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Reduction of cement consumption by producing smart green concretes with natural zeolites

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Abstract. This study was carried out to evaluate the natural zeolite in producing green concrete as an effort to prevent global warming and environmental impact problems of cement industries. To achieve this target, two types of natural zeolites applied to study physical, chemical and compressive strength of concrete containing different percentages of zeolites. The results in comparison with control samples indicate that compressive strength of zeolites mixes increases with the 15% replacement of zeolite instead of cement in all types of samples. In the water-cement ratio of 0.6, results showed an increase in the compressive strength of all percentages of replacement. This results indicate that using natural zeolites could be produced a green concrete by a huge reduction and saving in the consumption of cement.

Keywords: cement; natural zeolite; green concrete; compressive strength

1. Introduction

Concrete, the most destructive material on earth is the single most widely used material in the world. It is an important construction material used extensively in buildings, bridges, roads and dams (Luo *et al.* 2019, Xie *et al.* 2019, Shariati 2008) (Shariati 2008). Its uses range from structural applications, to paviors, kerbs, pipes and drains (Nosrati *et al.* 2018, Sajedi and Shariati 2019). Concrete as a composite material has different range of preparation depending on its application like its usage as high strength concrete (Arabnejad Khanouki *et al.* 2011, Shariati *et al.* 2011b, Shariati *et al.* 2012a, Shariati *et al.* 2012c, Shariati *et al.* 2012d, Shariati 2013, Mohammadhassani *et al.* 2014a, Mohammadhassani *et al.* 2014b, Mohammadhassani *et al.* 2014c, Shariati 2014, Shariati *et al.* 2014a, Shariati *et al.* 2014b, Khorramian *et al.* 2015, Shariati *et al.* 2015, Khanouki *et al.* 2016, Shahabi *et al.* 2016, Shariati *et al.* 2016, Khorramian *et al.* 2017, Nguyen-Sy *et al.* 2017, Hosseinpour *et al.* 2018, Nasrollahi *et al.* 2018, Shao *et al.* 2018, Ziaei-Nia *et al.* 2018) or light weight concrete (Shariati *et al.* 2010, Hamidian *et al.* 2011, Shariati *et al.* 2011a, Shariati *et al.* 2011c, Shariati *et al.* 2012b, Shariati *et al.* 2017, Vo-Duy *et al.* 2017, Dinh-Cong *et al.* 2018, Ho-Huu *et al.* 2018, Davoodnabi *et al.* 2019, Shao *et al.* 2019) which are commonly used concrete in structures. Application of this important material in structure with special characteristics is extended to some other important usage in the life like pavement and roads (Abedini 2017 and Abedini 2019). Pervious concrete (no-fines concrete) is one of them (Toghroli *et al.* 2017, Li *et al.* 2019).

It's a mixture of hydraulic cement, smaller coarse aggregates, admixtures and water, percolated by the concrete into subbase recharging the under-ground water levels (Sinaei *et al.* 2011, Ghassemieh and Bahadori 2015, Ghosh *et al.* 2015, Bahadori and Ghassemieh 2016).

Annually, over 10 billion tons concrete are used in the world (Juenger *et al.* 2011). To produce this amount, approximately 3 billion tons of Ordinary Portland Cement (OPC) is required. Estimates for global demand for OPC show that cement consumption will reach nearby 6000 Mt/year in the next 40 years (Pacheco-Torgal *et al.* 2012). Some authors have estimated that the process of producing 1 ton of OPC generates about 0.94 ton of carbon dioxide.

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The cement industry emissions are about 7% of total worldwide carbon dioxide. Thus, it is known to be one of the most pollutant industries (Gartner 2004, Pacheco-Torgal *et al.* 2012). It is well documented that the production of OPC generates huge amount of carbon dioxide (Damtoft *et al.* 2008). Since, the cement industry is one of the biggest polluters and consumers of natural sources this industry should use some other cement supplementary. Reducing carbon dioxide emissions of this industry gives us incentives to investigate supplementary cementitious materials including natural pozzolans, such as zeolite, fly ash and silica fume (Ahmadi and Shekarchi 2010). Mix of zeolite-lime tuff was used in construction widely from ancient times. Nowadays more than 50 types of natural zeolite and 150 types synthetic zeolite have been known and are used in many industries (Feng and Peng 2005). Recently in some countries, natural zeolites are widely used as pozzolanic materials in concrete, such as China which is allocated in the first place by consumption of approximately 30 million tons per a year. Also the consumption of zeolite has been reported in Russia, Spain, Serbia, Cuba and Slovenia (Feng and Peng 2005, Colella 2007).

Natural zeolite has a three-dimensional structure and classified as alkali hydrated Aluminosilicates and cations Alkaline. Total surface of zeolite (internal and external) is 35 to 45 m^2/gr (Mumpton 1977). Microscopic structure of spongy zeolite has possibility of absorption and confined chemical substances including heavy metals, ammonia, calcium, anionic, cationic, gases and liquids (Englert and Rubio 2005). So, these features of zeolite which can be absorb or exorcise water as much as 30% of their dry weight is well known. Moreover, the main component of positive ions can be changed without any significant changes in the structure (Cejka and van Bekkum 2005). Zeolites aluminosilicate great surface (internal and external) is the base of high activity and reactivity (Mostafa *et al.* 2001). Active pozzolanic materials such as fly ash, silica fume and natural pozzolans are glassy and amorphous. Although natural zeolite crystals are crystalline and transparent, they can act as pozzolanic materials (Mindess 1981).

Literature review indicates that natural zeolite is one of the finest cement supplements. Plenty of active SiO2 and Al2O3 in the zeolite improve the microstructure of the hardened cement. SiO2 and Al2O3 chemically combine with calcium hydroxide from cement hydration process. This process makes C-S-H gel and additional aluminate (Ortega *et al.* 2000, Perraki *et al.* 2003, Caputo *et al.* 2008, Shao and Vesel 2015).

2. Experimental procedures and methods

2.1 Materials

According to ASTM C 150 (ASTM), cement used in this study is Portland cement type II. Zeolites were selected from two separate mine with two different sources to compare with each other. First zeolite pellets are prepared from Semnan mine, while the second was prepared from Miyane mine. These materials were micronized and milled smaller than cement particles. Physical characteristics of cement and two types of zeolite are presented in Table 1.

Based on ASTM C 618 (Testing *et al.* 2005), in natural pozzolans sum of SiO_2, Al_2O_3 and Fe_2O_3 should be at least 70% of the constituent elements of substances.

Table 1 Physical properties of cement and zeolite

Materials	Specific weight gr/cm³	Specific surface m²/kg
Portland cement type II	3.12	288
Semnan zeolite	2.18	472
Miyane zeolite	2.16	827

Table 2 Chemical properties of cement

Type (%)	SiO_2	Al_2O_3	Fe_2O_3	CaO	Na_2O	K_2O	MgO	C_3S	C_2S	C_3A	C_4A
OPC type II	21.91	4.85	3.46	64.56	0.34	0.97	2.38	49.5	25.47	7	10.53

Table 3 Chemical properties of zeolites

Type (%)	SiO_2	Al_2O_3	Fe_2O_3	CaO	Na_2O	K_2O	MgO	TiO_2	MnO	P_2O_5
Semnan	68.95	10.75	1.05	1.29	2.86	1.51	0.91	0.139	0.025	0.003
Miyane	68.69	10.89	0.83	2.22	1.78	1.46	0.66	0.139	0.003	0.037

Table 4 Physical properties of aggregates

Aggregate type	Fineness modulus	Absorption%	Specific weight gr/cm³
Crushed gravel	-	1.98	2.68
Lake sand	3.2	2.89	2.71

According to the XRF[1] analysis, sum of these chemical compounds in zeolite of Semnan is 80.75% while it is 80.41% for zeolite of Miyane. The chemical composition of cement and XRF analyzes are presented in Table 2 and 3.

According to the XRD[2] analysis, zeolite of Semnan has 55.3% Clinoptilolite and 25% Cristobalite compositions with the chemical formula of SiO_2. Zeolite of Miyane has 71.8% Clinoptilolite and 15.4% Opal compositions with the chemical formula of $SiO_2.nH_2O$. Chemical composition of Clinoptilolite has high participation and Cristobalite and Opal have medium participation in the pozzolonic activities. XRD analyses of two types of zeolites are presented in Figs. 1 and 2.

Aggregates were prepared from local source. Physical characteristic of consumed sand and gravel is presented in Table 4. Urban water with the pH 7.7 has been used in this research. This water did not have any smell and color.

2.2 Mix design

In this study, 48 different mix designs were planed based on ACI-211 (ACI 1998). Detailed results are shown in Tables 5-8. Proportion of mix designs were designed

[1] X-ray Fluorescence
[2] X-ray powder Diffraction

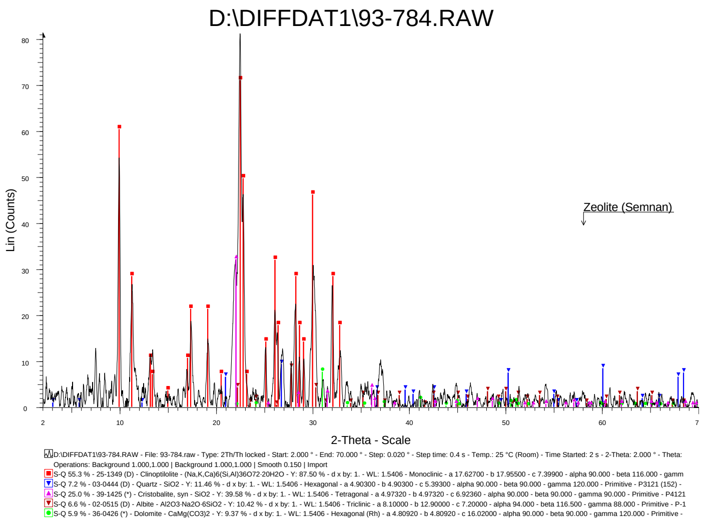

Fig. 1 XRD analysis of Semnan zeolite

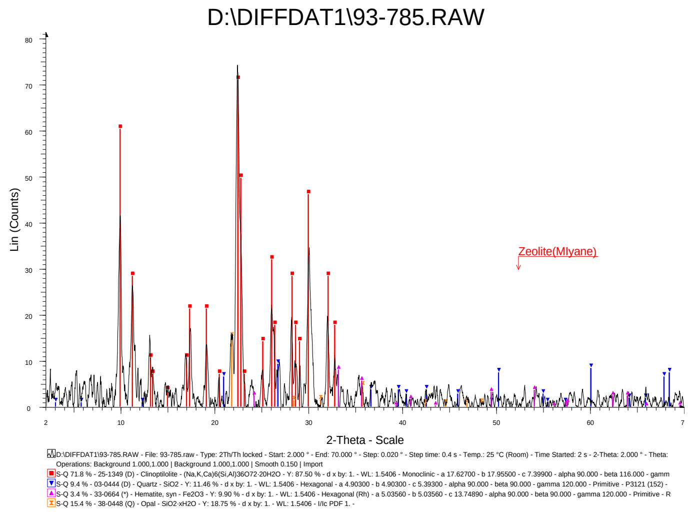

Fig. 2 XRD analysis of Miyane zeolite

Table 5 Mix design of control specimens

Specimens code	Cememt kg/m^3	Zeolite kg/m^3	W/C	Sand kg/m^3	Gravel kg/m^3
ZS300/0.4/0 ZM300/0.4/0	300	-	0.4	1150	908
ZS300/0.5/0 ZM300/0.5/0	300	-	0.5	1069	908
ZS300/0.6/0 ZM300/0.6/0	300	-	0.6	988	908
ZS350/0.4/0 ZM350/0.4/0	350	-	0.4	1053	908
ZS350/0.5/0 ZM350/0.5/0	350	-	0.5	958	908
ZS350/0.6/0 ZM350/0.6/0	350	-	0.6	922	908

Table 6 Mix design of concrete specimens with 15% replacement

Specimens code	Cenemt kg/m^3	Zeolite kg/m^3	W/C	Sand kg/m^3	Gravel kg/m^3
ZS300/0.4/15 ZM300/0.4/15	255	45	0.4	1150	908
ZS300/0.5/15 ZM300/0.5/15	255	45	0.5	1069	908
ZS300/0.6/15 ZM300/0.6/15	255	45	0.6	988	908
ZS350/0.4/15 ZM350/0.4/15	297.5	52.5	0.4	1053	908
ZS350/0.5/15 ZM350/0.5/15	297.5	52.5	0.5	958	908
ZS350/0.6/15 ZM350/0.6/15	297.5	52.5	0.6	922	908

Table 7 Mix design of concrete specimens with 20% replacement

Specimens code	Cenemt kg/m^3	Zeolite kg/m^3	W/C	Sand kg/m^3	Gravel kg/m^3
ZS300/0.4/20 ZM300/0.4/20	240	60	0.4	1150	908
ZS300/0.5/20 ZM300/0.5/20	240	60	0.5	1069	908
ZS300/0.6/20 ZM300/0.6/20	240	60	0.6	988	908
ZS350/0.4/20 ZM350/0.4/20	280	70	0.4	1053	908
ZS350/0.5/20 ZM350/0.5/20	280	70	0.5	958	908
ZS350/0.6/20 ZM350/0.6/20	280	70	0.6	922	908

Table 8 Mix design of concrete specimens with 30% replacement

Specimens code	Cenemt kg/m^3	Zeolite kg/m^3	W/C	Sand kg/m^3	Gravel kg/m^3
ZS300/0.4/30 ZM300/0.4/30	210	90	0.4	1150	908
ZS300/0.5/30 ZM300/0.5/30	210	90	0.5	1069	908
ZS300/0.6/30 ZM300/0.6/30	210	90	0.6	988	908
ZS350/0.4/30 ZM350/0.4/30	245	105	0.4	1053	908
ZS350/0.5/30 ZM350/0.5/30	245	105	0.5	958	908
ZS350/0.6/30 ZM350/0.6/30	245	105	0.6	922	908

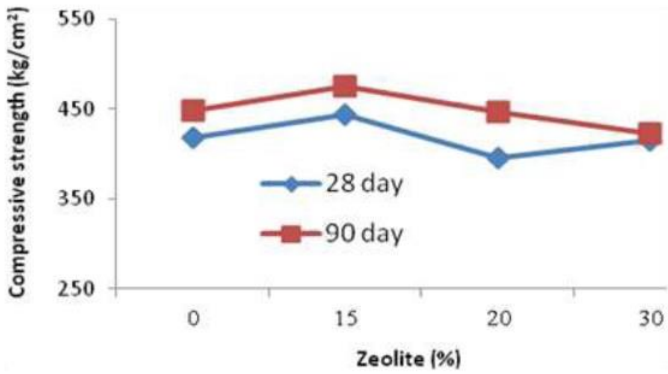

Fig. 3 Compressive strength of concrete with Semnan zeo., 0.4, 300 kg/m^3

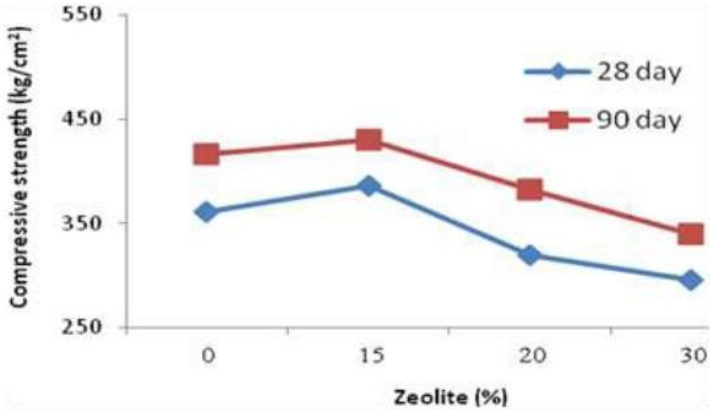

Fig. 4 Compressive strength of concrete with Semnan zeo., 0.5, 300 kg/m^3

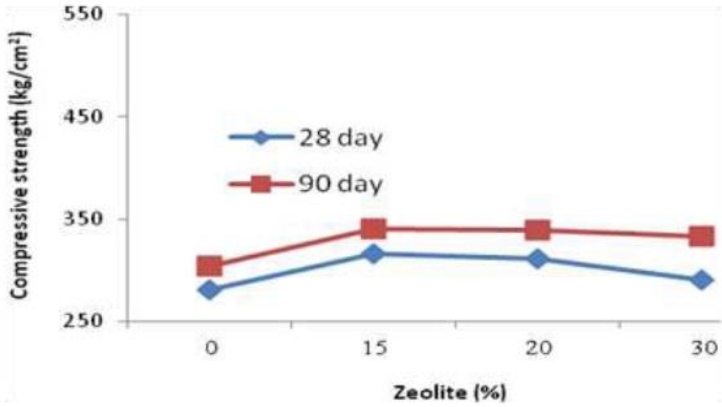

Fig. 5 Compressive strength of concrete with Semnan zeo., 0.6, 300 kg/m^3

with two cement content of 300 and 350 kg/m^3, two types of zeolite, Semnan and Miyane, three water-cement ratio of 0.4, 0.5 and 0.6 and with three replacement of zeolite instead of cement, 15%, 20% and 30%. This amount of zeolite replacements and different types of water-cement ratio in different cement contents is based on previous studies and also initial experimental designs. It should be mentioned that mix designs without zeolite have been designed to control for each series of mix designs.

ZSx/y/z and ZMx/y/z refer to zeolite of Semnan and Miyane fabricated using with x cement content and y water-cement ratio and z replacement of zeolite instead of cement.

2.3 Method of sample preparation and testing

For fabrication of specimens, the gravel and sand have

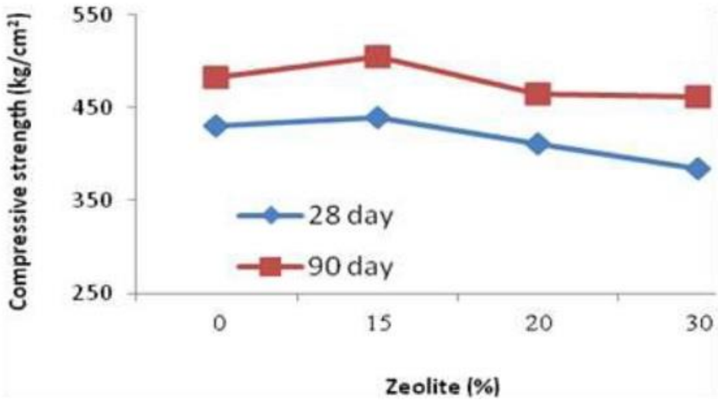

Fig. 6 Compressive strength of concrete with Semnan zeo., 0.4, 350 kg/m^3

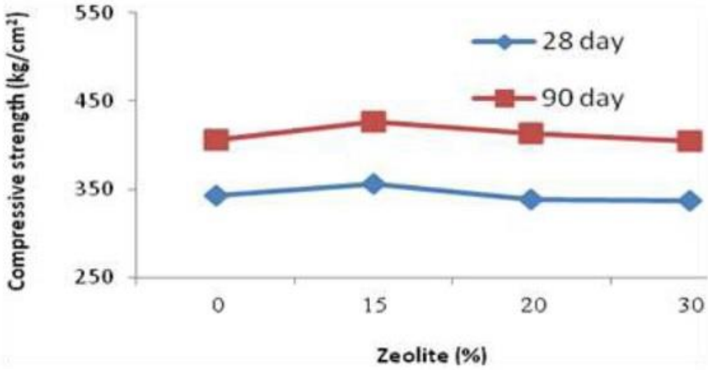

Fig. 7 Compressive strength of concrete with Semnan zeo., 0.5, 350 kg/m^3

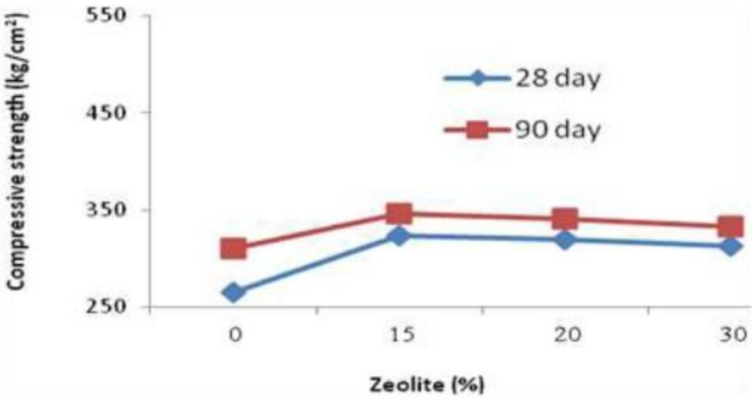

Fig. 8 Compressive strength of concrete with Semnan zeo., 0.6, 350 kg/m^3

been used in saturated surface-dry (SSD) condition and surface moisture condition, respectively. The sand, gravel, cement, and finally zeolite poured into mixer. They were mixed for 30 seconds. The water was added to the mixture in two phases and the total mixing time was 2 minutes. Then, 3 cubic specimens for each group were sampled. Filling the mold was performed according to ASTM C 31 ((ASTM)). After 24 hours, the samples extracted from the mold. Based on standard ASTM C 192 (ASTM 1998), the specimens were placed in the water with 23±1°C temperature for curing. After reaching samples to the intended age, compressive strength of samples was determined according to ASTM C39 (Standard 2010).

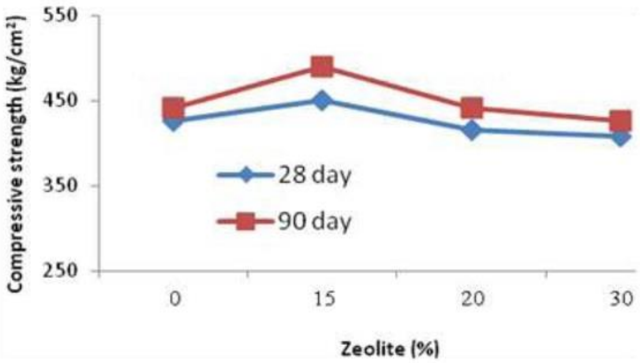

Fig. 9 Compressive strength of concrete with Miyane zeo., 0.4, 300 kg/m^3

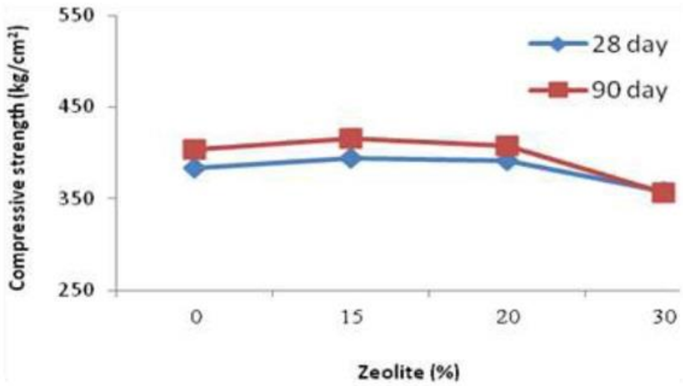

Fig. 10 Compressive strength of concrete with Miyane zeo., 0.5, 300 kg/m^3

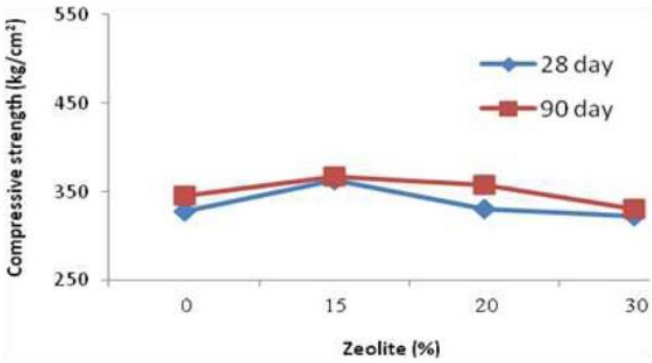

Fig. 11 Compressive strength of concrete with Miyane zeo., 0.6, 300 kg/m^3

3. Experimental results and discussion

The average of three concrete tests results with zeolite of Semnan in cement content of 300 and 350 kg/m^3 at 28 and 90 days are presented in Figs. 3-8. Corresponding results for zeolite of Miyane are presented in Figs. 9-14.

From Figs. 3-8, the zeolite of Semnan showed an increase in the compressive strength in all cases with 15% replacement of zeolite instead of cement. The compressive strength of concrete with all replacement of zeolite rates has been increased in water-cement ratio of 0.6 in both cement content. A slight decrease in the compressive strength has been observed in water-cement ratio of 0.4 and 0.5 with replacement of 20% and 30%. The zeolite of Semnan has increased the compressive strength of concrete about 22% in cement content of 350 kg/m^3 with 15% replacement of zeolite and water-cement ratio of 0.6. The 28-day compressive strength of concrete with 30% replacement of zeolite instead of cement was 283 kg/cm^2 and 90-day compressive strength was 321 kg/cm^2. Therefore, 13% of increase was observed for strength of concrete in the period of 28 to 90 days. According to this result, 30% reduction of

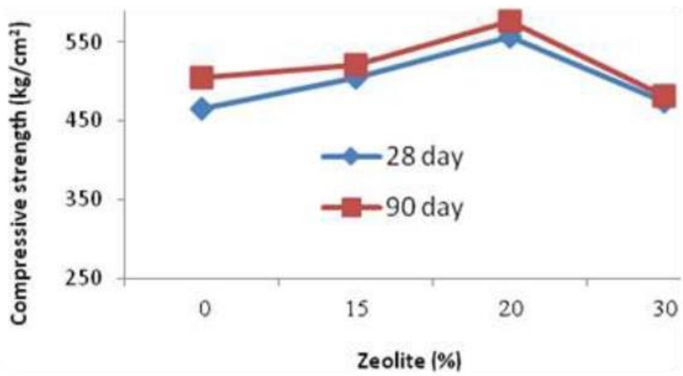

Fig. 12 Compressive strength of concrete with Miyane zeo., 0.4, 350 kg/m^3

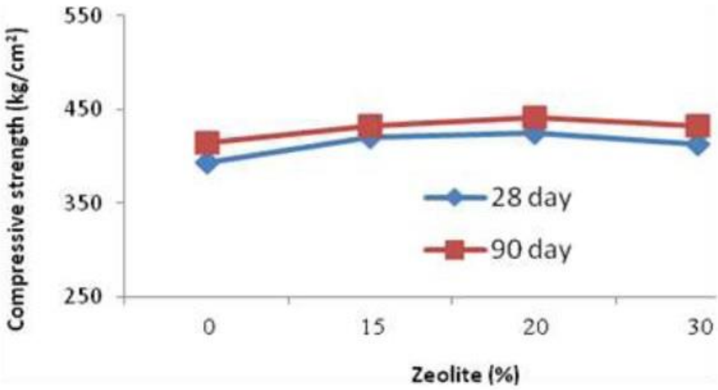

Fig. 13 Compressive strength of concrete with Miyane zeo., 0.5, 350 kg/m^3

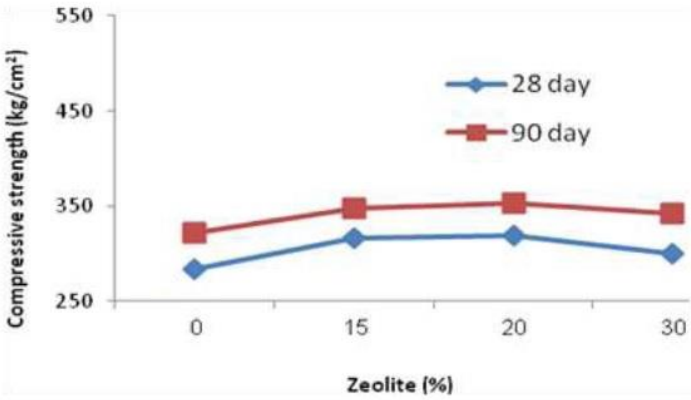

Fig. 14 Compressive strength of concrete with Miyane zeo., 0.6, 350 kg/m^3

cement consumption by obtaining desirable strength was achieved. It means that it is possible to reduce emission and the cost of cement consumption, especially in massive and huge structures.

Zeolite of Miyane results showed that compressive strength has been increased with 15% replacement in all cases. This type of zeolite at cement content of 350 kg/m^3 with water-cement ratio of 0.5 and 0.6 in all percent of replacements levels has been showed increases in compressive strength. Generally, it can be said that the zeolite of Miyane in concrete at cement content of 350 kg/m^3 has been increased the compressive strength of all cases.

Actually, comparison between the effect of Miyane and Semnan zeolites showed that compressive strength increases by increasing the amount of Miyane zeolite but in Semnan zeolite relative reduction in strength was observed in high percentage of replacement. This increase in strength is due to the high specific surface area of Miyane zeolite particles and therefore according to the tiny grains of zeolite and surrounded cement by high Blaine particles there will be desirable chemical and pozzolanic reaction. Thus, desired compressive strength can be considered by increases in Blaine in zeolite but appropriate Blaine to reduce the cost of using zeolite powder can provide structural strength requirements.

4. Conclusions

Summary of the effect of natural zeolite to reduce the consumption of cement is provided below:

1. Increase in the strength about 7% to 22% was obtained by using Semnan zeolite in concrete with 0.6 water-cement ratio in all replacement rates.

2. The best replacement of Semnan zeolite as pozzolan in concrete with cement content 300 and 350 kg/m^3 is 15% because this replacement in all ages and water-cement ratios increased strength.

3. Miyane zeolite in concrete with cement content 350 kg/m^3 has been increased strength about 3% to 23% in all cases. For this reason we can say this type of zeolite as pozzolan in concrete with cement content 350 kg/m^3 can significantly reduce cement consumption.

4. Miyane zeolite such as Semnan in 15% replacement in all cases increases the strength, so best replacement percentage of these two types of zeolite as pozzolan in concrete seems to be 15%.

5. Semnan zeolite in 0.6 water-cement ratio showed better performance than the Miyane zeolite and Miyane zeolite performance in the long-term strength is better than Semnan zeolite.

6. As regards in important projects and infrastructures such as dams, bridges and huge foundations design compressive strength can be in the range of 250 to 350 kg/cm^2. So that 15% to 30% replacement of zeolite instead of cement can achieve high compressive strength meanwhile can save significant usage in cement and cost of project and moreover, reducing cooling costs in order to reduce the temperature of hydration is the other benefit of replacement of zeolite instead of cement which these actions to reduce carbon dioxide emissions and produce green concrete in its true sense is completely effective.

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