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Influence of Cracking on the Physical and Chemical Properties of Concrete Containing Wastes

A. M. EL – Dakroury

Atomic Energy Authority, Hot Lab. Center, Cairo, Egypt

ABSTRACT

Fly ash, which is a by-product from combustion of pulverized coal, can partly replace the cement in concrete. The performance of concrete with fly ash is in many situations improved compared to Portland cement concretes, but there are fears regarding freeze/thaw durability. The study aims at investigate the properties of concrete with fly ash, mixed with waste with special focus on freeze/thaw durability. Cement and concrete are used extensively to isolate waste materials from the environment and to control groundwater flow rates in mining, waste disposal and site remediation activities. High quality concrete is a very low permeability material; however it is brittle and subject to cracking. In practice, the permeability of concrete is controlled by the fractures or cracks which form in the structures. The study of micro cracks in concrete is very important to understand its behavior under load and to improve this material. Damaging by using freeze-thaw exposure is often manifested in two ways. D-line cracking, or deterioration line cracking, is defined as a series of cracks in concrete near and roughly parallel to joints, edges and structural cracks. The effects of cracking on radionuclide migration and compressive strength were investigated. The results of the freeze-thaw tests, specifically the relative dynamic modulus and relative weight of each concrete mixture as a function of freeze-thaw cycles of exposure resulted that the lowest relative dynamic modulus was fairly consistently seen for the Portland cement concrete without fly ash comparing for cement with fly ash and the rate of radionuclide migration was increased. The increased radionuclide migration due to cracking could be predicted fairly.
KEYWORDS : Cementitious Materials ; Freeze- Thaw Durability ; Pozzolanic Materials.

INTRODUCTION

Current repository designs for the disposal of radioactive waste envisage extensive use of cement based materials (i.e., grouts, concrete). [1-4]. These materials may be used for the waste form matrix, , as construction materials for walls and floor, as a grout to fill in gaps between waste packages, and to seal boreholes, shafts, tunnels, and natural fractures or fractures generated during vault excavation . The cement based materials will fulfill different functions in each of these applications. Cementitious material used in these applications due to its inherent strength, durability, and versatility. Cement based grouts have been identified as potential grouting materials. The required performance of cement based grouts includes not only an ability for the material to penetrate and seal very fine fracture , but also a potential for long – term mechanical and chemical stability in disposal vault environment. Under many conditions, the deterioration of cement based materials is not attributed to any single cause, but arises from the combined action of a number of potentially destructive agents. There are many possible environmental factors and processes by which the design properties of the cement based grouts may change over long periods of time. These include reaction with the environment factors and processes by which the design properties of the cement based grouts may change over long periods of time. These include reaction with the environment through the aqueous phases, internal microstructure change, temperature, action of microorganism, and radiation. It is commonly held that the hydraulic performance of cement based grouts would. Change as water passed through the pores in the materials. Cracks in cement structures can have negative effects on important properties such as permeability, rate of radionuclide migration and compressive strength [5].

Fly ash, (FA) the most widely used supplementary cementitious material in concrete, is a byproduct of the combustion of pulverized coal in electric power generating plants. Upon ignition in the furnace, most of the volatile matter and carbon in the coal are burned off. During combustion, the coal's mineral impurities (such as clay, feldspar, quartz, and shale) fuse in suspension and are carried away from the combustion chamber by the exhaust gases. In the process, the fused material cools and solidifies into spherical glassy particles called fly ash . The fly (FA) ash is then collected from the exhaust gases by electrostatic precipitators or bag filters. Fly ash is a finely

***Corresponding Author:** A. M. EL – Dakroury, Atomic Energy Authority, Hot Lab. Center, Cairo, Egypt,
Email: n.h.mansoub@gmail.com

divided powder resembling Portland cement (P) . Most of the fly ash particles are solid spheres and some are hollow cenospheres. Also present are plerospheres, which are spheres containing smaller spheres. Ground materials, such as Portland cement (P), have solid angular particles. The particle sizes in fly ash vary from less than 1 µm (micrometer) to more than 100 µm with the typical particle size measuring less than 20 µm. Only 10% to 30% of the particles by mass are larger than 45 µm. The identification of a universal model for the hydration of Portland cement and fly ash mixtures requires the study of the mechanisms of the hydration of Portland cement and fly ash, as well as their interaction during the process. Hydration of PC is consists of a series of simultaneous chemical reactions related to the hydration of the individual materials constituting PC. Very often, for the purpose of presenting a model for the process of Portland cement hydration, the reaction of alite (C_3S) and water is used, *i.e.,* alite hydration.[6]. On the basis of the simplified model of alite hydration [7] and the principles of the progress of pozzolanic3 reactions, the process of (PC+FA) hydration is presented in Fig. 1 as a multiphase process, where an: early, medium and late period [12]

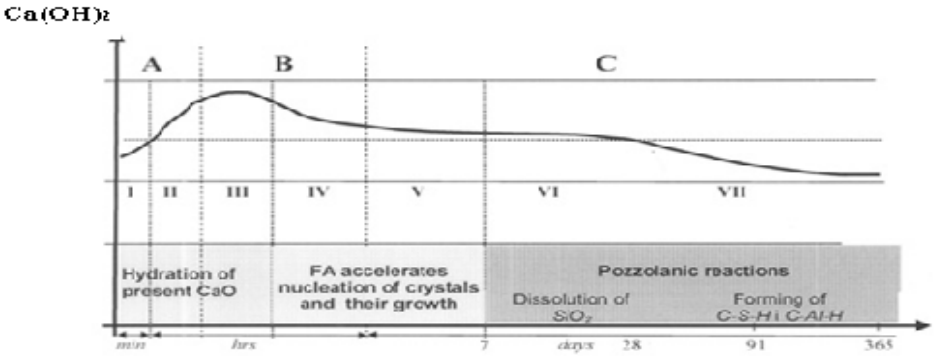

Fig.1 Physical model of the hydration of PC+FA mixture[12]

Early period

I preinduction- hydration of portland cement and creation of C-S-H, C-Al-H, and $Ca(OH)_2$ products

II Induction- stagnancy period

Medium period

III acceleration phase

IV nucleation phase

Late period

V crystal growth

VI dissolution of amorphous SiO_2

VII pozzolanic reactions

Hydration can be differentiated. A parameter of the progress of the hydration reaction of a PC and FA mixture is the change of the concentration of $Ca(OH)_2$. In the earliest period, after several minutes, the first hydration of alite and belite (C_2S) from Portland cement with the release of hydration heat occurs:[7]

2(3CaO·SiO2) + 6H2O = 3Ca·2SiO2·3H2O + 3Ca(OH)2 + _*H*1 (C3S2H3) (1)

2(2CaO·SiO2) + 4H2O = 3CaO·2SiO2·3H2O + Ca(OH)2 + _*H*2 (C3S2H3) (2)

As a product of the cement hydration reaction $Ca(OH)_2$ is formed. The higher presence (concentration) of $Ca(OH)_2$ in cement paste causes an instability of concrete exposed to the action of soft water and acidic solutions, as well as to the action of high temperatures. After the early period of setting, there is a stagnation period, during which the emission of hydration heat is relatively low. The physical changes in the cement paste during this period can be seen in its gradual hardening. After the period of stagnacy, in the medium phase, the reaction accelerates and a new hydration of the cement occurs. The maximum is reached [9–10] h after the beginning of the reaction. Due to the increase of the OH– ion concentration, as a result of the generation of $Ca(OH)_2$, the environment becomes progressively alkaline. Fly ash which is negligibly activated in this early period, acts as an inert material which accelerates the setting of the cement paste by acting as a nucleus for the sedimentation of C–S–H, C–Al–H and $Ca(OH)_2$, which appear after cement hydration.[8,9] In the nucleation phase, the final formation of the structure of the cement paste occurs. In the late period, the cement paste hardens, while the pH value increases in the pores of

the cement paste, which affects the dissolution of the molecules of amorphous SiO_2.[10] $Ca(OH)_2$, created by the hydrolysis of alite and belite, behaves as an activator of the latent hydraulic properties of fly ash and reacts, in so-called pozzolanic reactions, with the active part of the fly ash (SiO_2, Al_2O). In this way, the negative influence on the quality of cement can be reduced or entirely neutralized:

$$2SiO_2 + 3Ca(OH)_2 = 3CaO{\cdot}2SiO_2{\cdot}3H_2O\ (C_3S_2H_3) \qquad (3)$$

$$3CaO{\cdot}Al_2O_3 + CaSO_4 \cdot 2H_2O + 10H_2O = 4CaO{\cdot}Al_2O_3{\cdot}SO_3{\cdot}12H_2O(C_4A\_H_{12}) \qquad (4)$$

$$Al_2O_3 + CaSO_4 \cdot 2H_2O + 3Ca(OH)_2 + 7H_2O = 4CaO{\cdot}Al_2O_3{\cdot}SO_3{\cdot}12H_2O(C_4A\_H_{12}) \qquad (5)$$

$$Al_2O_3 + 4Ca(OH)_2 + 9H_2O = 4CaO{\cdot}Al_2O_3 \cdot 13H_2O\ (C_4A\_H_{13}) \qquad (6)$$

As the pozzolanic reactions progress, the fly ash particles lose their spherical form and become increasingly coated by a layer of the product, and after a period of [11] months they can no longer be identified.[11] During the hydration of cement with added FA, the same hydration products appear as during the hydration of Portland cement. There are data[12] supporting the fact that many cements with added fly ash have better mechanical properties than PC itself. The increased resistance to corrosion of cements with added FAis accounted for by the lower content of $Ca(OH)_2$ in the cement paste, and thus in the concrete. For the same reasons, Portland cement with added FA behaves better when exposed to high temperatures.[13.14]

Objectives

The objectives of this research project are:

Investigating properties of concretes containing fly ash, with special focus on the freeze/thaw durability. The results from the research are expected to improve the knowledge regarding freeze/thaw durability of concretes with additions of fly ash .To analyze the chemical components of the type of fly ash produced in Egypt ,to provide the appropriate concrete mix that can utilize fly ash efficiently, To give an overview of possible applications in which fly ash could be utilized in concrete mixes used for immobilization of radio wastes and to provide a cost analysis for replacing concrete with fly ash.

Materials mixing and specimens

Cement mixtures with W/C =0.4 with fly ash (15%) and without any additives. The tests were conducted using ordinary Portland cement which is kindly supplied from Suez Cement Co., Egypt, the measured Blain surface area was ~ 0.350 cm^2/kg with W / C = 0.4 ,and mixed with and C_s^+ (as radioactive waste) Control specimens were prepared without any stress of freez- thaw cycles,. First, the specimens with 7x7x7 cm, for measuring compressive strength and cylindrical (Φ2.5 x5) mm in size were made to measure diffusion coefficient. The cylinders were stripped after 48 hr. and cured in lab. (~90% relative humidity and 28±2°C)for 28 days , and for one month prior to the heating and cooling testing .Table (1) gives the chemical composition of Ordinary Portland Cement and fly ash .

Table 1. Chemical Composition of Materials (%).

Contents	Cement (OPC)	Fly ash
*LOI	2.67	3.5
SiO_2	21.14	59.64
CaO	61.2	1.98
MgO	2.67	0.85
Fe_2O_3	3.92	4.38
AL_2O_3	4.8	27.0
SO_3	2.08	-
Na_2O	.2	0.23
K_2O	0.80	1.28
CI	.008	0.01
P_2O_3	0.32	0.45
TiO_2	0.10	traces
Mn_2O_3	0.07	traces
**IR	3.06	90.41
Free CaO	0.5	-
Sulphide	-	-
Glass content	-	32
Density	3.14	2.25
Fineness (cm^2/ gm)	3044	3650

***LOI, Loss of ignition at 1000 C ; **IR, insoluble residue**

Freeze-Thaw Resistance Testing

All of the freeze-thaw resistance testing in this study was performed in substantial accordance with ASTM C 666. In a constant temperature water bath, the test specimens were initially brought to the control temperature of 90±2 °F after curing for 12 days. Once the control temperature had been reached, the specimens were removed from the bath one at a time and initial readings of transverse and longitudinal frequency were taken, as well as the weight and dimensions. The specimens were then returned to the freeze-thaw cabinet and the test was begun. A central reference specimen was monitored to record the number of freeze-thaw cycles and temperatures between testing intervals. Each sample was then periodically removed from the cabinet for measurement of transverse frequency in intervals that were not to exceed 36 cycles of freezing and thawing. According to ASTM Test Method C 666, the freeze-thaw resistance test is considered complete when either the specimen has been subjected to 120 freeze- thaw cycles.

Table 2. Proportioning Of Mixed Tested.

Mix no.	Cement %	Fly ash %	Mixed with radionuclide
C	100	0	no
CC_S	100	0	Mixed with Cs^{+137}
CF	85	15	no
CCsF	85	15	Mixed with Cs^{+137}

Measurements of Crack Density.

The crack density caused by rabid freeze / thaw exposure was measured on polished sections impregnated with a red dye, [15]. The impregnation procedure involves no drying that may increase the micro cracking. Two polished sections were made at each level of deterioration freeze / thaw cycles. Crack density was measured by counting number of cracks traversing parallel lines of approximately 5 mm distance. 1600 mm traversing length was counted for each level of deterioration.

Leaching Rate Mechanism (leaching Model)

To study the mechanism of leaching, it is very important to estimate the longer period of time that will be the leaching rate in the future period till the at very low radioactive. The leaching usually follows diffusion type mechanism especially of "Fick's" equation which follows the equation [16]

$$\left(\frac{\sum A_i}{A_0}\right).\frac{v}{s} = 2\left(\frac{D_e}{\pi}\right)^{1/2}\left(\sum t_i\right)^{1/2} \qquad (1)$$

$$F = \frac{2s\sqrt{Dt}}{v\sqrt{\pi}} \qquad (2)$$

where F = The fraction leached at time t ;
D_e= The diffusion coefficient cm^2/day;
S = Surface area of specimen cm^2;
V = Volume of specimen cm^3.

The value of D_e can be calculated from the slope m of the linear relation between F and $t^{1/2}$

$$m = \frac{2(D_e/\pi)^{1/2}}{v/s} = \frac{2s\left(\frac{D_e}{\pi}\right)^{1/2}}{V} = 2S\sqrt{D_e}/v\sqrt{\pi}$$

$$D_e = \pi m^2 v^2/4s^2$$

The amount of a radionuclide leached from the waste composites over a given period can be predicted from equation (2).

RESULTS AND DISCUSSION

Durability data The fundamental frequency and weight of each specimen were recorded initially and approximately every 12 cycle until failure. a corresponding weight was obtained for each fundamental frequency reading . One set

of reading for some typical specimens can be found in fig.2,3. only weight change and to failure were recorded for these specimens. The transverse dynamic modulus of elasticity was calculated using the equation given in ASTM C 215 [14] , as shown below.

$$\textbf{Dynamic E} = CWn^2 \quad (1)$$

Where
W = weight of specimen, Ib
n = fundamental transverse frequency, Hz

$$C = 0.00245 \{L^3 T / bt^3\} \{sec^2 / in.^2\} \quad (2)$$

L = length of specimen, in. d = diameter of cylinder, in;
t, b = dimension in driving direction and other cross- sectional dimension , respectively;
T = correction factor based on ratio of radius of gyrations length of specimen and on Poisson;
Ratio

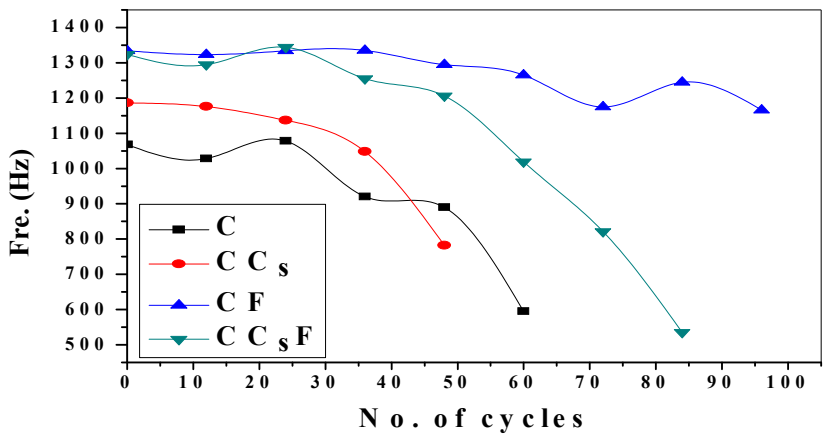

Fig 2 fundamental Frequency reading (Hz) with No. of cycle

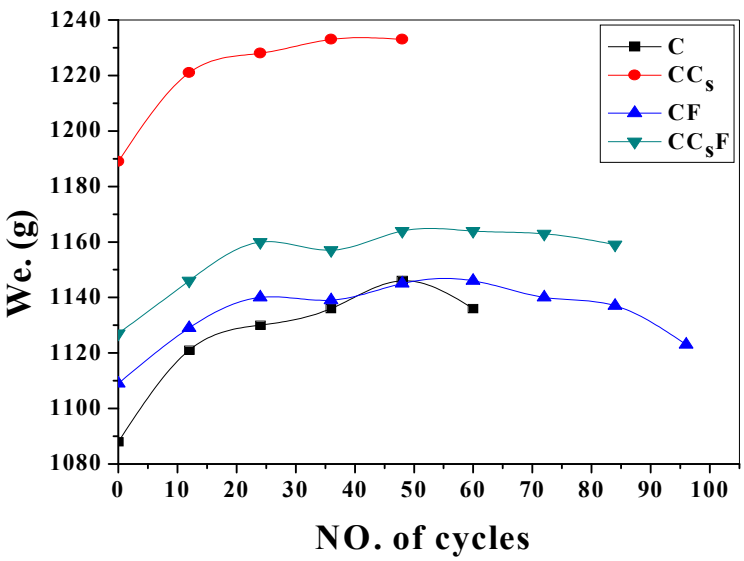

Fig3 Weight (g) with No. of cycle

The radius of gyration K is equal to d / 4 for a cylinder and 1 /(3.464) for a prism. Fig. 4 show the values of T agented K / L. The decrease in dynamic modulus and weight were used to detect internal cracking due to freeze – thaw cycling ; values calculated for the specimens (CF) are show in fig 5 The change in dynamic modulus was calculated for each reading by dividing the value of the dynamic modulus for that reading by the initial dynamic modulus of the specimen

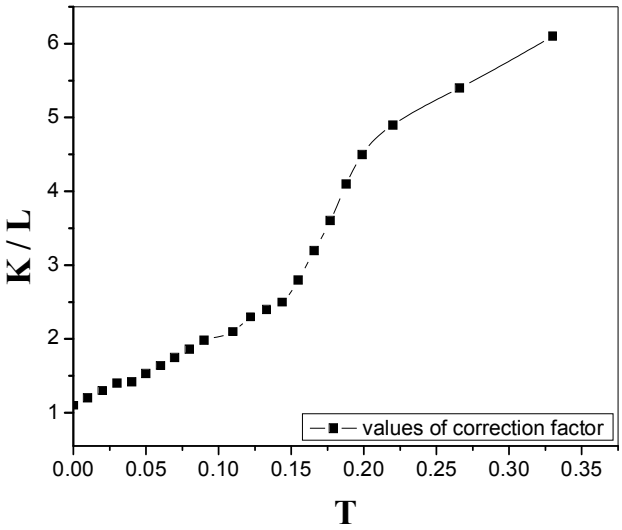

Fig. 4. Values of Correction Factor T"

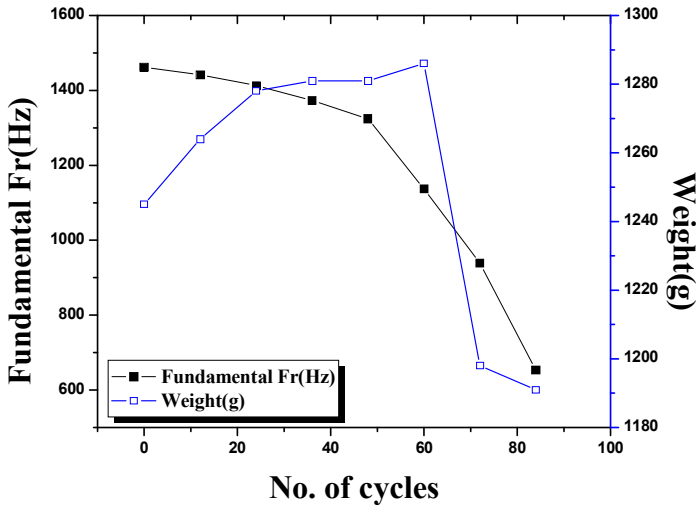

Fig. 5. Dynamic modulus of elasticity and weight for Specimen of 85 % cement With 15% fly ash (CF)

The percentages for specimen cement can be seen in table 3. Three different durability factors [DF", DF(60) , and DF (84)] were used to calculated each of the specimens . Durability Factor (DF) was calculated as the number of cycles at which the unites reached 60 percent of their original dynamic modulus or the number of cycles at which the specimen failed if the specimen failed before reaching 60 percent of its original modulus. Since readings were taken only every 9 to 15 cycles, linear interpolation was used to determine the number of cycles at which exactly 60

percent of the modulus was reached. For specimen 2-11, the durability factor (DF) was calculated as:

$$\mathbf{DF = 67 + \{61 - 60 / 61 - 40\} (77 - 67) = 67.5 \text{ cycles}} \qquad (3)$$

The durability factor DF (60) was calculated using the method given in ASTM C 666. For specimen 2-11 using 60 percent of the initial dynamic modulus as a failure criterion and 100 cycles as a criteria for passing, the relative dynamic modulus is.

Table 3. Percent of original dynamic modulus and percent of original dynamic modulus psi Specimen of 85 % cement with 15% fly ash (CF)

No. of cycles	Dynamic modulus psi	% of original dynamic modulus psi
0	2100000	100
12	2100000	96
26	2300000	104
38	2000000	93
48	1800000	85
60	1300000	62
72	820000	41
84	380000	15

$$\mathbf{P = \{(1{,}300.000)^2 / (2.100.000)^2\} (100) = 61.9} \qquad (4)$$

The durability factor Df (60) was calculated as

$$\mathbf{DF\ (60)_{2\text{-}11} = 61.\ (67) / 100 = 41.5} \qquad (5)$$

The same procedure was used to calculate the durability factor DF (80), but 80 percent of the dynamic modulus was used as a failure criterion. For specimen (cement .The changes in modulus for the specimens in Table 3.

$$DF\ (80) = \{(1{,}300.000)^2 / (2.100.000)^2\}\ (100)\ (49) / 100 = 36.0 \qquad (6)$$

Absorption measuring

Absorption testing was carried out in accordance with ASTMC140 [14] . Each unit was weighed while saturated, both in air and while suspended in water, and again after oven drying. After converting weight measurements to pounds, the density, net volume, and absorption were calculated using the equations given in ASTMC140. Average values for these properties for the mixes tested can be found in table 7 . An example of the calculation is given below for one of the specimens

Saturated weight while suspended in water (C) = 3462.9 g = 7.6 Ibs

Saturated surface dry weight in air (A) = 456 g = 20.Ibs

Oven dry weight in air (B) = 8315.6 = 18.3 Ibs

Density = {B / (A-C)} (62.4)

$$= 18.3\ (62.4) / 20. - 7.6 = 87 \qquad (7)$$

$$\text{Net volume} = B/ D = 18.9 / 87 = 0.21\ ef \qquad (8)$$

$$\text{Absorption} = \{(A-B) / (A-C)\}\ 62.4$$

$$\{[(20.9 - 18.3) / (20.9 - 7.6)\}\ (62.4) = 11.87\ pef \qquad (9)$$

Table 4. Results of absorption tests.

Material	No. of cycle	Density , pef	Adsorption	Material	No. of cycle	Density , pef	Adsorption
C	0	70	17.91	CF	0	87	11.51
	12	63	19.2		12	80.	12.3
	67	57	21.3		67	71.	13.6
	88	46	23.5		88	62	15.2

compressive strength

Fig 6 shows the compressive strength, as affected by freeze / thaw. For cement without additives freeze / thaw cycles reduced the strength to 68 – 40 %.

After using fly ash as 15 % the reduction of compressive strength decrease to 25% .These may be attributed to chemical components of fly ash contributing to this reaction include tetra calcium trialuminate sulfate, tricalcium aluminates anhydrate, amorphous silica glass, and lime.

When water is added, these substances and other trace minerals combine in various reaction to form monosulfoaluminate, calcium silica hydrate (tobermorite gel) , calcium aluminates ferrite silicate (hydrogamet)

and other compounds which increase the CSH leads to a reduction in capillary pores and increase impermeability. This dual effect gives a concrete with increased resistance to freezing and thawing cycles. [18-19]

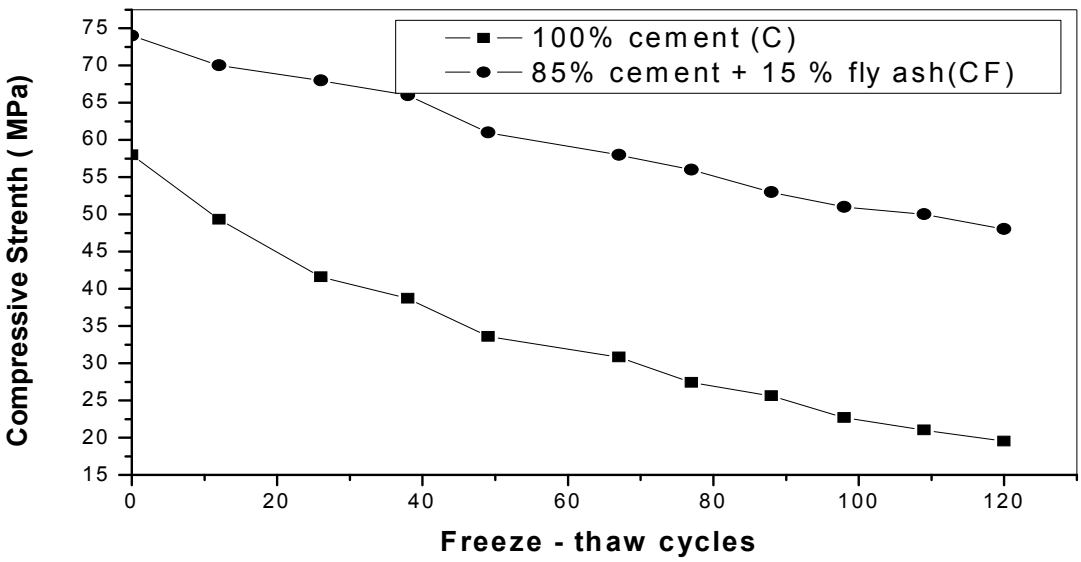

Fig 6. the compressive strength, as affected by freeze/ thaw after cycles 28 days

Volume, Mass, Density and Absorption

Table 4 shows changes in volume, mass, density and water absorption of the specimen of cement without fly ash during freeze / thaw exposure. The values are significant, and indicate that the created crack volume is filled with water, since the volume increase equals volume of water absorbed. The volume increase due to internal cracking is in good accordance with previous measurements [19]. Due to the increase volume and absorption of water, the density was relatively reduced.

Table 8. Changes in volume, mass, density and absorption of cement specimen without fly ash during freeze / thaw exposure

Parameters	Exposure cycles					
	0	12	38	67	98	120
$\Delta V / V_o$	0	0.3	0.7	1.7	2.9	4.5
$\Delta m / m_o$	0	0.45	1.0	1.9	2.9	5.1
Density(kg/ m^3)	2.479	2.471	2.253	2.11	2.0	1.89
Absorption data	10.43	11.41	13.36	14.2	14.9	17.91

Table 9. Changes in volume, mass, density and absorption of cement specimen of cement with fly ash as 15 % during freeze / thaw exposure.

Parameters	Exposure cycles					
	0	12	38	67	98	120
$\Delta V / V_o$	0	0.2	0.57	1.2	1.8	4.0
$\Delta m / m_o$	0	0.34	.087	1.8	2.1	4.9
Density(kg/ m^3)	2.623	2.542	2.398	1.9	1.7	1.23
Absorption data	9.2	10.0	11.54	12.78	13.34	15.87

RADIONUCLIDE DIFFUSION

Leaching tests are used as a tool to estimate the release potential of constituents from waste materials over a range of possible waste management activities, including during recycling or reuse, for assessing the efficacy of waste treatment processes, and after disposal. .They may also be used to develop end points for remediation of contaminated soils and the source term for remediation of contaminated soils and the source term for environment risk characterization Cs being the most mobile water-soluble species in nuclear waste streams. Further reduction in

leaching of all radionuclides can be expected when careful consideration is given to matrix modification and developed for structural applications by using admixture like fly ash were primarily Samples for leachability determination were prepared according to the IAEA standard procedure [16]. Non-radioactive CsC1 was used in this to sudsy. Cs was selected due to its known high leachability from unmodified cement. All ingredients were mixed in a small mixer, and then cast into cylindrical moulds of diameter 3.8 cm (1.5 inch) and height 3.8 cm (1.5 inch). The specimens were demoulded after 24 h curing at room temperature and 90% relative humidity. Subsequently, the specimens were placed in 1L polyethylene containers filled with 500ml deionized water. The water volume to specimen surface area ratio is 10. At fixed time intervals (every 2 days), the concentration of Cs in the leachate was measured by using atomic absorption spectroscopy. After sampling for analysis, the leachate was totally replaced with fresh deionized water. The test was performed at 25 °C ± 2 from 1 day up to 35 days. Three specimens were prepared for each material, with the average value reported here. The quantity of Cs leached and the leach rate are plotted in Figs 7 and 8, respectively. The total amount of Cs leached at 35 days is greatest in the mortar at 120 cycles, followed by the 80cycles, and 0cycles as a result of reduction in the number of cracks and the crack widths (Fig. 7) However, when cracks are larger and widely spaced, resistance through the adjacent porous medium (i.e., cement) governs flow rates through cracks. The reductions in total quantity of Cs leached from CCsF (paste), compared to the CCs (paste), are 69%. Fly ash proved to be an effective sorbent for Cs, (Fig 8)[20,21]

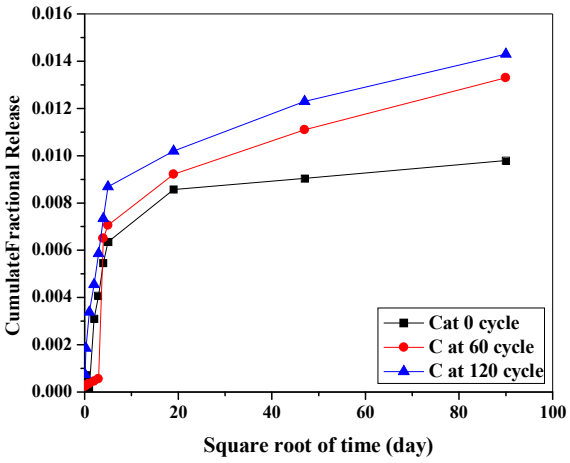

Fig. 7 Effect of cracking on the leaching rate of ^{137}Cs at different cycles of freeze/thaw cycle for cement.

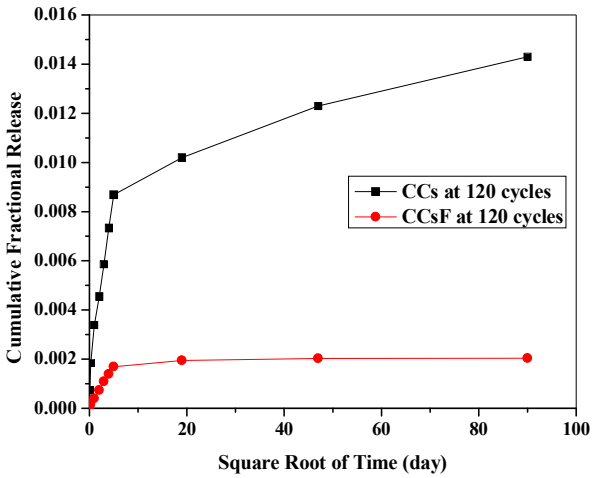

Fig. 8 Comparison between CCs and CCsF for the leaching rate Cs ion under the effect of freeze/thaw.

CONCLUSION

The freezing and thawing cycles that can cause internal cracking for cement It is commonly accepted that there are two basic forms of deterioration induced by freezing and thawing: internal cracking due to freezing and thawing cycles, and surface scaling, generally due to freezing of water to ice and the accompanying

expansion causes deterioration either of the hardened paste, aggregate, or both. Hydraulic and osmotic pressure develop in the pores when water freezes and expands. Water migrates to locations where it can freeze and ice develops in cracks and crevices that act to pry the cracks open wider. The magnitude of the pressure depends on the rate of freezing, degree of saturation, permeability of the concrete, and the length of the flow path to the nearest place for the water to escape. Concrete should be resistant to damage from freeze-thaw cycles if the concrete has gained sufficient compressive strength by adding mineral additives like fly ash which increase the CSH leads to a reduction in capillary pores and increase impermeability. This dual effect gives a concrete with increased resistance to freezing and thawing cycles.

Incorporation of pozzolanic and supplementary cementitious materials like., fly ash,. as partial replacement of portland cement) are known to increase the density, impermeability, and other properties of concrete microstructure by a combined effect of: (a) grain-size refinement (replacement of calcium hydroxide component of portland cement hydration to calcium silicate hydrate by pozzolanic reaction) and (b) pore-size refinement (densification of overall microstructure by filling of capillary pores in the paste and in various interfaces by the calcium silicate hydrate pozzolanic reaction product

Leaching tests are used as a tool to estimate the release potential of constituents from waste materials over a range of possible waste management activities, including during recycling or reuse, for assessing the efficacy of waste treatment processes, and after soils and the source term for remediation of contaminated soils and the source term for environment risk characterization

Cesium interacts with concrete, and may be incorporation into calcium-containing hydrated phases or sorption onto high surface area C-S-H. .This interaction is affected by the presence of micro cracks. The results indicate that leaching rates of wastes affect when exposure to freeze/thaw cycle.

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