximum sequestration of CO_2 is possible in CLSM (Mixture CLSM S-1) followed by concrete with 50% cement replaced with ASTM Class C fly ash (Mixture 50). The results indicated the maximum potential for carbon credits earned per ton of cement used in CLSM followed by concrete containing 50% cement replaced with fly ash.

This confirms the possibility to sequester carbon dioxide gas in simple and practical way. The following other major conclusions were reported by them;

- ASTM Class C fly ash is very effective in sequestration of carbon dioxide in cementitious materials such as concrete and controlled low strength materials (CLSM).

Fig. 2. 28-Day Carbonation Depth of CLSM

Fig. 3. 56-Day Carbonation Depth of CLSM

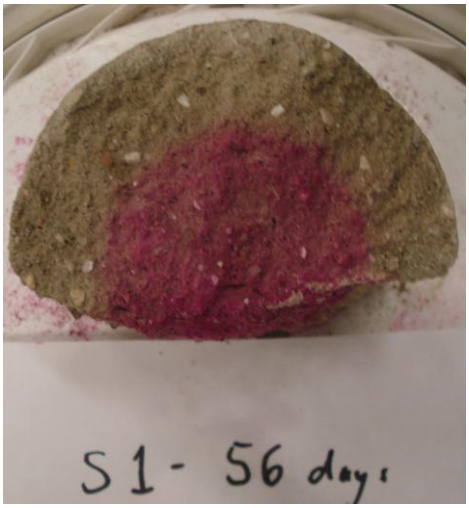

Fig. 4 . Another View of 56-Day Carbonation Depth of the CLSM

Fig. 5. 91-Day Carbonation Depth of CLSM

- The depth of carbonation increases with an increase in ASTM Class C fly ash content in concrete.
- Rate of carbonation of concrete can be increased up to three times in comparison to the concrete without fly ash, by replacing 50% cement with ASTM Class C fly ash. Therefore, such concrete has immense potential to sequester CO_2.

- A higher percentage replacement of cement by ASTM Class C fly ash causes concrete to carbonate at an earlier age (i.e. 28-day).
- CO_2 sequestration in concrete containing high amount of fly ash, i.e., 40% and 50% by mass of cement, is possible at much higher rate in comparison with portland cement concrete only.
- CLSM carbonates at a faster rate by about three to five times in comparison to concrete.

Fig. 6. Concrete Test Cylinder Showing Depth of Carbonation

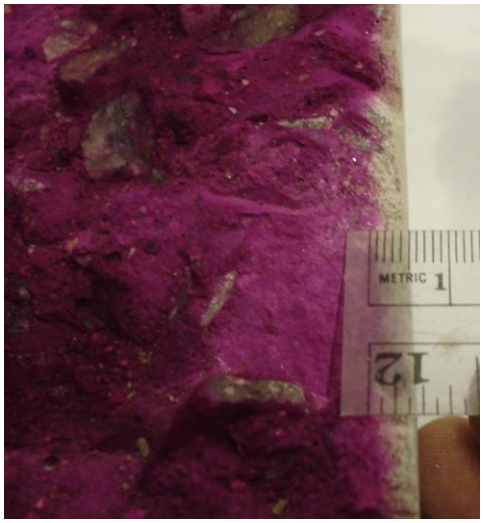

Fig. 7. Another View Showing Carbonation Depth in a Concrete

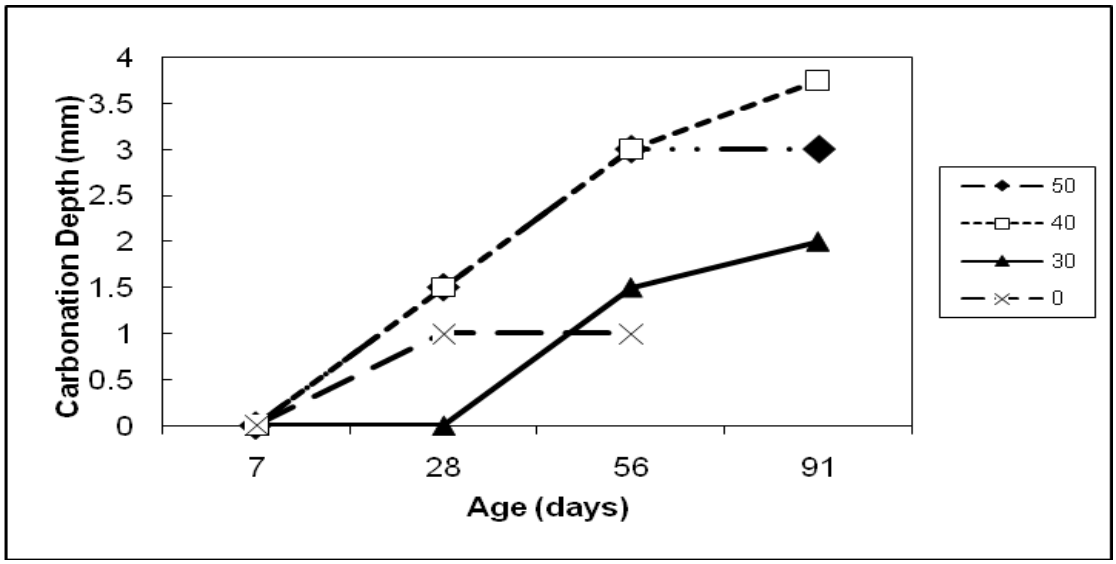

Fig. 8. Carbonation Depth of Concrete At Different Test Ages

Table 2. Carbon Dioxide Sequestered Per Ton of Cement Used

Mixture	Carbonation depth (mm)	Volume of cylinder carbonated (%)	CO_2 absorbed by cement used in the mixture (lbs)	CO_2 sequestered per ton of cement (lbs)
40	3.0	12	4.9	40
50	3.75	14	7.4	49
CLSM S-1	35	91	7.7	310

Note: 1 ton = 0.907 metric ton, 1 metric ton = 2 204.6 lbs

SUMMARY AND CONCLUSIONS

Increasing CO_2 concentration in the Earth's atmosphere has raised concerns about global warming, climate change, and effects on human health. Therefore, there is an urgent need for carbon dioxide contributing industries to develop technologies that could help in reducing CO_2 concentration in the atmosphere. Cement industries and coal-fired power plants are among major contributors of carbon dioxide in the Earth atmosphere. It is apparent from the information presented in this paper that the technology of the accelerated carbon dioxide curing gives higher rate of carbonation, consequently a quicker means for carbon dioxide sequestration in cement-based materials. Favorable conditions for rapid carbonation, even without a significantly higher concentration of carbon dioxide, allow concrete with fly ash or CLSM to sequester a significant amount of carbon dioxide through mineralization. This method of direct sequestration of carbon dioxide in cement-based materials, even during the production stage of such materials appears to be an efficient, economical, and viable. Furthermore, this technology of carbon dioxide sequestration would help cement, coal-burning power plants, concrete, precast products, and other similar industries to reduce carbon dioxide emission coming from them to a large extent. Rapid carbonation is a developing technology that has several benefits including permanent sequestration of carbon dioxide. Some of the possible opportunities from the use of the rapid carbonation technology in manufacturing of the cement-based products are.

- Safe and easy method for permanent carbon dioxide sequestration by mineralization;
- Sustainable by-products management, including ash utilization;
- Increasing productivity of pre-cast industry;
- Saving in curing water; and,
- Earning of carbon credit.

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