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Industrial wastewater and zeolite Interaction on compressive strength of plain concrete in different cement curing ages

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ABSTRACT
In the present study interactions of three levels of water quality (tap water, treated industrial wastewater, and a mixture of their equal ratio), four levels of zeolite application (the use of 0 %, 10 %, 20 % and 30 % of zeolite instead of cement) and two levels of cement content (250 and 350 kg m^{-3}*) on compressive strength of plain concrete over seven curing ages (3, 7, 28, 56, 90, 180, and 365 days) was investigated. For this, 504 concrete cube samples were constructed and data were analyzed using tests of analysis of variance and means comparison. Based on the statistical analysis of data, all two-, three- and four- way interactions of curing age with cement content, water type and zeolite percentage were insignificant, indicating that the variation rate of concrete compressive strength by time was not affected by mixing water type and application of zeolite and also all possible combinations of studied factors. Moreover, the results showed that the three-way interaction of cement content × water type × zeolite percentage on the compressive strength of concrete was significant. Therefore, the selection of best zeolite percentage in the mix design should be according to the mixing water type and cement content. Accordingly, simultaneous use of unconventional water as mixing water with zeolite up to 10 % in low cement content (250 kg* m^{-3}*) and also with zeolite up to 20 % in the high cement content (350 kg* m^{-3}*) can be recommendable.*

Keywords: *Analysis of variance (ANOVA) LSD;Means comparison Mix design;Mixing water Statistical analysis Unconventional Water;*

I. INTRODUCTION

Water is one of the most important materials used in cementitious mixtures and occupies 14 %–21 % of their total volume [1]. Concrete is one of the most important materials in the construction industry, consuming ≈150–210 l of water per cubic meter [2–4]. It was estimated that more than 10 billion m^3 of concrete is produced annually and about 2.1 billion m^3 (cubic meters) of water is needed as batch water regardless of water required for curing and other consumptions associated with concrete production [3,5]. Iran is located in the arid and semi-arid region of the world. Concrete production in Iran is estimated to be 250–500 Mt (megatons) during 2012–2050 which consumes 0.5–1 percent of annual renewable water resources [5]. The use of unconventional water such as reuse of industrial treated wastewater is one of the great solutions to deal with the problem of water shortage [6]. In the near future, developed and developing countries have to reused treated and partially treated wastewater in huge quantity in the concrete industry [7]. It is estimated that annual volume of industrial wastewater in Iran reaches 1088 million m^3 by 2022 [8] and the construction industry (for the production of cement and concrete) can be introduced as one of the potential consumers of this water. Water contaminated with industrial wastes, but free of suspended solids, can be generally suitable at low concentrations for concrete construction [9]. However, Industrial wastewater has a wider range of quality than domestic and agricultural ones; hence, they should be reused with more qualitative and environmental considerations [8] and precise management as well.

Inorganic and organic compounds in mixing water can interfere with the hydration of cement. These chemicals delay the initial and final cement setting time, trap more air in the concrete and affect the pore structure, and compressive strength of concrete [6,10,11]. Nevertheless, the setting time for the cement paste prepared with both industrial and domestic treated wastewater and wash water was within the acceptable limits of the ASTM and BS standards [10,6,11,12]. Presence of more voids among the aggregates and low particles density in the concrete made by domestic wastewater compared with drinking water was reported [6]. However, the mixing water contaminants have various effects on concrete, but possibly all of them do not have destructive effects; therefore, some of them can be harmless and even result in the improvement of concrete properties. In most cases, there are virtual limits for the amount of impurities in concrete mixing water, while some impurities can be harmless [13]. In a study by Mehrdadi et al. [13], the compressive strength of concrete at curing ages 7 and 28 days in treated domestic wastewater was lower than drinking water because of the influence of BOD (days biological oxygen demand) and COD (chemical oxygen demand) contents of wastewater. However, this reduction was lower than 10 % when compared with drinking water. Similar successful usage of industrial and domestic treated wastewater as concrete mixing water was reported by Asadollahfardi and Mahdavi [10] and Asadollahfardi et al. [6].

The global air pollution by greenhouse gases has amplified the need to use environmental friendly materials in the concrete industry [14]. Worldwide, about 3.8 Gt (Giga ton) of cement, annually, is using by the concrete industry [3]. Averagely, the production of 1 m^3 of concrete and 1 ton of cement emits 240–320 and 900 kg CO_2 (carbon dioxide) into the atmosphere, respectively [3,15]. Consequently, the cement industry was introduced as a responsible for 6 %–8.6 % of global CO_2 emissions [5,16,17]. Partially utilize the natural zeolite instead of cement in the concrete mix design is one important solution to mitigate this problem; in the meantime, it can effectively strengthen the stability of the final concrete product [16–18]. In this regard, it was reported that substituting 10 %–30 % of cement by zeolite can reduce the global warming index by 60 %–70 % [16].

Natural zeolites have been used in diverse fields including the construction industry (as pozzolanic additive as well as lightweight aggregate), water and wastewater treatment, adsorption, catalysis, gas purification, agriculture, soil remediation and energy [18–20]. The use of natural zeolites and its modified forms for removal of ammonium, heavy metals, cations, anions and organics from water and wastewater due to its high cation exchange capacity[1] (CEC) is a promising technique in environmental cleaning processes [20–22] so that wastewater treatment facilities that utilize of natural zeolites are already in operation in many countries [22]. For instance, the rate of ammonium adsorption on natural zeolite is 2.7–30.6 mg g^{-1} [20]. The pozzolanic property of zeolite can be recognized with the high amounts of reactive silicon dioxide (SiO_2) and aluminum oxide (Al_2O_3) in the zeolite which react with the calcium hydroxide ($Ca(OH)_2$ or CH) of cement in the presence of water (0.555 g of cement CH react with 1 g of zeolite) and create cementitious products such as calcium silicate (Ca_2O_4Si) and calcium aluminosilicate ($CaAl_2Si_2O_8$) [18,19,23]. Above products can fill more cavities, reduce the voids and form a dense microstructure in the hardened cement paste and concrete [14,18]. Tran et al. [18] reviewed the results of many studies about zeolite usage in concrete. They reported that the range of natural zeolite added into concrete mix design was generally from

2.5 %–30 % of cement content and concluded that the ratio of compressive strength in the zeolite-containing concrete to reference concrete generally was larger than 1. This ratio was 1.07, 1.11 (between 1.01 and 1.18), 1.12, 1.11 (between 0.85 and 1.26), 1.09, 1.04 (between 0.65 and 1.23), 1.07 (between 0.92 and 1.25) and 0.87 (between 0.76 and 1.05) in the zeolite levels 2.5, 5, 7.5, 10, 12.5, 15, 20 and 30 percent. While many researchers concluded that the use of natural zeolite had almost no negative effects on compressive strength of concrete, even showed its improvement, adverse impacts of zeolite on the concrete compressive strength also were reported [14,18]. For instance, results of Vejmelková et al. [19], Najimi et al. [17] and Vejmelková et al. [24] showed that substituting cement by zeolite in the rate of 10 %–40 %, 15 % and 30 % and 10 %–40 % led to the reduction of compressive strength of concrete in all curing ages up to 365. These conflicting results were attributed to the differences in water to cement ratio and the amount of natural zeolite used and the variety of natural zeolites in terms of structure, chemical composition, reactivity and purity [14,17,18].

Very general and broad tolerant limitations can be seen in the literature on concrete mixing water. On the other hand, mixing water limitations in concrete are strict. The research literature on the use of wastewater in concrete reveals that much work is needed in this regard. Therefore, it is stated that presently available information is not adequate to set up rigid specifications for maximum amounts of impurities that may present in mixing water [7]. Moreover, since the proven ability of natural zeolites in the removal of water pollutants as a potential to decrease of negative effects of impurities of mixing water in the concrete were not investigated, the current study was conducted in order to statistically examine the interaction of water quality, zeolite percentage and cement content on the compressive strength of concrete in the different curing ages.

II. MATERIALS AND METHODS

2.1 Treatments and statistical design

The current study was conducted to investigate the possibility of treated industrial wastewater usage plus zeolite for different cement contents to produce plain concrete. The experiment was carried out as a four-factor factorial based on completely randomized design with three replications by making 504 concrete specimens relied on 168 treatments including three water types (W1: tap water, W2: an equal ratio of tap water and treated industrial wastewater, and W3: treated industrial wastewater), four zeolite levels (substituting Z1: 0 %, Z2: 10 %, Z3: 20 %, and Z4: 30 % of cement by zeolite), two cement contents (C1: 250 and C2: 350 kg m^{-3}) and seven curing ages (d1: 3, d2: 7, d3: 28, d4: 56, d5: 90, d6: 180 and d7: 365 days).

In the present study, four studied factors (including water, quality, zeolite percentage, cement content and curing age) and 11 possible interactions between them (including six two-way interactions, four three-way interactions as well as one four-way interaction) can be as sources of variation of concrete compressive strength. In the previous similar studies, the changes of concrete compressive strength affected by water type or zeolite were presented and analyzed only in term of variation rates or values, and the statistical analysis was not done to investigate whether these variations are significant or not. In the present study, the statistical tests such as the of analysis of variance (ANOVA) and means comparison (based on least significant difference- LSD at 5 % probability level) were carried out on the compressive strength data in order to study the significance of these variations. These statistical analyses were conducted using SPSS 22 (2016) software.

2.2 Mixing water

The wastewater was collected from the output of sewage treatment plant of Agh-Qalla Industrial Park located in about 15 km away from Gorgan City, the capital of Golestan Province, Iran. The treatment plant is located at 36° 58’48’’ Northen latitude and 54° 27’18’’ Eastern longitude, on about 4 km from South of Agh-Qalla City. The activated sludge method is used in the treatment plant. The qualitative parameters of studied mixing waters were measured according to APHA [25]. Also, since present research studies the non-potable water as concrete mixing water, a detailed discussion on permissible limits of constituents in mixing water based on existing codes is essential [7]. Therefore, an extensive review of national and international codes was conducted in order to compare permissible limits of mixing water impurities of concrete and the quality of studied mixing waters with those.

2.3 Cement and zeolite

In the present study a type II Portland cement with anti-sulfate properties and clinoptilolite type of powdery zeolite, purchased from Shahroud Cement Company and Semnan Negin Powder Company, respectively, were used. Their chemical specifications are shown in Table 1. The specific gravity of cement and zeolite was 3110 and 2100 kg m^{-3}. The clinoptilolite is one of the most common natural zeolite. It is used also most commonly in water and wastewater remediation because it has high pore volume (0.34 cm^3 g^{-1}) and CEC, in the range of 2200—2600 meq kg^{-1} [21].

2.4 Experiment

The concrete mix was designed based on the double-washed fine and coarse aggregates with the specific gravity of 2550 and 2620 kg m^{-3} and water absorption 3.3 % and 2.1 %, respectively. The grading test of aggregates was performed based on ASTM C136 [26]. The grading curves of fine and coarse aggregates and the extracted findings showed that the maximum size of aggregates was 9.5 and 19 mm, respectively, whereas the fine aggregate fineness modulus was 2.9 as well. Therefore, the requirements of ASTM C33 [27] were passed.

Concrete samples with a dimension of 150 150 50 mm were made according to ASTM C192 [28]. Sampling was performed based on ISO 1920-1 [29]. The concrete mix in the current study did not design based on a specified compressive strength because it was considered that the data will compare with each other relatively. However, in order to increase the test accuracy, 54 concrete cubes were made based on 18 different mix designs with three replication including three ratios of fine to coarse aggregates, two ratios of water to cementitious materials, and three values of concrete specific gravity. Finally, the mix design was selected based on the maximum 3-days compressive strength of concrete samples that was specific

Table 1
Chemical composition of cement and zeolite.

Composition	Cement	Zeolite	Composition	Cement	Zeolite	Composition	Cement	Zeolite
SiO_2	21.11	69.28	CaO	63.36	3.56	MgO	1.51	0.50
Al_2O_3	4.42	10.43	Na_2O	0.38	0.73	SO_3	2.61	0.005
Fe_2O_3	3.96	0.49	K_2O	0.51	1.27	LOI	2.98	12.97

Table 2
Details of mix design (kg per cubic meter of concrete except for SP).

Material	C1Z1	C1Z2	C1Z3	C1Z4	C2Z1	C2Z2	C2Z3	C2Z4
Water	112	112	112	112	158	158	158	158
Cement	250	225	200	175	350	315	275	245
Zeolite	0	25	50	75	0	35	75	105
Sand	969	969	969	969	896	896	896	896
Gravel	969	969	969	969	896	896	896	896
SP (% of cementitious materials)	0.80	0.94	1.29	1.75	0.66	0.78	1.03	1.51

C: cement content (1: 250 & 2: 350 kg m^{-3}) Z: zeolite level (1: 0; 2: 10; 3: 20 & 4: 30 %) SP:

superplasticizer.

gravity of 2400 kg m^{-3}, fine to coarse aggregates ratio of 1:1, and water to cement materials ratio of 0.45. The mix design details in the different treatments are shown in Table 2.

It was reported that the use of natural zeolite in concrete production decreases the workability of the concrete. Accordingly, the results of several studies showed that the addition of a superplasticizer to concrete mix design in order to compensate of reduction of concrete workability by zeolite and maintain of the concrete slump is necessary so that its required volume increases when the amount of zeolite increases [18,19]. In the present study, a commercial carboxylate- based superplasticizer with a density 1070 kg m^{-3} was used. Its required dosage (between 0.65 %–1.75 % of cementitious materials depended on cement content and zeolite percentage) was obtained by the trial-and-error method in the preliminary mix design tests based on the slump range 6 8 cm. This criterion was used by Kaboosi et al. [14], Khoshroo et al. [15] and Ahmadi et al. [30], as well. In order to control of desirable workability of fresh concrete during the experiments, the slump test was repeated at least once for each mix design (treatment).

III. RESULTS AND DISCUSSION

2.5 Investigating the mixing water quality

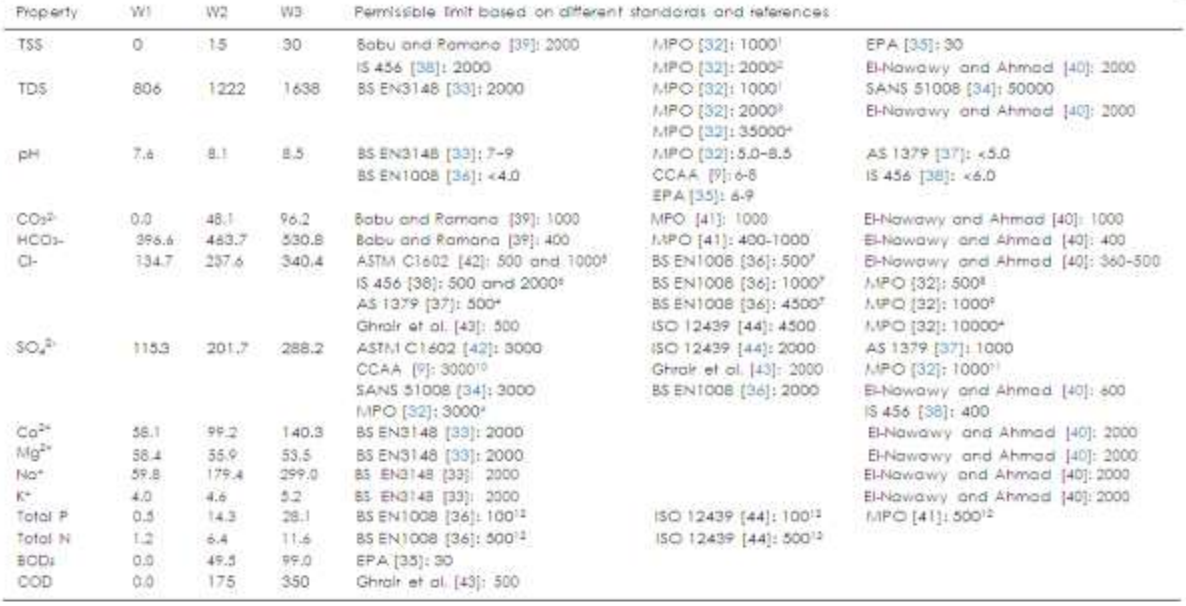
Different properties of three types of studied water, as well as different national and international codes about concrete mixing water, are presented in Table 3. The comparison of the permissible limit of total suspended solids (TSS) in different standards shows that codes have good accordance together in the basis of TSS 2000 ppm, except in EPA limit (30 ppm). In term of total dissolved solids (TDS), the permissible limits in different codes are very different in the range of 1000 and 50,000 ppm. The difference between MPO [32] limits can be partly reasonable because of concrete type (plain and reinforced concrete), although the difference of plain concrete and reinforced concrete limits are very high. Meanwhile, a very high difference can be seen between the limits of British standard [33] and South African standard [34], that are 2000 and 50,000 ppm, respectively. Also, there is considerable diversity between the different codes regarding the pH of mixing water, so that the lower and upper limits of all standards are different. While the BS EN3148 [33], MPO [32], CCAA [9] and EPA [35] codes determined a specific range (generally about 6–9) for pH of concrete mixing water, BS EN1008 [36], AS 1379 [37] and IS 456 [38] did not set a certain upper limit.

However, it is proven that sever acidic waters are not suitable as mixing water, though even, the definition of acidic water reduce setting time, durability and strength of concrete [41], international standards did not pay attention to these specifications. However, the similar limits were presented by the Iranian concrete code [41] and some references in this regard.

In term of the chloride concentration, it is reveals that the different standards have proposed two or three limits based on the type of concrete (prestressed, reinforced or plain concrete). The comparison of these limits showed that the permissible range of chloride in mixing water for prestressed and reinforced concrete within different codes mostly is close together but those have the significant difference for plain concrete in the very wide range of 500 [37], 4500 [36] and 10,000 [32]. Meanwhile, some codes and references such as ISO 12,439 [44], Ghrair et al. [43] and El-Nawawy and Ahmad [40] did not distinguish between the types of concrete in the regard of permissible content of chloride in the mixing water. Chloride can penetrate into the concrete and cause accelerated corrosion of the reinforcement [45]. On the other hand, however, it was reported that the presence of a relatively high amount of chloride in the concrete mixing water has a positive effect on early strength of concrete [46], especially in early curing ages [41].

Regarding the permissible content of sulphate in mixing water, comparison of different codes shows that they have not good accordance together so that the permissible limit of sulphate based on those codes can be divided into four limits 600, 1000, 2000 and 3000 ppm. Therefore, it is needed that these limits be effectively studied and matched. The most common

Table 3
Mixing water properties vs. permissible limits of its impurities for concrete (as ppm except for pH) [†].



Property	W1	W2	W3	Permissible limit based on different standards and references		
TSS	0	15	30	Babu and Ramana [39]: 2000 IS 456 [38]: 2000	MPO [32]: 1000[1] MPO [32]: 2000[2]	EPA [35]: 30 El-Nowawy and Ahmad [40]: 2000
TDS	806	1222	1638	BS EN3148 [33]: 2000	MPO [32]: 1000[1] MPO [32]: 2000[3] MPO [32]: 35000[4]	SANS 51008 [34]: 50000 El-Nowawy and Ahmad [40]: 2000
pH	7.6	8.1	8.5	BS EN3148 [33]: 7–9 BS EN1008 [36]: <4.0	MPO [32]: 5.0–8.5 CCAA [9]: 6-8 EPA [35]: 6-9	AS 1379 [37]: <5.0 IS 456 [38]: <6.0
CO_3^{2-}	0.0	48.1	96.2	Babu and Ramana [39]: 1000	MPO [41]: 1000	El-Nowawy and Ahmad [40]: 1000
HCO_3^-	396.6	463.7	530.8	Babu and Ramana [39]: 400	MPO [41]: 400-1000	El-Nowawy and Ahmad [40]: 400
Cl^-	134.7	237.6	340.4	ASTM C1602 [42]: 500 and 1000[5] IS 456 [38]: 500 and 2000[6] AS 1379 [37]: 500[4] Ghrair et al. [43]: 500	BS EN1008 [36]: 500[7] BS EN1008 [36]: 1000[7] BS EN1008 [36]: 4500[7] ISO 12439 [44]: 4500	El-Nowawy and Ahmad [40]: 360-500 MPO [32]: 500[8] MPO [32]: 1000[9] MPO [32]: 10000[4]
SO_4^{2-}	115.3	201.7	288.2	ASTM C1602 [42]: 3000 CCAA [9]: 3000[10] SANS 51008 [34]: 3000 MPO [32]: 3000[4]	ISO 12439 [44]: 2000 Ghrair et al. [43]: 2000 BS EN1008 [36]: 2000	AS 1379 [37]: 1000 MPO [32]: 1000[11] El-Nowawy and Ahmad [40]: 600 IS 456 [38]: 400
Ca^{2+}	58.1	99.2	140.3	BS EN3148 [33]: 2000		El-Nowawy and Ahmad [40]: 2000
Mg^{2+}	58.4	55.9	53.5	BS EN3148 [33]: 2000		El-Nowawy and Ahmad [40]: 2000
Na^+	59.8	179.4	299.0	BS EN3148 [33]: 2000		El-Nowawy and Ahmad [40]: 2000
K^+	4.0	4.6	5.2	BS EN3148 [33]: 2000		El-Nowawy and Ahmad [40]: 2000
Total P	0.5	14.3	28.1	BS EN1008 [36]: 100[12]	ISO 12439 [44]: 100[12]	MPO [41]: 500[12]
Total N	1.2	6.4	11.6	BS EN1008 [36]: 500[12]	ISO 12439 [44]: 500[12]	
BOD_5	0.0	49.5	99.0	EPA [35]: 30		
COD	0.0	175	350	Ghrair et al. [43]: 500		

W1, W2 and W3 show tap water, combined water made with the equal ratio of tap and treated industrial wastewater, and treated industrial wastewater, respectively.
TSS: total suspended solids; TDS: total dissolved solids; BOD_5: 5-days biological oxygen demands; COD: chemical oxygen demands.

[†] The BS EN3148 [33] code was superseded and replaced by BS EN1008 [36] because it did not give any information regarding the long-term durability of concrete and summarized only the knowledge about suitability of mixing water. Also, mixing water specifications of concrete in ASTM C1602 [42] and ASTM C94 [54] are same. Moreover, EPA [35] values are presented for restricted reuse of wastewater after secondary treatment and disinfection.

[1] For reinforced concrete in sever environmental condition and prestressed concrete.

[2] For reinforced concrete in mild environmental condition and plain concrete.

[3] For reinforced concrete in mild environmental condition.

[4] For plain concrete and concrete without embedded metal.

[5] 500 in prestressed concrete or in bridge decks and 1000 in other reinforced concrete, in moist environments or containing aluminum embedments or dissimilar metal or with stay-in-place galvanized metal forms.

[6] 500 for reinforced concrete and 2000 for concrete not containing embedded steel.

[7] 500 in prestressed concrete or grout, 1000 in concrete with reinforcement or embedded metal and 4500 in concrete without reinforcement or embedded metal.

[8] For reinforced concrete in sever environmental condition, prestressed concrete and concrete of bridges deck.

[9] For other types of reinforced concrete.

[10] Water with higher content has been used satisfactorily, as well [9].

[11] For reinforced concrete and prestressed concrete.

[12] P as Phosphate (P_2O_3) and N as Nitrate (NO_3^-).

form of sulfate attack involves the reaction of sulfate ions with calcium hydroxide ($Ca(OH)_2$ or CH) and tricalcium aluminate hydrates in the cement paste leading to the formation of gypsum (CaSO4.2H2O) and massive ettringite (calcium sulfoaluminate) [47]. This reaction is normally accompanied by a considerable increase in volume, which initiates the expansion of the concrete and leads to an irregular pattern of cracks and a subsequent loss in strength [46]. In general, structures affected by sulfate attack usually exhibit large deformations caused by swelling leading to crack formation [47]. Similar to carbonate and bicarbonate, permissible limits of calcium, magnesium, sodium, potassium, phosphorus and nitrogen in mixing water did not study in details. It seems that those have to be studied extensively because a few references, including one superseded code [33], have set a specific limit for these constituents. The high content of inorganic substances beyond the recommended limits may extend the setting time of the fresh concrete and possibly result in a significant reduction in the concrete strength by retardation of cement hydration [46]. Sodium and potassium ions may produce or intensify the alkali-aggregate reaction if reactive types are used, and sulfate and magnesium ions cause a weakening action on the cement paste [45]. Also, while phosphorus increases the setting time of cement paste [41], nitrate-containing admixtures such as $Ca(NO_3)_2$ (calcium nitrate) is used as a setting accelerator [48].

The BOD_5 and COD are two parameters that represent degradable and total organic matters in the water, respectively. The ratio of BOD_5 to COD shows the biodegradable content of water. When this ratio is high, the effluent is easily biodegradable. Despite the importance of the degradable organic matter in mixing water that can reduce the compressive strength of concrete, almost no standards, except EPA [35], has determined a permissible limit of BOD_5 of mixing water. However, some codes such as IS 456 [38] specified a certain limit in term of organic matter in the concrete mixing water.

The above results showed that the concrete mixing water standards vary based on the country and international organization that promotes the standard. Therefore, it seems that further and detailed studies on existing permissible limits are essential in order to uniformize these codes. A similar result was reported by Babu et al. [7]. Meanwhile, the effect of some constituents such as heavy metal, fluoride, detergent and emerging contaminants in mixing water on the compressive strength of concrete is not studied in details and therefore, no permissible limit is presented in different national and international codes. Also, while there are some signs that show the effect of concrete curing water type is important [6,13,49], almost all codes were focused on the quality of concrete mixing water and no attention was paid to concrete curing water. Another important issue is that in order to assess the suitability of concrete materials, cumulative amount of impurities in all materials including water, aggregates, cement and additives should be measured. For this reason, some codes such as AS 1379 [37] presented the permissible limit of some impurities as a total limit per unit concrete volume (as kg of substance per cubic meter of concrete). Besides, almost all standards proposed two important criteria: 1- gaining a specified compressive strength by concrete or mortar samples that made with suspected water compared to control ones; 2- limiting the change of initial or final setting time of cement paste affected by unstudied water [9].

The comparison of properties of the used mixing waters with different codes showed that although pH of tap water and combined water was fall within the recommended limits based on different standards, its value for treated industrial wastewater were slightly more than CCAA [9] limit. It was reported that alkaline mixing waters normally have an effect on the maturation reactions and compressive of concrete [1,46]. However, because the pH value of concrete normally varies between 11 and 13 [46], the slight increase of mixing water pH in the present study might not cause any significant effect. Table 3 also shows that the concrete durability made with the studied treated industrial wastewater and combined water likely will not be adversely affected due to mixing water quality because the levels of different ions and substances, based on existing standards and references, were within the permissible limits for concrete mixing water, except for BOD_5 that was 1.65 and 3.3 times of allowable limit of EPA [35] in combined water and wastewater, respectively. A similar finding was reported by Asadollahfardi and Mahdavi [10], Kaboosi et al. [14], Asadollahfardi et al. [6], Alqam et al. [49], Su et al. [12] and Lee et al. [11] that studied treated industrial wastewater, greywater, treated domestic wastewater, recycled household greywater, wash water (from ready-mixed concrete plant) and treated municipal effluent as mixing water of concrete, respectively. In the regard of phosphates and nitrates hazard, it is recommended that if their qualitative tests show positive results relative to the standard, either the quantity of the substance concerned shall be determined or tests for setting time and compressive strength shall be performed [36]. Therefore, in the present study, with respect to the mixing water qualities (Table 3), it was assumed that the effect of these substances on concrete in all types of studied water is not considerable. Nevertheless, while the results energy dispersive x-ray test (EDX) indicated that the contents of sodium, chloride and sulfur in concrete samples made with the treated domestic wastewater were increased slightly compared to ones made with drinking water [6], the weight percent of these elements in concrete made with drinking water and treated industrial wastewater were equal approximately [10]. Also, Al-Saleh [50] showed that the effect of chloride content of mixing water on the total chloride content of concrete was minimal to moderate for water to cement ratio up to 0.4 and chloride content of mixing water could be higher than ASTM specified limits for mix designs with low water to cement ratios. In addition, they found that the concrete aggregates, no mixing water, are the critical ingredient in term of determining the presence of chloride in concrete. However, in order to a better conclusion, the durability tests are recommended to more investigate the potential of corrosion and sulfate attack by chloride and sulfate, respectively.

Table 4
The result of ANOVA test on data of concrete compressive strength.

Source of Variation	d.f	Mean square	F	P value
Cement content (C)	1	42920.8	4215.32	0.000
Water type (W)	2	18.1	1.78	0.171

Zeolite level (Z)	3	1009.9	99.19	0.000
Curing age (d)	6	1283.5	126.05	0.000
Interaction of C × W	2	177.3	17.41	0.000
Interaction of C × Z	3	180.6	17.74	0.000
Interaction of C × d	6	229.3	22.52	0.000
Interaction of W × Z	6	75.5	7.41	0.000
Interaction of W × d	12	3.3	0.33	0.984
Interaction of Z × d	18	12.3	1.21	0.252
Interaction of C × W ×	6	83.3	8.18	0.000
Z	12	7.3	0.71	0.737
Interaction of C × W ×	18	4.7	0.46	0.971
d Interaction of C × Z	36	9.6	0.94	0.568
× d Interaction of W ×	36	12.9	1.27	0.149
Z × d Interaction of C	336	10.2		–
× W × Z × d Total				
Error				

2.6 Means comparison (LSD) results

When the result of ANOVA test shows the significant simple effect or interaction, post hoc tests like LSD are used as an complementary part in order to explore the differences between means of multiple groups. Also, in order to the better conclusion, it is recommended that if the interaction of independent factors on the dependent factor was significant, the main attention was paid to the means comparison of interactions of studied factors instead of their simple effects. Accordingly, in the following sections results of means comparison are presented only for sources of variation which had a significant effect on compressive strength of concrete, based on the results of the ANOVA test. Meanwhile, it is necessary to mention that in each means comparison, treatments with at least one similar sign (letter) did not have a significant difference with each other at 5 % statistical level.

2.6.1 Means comparison of simple effects

The means comparison results of the simple effects of four studied factors on compressive strength of concrete are shown in Fig. 1. As expected, increasing the cement content from 250 to 350 kg m^{-3} caused a significant increase (151 % on average) in the compressive strength of concrete. Also, the rate of increasing the compressive strength of concrete in each curing age compared with the aforetime curing age was 25 %, 21 %, 10 %, 5 %, 1 %, and 2 % for curing ages 3 up to 365 days, respectively. This rate was significant for curing ages 3 up to 90 days, while it was not significant for curing ages 180 and 365 days (Fig. 1). The results showed that there was no significant difference between all types of mixing water in term of compressive strength of concrete (Fig. 1). Therefore, it seems that using the treated industrial wastewater has no significant destructive effect on compressive strength of plain concrete. The use of treated industrial wastewater (W3) although slightly reduced (approximately only 2 %) the concrete compressive strength compared with tap water (W1), this decrement was not statistically significant (Fig. 1) and was within the permissible limit less than 10 % according to ASTM C1602 [42], ISO 12439 [44], BS EN1008 [36], MPO [32] standards. This finding is completely in accordance with the results of Asadollahfardi and Mahdavi [10] about industrial treated wastewater. Asadollahfardi and Mahdavi [10] and Asadollahfardi et al. [6] by comparison of the different SEM tests found that the distance of the crystals were greater in the concrete made with both industrial and domestic treated wastewater than ones made with drinking water and then concluded that this further vacant space could be one of the reasons for reducing the compressive strength of the concrete made with treated industrial wastewater. However, the use of combined water (W2) resulted in a slight insignificant increase in compressive strength of concrete compared with tap water (0.9 %) and treated industrial wastewater (3.0 %). In this regard, results of Noruzman et al.

[46] and Lee et al. [11] showed that the compressive strength of concrete made with treated effluent of heavy industries and treated domestic wastewater, respectively, was greater than potable water due to higher concentration of sodium chloride and fine solids. Fine solids of wastewater could fill voids in the concrete matrix. The reduction in the number of voids normally has a significant impact on the strength properties [46]. The higher concentration of sodium and calcium chlorides also was reported as a reason for increasing the early strength of concrete [11,46]. Calcium chloride increases the rate of heat

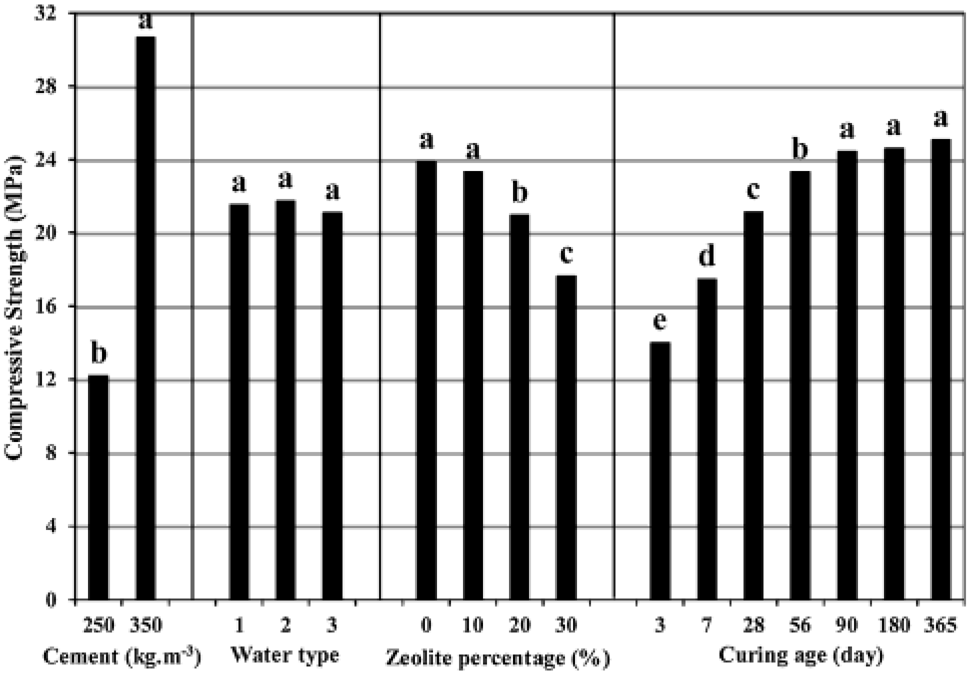


Fig. 1. The simple effect of cement content, water type (1: tap water; 2: combined water & 3: treated industrial wastewater), zeolite percentage and curing age on the compressive strength of concrete.

liberation during the first few hours after mixing and acts as catalyst in the reaction of hydration of C_3S and C_2S [11]. Also, Asadollahfardi and Mahdavi [10] and Asadollahfardi et al. [6] reported that the surface electrical resistivity of concrete samples, as an electrical indicator of permeability, made with both types of domestic and industrial treated wastewater was within a very low chloride ion penetration range, similar to ones made with drinking water. Their results as well as findings of Noruzman et al. [46] showed that the use of these types of unconventional water did not affect the concrete water absorption percentage in comparison with using drinking water. These results indicated that two indicators of concrete durability did not significantly decrease by unconventional water usage compared to drinking water.

The results showed that substitution of cement with 10 %, 20 %, and 30 % of zeolite led to the reduction of compressive strength of concrete compared to the control treatment (non-zeolite) by 2.4 %, 12.3 %, and 26.1 %. However, the reduction was not statistically significant in the substitution of 10 % (Fig. 1). These results were in accordance with the findings of Kaboosi et al. [14], Vejmelková et al. [19], Najimi et al. [17] and Vejmelková et al. [24]. Their results showed that substitution of cement with zeolite in the rate of 10, 20 and 40; 10, 20 and 30; 15 and 30 as well as 10, 20 and 40 percent, averagely on different curing age, reduced the compressive strength of concrete by 12.8, 15.1 and 38.6; 5.6, 7.2 and 20.0; 7.5 and 22.6 and 11.9, 27.2 and 51.2 percent, respectively.

Finally, it should be emphasised that regarding the some significant interactions of studied factors with each other (Table 4) final conclusion cannot be based on means comparison of simple effects. So, results of two- and three- way interactions should be considered.

2.6.2 Means comparison of two-way interactions

The means comparison of the two-way interactions of cement water type and cement content zeolite percentage are presented in Fig. 2. The results indicated that in the cement content 250 kg m^{-3}, compressive strengths of concrete samples made with combined water (C1W2) and treated industrial wastewater (C1W3) were significantly higher than ones made with tap water (C1W1) by 19.3 % and 9.6 %, respectively. The similar results were reported by Kaboosi et al. [14] and Su et al. [12]. They showed that compressive strength of concrete made with greywater and wash water in the cement content 220–300 and 250 kg m^{-3} was greater than tap water, respectively. Su et al. [12] stated that it was probably due to the high alkalinity of wash water. However, in the cement content 350 kg m^{-3}, the use of combined water (C2W2) and treated industrial wastewater (C2W3) resulted in a significant decrease in the compressive strength of concrete compared with tap water (C2W1) by 5.5 % and 6.1 %, respectively. Therefore, the use of treated industrial wastewater and combined water in this cement content did not lead to the reduction of concrete compressive strength more than allowable range (less than 10 %). These results were completely in accordance with the findings of Asadollahfardi and Mahdavi [10]Kaboosi et al.

[14]Meena and Luhar [2] and Asadollahfardi et al. [6]. The results of Asadollahfardi and Mahdavi [10]Meena and Luhar [2] and Kaboosi et al. [14] showed that although the use of treated industrial and domestic wastewater and greywater as mixing water in the high cement contents (350—450 kg m^{-3}) led to the reduction of compressive strength of concrete compared

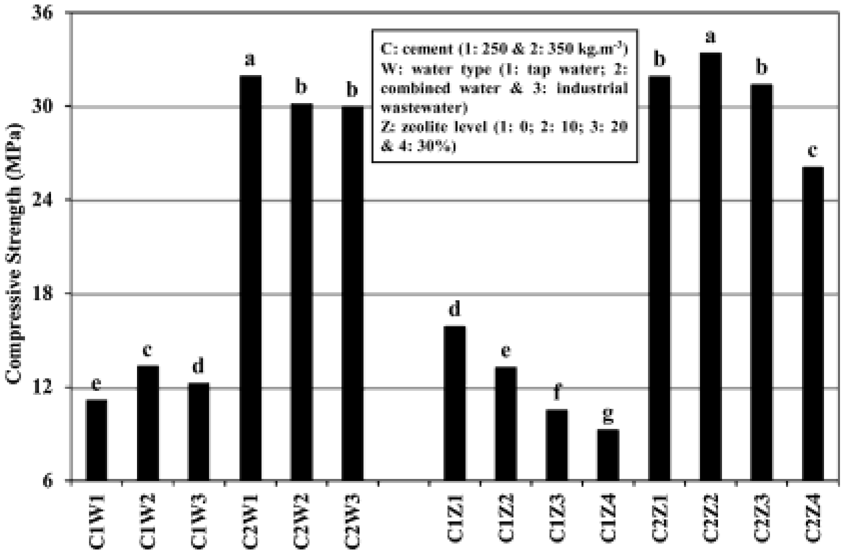


Fig. 2. Means comparison of two-way interactions of cement content × water type and cement content × zeolite percentage on the compressive strength of concrete.

with the drinking water, the reduction was less than permissible limit 10 %. Also, Asadollahfardi et al. [6] reported that treated domestic wastewater resulted in slight reduction (6 %) in the 28-days compressive strength of concrete in high cement content (300 400 kg m^{-3}) likely due to the BOD_5 and COD contents of wastewater. Meanwhile, the study of Ismail and Al-Hashmi [51] showed that using the polyvinyl acetate resin wastewater instead of fresh water in the concrete with high cement content (666 kg m^{-3}) slightly increased the compressive strength of concrete. It was while the wastewater has very high contents of COD, TDS and TSS. Therefore, it seems that using treated industrial wastewater and its equal combination with tap water as mixing water in both cement contents can be recommended since the reduction of concrete compressive strength was within the allowable range less than 10 %.

The means comparison of two-way interaction of×cement content zeolite percentage (Fig. 2) showed that in the cement content 250 kg m^{-3}, all levels of zeolite application resulted in the significant reduction in compressive strength of concrete compared to the non-zeolite content. However, in the cement content 350 kg m^{-3}, zeolite application not only caused no significant reduction (only about 1.6 %) in the compressive strength of concrete in the zeolite level 20 %; but also it resulted in a significant increase (about 4.7 %) in the compressive strength of concrete in the zeolite level 10 %. Similar findings were reported by Kaboosi et al. [14] and Ahmadi et al. [52]. The increase of compressive strength of concrete in the high cement content by zeolite application was attributed to the reaction of zeolite with calcium hydroxide (CH) of cement. In the high cement content, more volume of CH reacts with the zeolite and produces more volume of calcium silicate hydrate (CSH) gel. It results in more filled voids and improves the concrete microstructure and finally increases the compressive strength of concrete [52]. However, application of 30 % of zeolite significantly decreased the compressive strength of concrete in both cement contents. Moreover, the variation rate in the compressive strength of concrete samples affected by the zeolite usage compared with ones made without zeolite was much less in the cement content 350 kg m^{-3} than 250 kg m^{-3} since this rate for 10 %, 20 %, and 30 % of zeolite usage instead of cement in the mix design was 16.6 %, 33.6 %, and 41.7 % in the cement content 250 kg m^{-3} and +4.7 %, 1.6 %, and 18.3 % in the cement content 350 kg m^{-3}, respectively. These rates and this trend were reported by Kaboosi et al. [14]. Therefore, it seems that the application of zeolite at the optimum proportion of 20 % in the concrete mix design with high cement content did not create special restriction while its usage in the mix design with low cement content cannot be recommended. Moreover, since interaction of cement content water type zeolite percentage was significant; more attention should be paid to this three-way interaction (Section 3.3.3) instead of two above two-way interactions (Fig. 2).

The means comparison of the two-way interaction of cement×content curing age (Fig. 3) indicated that the increase rate of compressive strength of concrete by time was significantly different in two cement contents; so that, increase of concrete compressive strength at curing ages 7, 28, 56, 90, 180, and 365 days compared with 3-day curing age was 22.9 %, 48.9 %, 61.4 %, 72.8 %, 75.3 %, and 84.3 % in the cement content 250 kg m^{-3}, while it was 25.7 %, 51.8 %, 68.4 %, 75.3 %, 76.0 %, and 77.0 % in the cement content 350 kg m^{-3}, respectively. Therefore, except for curing age 365 days, this rate was greater in the cement content 350 than 250 kg m^{-3}. Similar rates and trend were reported by Kaboosi et al. [14].

The means comparison of two-way interaction of water type   zeolite percentage (Fig. 4) showed that the highest compressive strength was obtained in the mix design of combined water without zeolite content (W2Z1) and the treated industrial wastewater with zeolite 10 % (W3Z2) so that their superiority were statistically significant compared with the other mixes. Also, the lowest compressive strength with a significant difference compared to the others was obtained in all three water types with zeolite 30 %. Totally, the use of treated industrial wastewater or combined water plus zeolite   e 10 % and tap water plus zeolite 20 % resulted in no significant reduction in the concrete compressive strength compared with the control mix design (W1Z1: tap water without zeolite content), and even led to a significant increase of this property in some cases. Above results are very similar to Kaboosi et al. [14] when investigating the simultaneous use of greywater and zeolite as mixing water and pozzolanic material in the concrete mix design, respectively. These findings also in accordance with the results of Su et al. [12] that reported the alkaline nature of unconventional water such as wash water of ready-mix concrete plant not only accelerate the cement hydration but also activate the pozzolanic reaction of mineral admixtures. Therefore, it enhances the compressive strength of concrete made with these types of water. Ion charges and size are two factors that may affect the their immobilization process. The immobilizing of organic pollutants may be grouped into three classes, including 1- direct immobilization of organic pollutants; 2- direct immobilization of organic pollutants after adsorption; 3- immobilization of organic pollutants by applying oxidizing and reducing agents [6]. Natural zeolites have valuable physicochemical properties, such as high specific surface area (300   700 m^2 g^{-1}), low specific gravity, microporous structure and numerous pores (porosities between 0.1 and 0.35 cm^3 g^{-1} with pore sizes between 0.3 and 1 nm), large channel and cavity system, electrical imbalance (negative charges) due to the presence of Al-O tetrahedroids which results in its high cation exchange capacity (0.6 and 2.3 meq g^{-1}), high adsorption capacity (nearly 40 % of its own weight), high ability to

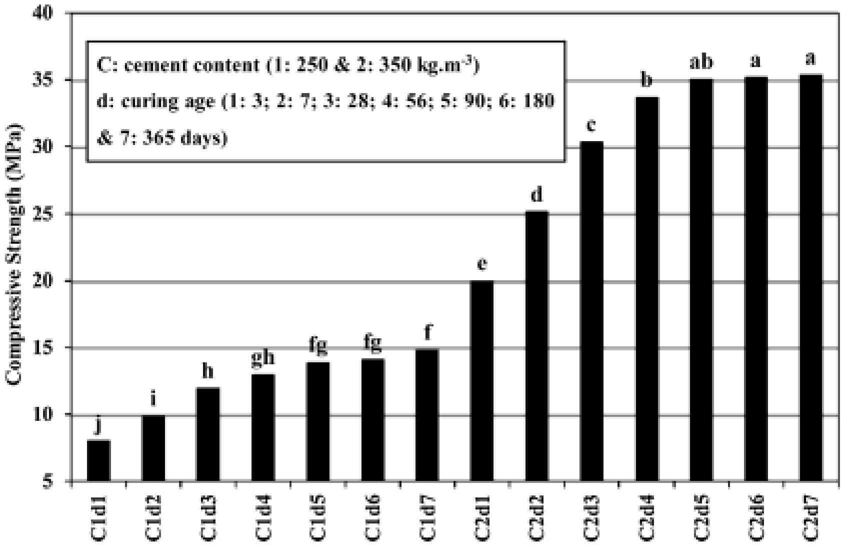


Fig. 3. Means comparison of two-way interaction of cement content × curing age on the compressive strength.

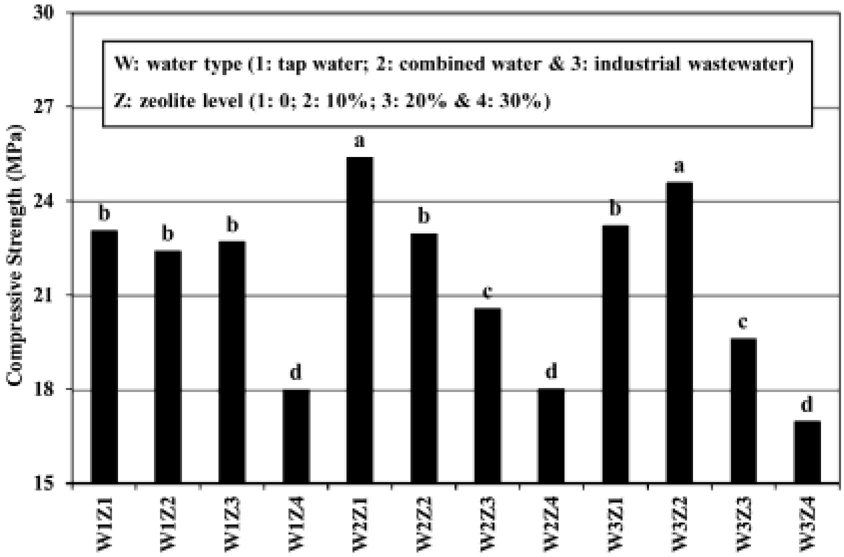


Fig. 4. Means comparison of two-way interaction of water type × zeolite percentage on the compressive strength.

internal curing by water adsorption and desorption, molecular sieving, catalysis and sorption [14,18,20,21,23]. These properties result in natural zeolites exhibit molecular sieving properties under appropriate conditions which can efficiently chloride and sulfate in the unconventional studied water on one hand and direct immobilization of organic pollutants after adsorption by zeolite (immobilization class 2) due to high specific surface area and CEC and molecular sieving properties of zeolite on the other hand, adverse effect of impurities of studied unconventional mixing water in the concrete compressive strength was not significant. In this regard, the results of Vejmelková et al. [19] showed that the use of zeolite in concrete mix design instead of cement led to a clear positive effect on the concrete resistance against corrosive salts including Na_2SO_4, $MgCl_2$, and NH_4Cl. The review study of Tran et al. [18] also showed that concrete permeability to water and chloride very apparently decreased when 5 %–30 % of the natural zeolite was added into concrete mix design. However, the use of studied unconventional waters with zeolite 20 % and 30 % significantly reduced the compressive strength of concrete compared with the control treatment (W1Z1). Similarly, it was reported that substituting cement by zeolite 20 % caused no reduction in the compressive strength of concrete when tap water was used as mixing water [15,53], which was closely accordance with the results of the present study. Also, the results showed that the average reduction rate of concrete compressive (C1Z1W1: tap water without zeolite usage) while it did not significantly vary in the treatments of industrial wastewater without zeolite (C1Z1W3), combined water with 10 % of zeolite (C1Z2W2) and industrial wastewater with 10 % of zeolite (C1Z2W3). However, in the cement content 350 kg m^{-3}, compressive strength of concrete in the treatments of tap water with 10 % of zeolite (C2Z2W1), industrial wastewater with 10 % of zeolite (C2Z2W3), and tap water with 20 % of zeolite (C2Z3W1) were significantly greater than the control treatment (C2Z1W1: tap water without zeolite usage) by 8.3 %, 11.3 %, and 9.4 %, respectively. In addition, the reduction of compressive strength in no treatment in this cement content was statistically significant except in the treatments of zeolite 20 % with combined water (C2Z3W2) as well as zeolite 30 % with tap water (C2Z4W1), and with combined water (C2Z4W2), and also with the wastewater (C2Z4W3) when compared with the control treatment (C2Z1W1). Therefore, the results indicated that the use of zeolite 20 % with the treated industrial wastewater or with the combined water not only resulted in no significant reduction in the compressive strength of concrete; but also significantly increased it in some cases, particularly in high cement content. These results were very close to findings of Kaboosi et al. [14] when investigating the effect of zeolite and greywater in different cement contents on compressive
ted wastewater < tap water.

2.6.3 Means comparison of three-way interaction

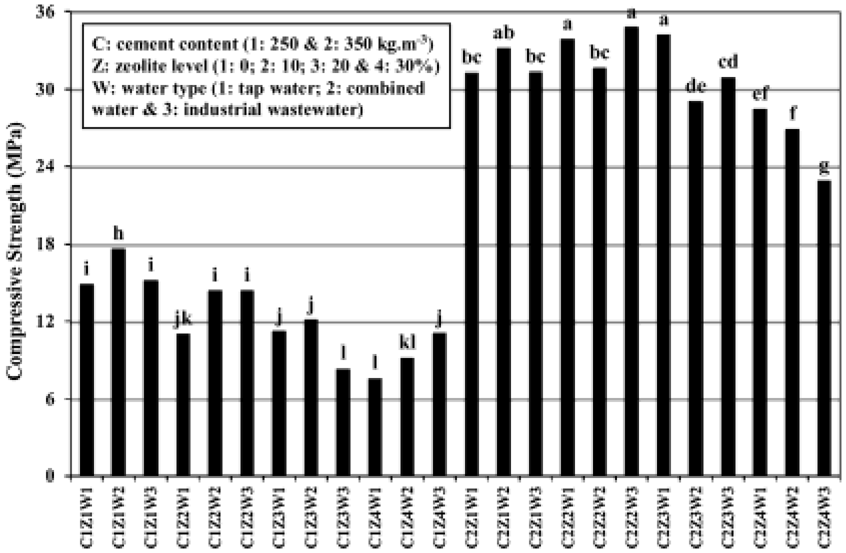


Fig. 5. Means comparison of three-way interaction of cement content × zeolite percentage × water type on the compressive strength of concrete.

strength of plain concrete. It was reported that the mix design with highest cement content is preferable as far as chloride hazard in mixing water is concerned [50]. Also, these results were in accordance with the findings of Wegian [45] who reported that resistance against all possible forms of deterioration is distinctly improved by using higher cement contents. Increasing the cement content due to more cement matrix exposed to salts, better workability to the mix, enhancement of the bond strength among concrete components and retarding the strength loss lead to the construction of concrete with higher resistance to the attack of chemical solutions and salts [45]. In this regard, it was also reported that due to more resistance to sulfate attack and chlorine penetration in the zeolite-containing concrete, improvement in the compressive strength was happen in the zeolite levels up to 20 % compared with non-application of zeolite, although this superiority gradually decreased when the added zeolite exceeded 10 % level [18]. Generally, it can be elicited from the findings of present study that the simultaneous use of unconventional water as mixing water with zeolite up to 10 % in low cement content and also with zeolite up to 20 % in the high cement content can be recommendable for construction of plain concrete.

IV. CONCLUSION

1) The results showed that there is a need to further and detailed studies on existing mixing water standards in two points of view: 1- uniforming the existing codes in term of permissible limit of impurities in the concrete mixing water; 2- investigation of the effect of some constituents such as heavy metal, fluoride, detergent and emerging contaminants in mixing water on the compressive strength of concrete.

2) In terms of specifications of mixing water, the results generally indicated that the treated industrial wastewater and combined water can be used satisfactorily in the construction of plain concrete. However, due to higher contents of chloride and sulfate in these types of unconventional water than tap water, it could raise concerns about the corrosion potential in the reinforced concrete and sulfate attack and cracking in the concrete, respectively. Therefore, it is recommended that durability tests be conducted to investigate the potential of corrosion and sulfate attack.

3) It seems that the particular properties of natural zeolites such as high specific surface area and cation exchange capacity and also its molecular sieving can enable zeolite to effectively adsorb different contaminants of unconventional mixing waters and consequently it mitigates the adverse effects of these substances on compressive strength of concrete.

Declaration of Competing Interest

The authors declare that there is no conflict of interest regarding the publication of this article.

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