

CO₂ Sequestration in Non-air Entrained Concrete

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ABSTRACT

This study deals with a laboratory investigation conducted for the development of a technology for the carbon dioxide (CO₂) sequestration in non-air entrained concrete. Several experimental factors such as replacement of cement with ASTM Class C fly ash, different replacement levels of cement with fly ash, and different environmental exposures (i.e., relative humidity and carbon dioxide concentration) were adopted in this study. Carbonation test was performed to determine the depth of carbonation indicating potential of CO₂ sequestration in non-air entrained concrete. Compressive strength, splitting tensile strength, flexural strength, and abrasion resistance of the concrete were also determined to evaluate the effects of carbonation on these properties of concrete. The study revealed an increase in CO₂ sequestration potential with increase in the replacement level of cement by Class C fly ash. The study further suggested no adverse effect of CO₂ sequestration on the measured mechanical properties of concrete.

INTRODUCTION

Global warming has attracted increasingly serious interest among scientists, policy makers, politicians, journalists, and the general public around the world. Popular and academic articles, books, web sites, and television news and documentaries all provide dramatic and detailed examples of the serious consequences of ignoring the issue of global warming. One of the most prevalent greenhouse gases responsible for the global warming is the carbon dioxide (CO₂). 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 and carbon dioxide gas. Each of the primary ingredients of concrete (i.e., cement, aggregate, and water) has some adverse environmental impacts [Mehta, 2001 and 2002]. Manufacturing of portland cement, the key ingredient of concrete, is not very eco-friendly because each ton of its production releases about one ton of CO₂ and other green-house gases (GHG) into the atmosphere. Cement industry contributes about 5% of total anthropogenic CO₂ emission [Hendriks et al., 2004]. Since 1990, global CO₂ emissions from fossil fuel use and cement production has increased by about 34% [PBL, 2008]. Therefore, the cement industry is held responsible for global warming to some extent. Scientists, engineers, researchers, environmentalist, geologists along with carbon dioxide contributing industries and others, are making tireless effort to develop efficient and viable technologies in their respective areas of interest that could help in reducing carbon dioxide concentration in the atmosphere. Thus, there is a critical need that concrete construction industry and concrete technologist should evolve

some viable and economical ways for CO₂ sequestration in concrete products. This paper describes one of the ways that CO₂ can be sequestered in regular, everyday production of non-air entrained concrete.

Cement-based materials including concrete absorb carbon dioxide through a chemical phenomenon known as carbonation reaction. Carbonation reaction of cement-based materials increases the chances of corrosion of the reinforced steel by reducing their pH from 12.5. The carbonation reaction of cement-based materials results in mineralization of alkali compounds in them. About 19% of the carbon dioxide produced during manufacture of cement is reabsorbed by the concrete over its lifecycle i.e. during its service life and secondary life following crushing and reuse [CO₂ and Carbonation, 2008; Gajda, 2001]. Today, such mineralization of CO₂ in concrete and other cement-based products appears to possess an economical and viable technology for sequestration of CO₂ for reducing global warming. PCA reported that portland cement consumption in U.S. is about 100 million metric tons (110 million tons) per year; and, it has a potential for 17.3 million metric tons (19.1 million tons) of sequestration of CO₂ per year in concrete; or, a market value of about 350 million dollars (at about \$20 per metric ton of CO₂) [Gajda, 2001]. However, the natural process of carbonation in conventional concrete is very slow and its average rate is about only one mm/year [Vasburd et al. 1997]. The rate of carbonation of concrete and other cement-based materials depend on many factors, for example, the type of cement, quality of concrete, environmental conditions, and permeability of concrete [Fattuhi, 1986]. Therefore, specially designed methods, such as early age carbonation curing, porous micro structure of concrete, and other similar ideas are being tried to enable faster rates of mineralization of carbon dioxide in such materials without compromising their mechanical properties. There are some recent studies available [Shah 2005; Ramme, 2007; Chun et al 2007; shao et al. 2006; Shao et al. 2008; Naik et al, 2009] dealing with carbon dioxide sequestration potential in concrete and other cement-based products. Studies [Vasburd et al. 1997; Shah 2005; Chun et al 2007; Monkman et al. 2006] have shown many advantages such as early strength gain, increase in productivity, shortening in production time, and improvement in abrasion resistance, due to early age carbonation curing of the cement-based materials. By adopting an early age (two hours) carbonation curing chamber at 0.5 MPa pressure and at ambient temperature (23 °C) in a 100% concentration of CO₂ environment, a sequestration of 9 to 16% CO₂ by mass of the portland cement was reported [Shao et al. 2008]. The test specimens used were press-formed concrete, prepared by pressing them under a constant pressure of 8 MPa. Preconditioning of specimens at a relative humidity about 55 ± 10% at 22 ± 3 °C increases CO₂ consumption compared with the specimens pre-conditioned in moist environment with relative humidity greater than 95% at 22 ± 3 °C. Concrete with fly ash as a replacement of cement showed a higher carbonation rate than concrete without fly ash, primarily due to reduction of the amount of calcium ions to be carbonated in such concrete [Nagataki 1986, Paillere 1986; Lagerblad 2005; Shah 2005]. However, such increased carbonation effect depends on the type and amount of fly ash. Therefore, in these study effects of replacement of cement with ASTM Class C fly ash, levels of replacement, different curing environments (i.e., relative humidity) and CO₂ concentration on the carbon dioxide sequestration are discussed.

EXPERIMENTAL STUDY

Materials

ASTM Type I portland cement and one ASTM Class C fly ash fly were used in this study.

The cement and Class C fly ash met the requirements of ASTM standard C 150 and ASTM C 618, respectively. Natural sand and crushed quartzite stone of maximum size of ¾ inches (19 mm) were used as a fine aggregate and coarse aggregate, respectively were evaluated as per ASTM C 33. The specific gravity, bulk density, SSD water absorption, and void content of sand and coarse aggregates were 2.66, 1790 kg/m³ (112 lb/ft³), 1.37%, and 33% and 2.66, 1550 kg/m³ (97 lb/ft³), 0.42%, and 42%, respectively.

Mixture proportions

Nine non-air entrained concrete mixtures from three series of mixtures, Series 1, 2, and 3, were used in this study. ASTM Class C fly ash was used to replace the cement in these mixtures. The cement replacement levels were 0, 15, and 30 %. The replacement ratio of cement to fly ash was 1:1.25. Free water content was determined for trial mixtures to achieve a slump of concrete in the range of 2 to 4 inches without using a water-reducing admixture. The water to cementitious materials ratio was kept intentionally higher so that concrete matrix would be more porous in nature to allow easy ingress of CO₂ inside these concrete

Table 1. Mixture Designations

Series number	% Cement replacement	Curing Type		
		100 % RH & 0.15 % CO ₂ concentration	50 % RH & 0.15 % CO ₂ concentration	50 % RH & 5 % CO ₂ concentration
		Mixture designation		
Series 1	0	F1	F2	F3
Series 2	15	F4	F5	F6
Series 3	30	F7	F8	F9

Table 2. Mixture Proportions and Fresh Concrete Properties of Series 1 Concrete Mixtures (0% Cement Replacement)

Mixture designation	F1	F2	F3
Curing environment	100 % RH & 0.15 % CO ₂ concentration	50 % RH & 0.15 % CO ₂ concentration	50 % RH & 5 % CO ₂ concentration
Cement, lbs/yd ³	501	508	503
Fly ash, lbs/yd ³	0	0	0
% Cement replacement	0	0	0
Sand, SSD, lbs/yd ³	1490	1515	1505
¾" Aggregates, SSD, lbs/yd ³	1755	1785	1775
Water, lbs/yd ³	265	265	260
Water to cementitious material ratio, W/Cm	0.53	0.52	0.52
Slump, inch	3	2-¾	2
Air content, %	2.2	2.6	1.7
Air temperature, °F	70	70	69
Concrete temperature, °F	71	72	70
Concrete density, lb/ft ³	148.6	150.7	149.8

Note: 1 pound/cubic yard = 0.593kg/cubic meter, 1 inch =25.4 mm

Table 3. Mixture Proportions of Series 2 Concrete Mixtures (15% Cement Replacement)

Mixture designation	F4	F5	F6
Curing environment	100 % RH & 0.15 % CO ₂ concentration	50 % RH & 0.15 % CO ₂ concentration	50 % RH & 5 % CO ₂ concentration
Cement, lbs/yd ³	426	426	425
Fly ash, lbs/yd ³	94	94	94
% Cement replacement	15	15	15
Sand, SSD, lbs/yd ³	1520	1520	1515
3/4" Aggregates, SSD, lbs/yd ³	1765	1765	1760
Water, lbs/yd ³	270	270	269
Water to cementitious material ratio, W/Cm	0.52	0.52	0.52
Slump, inch	2¾	2½	3
Air content, %	1.5	1.5	1.2
Air temperature, °F	69	69	70
Concrete temperature, °F	71	70	70
Concrete density, lb/ft ³	151.0	150.9	150.4

Note: 1 pound/cubic yard = 0.593kg/cubic meter, 1 inch =25.4 mm

Table 4. Mixture Proportions of Series 3 Concrete Mixtures (30% Cement Replacement)

Mixture designation	F7	F8	F9
Curing environment	100 % RH & 0.15 % CO ₂ concentration	50 % RH & 0.15 % CO ₂ concentration	50 % RH & 5 % CO ₂ concentration
Cement, lbs/yd ³	355	354	353
Fly ash, lbs/yd ³	191	190	189
% Cement replacement	30	30	30
Sand, SSD, lbs/yd ³	1523	1520	1515
3/4" Aggregates, SSD, lbs/yd ³	1770	1765	1760
Water, lbs/yd ³	258	257	256
Water to cementitious material ratio, W/Cm	0.47	0.47	0.47
Slump, inch	3	3½	3¾
Air content, %	1.2	1.4	1.6
Air temperature, °F	70	68	70
Concrete temperature, °F	71	70	71
Concrete density, lb/ft ³	151.7	151.4	150.8

Note: 1 pound/cubic yard = 0.593kg/cubic meter, 1 inch =25.4 mm

mixtures. Mixing procedure was followed in accordance with ASTM C192. The mixture designations of the concretes are given in Table 1. The mixture proportions and properties of fresh concrete for Series 1, 2, and 3 concrete mixtures are shown in Tables 2, 3, and 4, respectively.

Specimen preparation

Concrete cylinders of 100 mm (4 inches) diameter and 200 mm (8 inches) length were prepared for evaluation of compressive and splitting tensile strengths while 12" x 4" x 3" (300 x 100 x 75- mm) beam specimens were prepared to determine the flexural strength of the mixtures. The specimens were de-molded approximately 24 hours after casting of the test specimens. After de-molding specimens, they were placed in different curing environments (100 % RH & 0.15 % CO₂ concentration, 50 % RH & 0.15 % CO₂ concentration, and 50 % RH & 5 % CO₂ concentration as given in Table 1) until the time of test.

RESULTS AND DISCUSSION

Carbon dioxide sequestration in concrete

Carbon dioxide sequestration potential of concrete can be expressed in the term of the depth of carbonation. The mechanism involved in sequestration of CO₂ in cement-based materials is conversion of unstable cement hydrates (e.g., Calcium Hydroxide – Ca(OH)₂) to stable carbonates with the help of carbon dioxide gas. Carbon dioxide does not react with cement hydrates in the gas form. First CO₂ dissolves in the pore solution to make a weak acid, carbonic acid, which in turns reacts with cement hydrates to precipitate as calcium carbonates. Carbonation depth in concrete is measured in accordance with the recommendations of RILEM CPC-18. Standard phenolphthalein, a pH indicator solution, was sprayed on the freshly fractured surface of the concrete test specimen after conducting splitting tensile strength testing. After spraying this solution on the fractured surface, the non-carbonated portion turned “pink” while the carbonated portion showed no color change and remained gray in color. This made it possible to measure carbonation depth in concrete. Figure 1 shows concrete specimens that were tested for depth of carbonation at the age of 28 days.

Figure 2 shows carbonation depth of the concrete mixtures. From Figure 2 it is evident that concrete with or without fly ash, cured in 100 % relative humidity and 0.15 % CO₂ concentration (Mixtures F1, F4, and F7) did not show any carbonation depth. This is due to fully saturated pores of the concrete, which did not allow CO₂ to penetrate inside the



Fig. 1. 28-Day Depth of Carbonation of Series 1 Mixtures

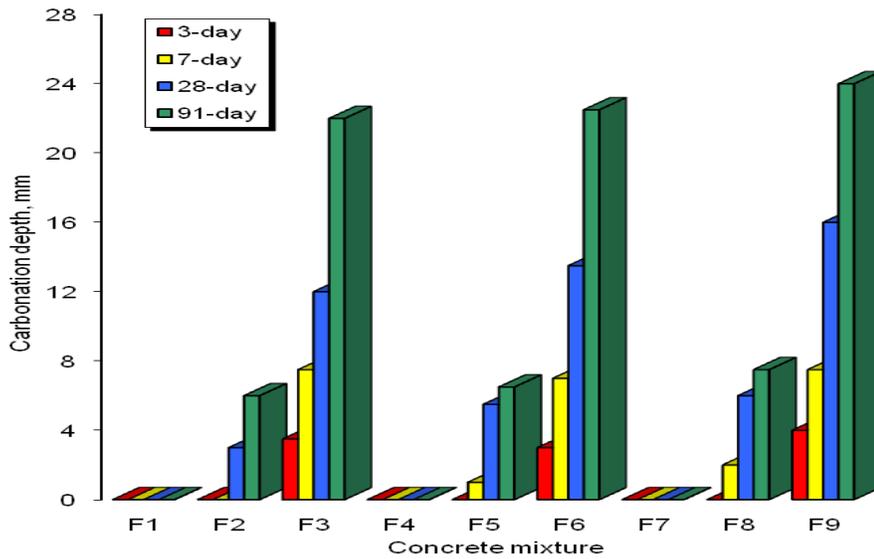


Fig. 2. Depth of Carbonation of Concrete Mixtures

concrete. Mixture F2 showed 0, 0, 3, and 6 mm depth of carbonation at the test ages of 3, 7, 28, and 91-day, respectively. Mixtures F1 and F2 were similar except for the 50% relative humidity and 0.15 % CO₂ concentration curing environment for Mixture 2. It is evident from Figure 2 that a reduction of relative humidity from 100% to 50% increased the carbonation depth for Mixture F2. Mixture F3 showed depth of carbonation of 3.5, 7.5, 12, and 22 mm at 3, 7, 28, and 91-day, respectively. Mixture F3 showed carbonation from the early age of 3 days. It had maximum depth of carbonation among Mixtures F1, F2, and F3. It is apparent from the results that for Mixture F3, which was exposed to CO₂ concentration of 5% at a RH of 50%, had a profound effect on the carbonation rate of concrete. Mixture F5 showed 0, 1, 5.5, and 6.5 mm depth of carbonation and Mixture F6 showed 3, 7, 13.5, and 22.5 mm depth of carbonation at 3, 7, 28, and 91-day, respectively. Mixture F6 showed significant depth for carbonation from the age of 3 days. Similar to Mixture F6, Mixture F9 also showed significant depth of carbonation from three days onwards. Higher depths of carbonation were observed in concrete Mixtures F9, F6, F3 followed by F8, F5, and F2, respectively, which indicated significant effect of CO₂ concentration in the curing environment than the replacement levels of cement with fly ash. It is also evident from Figure 2 that an increase in replacement level of cement by Class C fly ash slightly increased depth of carbonation in concrete cured at 50 % relative humidity with 0.15 % CO₂ concentration, as well as in 50 % relative humidity with 5 % CO₂ concentration. Also, concrete cured in high-CO₂ concentration of 5 % at 50 % relative humidity showed much higher carbonation depths than the concrete cured in 50 % relative humidity with 0.15 % CO₂ concentration. It is also evident from Figure 2 that carbonation was not noticeably accelerated in concrete with cement replacement levels of 30% compared to that of 15% fly ash at 50% relative humidity and 0.15% CO₂ concentration. However, increased replacement level of cement with fly ash made concrete to carbonate at earlier age at higher rates compared with concrete with less fly ash. Carbonation was accelerated considerably when CO₂ concentration was increased from 0.15 to 5 % at the relative humidity of 50 %.

Compressive strength

Average compressive strength obtained after testing three specimens at each test age for each mixture at 3, 7, 28, and 91 days age are presented in this Figure 3. Figure 3 showed that compared to the concrete, with or without fly ash, cured in 100 % relative humidity at 0.15 % CO₂ concentration (Mixtures F1, F4, and F7), concrete with the same mixture proportions but cured in 50 % relative humidity and 0.15 % CO₂ concentration (Mixtures F2, F5, and F8) developed relatively lower strength, while concrete with the same mixture proportions but cured in 50 % relative humidity and at 5 % CO₂ concentration (Mixtures F3, F6, and F9) developed similar compressive strengths.

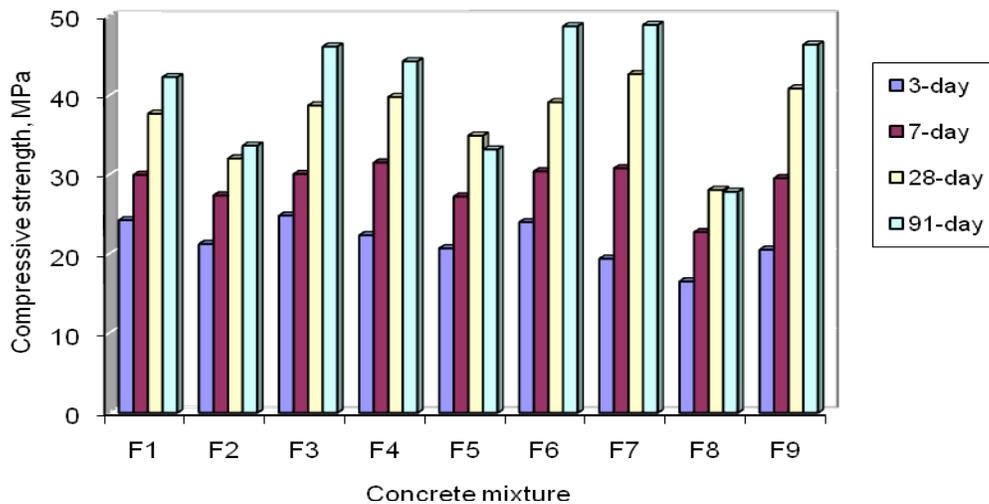


Fig. 3. Compressive Strength of Concrete Mixtures

After the 28-day age, concrete with or without fly ash and cured in 100% relative humidity and 0.15 % CO₂ concentration, or cured in 50 % relative humidity and about 5 % CO₂ concentration, continued to gain compressive strength. However, the concrete with or without fly ash but cured in 50 % relative humidity and 0.15 % CO₂ concentration practically ceased to gain compressive strength. It is most likely due to the lack of favorable hydration condition for cement particles in the case of Mixtures F2, F5, and F8 (cured in 50 % relative humidity and 0.15 % CO₂ concentration). However, in case of Mixtures F3, F6, and F9 (cured in 50 % relative humidity and at 5 % CO₂ concentration) water liberated due to carbonation reaction might have provided favorable condition for the concrete to continue to gain in strength beyond the 28-day age. Figure 3 also showed that in 100% relative humidity and 0.15 % CO₂ concentration curing environment, as the cement replacement level increased from 0 to 30 %, the 28-day and 91-day compressive strength of concrete Mixtures F1, F4, and F7 increased but the earlier age strengths of these mixtures were not significantly affected. On the other hand, in 50 % relative humidity and 0.15 % CO₂ concentration curing environment, the compressive strength of concrete increased marginally as the cement replacement level increased from 0 to 15 %, which may be due to the filling affect of fly ash leading to denser microstructure; and, decreased considerably when the cement replacement level was increased from 15 to 30 % (Mixtures F2, F5, and F8) due to the lower relative humidity curing at 50%. In the curing environment with 50 % relative humidity and 5 % CO₂ concentration, the compressive strength of concrete (Mixtures F3, F6, and F9) did not

significantly get affected by increased levels of cement replacement. The concrete mixtures in the environments as used in this study did not show improved compressive strength due to the fact that a concentration of up to 5% of CO₂ takes several days to produce carbonation, even in few millimeters at the surface of the specimens that would affect mechanical properties of concrete [Castellote and Andrade 2008].

Splitting tensile strength

Figure 4 shows average splitting tensile strength of concrete mixtures used in the study. Splitting tensile strength trends were similar to the trends for the compressive strength of concrete mixtures. It can also be seen that concrete with 30 % cement replacement and cured in 50 % relative humidity and 0.15 % CO₂ concentration (Mixtures F8) developed lowest splitting tensile strength. Concrete with 15 and 30 % cement replacement with fly ash, cured

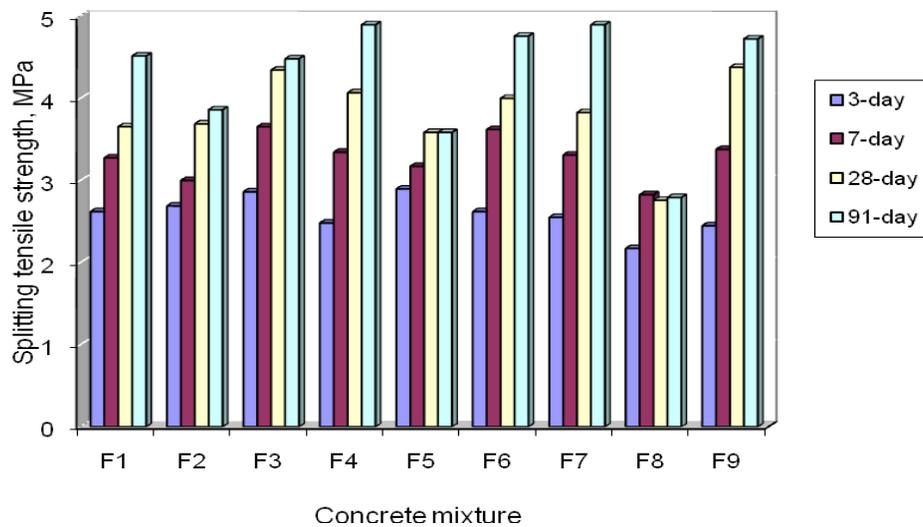


Fig. 4. Splitting Tensile Strength of Concrete Mixtures

in 100 % relative humidity with 0.15 % CO₂ concentration and 50 % relative humidity (Mixtures F4 and F7) showed slightly higher splitting tensile strength than concrete mixtures cured in the same type of environments. It indicated negligible effect of curing environments on the splitting tensile strength of concrete which is due to insignificant change in microstructure of surface concrete for these ages of concrete at these CO₂ concentrations [Castellote and Andrade 2008].

Flexural strength

The flexural strength concrete was evaluated by testing 12" x 4" x 3" beam specimens as per ASTM C78. The average flexural strength of concrete mixtures at 7, 28, and 91-day is shown in Figure 5. The trends of flexural strength of concrete mixtures were observed similar to those of the trends of compressive strength and splitting tensile strength of concrete. Figure 5 indicated negligible effect of curing environments on flexural strength of concrete due to the fact that a concentration of up to 5% of CO₂ takes several days to give

carbonation depth even in few millimeters at the surface of the specimens that would affect mechanical properties of concrete [Castellote and Andrade 2008] .

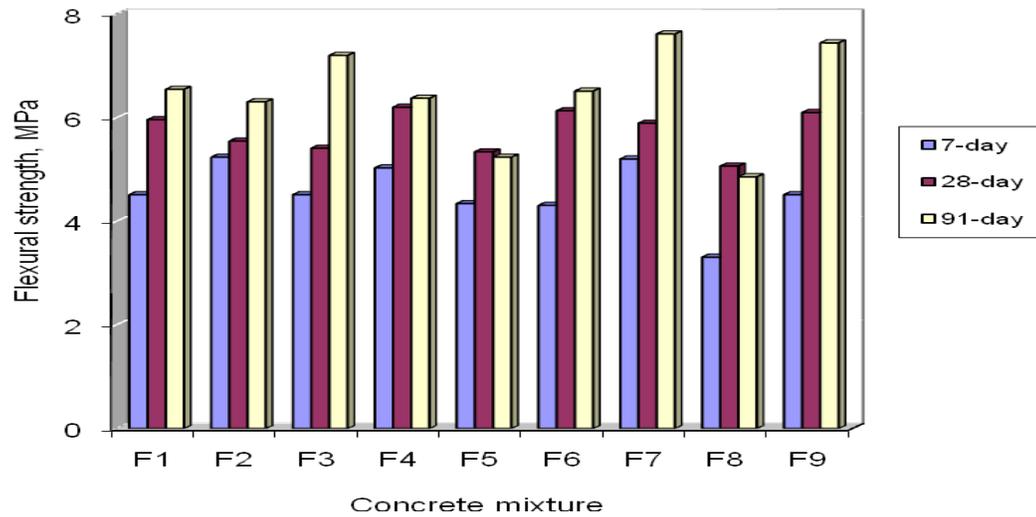


Fig. 5. Flexural Strength of Concrete Mixtures

Abrasion resistance of concrete

Abrasion resistance tests were performed in accordance with ASTM C944. To determine the abrasion resistance, 6" x 12" cylindrical specimens were used. Abrasion test specimens were cut from the top of the cylinders to obtain the specimen height of 1¾". Before performing tests, specimens were kept in the open-air environment for at least 24 hours to keep all test specimens in equivalent moisture condition at the start time of abrasion testing. Specimens were abraded for three cycles of two minutes each (total of six minutes) at 44 lbf load. Figure 6 shows abrasion resistance test results of all the concrete mixtures of Series 1, 2, and 3.

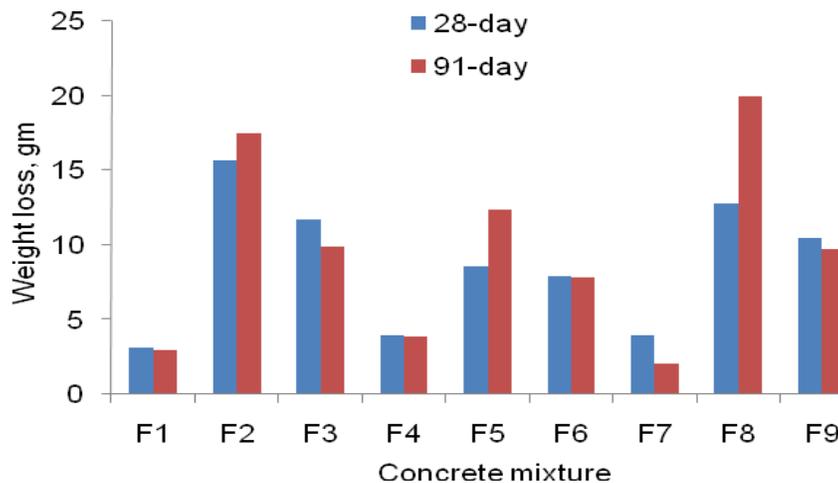


Fig. 6. Abrasion Resistance of Concrete Mixtures

Irrespective of the cement replacement levels, it was observed that concrete cured in 100 % relative humidity with the CO₂ concentration of 0.15 % (Mixture F1), showed the highest resistance to abrasion followed by concrete Mixtures F4 and F7. Concrete cured in 50% relative humidity and 0.15 % CO₂ concentration (Mixtures F2, F5, and F8) showed the least abrasion resistance. This indicated a reduction in abrasion resistance of concrete in curing environment of 50 % relative humidity and 0.15 % and 5 % CO₂ concentration that might be due to insignificant change in microstructure of the surface concrete for these ages of concrete at these low CO₂ concentrations [Castellote and Andrade 2008].

CONCLUSIONS

The following important conclusions emerged from the above study;

- The relative humidity and carbon dioxide concentration are two key factors governing CO₂ sequestration in concrete.
- Concrete with or without Class C fly ash (Mixtures F1, F4, and F7), cured in 100% relative humidity and 0.15% CO₂ concentration, did not show any depth of carbonation.
- A reduction of relative humidity from 100% to 50% at 0.15% CO₂ concentration increased the carbon dioxide sequestration potential of concrete.
- Replacement of cement with Class C fly ash enhanced the carbon dioxide sequestration potential of concrete when exposed to an environment of 50% relative humidity and 0.15% CO₂ concentration.
- At 50% relative humidity, an increase of CO₂ concentration from 0.15% to 5% in the curing environment of concrete, enhanced carbon dioxide sequestration potential of concrete considerably irrespective of the replacement level of cement with fly ash.
- All concrete (Mixtures F3, F6, and F9) cured at 50% relative humidity and 5% CO₂ concentration developed similar potential for CO₂ sequestration irrespective of the cement content.
- Concrete with Class C fly ash, when cured at lower relative humidity of 50 %, exhibited lower strength levels than concrete without fly ash.
- Replacement of cement with Class C fly ash in concrete by 15 and 30 % showed lower compressive and splitting tensile strength of concrete at three-day age and but showed higher compressive strength at 7, 28, and 91-day ages compared concrete without fly ash.
- Curing of concrete at 50 % relative humidity resulted in poor strength development and reduced resistance to abrasion.

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