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Effect of Incorporating Natural Zeolitic Tuffs in Concrete Mixed and Cured Using Seawater

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Abstract: Concrete production has increasingly used seawater to overcome the challenge of freshwater scarcity. Although the use of seawater in concrete still has a controversial reputation, it is a promising application, particularly when combined with mineral admixtures such as natural zeolitic tuffs (ZT). This paper aims to investigate the effect of using locally quarried ZT on the strength of unreinforced concrete mixed and/or cured using seawater. The mix proportions of the concrete were selected to obtain the optimum combination for the M20 grade of concrete with a water-to-cement ratio of 0.69. Moreover, 150mm-cubes and cylinders of 100 mm diameter by 200mm height were cast from the concrete mixtures, which contain 0%, 5%, 7.5%, 10%, and 25% of ZT as a partial replacement of silica sand. Splitting tensile tests and compressive strength tests were conducted on these specimens at 7, 28, and 90 days. The results show the harmful effect of seawater on the strength of plain concrete (without ZT) at 7, 28, and 90 days of curing, especially when seawater is used in both mixing and curing of the concrete. However, adding ZT in seawater-based concrete improved its strength apparently, especially at early curing ages. For example, using 10% of ZT as a partial replacement of silica sand increased the compressive strength of seawater based-concrete by 105.4%, 28.3%, and 34.6% after 7, 28, and 90 days of curing, compared with concrete without ZT and produced using seawater. These results contribute to the enhancement of the sustainability of both freshwater and concrete material through the use of ZT in producing concrete, particularly in areas where freshwater is scarce or expensive.

Keywords: zeolitic tuffs; seawater; curing; compressive strength tests; sustainability

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1. Introduction

Due to its outstanding properties, concrete is the most versatile and widespread construction material in the world. The world produces over 30 billion metric tonnes of concrete annually [1]. The ever-growing demand for concrete worldwide leads to a substantial increase in water requirements for concrete production. However, in some regions, water scarcity is a significant and pressing concern, while the concrete industry contributes to challenges posed to sustainable water management. A shortage of water can be considered the most critical problem in Jordan, as it has been ranked number ten among the world's most water-scarce countries [2]. The average precipitation in about 96% of the country is less than 300 mm per year [3]. To alleviate this serious problem, reducing the usage of freshwater in the concrete industry as well as in other sectors is a necessity. One possible way to reduce the consumption of freshwater in the construction industry is by using non-conventional water supplies such as wastewater and seawater. Therefore, it is of significant importance to conduct continuous investigations on using unconventional water resources and optimizing conditions for their usage in the construction industry.

Several studies have investigated the effect of seawater on concrete, as reviewed by [4,5]. The high chloride content in seawater can penetrate the concrete cover and cause the corrosion of reinforcing steel bars, which leads to cracking, spalling, and the loss of structural integrity of the concrete over time. To deal with the corrosion problem, fiber-reinforced polymer (FRP) and stainless-steel reinforcement can be utilized as alternatives to traditional steel rebars to reinforce seawater concrete [6]. For non-reinforced concrete, research studies have shown that seawater leads to changes in the fresh and hardened properties. The use of seawater accelerates the cement hydration process due to the presence of chloride ions, leading to an earlier production of calcium hyrdosilicates (C-H-S gel). The salt NaCl can react with $Ca(OH)_2$ existing in concrete to produce calcium chloride $CaCl_2$, which serves as a catalyst for cement during the early hydration stage [5].

$$Ca(OH)_2 + 2NaCl \rightarrow CaCl_2 + 2Na^+ + 2OH^-$$

The acceleration in the hydration reaction causes a reduction in the workability of fresh concrete and thereby decreases the initial and final setting times [7,8]. Moreover, the results of research studies on seawater in concrete have shown a slight increase in early strength (at the curing age of 7 days), but at later ages, such as 90 days, the strength fell by 3.8–14.5%, compared with freshwater concrete [7,9]. According to the literature, the increase in the early strength of seawater concrete could be due to the presence of NaCl, which accelerates the cement hydration process. This acceleration leads to a reduction in the porosity and enhancement in the microstructure due to the formation of more hydration products [7,10]. At the same time, the lower long-term strength could be due to the presence of magnesium sulfate ($MgSO_4$) in seawater, which reacts with calcium hydroxide to form expansive products and hence induce microcracks and reduce long-term strength [5]. In addition, the formation of salt crystallization affects strength gain [9], particularly when seawater-mixed concrete is produced with a high water-to-cement ratio. Various concrete standards have provided recommendations for utilizing seawater in the production of concrete and proposed threshold values for chlorides and sulfate contents.

Generally, concrete resistance to aggressive environments can be significantly improved by introducing supplementary cementitious materials (such as fly ash and natural pozzolanic materials) in concrete with appropriate proportions and using a low w/c ratio [11–16]. Natural Zeolites are one of the pozzolanic materials that contain hydrated aluminosilicates of alkali and alkaline earth elements [17]. Due to its cementing characteristics, natural zeolite has been utilized in construction since ancient times. In recent decades, the utilization of natural zeolite in the production of cement has gained significant attention [18–20]. The microstructure of hardened cement improves significantly due to the pozzolanic reaction of natural zeolites with the calcium hydroxide $Ca(OH)_2$ produced during the hydration process of cement. The natural zeolites, which consist of reactive SiO_2 and AL_2O_3, combine with $Ca(OH)_2$ to form an extra amount of dense calcium hyrdosilicates (C-S-H gel). In Jordan, zeolites are mainly associated with volcanic tuff and are thus called zeolitic tuff (ZT). Huge reserves of ZT are found in various locations within Jordan and are used mainly in agriculture and wastewater treatment plants [17,21,22].

Basically, ZT can be added directly to the concrete mixtures as a partial replacement for cement, fine sand, or coarse aggregates. Concrete mixes incorporating natural zeolite as an admixture have lower early strength due to their pozzolanic activity and enhanced durability properties because of a reduction in the permeability to water and chloride [23–28]. Nevertheless, the experimental results may differ according to the type, structure, and purity of the used zeolite, and in some cases, the results were in contradiction [23,24]. However, limited studies have been conducted to investigate the effect of utilizing Jordanian ZTs in concrete [24–35]. In addition, there are very few studies that have studied the effect of adding zeolite to concrete mixtures produced using non-freshwater. Koobsi et al. [36] have investigated the effect of using greywater in the mixing and curing of concrete that contains natural zeolite. They found that zeolite can efficiently adsorb the contaminants in the concrete mix design and mitigate their negative effects on concrete [28,37].

This paper investigates the feasibility of incorporating locally quarried ZT in concrete produced using seawater and compares the results with concrete produced using freshwater. The main objective of this study is to investigate the effect of adding ZT on the strength of concrete mixed and/or cured using seawater. Concrete mixtures containing 0%, 5%, 7.5%, 10%, and 25% of ZT as partial replacement of fine sand were prepared using two types of mixing water: freshwater and seawater. Concrete specimens produced by using freshwater were divided into two groups: one group cured in freshwater and one group cured in seawater, while all specimens produced by using seawater were cured in seawater. Splitting tensile tests were conducted on cylindrical specimens, and compressive strength tests were conducted on cubic specimens at curing ages 7, 28, and 90 days.

2. Materials and Methods

2.1. Materials

The materials used in preparing concrete mixtures in this research include crushed limestone, silica sand, white cement, locally quarried zeolitic tuff, tap water, seawater, and superplasticizers. Seawater was created in the laboratory by adding 35 g of non-iodized sea salt per liter of water to achieve a salinity of 3.5% that mimics the salinity of natural seawater. The maximum size of the coarse aggregate used was 20 mm. Two types of fine sand were used: crushed limestone and silica sand, in addition to ZT, which was added as a partial replacement for silica sand. The ground zeolitic tuffs have the same fineness modulus as silica sand, which is valued at 1.67. The source of ZT was a mine in Tafelah City, located in the south of Jordan. As shown in Figure 1, ZT has a reddish-brown color. The X-ray diffraction technique was used to identify and quantify the chemical composition of ZT, and it is presented in Table 1. The chemical composition of the used white cement and silica sand is also presented in Table 1.

Figure 1. Ground zeolitic tuffs.

Table 1. Chemical composition of zeolitic tuff and white cement.

Item	Zeolitic Tuff	Silica Sand	White Cement
Fe_2O_3	9.72%	0.05%	0.5%
MnO	0.16%	0.02%	-
TiO_2	3.07%	0.14%	-
P_2O_5	0.15%	0.01%	-
SiO_2	39.36%	98.9%	20–25%
Al_2O_3	11.42%	0.46%	3–6%
MgO	9.88%	0.01%	1–3%
Na_2O	2.51%	0.09%	-
CaO	9.29%	0.06%	63–68%
K_2O	1.57%	0.01%	2%
Mn_2O_3	-	-	0.25%
SO_3	-	0.05%	2–4%

2.2. Mix Proportions

In the current study, eight concrete mixtures have been prepared to analyze the effect of incorporating ZT in seawater-mixed and/or cured concrete. These mixtures can be divided into two groups based on mixing water: the first group consists of five mixtures that were mixed with freshwater, and each mix contains 0%, 5%, 7.5%, 10%, and 25% of ZT as a partial replacement for silica sand. The specimens cast from this group were either cured in freshwater or in artificial seawater until the day of testing. The second group consists of three mixtures that were mixed and cured in seawater, incorporating 0%, 5%, and 10% silica sand, and replaced with ZT. Table 2 shows the details of the mix design. The mixtures were named according to the percentage replacement ratio of silica sand by ZT, followed by two letters indicating the type of mixing water and curing water, where F stands for freshwater, and S stands for seawater. For example, specimens prepared from mixture M5ZFS have 5% ZT as a partial replacement of silica sand and are mixed with freshwater but cured in seawater.

Table 2. Mixture design of concrete specimens.

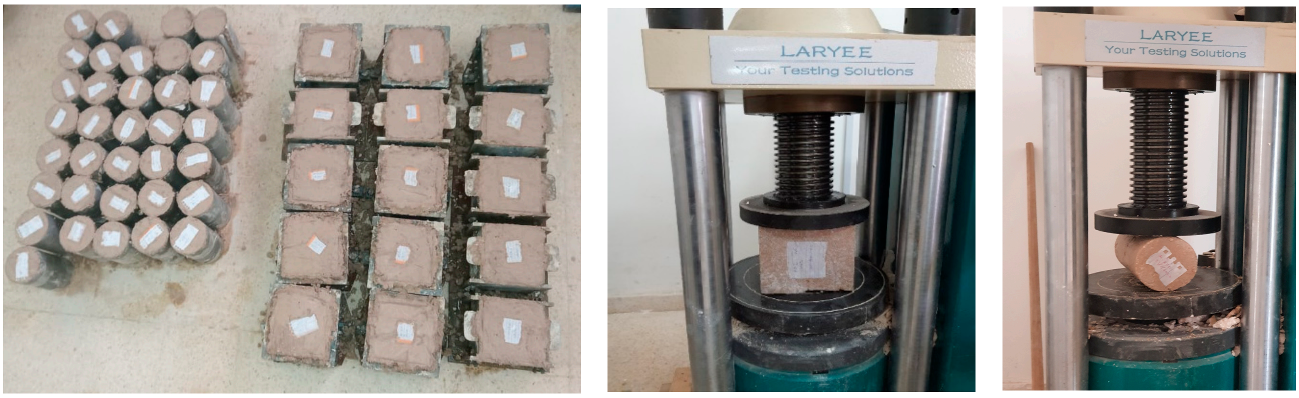
Mix No.	Zeolitic Tuff % [a]	Mixing Water	Curing Water	Silica Sand kg/m^3	Zeolitic Tuff kg/m^3
M0ZFF	0%	Freshwater	Freshwater	834	0
M5ZFF	5%	Freshwater	Freshwater	792.3	41.7
M7.5ZFF	7.5%	Freshwater	Freshwater	771.45	62.55
M10ZFF	10%	Freshwater	Freshwater	750.6	83.4
M25ZFF	25%	Freshwater	Freshwater	625.5	208.5
M0ZFS	0%	Freshwater	Seawater	834	0
M5ZFS	5%	Freshwater	Seawater	792.3	41.7
M10ZFS	10%	Freshwater	Seawater	750.6	83.4
M25ZFS	25%	Freshwater	Seawater	625.5	208.5
M0ZSS	0%	Seawater	Seawater	834	0
M5ZSS	5%	Seawater	Seawater	792.3	41.7
M10ZSS	10%	seawater	Seawater	750.6	83.4

[a] % of ZT add to concrete mixture as a partial replacement of silica sand.

The mix proportions of the concrete were selected to obtain the optimum combination for the M20 grade of concrete according to ACI standards 211 [38], with a water-to-cement ratio of 0.69. This strength grade is commonly used in Jordan for the construction of several concrete structures. All mixtures in this experiment have the same cement content (275 kg/m^3) and the same coarse aggregate content (1056 kg/m^3). The incorporation of ZT in the concrete mixtures increases the demand for water because of the high porosity of the ZT and its platy microstructure [23]. Therefore, a dosage of superplasticizer was added to concrete mixtures to keep the workability constant, with a slump value ranging between 30 and 60 mm.

2.3. Preparation and Testing of the Specimens

Casting, curing, and testing procedures were conducted according to ASTM C 192 [39]. To prepare the concrete mixtures, dry ingredients were added first to a laboratory batch mixer and mixed for a minute, then water and superplasticizer were added gradually while the mixer was running. Immediately after mixing, the slump test was conducted, and the specimens were cast and compacted using a vibrating table. The specimens were left in the laboratory environment for 24 h. Then, they were demolded and cured either in freshwater or seawater at room temperature until the day of testing (Figure 2a). Compressive strength tests were conducted on 150-mm cubic specimens (Figure 2b), according to the standard test method for compressive strength ASTM 39/C 39M [40]. The splitting tensile tests were performed on cylindrical specimens 100 mm in diameter and 200mm in length (Figure 2c), according to the ASTM C496/C496M-11 [41] test method. These tests were carried out at the ages of 7, 28, and 90 days.



(**a**) (**b**) (**c**)

Figure 2. (**a**) Molded concrete specimens. (**b**) Compressive strength test. (**c**) Splitting tensile test.

3. Results and Discussion

The strength of all concrete specimens showed a continuous strength development over the period of curing, as shown in Tables 3–5. The values in these tables are the average of the results from three specimens of each mixture. It can be seen that the desired grade of strength was achieved by obtaining a compressive strength of 24.31 MPa for the plain concrete M0ZFF. In general, incorporating ZT reduces the compressive strength of concrete specimens that were mixed with freshwater at all ages. Either these specimens were cured in freshwater, as seen in Table 3, or in seawater, as seen in Table 4. Similar findings have been reported by other research studies in the literature [23,42,43]. In this regard, Najimi et al. [23] have discussed the negative effect of natural zeolite on the compressive strength of concrete with high water-to-cement ratios. Chan and Ji [42] have found that using a water-to-cement ratio higher than 0.45 reduced the compressive strength of concrete containing natural zeolites, compared with control concrete without natural zeolite. Whereas, when the water-to-cement ratio is lower than 0.45, the obtained compressive strength is higher than that for control concrete. In another study, Ahmadi et al. [25] have reported an increase in the compressive strength of concrete containing natural zeolites with a water-to-cement ratio of 0.4 at curing ages of 3, 7, 28, and 90 days. However, when the water-to-cement ratio increased to 0.5, the compressive strength decreased at all ages [23]. Poon [43] investigated the impact of the water-to-cement ratio on the compressive strength of cement paste that contains natural zeolite. Their findings revealed that zeolites significantly enhance the strength of the paste, particularly when the water-to-cement ratio is low. It is worth noting that the least reduction in strength was in the specimens that contained 25% of ZT (M25ZFF and M25ZFS) as a partial replacement of silica sand, indicating the positive effect of incorporating higher amounts of ZT rather than 5%, 7.5%, and 10% percentages. For example, the compressive strength of M25ZFS decreased by 22.5% at the age of 7 days, which is comparted with M0ZFS, but it only decreased by 8.4% and 10.5% at the ages of 28 and 90 days of curing in seawater, respectively. The reduction in compressive strength at 90 days decreased as the percentage of ZT increased in the concrete mixture. This finding contradicts the results reported in [18]. As mentioned above, experimental results may differ significantly according to the type, structure, and purity of the used zeolite [23,24]. The improvement in compressive strength with increasing age can be attributed to the decrease in permeability, which is more significant at later ages due to the pozzolanic reaction of ZT [44–46].

Table 3. Effect of incorporating ZT in concrete that was mixed and cured with freshwater.

Mix No.	Compressive Strength (MPa) (Change %)						Splitting Tensile Strength (MPa) (Change %)			
	7 Days	**COV**	**28 Days**	**COV**	**90 Days**	**COV**	**7 Days**	**COV**	**28 Days**	**COV**
M0ZFF (Ref)	20.10	0.040	24.31	0.087	27.62	0.031	1.31	0.061	1.64	0.074
M5ZFF	13.31 (−38.8)	0.115	17.60 (−27.6)	0.074	21.40 (−22.5)	0.111	1.36 (+3.8)	0.138	1.76 (+7.3)	0.108
M7.5ZFF	9.91 (−50.6)	0.152	17.72 (−27.1)	0.078	21.86 (−20.8)	0.128	0.95 (−27.4)	0.092	1.20 (−26.8)	0.066
M10ZFF	11.01 (−45.2)	0.036	16.70 (−31.3)	0.167	22.13 (−19.9)	0.0875	1.10 (−16.0)	0.105	1.34 (−18.3)	0.111
M25ZFF	13.81 (−31.3)	0.050	20.31 (−16.5)	0.102	23.80 (−13.8)	0.020	1.22 (−6.9)	0.080	1.48 (−9.8)	0.080

Table 4. Effect of using seawater in curing of concrete containing ZT and mixed with freshwater.

Mix No.	Compressive Strength (MPa) (Change %)						Splitting Tensile Strength (Change %)			
	7 Days	**COV**	**28 Days**	**COV**	**90 Days**	**COV**	**7 Days**	**COV**	**28 Days**	**COV**
M0ZFS (Ref)	18.60	0.041	24.13	0.072	26.60	0.018	1.03	0.077	1.77	0.098
M5ZFS	14.31 (−23.1%)	0.054	19.21 (−20.4%)	0.095	21.02 (−21.0%)	0.049	1.26 (+22.3%)	0.138	1.58 (−10.7%)	0.030
M10ZFS	11.82 (−36.5%)	0.047	15.62 (−35.3%)	0.131	17.54 (−34.1%)	0.071	0.92 (−10.6%)	0.057	1.36 (−23.6%)	0.093
M25ZFS	14.41 (−22.5%)	0.025	22.11 (−8.4%)	0.078	23.82 (−10.5%)	0.076	1.32 (+28.2%)	0.215	1.60 (−9.6%)	0.052

Table 5. Effect of using seawater in curing and mixing of specimens containing ZT.

Mix No.	Compressive Strength (MPa) (Change %)						Splitting Tensile Strength (Change %)			
	7 Day	**COV**	**28 Days**	**COV**	**90 Days**	**COV**	**7 Days**	**COV**	**28 Days**	**COV**
M0ZSS (Ref)	10.52	0.115	18.23	0.054	19.70	0.074	0.88	0.052	1.23	0.089
M5ZSS	15.51 (+47.4%)	0.042	19.20 (+5.3%)	0.059	22.13 (+12.3%)	0.011	1.20 (+36.4%)	0.086	1.38 (+12.2%)	0.131
M10ZSS	21.61 (+105.4%)	0.040	23.40 (+28.3%)	0.021	26.51 (+34.6%)	0.070	1.42 (+61.4%)	0.069	1.73 (+40.7%)	0.075

Table 3 shows that the splitting tensile strength of all specimens mixed and cured in freshwater and containing ZT is less than that for the reference specimens M0ZFF except M5ZFF, which showed an increase of 3.8% and 7.3% at 7 and 28 days, respectively. Whereas the specimens mixed with freshwater and cured in seawater shown in Table 4, an increase in splitting tensile strength at 7 days and a reduction at 28 days was observed, except M10ZFS, which showed a 10.6% and 23.6% reduction in splitting tensile strength at 7 and 28 days respectively.

The positive effect of incorporating ZT in concrete that was mixed and cured in seawater can be clearly shown in Table 5. Both compressive and splitting tensile increased significantly at all ages when replacing 5% and 10% of silica sand with ZT, especially at the age of 7 days. For example, the compressive strength of M10ZSS doubled after 7 days, compared with M0ZSS. These results suggest that the reduction in early age strength of concrete containing ZT can be contradicted by using seawater in the production of the concrete. However, further tests are required to prove this hypothesis and determine the optimum content of ZT in a concrete mixture.

The effect of using seawater in the mixing and/or curing of concrete mixtures containing different percentages of ZT on its compressive and splitting tensile strength at 7, 28, and 90 days is illustrated in Figures 3–7. A plain concrete mixture of M0ZFS that was mixed with freshwater and then cured in seawater had a 7.4%, 0.7%, and 3.7% decrease in compressive strength after 7, 28, and 90 days, respectively, compared with M0ZFF. Nevertheless, when using seawater in both the mixing and curing of plain concrete, the compressive strength decreased significantly to 47.7%, 25.0%, and 28.7% at 7, 28, and 90 days of curing, compared with M0ZFF. A similar trend can also be observed for the splitting tensile strength of plain concrete produced by using seawater, as seen in Figures 6 and 7. The reduction in strength at an early age (at 7 days) contradicts the findings observed in [9], who reported an increase in the compressive strength of plain concrete produced using seawater at ages less than 14 days. This can be attributed to the high water-to-cement ratio used in this study [11]. On the other hand, Wegian [9] observed a definite decrease in the strength of seawater concrete for ages greater than 28 days, which is consistent with the findings of this research. The noticeable decline in strength of seawater-mixed concrete can be attributed to the formation of salt crystallization, which adversely affects strength gain as indicated in [9]. In addition, the long-term strength could be reduced due to the reaction of magnesium sulfate presented in seawater with calcium hydroxide to form expansive products that induce microcracks within concrete [5].

When replacing 5% of silica sand with ZT, the use of seawater for mixing and/or curing has a slight effect on the compressive strength of the concrete specimens at all tested ages. However, the compressive strength of M10ZSS increased significantly, compared with M10ZFF and M10ZFS at all ages. The compressive strength of M10ZSS increased by 96.3%, 40.1%, and 19.8% at 7, 28 and 90 days of curing, compared with M10ZFF. A similar increasing trend can also be observed in the splitting tensile strength of M10ZSS. These findings are consistent with other studies performed on different pozzolanic materials such as silica fume, fly ash, and slag [47,48]. The higher strength can be attributed to the pozzolanic reaction between ZT and calcium hydroxide, leading to a significant decrease in its quantity. Thus, there are fewer chances for calcium hydroxide to react with magnesium sulfate existing in seawater to generate expansive products. The obtained results suggest that the reactivity of ZT was enhanced when using seawater in the mixing of concrete, leading to the consumption of a larger quantity of calcium hydroxide. This leads to forming more C-S-H gel, which fills the pores of the concrete matrix and produces a denser microstructure. However, further tests to characterize the microstructure of the seawater-mixed concrete incorporating ZT are recommended.

These results suggest the suitability of local ZT in improving the resistance of seawater-mixed concrete, as well as the marine concrete structures that are continuously submerged in seawater to harsh salinity environments. However, further tests should be conducted for incorporating ZT in seawater-mixed concrete to investigate its durability properties and corrosion of steel in reinforced concrete.

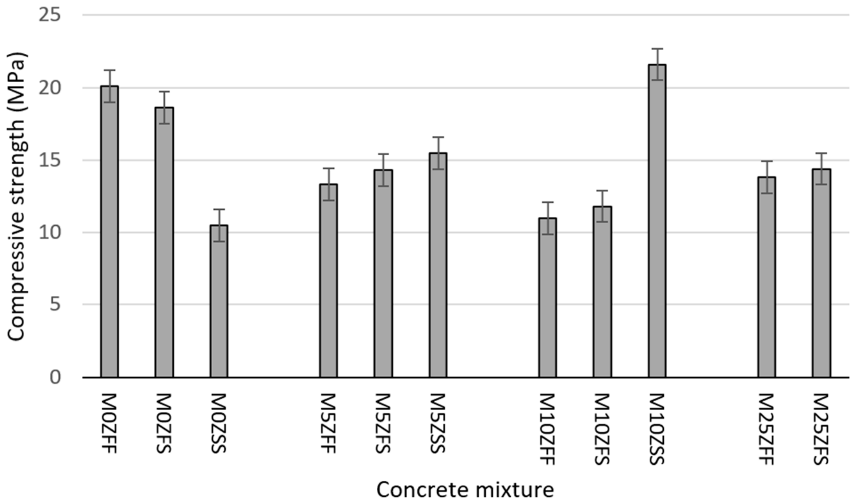


Figure 3. Compressive strength of concrete specimens cured and/or mixed with fresh and seawater incorporating ZT at age of 7 days.

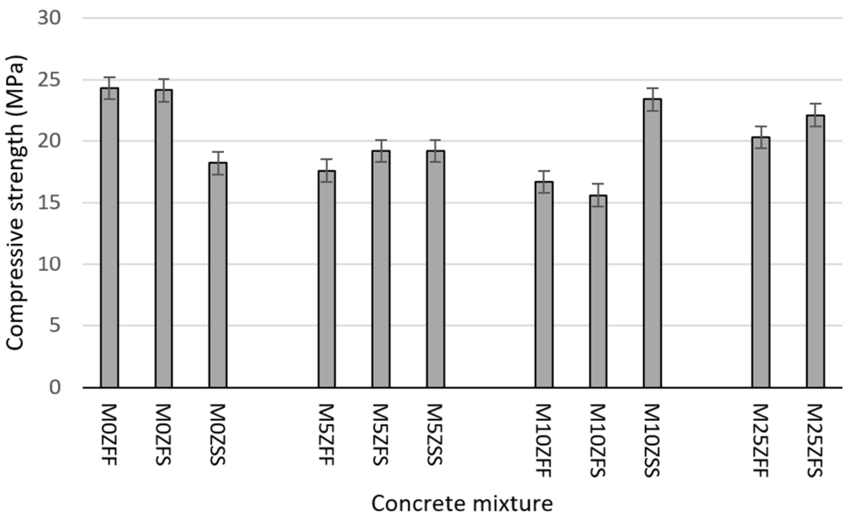


Figure 4. Compressive strength of concrete specimens cured and/or mixed with fresh and seawater incorporating ZT at age of 28 days.

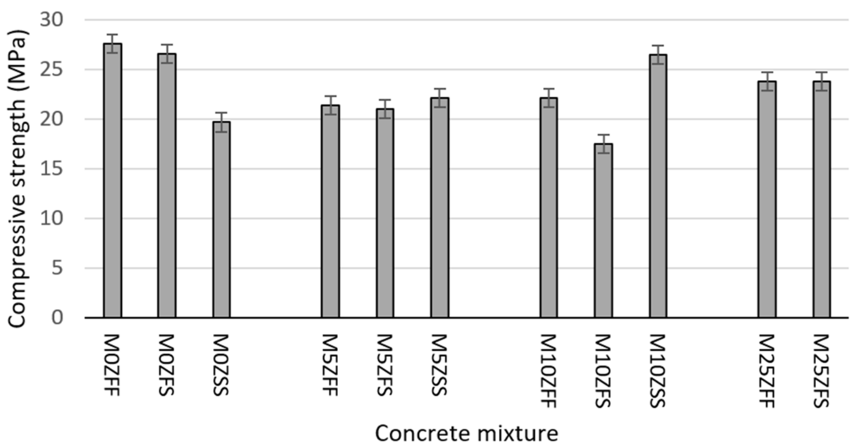


Figure 5. Compressive strength of concrete specimens cured and/or mixed with fresh and sea water incorporating ZT at age of 90 days.

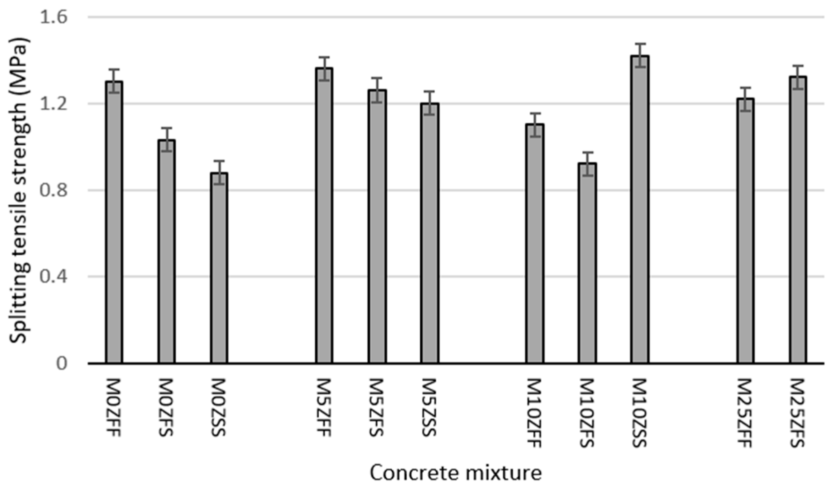


Figure 6. Splitting tensile strength of concrete specimens cured and/or mixed with fresh and seawater incorporating ZT at age of 7 days.

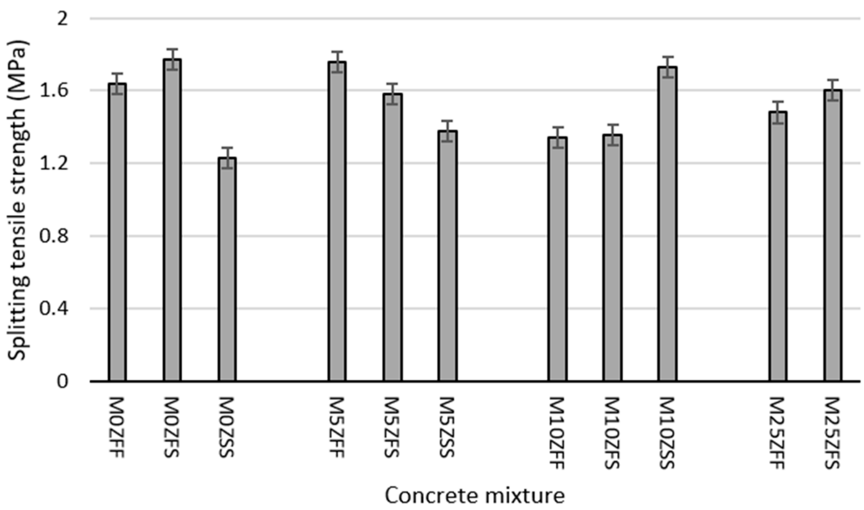


Figure 7. Splitting tensile strength of concrete specimens cured and/or mixed with fresh and seawater incorporating ZT at age of 28 days.

4. Conclusions

This paper has assessed the suitability of using locally queried ZT in the production of concrete cured and/or mixed with seawater. The results have shown that using ZT improves the strength of the seawater concrete even when a high water-to-cement ratio is used. The findings could have important applications in coastal areas and marine environments where seawater is easily accessible. In general, utilizing locally sourced ZT can support local industries. Moreover, adding ZT to concrete contributes to significant carbon and cost savings by lowering the use of Portland cement and reducing the need for long-term maintenance due to improvement in durability properties, which is associated with the pozzolanic properties of ZT. In addition, using seawater in concrete structures of marine environments and sourcing NZ from quarries and deposits close to the construction site could reduce the cost and emissions associated with transporting materials.

This should encourage engineers to adopt positive attitudes toward using ZT and seawater in concrete production. A comprehensive understanding of the performance of this kind of concrete paves the way toward developing more sustainable and cost-effective building materials for coastal areas. Therefore, we recommended conducting more tests to find the optimum ZT content in concrete mixtures mixed and/or cured with seawater. In addition, further tests were recommended to perform microstructure characterization, such as porosity measurements and SEM observations, and to investigate the durability properties of zeolitic-seawater concrete and its effect on the corrosion of steel bars in reinforced concrete.

Based on this experimental study, the following conclusions can be drawn:

1. Incorporating ZT in concrete mixtures with a water-to-cement ratio of 0.69, which were mixed with freshwater, reduces its compressive strength at 7, 28, and 90 days of curing. However, the reduction was lower at later ages due to pozzolanic activity.
2. Using seawater in the curing of concrete with and without ZT has little effect on its strength.
3. Using seawater as a mixing agent in plain concrete with a water-to-cement ratio of 0.69 reduced its strength significantly. However, incorporating ZT in concrete mixtures contradicts the negative effect of mixing with seawater.
4. Incorporating ZT in concrete has little effect on splitting tensile strength, but in some specimens, it has a positive effect.

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