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Effect of mixing water types on the timedependent zeta potential of Portland cement paste

Abstract: The measurement of zeta potential (ZP) has important applications in a wide range of industries including ceramics, pharmaceuticals, medicine, mineral processing, electronics, cement industry and water treatment. It provides new information on cement hydration, gel structure formation and the effects of chemical and mineral admixtures. In this study, ZP and pH measurements of Portland cement suspension prepared at wt% 1 cement/water ratio in the presence of four different types of waters, i.e., pure water, tap water, and salted (NaCl and CaCl₂) water, were carried out depending on the time. Also, vicat tests of Portland cement pastes prepared at a water-to-cement ratio of 0.34 were performed. After the vicat tests, X-ray diffraction analysis and scanning electron microscopy investigations were performed on the samples. The most important result obtained from this study is that setting time and time-dependent ZP values of Portland cement paste have different values depending on the used mixing water type. Among the water types, water with CaCl, gives the lowest ZP as absolute value, so this leads to minimum initial and final setting times for the cement paste prepared with this type of water.

Keywords: mixing water; pH; Portland cement; setting; zeta potential.

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1 Introduction

The zeta potential (ZP) of a substance, inorganic or organic, is used to explain the mechanism of dispersion and agglomeration (coagulation or flocculation) in a liquid phase and to identify the adsorption mechanisms of ions or molecules in a solid-liquid interface. Therefore, it plays an important role in a spectrum of applications including ceramics, food, mining, cement, paper, medicine, water and wastewater treatment, emulsions, biochemistry, and detergents [1-4]. Each mineral or solid particle in water carries electrokinetic charges constituting ZP depending on the properties of water such as pH and ionic strength [1, 4]. The change in ZP of any substance is due to either adsorption of ions present in the solution at its surface or dissociation of some functional surface groups (i.e., -SiOH, -AlOH, -COOH, -SO₂H, and -NH₂ if they are present at the particle surface) depending on the solution pH. These groups may be ionizable depending on the solution pH; a neutral surface would be charged either negatively at high pH values or positively at low pH values. Research studies in the literature showed that the stability of a cement paste increases whereas its viscosity decreases by increasing the absolute ZP values of cement particles; thus, the workability of concrete is influenced positively [5–10]. Of course, the hardened properties of concrete would be better, too [11]. The surface charge and, of course, the ZP of a cement or hydrated cement particle depend on the chemical nature of the phase and ions in the pore solution [12]. According to the literature, the potential determining ions for ordinary Portland cement (OPC) are Ca²⁺, Ba²⁺, SO₄²⁻, CO₃²⁻, F⁻, Ag⁺, and I⁻ [13].

When a cement is mixed with water, some hydration products are formed depending on the hydration periods [10, 14, 15]. In the initial hydration period (the first 15–20 min), calcium and hydroxyl ions are released into the solution and precipitation of calcium aluminate sulfate hydrates occurs [16]. Then, semi-crystalline calcium silicate hydrate gel (C-S-H) forms on the cement particle surface while the calcium and hydroxyl ion concentrations rise slowly [17, 18].

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The degree of hydration describes the process of hydration, and it is directly related to the fraction of the hydration products or porous structure in a hydration system in cement-based materials [19-22]. In contrast, the surface of C-S-H is mainly dominated by the >SiO⁻, >SiOCa⁺, and SiOH surface groups. In the C-S-H surface, the mean surface charge is the sum of >SiO⁻ and >SiOCa⁺. The magnitude and sign of the ZP of OPC depend on the above surface groups and their charges [12, 13, 23]. The second period is the induction period (minutes to several hours), and it is characterized by very slow reactivity. During this period, calcium silicate phases continue to dissolve and give OH ions to the medium. This leads to an increase in pH and it reaches to around 12.5. The third period is the main hydration period (hours to days). Dissolution of silicate-rich phases and precipitation of calcium silicate hydrates and calcium hydroxide lead to setting. This helps in the early strength development of the cement suspension. The last period is the continuous hydration period (days to years). This is the declaratory period in which hydration slowly continues in hardening the solid cement until the reaction is complete and strength development continues.

Positive ions such as Ca²⁺, Mg²⁺, and Na⁺ are released from the cement particle into the solution while keeping the SiO_{4}^{3} and AlO_{3}^{3} frame results in negative charge after the cement is mixed with water. Pore solution compositions of hydrated cement paste (HCP) are dominated by di- and monovalent ions such as Ca²⁺, Na⁺, K⁺, and OH⁻ [24, 25]. So, the type of mixing water determines the ZP of cement due to its chemical contents. This affects the dispersion and homogenization of cement paste and changes the physical and mechanical properties of hydrated cement [11, 26]. Some studies on ZP measurements of cementing materials are available. Zingg et al. [10] investigated the influence of polycarboxylate-based polyelectrolytes on the particle interaction among tricalcium silicate (C₂S, main clinker phase), calcium silicate hydrates (C-S-H), and calcium aluminate sulfate hydrates (ettringite). They concluded that the phases C₃S and C-S-H are positively charged in synthetic pore solution (liquid phase of hydrating cement suspension), whereas the ettringite is negatively charged. Elakneswaran et al. [24] studied the surface charge mechanism of HCP and its relations to adsorption of chloride ions. They indicated that HCP shows a net positive surface charge by dissociation and adsorption reactions. In HCP, chlorides bind as Friedel's salt (chemical binding) as well as adsorb on the surface of hydrates (physical binding). However, chloride ions neutralize the positively charged surfaces of portlandite and Friedel's salt by physical adsorption. The authors reported that C-S-H is the dominant phase in terms of chloride adsorption in HCP. The ZP of hardened, cementitious matrixes has been

determined and the optimum experimental parameters have been established by Castellote et al. [27]. In addition. the influence of the composition of the binder on the ZP of cement has been quantified and the results indicated that higher amounts of Al₂O₂ in the binder lead to higher positive values of ZP, while higher amounts of SiO₂ lead to negative ZP values. According to experimental results, carbonation increases the absolute value of the ZP toward more negative values; this behavior has been attributed to the influence of the specifically adsorbed Ca²⁺ on the negative sites of the surface in the non-carbonated samples. Pointeau et al. [28] performed ZP measurements on CEM I (OPC) and CEM V (blast furnace slag and fly ash added to OPC). The measurements showed that ZP increases from -17 to +20 mV for pH 13.3 to pH 12.65 (fresh HCP states) and decreases from 20 to -8 mV for pH 12.65 to 11 (degraded HCP states).

In this study, the effect of mixing water types on timedependent ZP of OPC was investigated and the obtained results were compared with its setting behavior.

2 Experimental program

2.1 Materials

The cement paste mixes were prepared with OPC (CEM I/42.5 R), produced according to the European Standards EN 197-1 [29] in Afyonkarahisar/Turkey by SET Cement Factory. The particle size and chemical properties of OPC are presented in Figure 1 and Table 1, respectively. The main components of OPC are 60.11% C₃S, 11.02% C₂S, 6.97% C₃A, and 9.95% C₄AF, respectively. The specific gravity and surface area (by Blaine) of OPC are 3.23 and 2994 cm²/g, respectively.

2.2 Mixtures and tests

To produce the cement suspensions and cement pastes, four different types of mixing water were used: pure water (distilled water), tap water, 5×10^3 M NaCl and 5×10^3 M CaCl₂ solution. NaCl and CaCl₂ (as CaCl₂·2H₂O; Carlo Erba Co.) were used to prepare the salted water. Specific conductivity values for pure water, tap water, NaCl, and CaCl₂ salted water were measured as 96.4, 1117, 1516, and 1664 µmhos/cm, respectively. The cement/water ratio was 0.1 wt.% for all the tests, except for the vicat test. On the cement suspension, time-dependent pH measurements from 0 to 480 min were performed using the Mettler Toledo (Europe) SevenEasy S20 pH meter with glass electrode

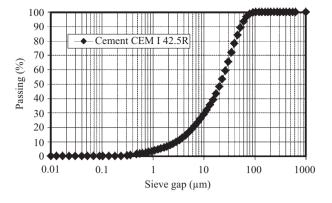


Figure 1 Particle size of the Portland cement used in the experiments.

Table 1 Chemical analyses of OPC (%).

SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	K ₂ 0	Na ₂ O	Free CaO
20.52	5.41	3.41	64.11	2.06	1.69	1.08	0.65	1.07

which has a sensitivity of ± 0.01 . With the same instrument, conductivity measurements of the used waters were carried out. In order to compare the relation between ZP and pH of cement, pH measurements were also carried out after 1440 min while mixing 100 ml of liquids on the magnetic stirrer. On the cement pastes, the initial and final setting times were determined by using an automatic vicat apparatus according to TS EN 196-3 [30] in order to establish the effect of mixing water types on setting time on cement pastes. Vicat experiments were carried out in laboratory condition at a temperature of 20±2°C and a relative humidity of 65%. The specimens were prepared at a water-to-cement ratio of 0.34. Totally, 90 insertions were performed within 10 h. The initial set is defined as the time at which the needle does not penetrate within a 5-mm distance the bottom of the mass, and the final set is defined as the time when there is no mark upon the surface from the needle. The frustum height was 40 mm.

To determine the time-dependent ZP of cement suspension on mixing water types and pH, electrophoretic mobility measurements were conducted using a zeta meter (Zetasizer ZS90, Malvern, UK) (Figure 2). This instrument determines the electrophoretic mobility of particles automatically by using Smoluchowski's equation and converts it to the ZP [31]. Separate cement suspensions were prepared for each measurement period (15, 30, 60, 90, 120, 150, 180, 240, 360, 480, and 1440 min). Each mixture was prepared by mixing 0.1 g of OPC and 100 ml of water. With four different types of water, totally 44 different mixtures were produced throughout the studies. Suspensions were mixed homogenously with a magnetic stirrer at 500 rpm. Also, they were dispersed by ultrasonic dispersion for 20 s before the ZP measurements. From the suspension, 0.75 ml was taken by an automatic pipette and then it was injected to the ZP measuring cell in the zeta meter instrument. All the ZP measurements were carried out at 23°C and, Merck quality NaOH and HNO, were used for pH adjustments.

In order to see the possible differences in the microstructure and crystallinity of the cement pastes prepared with different types of mixing waters, X-ray diffraction (XRD; Shimadzu XRD-6000, Japan) using Cu K α radiation (λ =1.5418 Å) and scanning electron microscopy (SEM; LEO VP-1431) using secondary electron image analysis were carried out. Before XRD analysis, the hydrated cement particles were ground to under 100 µm, and before SEM analysis, freshly fractured surfaces of mortar chips were coated with carbon in a vacuum evaporator.

3 Results and discussion

3.1 Setting time

The effect of mixing water types on the setting of cement was defined with a vicat apparatus, and needle

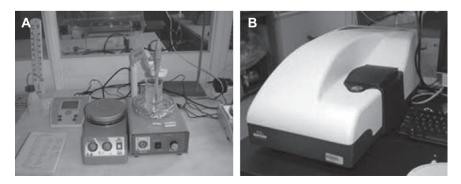


Figure 2 (A) pH measurements and (B) ZP measurements with Zetasizer ZS90.

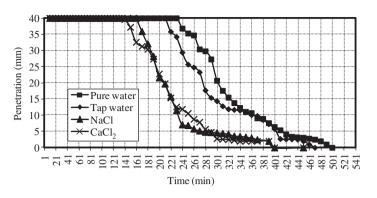


Figure 3 Needle penetration on the vicat apparatus for cement pastes.

penetration height in the vicat apparatus vs. time is presented in Figure 3. It shows that the penetration height of cement paste with all the mixing waters is about 40 mm during the first 2 h. Use of water with CaCl₂ considerably decreased the penetration height of 40 mm. After 240 min, the penetration height of vicat needle was 37, 30, 13, and 7 mm for cement pastes prepared from pure, tap, NaCl-, and CaCl₂-containing water, respectively. However, the needle penetration height was 4 mm for pure water, tap water, NaCl-, and CaCl₂-containing water after 440, 410, 340, and 301 min, respectively. The longest setting time was obtained in cement paste with pure water, while the shortest one was obtained in cement pastes with CaCl, solution. These may be explained by the effects of the flocculation process on the setting of cement. As seen in Figure 4, the cement particles in CaCl, salted water have the minimum ZP and this leads to an accelerated network formation between the cement particles so that the setting time of cement paste shortens.

The initial and final setting time of cement pastes are presented in Figure 5. Setting is due to early-stage calcium silicate hydrate (C-S-H) and ettringite formation. As seen in Figure 5, both the initial and final setting times of the pastes decrease when mixing water other than pure water was used. This is mainly due to the mono- and divalent

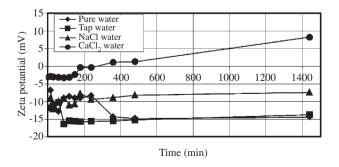


Figure 4 Time-dependent zeta potential variation of cement suspension prepared in different types of water.

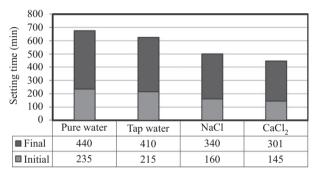


Figure 5 Setting time of the mixtures.

ions such Ca²⁺, Na^{+,} and Cl⁻ ions in the used mixing water. The initial setting time for cement pastes with pure, tap, NaCl-, and CaCl₂-containing water was found to be 235, 215, 160, and 145 min, respectively. For cement pastes with tap, NaCl-, and CaCl₂-containing water, the initial setting time diminished by approximately 20, 75, and 90 min, respectively. The reduction in final setting time was 30, 126, and 151 min for the same mixtures when they were compared to cement pastes with pure water. From the literature, it is known that NaCl and CaCl₂ are used as accelerators for the setting time of OPC [15].

3.2 Zeta potential and pH results

In the present research, the ZP and pH of cement suspension with different types of mixing water were measured to better understand their surface properties. The pH changes of the suspensions as a function of time are given in Figure 6. The cement suspension with tap water has the lowest pH values compared to the others. This may be attributed to the presence of bicarbonate (HCO_3) and carbonate (CO_3) ions in the tap water (Table 2). These ions have a significant effect on water chemistry [32], and they cause an increase in the solubility of CO_3 in water, which

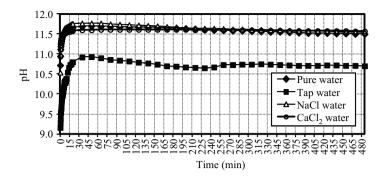


Figure 6 pH values of cement pastes vs. time.

Table 2 Chemical content of tap water (mg/l).

Cl	F	NO ₂	PO ₄	SO ₄	CO ₃	HCO ₃	К	Na	\mathbf{NH}_4	Ca	Mg
144	0.4	<0.1	<0.1	144	<10	439	9.6	93.6	<0.1	112	33.5

leads to an increase in the acidity of water. In contrast, pH values do not change so much after 15 min. Almost all of them were stable except for cement suspended with tap water. The cement suspensions with pure water, tap water, NaCl-, and CaCl₂-containing water had pH values of 11.69, 10.92, 11.77, and 11.60 at 30 min, and they had pH values of 11.5, 10.7, 11.54, and 11.57, respectively, after 8 h. While the pH value of each type of water was low (pH \approx 7–8) at the beginning of the measurements, they increased suddenly after the addition of cement into water, and the medium became basic (pH \approx 10–11) in a few minutes. This phenomenon is due to hydroxide ions that come from the dissolution of C₃S phase in water [7].

The measured value of the ZP of cement has contribution from all hydrated and un-hydrated phases of cementitious materials such as C-S-H, portlandite, and unhydrated cement. The ZP of any cement phase, either hydrated or unhydrated, depends on the concentration of the potential determining or specifically adsorbing ions present in the pore solution. Since the amount of those ions may vary with cement composition, water-to-cement ratio, time of hydration, etc., change in ZP values for different hydration phases and minerals can be expected [31]. The ZP variation of cement particles in different types of waters is given in Figure 4 as a function of time. It is clearly seen that ZP values are negative at the beginning of the hydration process. After 15 min, the cement paste with pure water has a ZP value of -6.75 mV, while the pastes with tap water, NaCl, and CaCl, have ZP values of -11.8, -9, and -2.92 mV, respectively. However, during the second period of cement hydration (i.e., at the 180th min), their ZP values change to -8.99, -15.7, -7.68, and -0.39 mV,

respectively, for the same mixtures. After 24 h, the cement with pure water, tap water, and NaCl-containing water still had negative ZP values, while it was a positive ZP for CaCl₂ (8.18 mV). In general, the ZP of cement particles in CaCl₂ salted water had the lowest absolute value compared to the others; therefore the initial and final setting times of cement pastes prepared with CaCl₂ salted water showed minimum values. According to the literature [33–36], the ZP of OPC is generally measured at approximately -15 to -20 mV.

Figure 7 shows the ZP change of cement particles present in various types of water vs. pH. It is clearly seen that, except for CaCl₂ salted water, in other types of waters the cement particles show negative ZP values for the whole studied pH range (pH 10.30–11.80). This can be attributed to the adsorption of various anions such as OH⁻, SO₄², and Cl⁻ onto cement particles which are released from cement into water during the dissolving process when the cement comes into contact with water [37]. Also, the adsorption capacity of sulfate ions is great, especially at high pH values. However, Ca²⁺ and Mg²⁺ions make a positive charge to the OPC particle surface [34–37].

It is clear that cement pastes with CaCl₂ solution have positive ZP values. It is thought that Ca²⁺ ions present in CaCl₂ solution are easily and strongly adsorbed onto

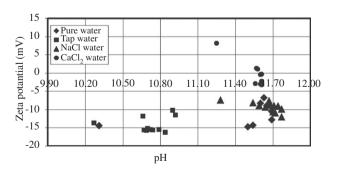


Figure 7 Zeta potential of cement particles in various types of water vs. pH.

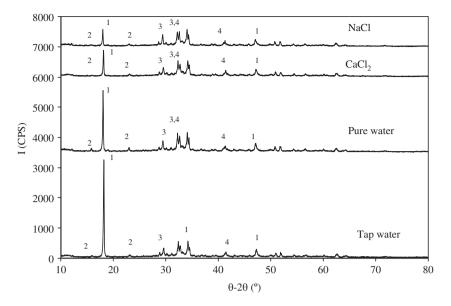


Figure 8 XRD spectrums of cement pastes prepared with different type of waters (1: portlandite; 2: ettringite; 3: alite; 4: belite).

negative charge centers on cement particle surfaces and are able to reverse the sign of the ZP value of the cement from negative to positive. In contrast, cement pastes with NaCl solution had negative ZP values, although the solution includes Na⁺ which gives a positive charge to ZP. As mentioned earlier, chloride ion plays an important role in the ZP of the particles of cement paste, and it gives a negative charge to the surface. However, the ZP values of cement pastes with NaCl and CaCl₂ are closer to the positive region than those of cement pastes with tap water and pure water. In contrast, the highest negative ZP values were obtained from cement pastes with tap water. The main reason for this phenomenon can be explained by the XRD analysis result shown in Figure 8. The main hydration product of cementitious materials is calcium silicate hydrates (C-S-H) and portlandite $[Ca(OH)_2]$. The portlandite-dominant products that give a negative charge to the surface of cement particles and the highest portlandite content were observed in mixtures having tap water. So, the ZP values of cement pastes with different types of mixing water also became close to zero or positive depending on portlandite content.

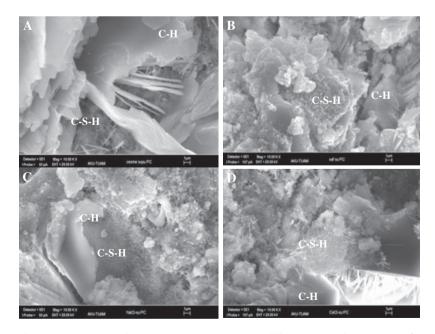


Figure 9 SEM image of cement pastes prepared with different types of mixing water (A: tap water; B: pure water; C: NaCl; D: CaCl₂) (×1000).

3.3 Microstructure

In Figure 9, the microstructure of cement pastes containing the different types of mixing water is presented. As clearly seen from the figure, all cement pastes are covered with portlandite (C-H) as supported by XRD spectrums (Figure 8). However, cement pastes prepared with pure and tap water have more C-H content than those prepared with CaCl₂- and NaCl-containing water. In contrast, C-S-H products can be seen in all the early-stage HCPs. Regarding the ZP of cement, C-S-H products are very important because calcium is a potential-determining ion for the C-S-H and other hydrated products [38]. In addition, several ettringite needles are seen growing from the inward. This indicates a renewal of the reaction of aluminate phases.

4 Conclusion

In the present investigation, the ZP of cement paste suspensions that contain different types of mixing water was measured by electrophoresis technique. Based on the results obtained in this research, the following conclusions can be drawn.

 In cement suspensions containing different types of mixing water, the particles usually show a negative ZP in water. However, cement pastes with CaCl₂ solution show a positive ZP due to calcium ions. Because calcium ions adsorb strongly onto the surface of cement particles, they change the ZP of cement from a negative to a positive value. For the OPC particles, the lowest ZP values were also obtained with CaCl₂ solution (as its absolute value) among all types of water.

- The initial and final setting times of the cement pastes are in the order of CaCl₂<NaCl<tap water<pure water. This indicates that the type of water used in mixing cement plays a critical role in the setting behavior of OPC. However, the most significant effect is seen on the setting of cement pastes prepared with salted water containing CaCl₂ as supported by the time-dependent ZP results.
- Even though the order given for the initial and final setting times was CaCl₂ water<NaCl water<tap water<pure water, the order of the ZP of cement pastes can be arranged as CaCl₂ water<NaCl water<pure water<tap water according to their negative values. This means that a relation between ZP values and the setting time of cement paste can be established.

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