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A Study on the Effect of Ceramic Polishing Powder on the Nucleation and Growth of Hydrates in Cement Paste

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Abstract: The production of cement leads to a large amount of CO₂ emission. Using industrial waste slag, such as ceramic polishing powder (PP), to replace part of Portland cement can reduce the pollution caused by the cement industry and solid waste disposal. In order to use PP as a replacement for cement, its effects on the properties of cement paste need to be clarified. In this study, the effect of PP on the nucleation and growth of hydrates in cement paste at very early ages was investigated. Quartz was used as a reference. The interactions of their surface with various ions in cement paste solution, which has an important effect on the nucleation and growth of hydrates, were studied by using the zeta potential test. The morphology of the nucleus and crystal of hydrates was investigated by using SEM. The zeta potential measurements showed that the affinity of the surface of PP and quartz to ions in the pore solution of cement paste is similar. The image of SEM indicated that there is also not much difference in the crystallization of hydrates on the surfaces of PP and quartz. These evidences suggested that PP has similar surface charge properties as quartz, and its effects on the properties of cement paste are the same as that of quartz. From the viewpoint of the effect on very early hydration, PP can be used in cement paste, similar to quartz power.

Keywords: quartz; ceramic polishing powder; nucleation; zeta potential; C-S-H; crystal; cement paste; ions interaction

1. Introduction

The mass production of cement causes the depletion of mineral resources and serious environmental pollution. According to statistics, the production of one ton of Portland cement consumes 4 to 5 GJ of energy and releases a large amount of CO₂ [1]. Meanwhile, the ceramic industry produces a large amount of solid waste (ceramic polishing powder) during the process of grinding and polishing ceramic products. Millions of tons of ceramic polishing powder are produced each year in southern China [2,3]. Such a large amount of ceramic polishing waste powder is mainly treated by landfill, causing serious pollution of the land and water. Ceramic is a product of calcination of clay at 1200 °C. The major constituents of polishing powder are SiO₂ and Al₂O₃ with some minor constituents

such as CaO, MgO, Na₂O, and K₂O. Considering its particle size, components, and surface chemical property, it is quite possible to use waste ceramic polishing powder as a supplementary cementitious material in cement production. Using the ceramic polishing powder (PP) to replace part of the Portland cement in concrete can reduce the pollution caused by the cement industry and solid waste disposal.

In order to use PP as a replacement for cement in concrete, its effect on the properties of cement paste need to be clarified, for example, nucleation or growth-inducing factor of hydrates on their surface in early-age concrete is important because it can affect the properties of late-age concrete. Many relevant studies [4–10] have been conducted. Tydlitát et al. [7] reported that using more than 20% ceramics in blended cement does not affect the compressive strength. In a study performed by Fatima et al. [9], concrete was made by replacing up to 30% of the cement with PP resulting in concrete that had good workability, proper compressive strength, split-tensile strength, flexural strength, and elastic modulus. A study by Cheng et al. [4] showed that the permeability resistance of concrete mixed with PP is superior to that of a control group, and the most desirable permeability resistance was achieved when the cement substitution rate was 30%. These studies mainly focused on the effects on macroscopic properties and microstructure of mortar or concrete. The effect of PP on the interactions of their surface with various ions in cement paste solution and the effect on the nucleation and growth of hydrates on their surface are rarely reported. It is crucial to understand the mechanisms of the effect of PP on the properties of a blended cement paste system.

Previous studies [11–17] have shown that the crystallization and growth of hydrates on the filler particle are mainly attributed to the surface properties of filler particles and the interaction with ions in cement paste solution. The affinity of filler particles for cement hydrates has a significant influence on the development of strength. The nucleation and growth of C-S-H on quartz are caused by absorbing the Ca²⁺ ion in the pore solution of cement paste on its surface through the electrostatic attraction forces. The surfaces of PP and quartz particles are rich in silicon phase and the aluminum phase in PP does not react in a low alkaline environment, therefore, it is assumed that PP has similar surface chemical properties as quartz which is commonly accepted as an additive in mortar and concrete. Therefore, it is also assumed that the interactions of PP's surface with various ions in cement paste solution, and the nucleation and growth of hydrates on their surface are also similar as quartz.

With this hypothesis, quartz was chosen as a reference. X-ray diffraction (XRD) was used to analyze the composition of quartz and ceramic polishing powder. The zeta potential of these particles in the model solution simulating pore solution of cement paste was studied in order to clarify the interactions between their surface and the ions. These interactions lead to the nucleation and growth of the hydrates on their surfaces. Parallel to the zeta potential test, the nucleation and growth of the hydrates on the surfaces of the two particles were micro-observed by scanning electron microscope (SEM). The influence of the chemical properties of PP on the very early hydration of blended cement was discussed based on these zeta potential tests and microscopic observations.

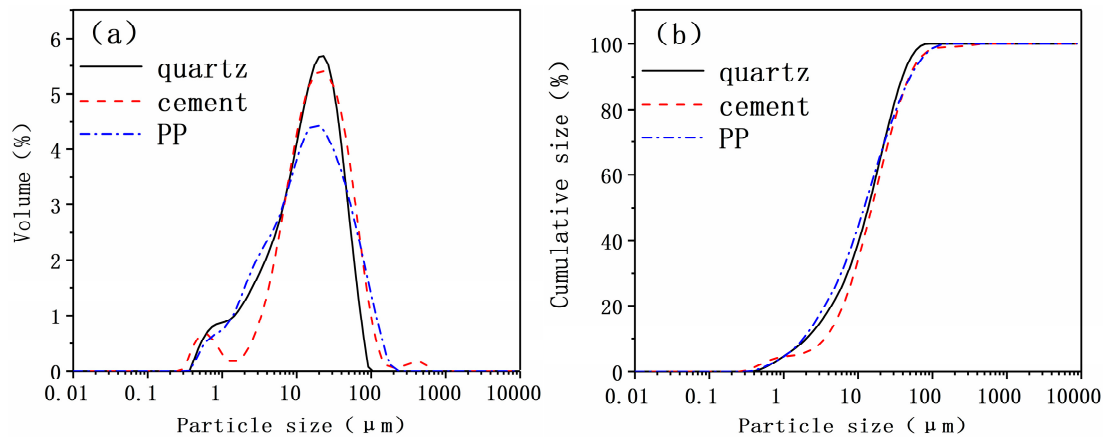
2. Materials and Methods

2.1. Materials

The cement used in the experiment was OPC CEM i 42.5, produced by Shijing Co. Ltd, Guangzhou, China. The quartz powder was bought from Tuoyi New Materials Co. Ltd. Guangzhou, China. The ceramic polishing powder (PP) was taken from Yujing ceramic factory in Foshan, China. The chemical composition of these materials is shown in Table 1. The specific surface area of cement, PP, and quartz powder is 0.86, 1.0, 0.96 m²/g, respectively. The particle size distribution (PSD) of these particles is shown in Figure 1.

Table 1. Chemical composition (%) of cement, quartz, and polishing powder.

Components	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	TiO ₂	K ₂ O	Na ₂ O	Loss
Cement	21.86	4.45	2.35	63.51	1.67	-	0.55	0.26	1.89
Quartz	99.5	0.2	0.03	0.02	-	-	-	-	-
PP	73.41	17.46	0.67	0.22	0.68	0.10	1.88	1.86	3.42

**Figure 1.** Volume distribution of particle size (a) and cumulative distribution of particle size (b) of quartz, cement, and ceramic polishing powder (PP).

2.2. X-ray Diffraction Analysis (XRD)

X-ray diffraction (XRD) was used to analyze the composition of quartz and ceramic polishing powder. XRD patterns were recorded at a scanning rate of 2° /min from 2 θ = 5° to 2 θ = 80° with Cu K α radiation (λ = 1.5418 Å) using a Bruker D max/RB diffractometer (Billerica, MA, USA).

2.3. Zeta Potential Test

A Malvern Zetasizer Nano (Malvern instruments Ltd.UK) was used to measure the zeta potential of the PP and quartz particles in model solution. The zeta potential is related to the surface charge of the particles which reflects the interactions between particle's surface and the ions in the solution. Three tests were conducted for each sample, with the average value being taken as the final result. Although this technique is appropriate, it has its limitations. When the ion concentration is too high, the electrical conductivity of the electrolyte increases and experimental error takes place. Therefore, it is necessary to control the concentration of ions in the solution.

The zeta potential test used five series of model solutions simulating the pore solution of cement paste, in order to evaluate the effect of different concentrations of Ca²⁺, Na⁺, K⁺, and SO₄²⁻, which are the main ions in pore solution of cement paste. The detailed chemical composition of the five model solutions is shown in Table 2. Solution 1 was used to test the particle surface potential at different pH values which ranged from 8 to 12. Solution 2 was used to test the effect of different calcium ion concentrations on particle surface potential. Solution 3 and solution 4 were prepared by mixing Ca(OH)₂ with NaOH and KOH, respectively. Solution 5 was the mixed solution of K₂SO₄ and Ca(OH)₂.

Table 2. Preparation of the model solutions (concentrations are shown in mmol/L units).

Solution 1		Solution 2		Solution 3		Solution 4		Solution 5	
NaOH	pH	Ca(OH) ₂	pH	Ca(OH) ₂	NaOH	Ca(OH) ₂	KOH	Ca(OH) ₂	K ₂ SO ₄
-	8	0.2	9.2	0.1	10	0.1	10	0.1	10
-	9	0.4	9.3	0.4	10	0.4	10	0.4	10
-	10	0.6	9.6	1	10	1	10	1	10
-	11	0.8	10.1	4	10	4	10	4	10
-	12	1	10.5	8	10	8	10	8	10
-	-	2	10.8	0.1	50	0.1	50	10	10
-	-	4	10.9	0.4	50	0.4	50	15	10
-	-	6	11	1	50	1	50	19.6	10
-	-	8	11.9	4	50	4	50	0.1	50
-	-	10	12	8	50	8	50	0.4	50
-	-	15	12.2	-	-	-	-	1	50
-	-	20	12.3	-	-	-	-	4	50
-	-	-	-	-	-	-	-	8	50
-	-	-	-	-	-	-	-	10	50
-	-	-	-	-	-	-	-	15	50
-	-	-	-	-	-	-	-	18	50

2.4. SEM Analysis

A Phenom ProX electron microscope was used to observe the hydration products on the surface of PP and quartz particles. Two groups of samples were prepared. One was PP blended cement paste and the other was quartz blended cement paste. In these samples, the mixing ratio of fillers to cement was 4:6, and the ratio of w/b was 0.4. At each required time (1, 4, and 7 h), about 1 g of the paste was taken to terminate hydration by solvent exchange with anhydrous alcohol. Then, the paste was filtered with filter paper and dried in a vacuum drying oven. The dried powder was then collected and stored under vacuum in a desiccator until used. The SEM observations were performed on the dried powder coated by gold.

3. Results

3.1. Morphology and X-ray Diffraction (XRD) Analysis

X-ray diffraction (XRD) data are shown in Figure 2. It shows that PP and quartz have similar XRD patterns. The crystalline phase of PP and quartz is mainly SiO₂ (quartz). The other compounds in the mineralogy of PP, such as mullite, are found in minor quantities. The SEM was used to observe the morphology of PP and quartz particles, as shown in Figure 3. The SEM figures show that there is not much difference from the viewpoint of the morphology.

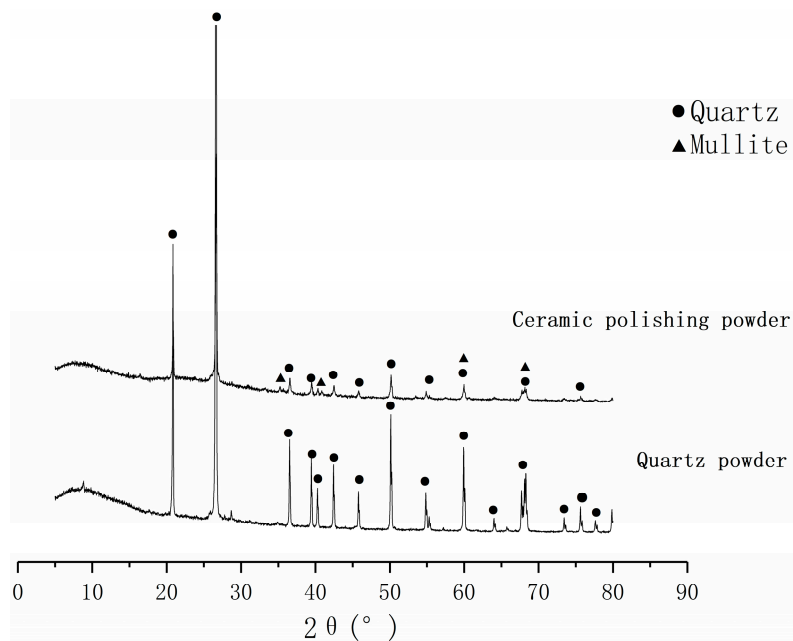


Figure 2. X-ray diffraction (XRD) analysis of ceramic polishing powder and quartz powder.

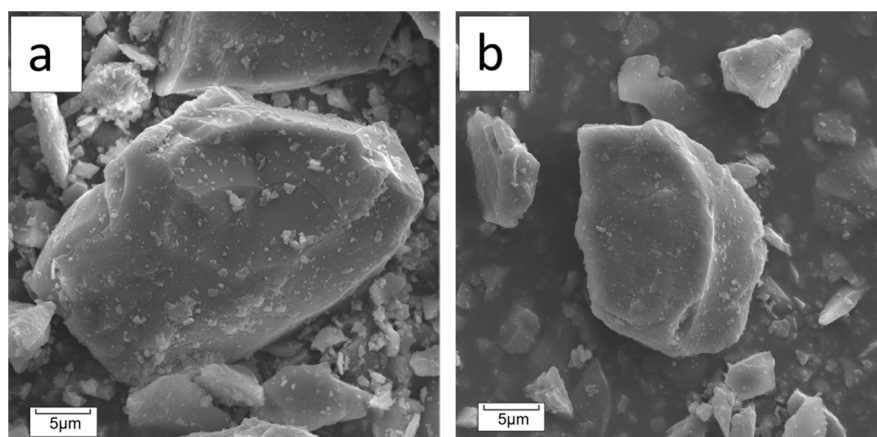


Figure 3. Morphology of quartz (a) and PP (b).

3.2. Ion Interaction

When the cement contacts with water, it releases ions rapidly and the concentrations of Na^+ , K^+ , Ca^{2+} , and SO_4^{2-} in the solution also increase rapidly. The liquid phase of cement paste at early stage mainly consists of Na^+ , K^+ , SO_4^{2-} , and Ca^{2+} [13]. The interactions of these ions with the surfaces of PP and quartz particles lead to the nucleation and growth of hydrates on their surfaces. Therefore, in order to study these interactions, five sets of model solutions, as shown in Table 2, were prepared. The zeta potential test was carried out to measure the interactions between particles' surface and the ions in the solution. The effect of pH and different concentrations of Ca^{2+} , Na^+ , K^+ , and SO_4^{2-} on the interactions is evaluated as follows:

3.2.1. Effect of pH

The zeta potential of PP and quartz particles tested in the solutions with different pH values (solution 1, as shown in Table 2) is presented in Figure 4. As can be seen from Figure 4, the zeta potential of the two particles is between -20.3 mV and -46.73 mV with the pH range from eight to 12. When the pH is in the range of eight to 10, the zeta potential of the two particles is approximately the same and remains relatively stable. After the pH equals 10, the potential value of the two particles decreases

rapidly due to the dissociation of the silanol groups at the interface. The dissociation equilibrium of silanol sites is due to the pH increase. The reaction is taken through the following equation [18–21]:



Therefore, the increase in the pH value in the solution promotes the ionization of silanol groups of the surface on PP and quartz particles, resulting in higher absolute values of the zeta potential, however, at pH equals 12, the zeta potential of the particles increases slightly, which is caused by the excessive conductivity of the solution due to the increase of ions in the solution. Overall, the zeta potential of PP and quartz is similar in the range of the whole concentration.

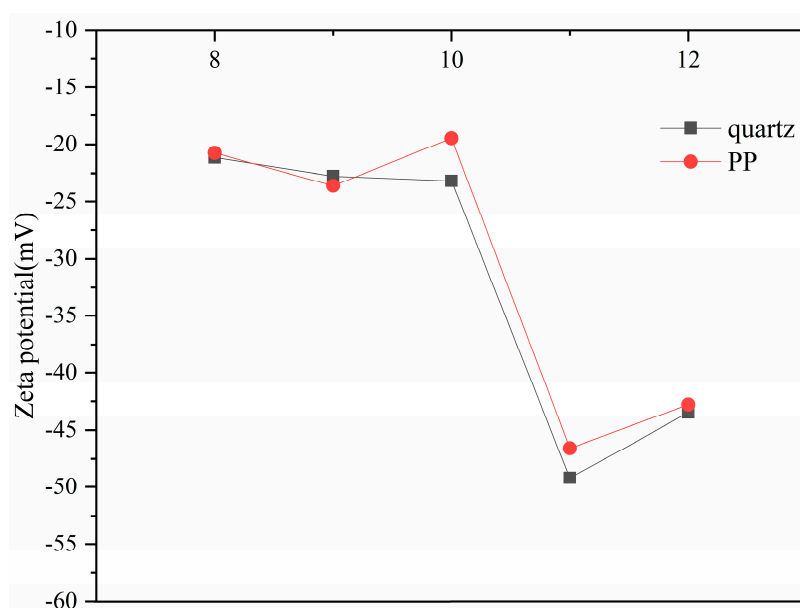


Figure 4. Evolution of the zeta potential of PP and quartz particles as a function of pH in NaOH solution.

3.2.2. Effect of Ca^{2+} Concentration

The zeta potential of PP and quartz particles in $\text{Ca}(\text{OH})_2$ solutions (solution 2, as shown in Table 2) is shown in Figure 5. It shows that PP and quartz particles have a similar iso-electric point, which is recorded at a Ca^{2+} ion concentration of about 2 mmol/L. This is because the surfaces of PP and quartz are rich in silicate phase. When the concentration of Ca^{2+} ion is low, more SiO^- groups are ionized on the particle surface, and the calcium ions adsorbed to the surface by static electricity are not enough to offset the negative charge on their surfaces, so the surface potential of all particles is negative before the concentration of Ca^{2+} is 2 mmol/L. When the concentration of Ca^{2+} is equal to or slightly less than 2 mmol/L, the potential on the surface of all particles is reversed due to the adsorption of enough Ca^{2+} . In the range of the tested concentrations, the zeta potential of PP and quartz particles is almost the same considering the reasonable fluctuation of measurements.

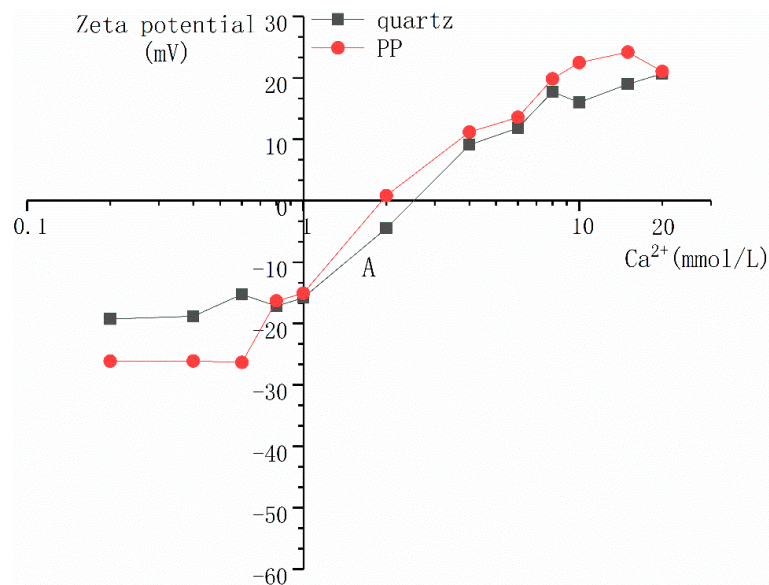


Figure 5. Evolution of the zeta potential of PP and quartz particles as a function of Ca^{2+} concentration in $\text{Ca}(\text{OH})_2$ solution.

3.2.3. Effect of Na^+ and K^+ Concentration

In addition to Ca^{2+} ions, Na^+ and K^+ ions are also important ions in pore solution of cement paste. In order to investigate the interactions of Na^+ and K^+ ions with the surfaces of PP and quartz particles, the zeta potential of the PP and quartz particles in $\text{Ca}(\text{OH})_2 + \text{NaOH}$ solutions, and $\text{Ca}(\text{OH})_2 + \text{KOH}$ solutions (solution 3 and 4, as shown in Table 2) were measured. The results are depicted in Figures 6 and 7 and show that the zeta potential of PP and quartz particles with the change of Ca^{2+} ion concentration in the same concentration of NaOH and KOH solution is almost the same. Meanwhile, in the same solution, the zeta potential values of PP and quartz particle is very close, and the iso-electric point (IEP) appears when the Ca^{2+} ion concentration is about 4 mmol/L, which is higher than that in the $\text{Ca}(\text{OH})_2$ solution without NaOH and KOH, because a higher Ca^{2+} ion concentration is needed to offset the more negative charge on their surfaces caused by higher pH with the addition of NaOH or KOH.

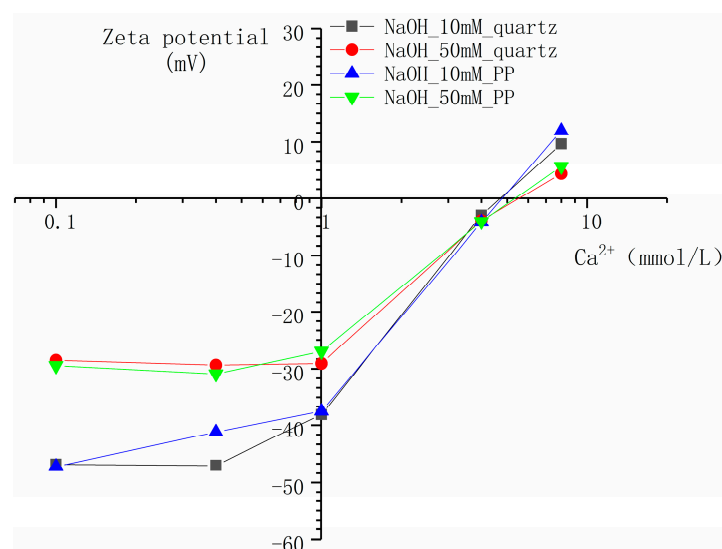


Figure 6. Evolution of the zeta potential of PP and quartz particles as a function of Ca^{2+} concentration in 10 mmol/L and 50 mmol/L NaOH solutions.

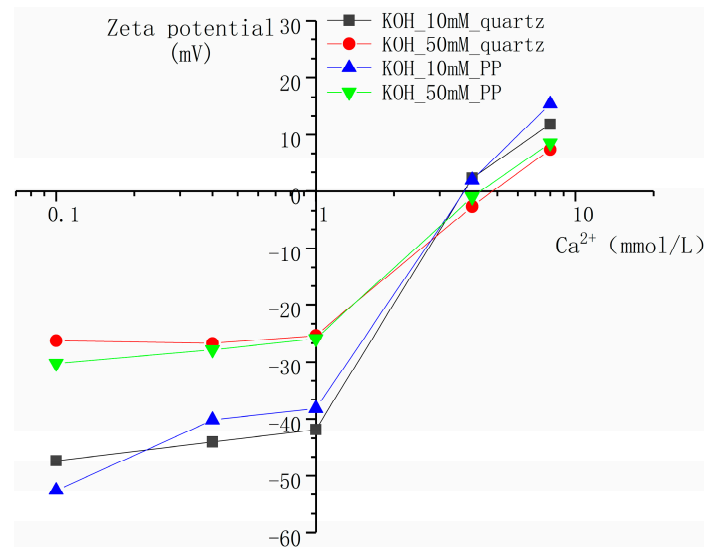


Figure 7. Evolution of the zeta potential of PP and quartz particles as a function of Ca^{2+} concentration in 10 mmol/L and 50 mmol/L KOH solutions.

3.2.4. Effect of SO_4^{2-} Concentration

As previously mentioned, SO_4^{2-} ion is also an important ion in the pore solution of cement paste. Therefore, the effect of SO_4^{2-} concentration is considered in this study. The evolution of the zeta potential of PP and quartz particles as a function of Ca^{2+} concentration in K_2SO_4 solution (solution 5, as shown in Table 2) is presented in Figure 8. It can be seen that the performance of PP and quartz particles is similar in the solution of K_2SO_4 with regard to the reasonable fluctuation of measurements. The zeta potential of PP and quartz particles in sulfate solution increases with an increase in the concentration of Ca^{2+} ion. With an increase of sulfate concentration, the iso-electric point (IEP) for PP and quartz particles moves toward higher Ca^{2+} concentration. At 10 mmol/L K_2SO_4 solution, the IEP for PP and quartz particles also appears at a Ca^{2+} concentration of about 10 mmol/L. This indicates that the adsorption of SO_4^{2-} and Ca^{2+} ions onto the surfaces of PP and quartz is similar. Therefore, at the 50 mmol/L K_2SO_4 solution, no IEP appears, because the highest Ca^{2+} concentration in the tested solution is 18 mmol/L, which is much lower than the concentration of K_2SO_4 .

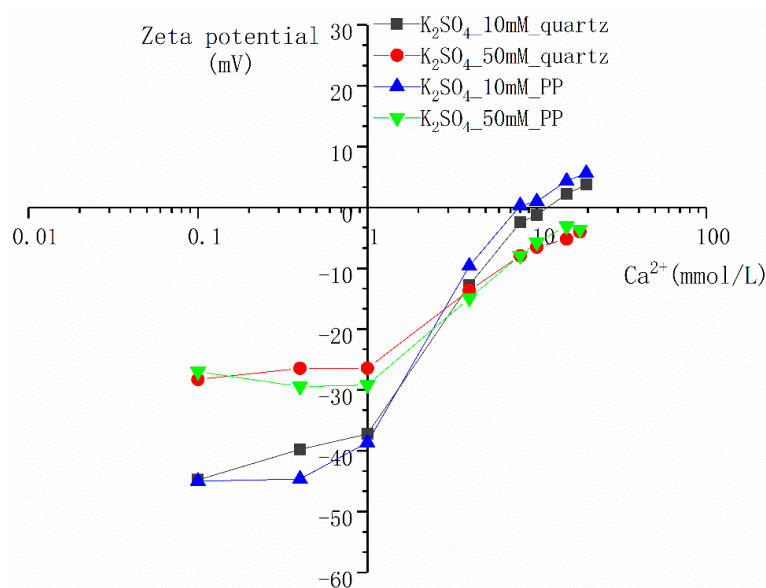


Figure 8. Evolution of the zeta potential of PP and quartz particles as a function of Ca^{2+} concentration in 10 mmol/L and 50 mmol/L K_2SO_4 solutions.

3.3. Analysis Morphology of Hydration Products on the Surface of PP and Quartz

The nucleation and growth states of the hydration products on the surface of PP and quartz particles at different time points are shown in Figure 9. Figure 9a,a₁,d,d₁ shows the surface hydrates on the surface PP and quartz particle after 1 h hydration, respectively. The surface of both particles is distributed with small and very sparse acicular hydration products, which is likely to be C-S-H formed at the early stage of hydration. Meanwhile, a small number of granular nuclei was observed, which were likely clusters of CH nuclei. The growth of the nuclei is not much different on the two types of particles at 1 h of hydration. Figure 9b,b₁,e,e₁ shows the surface hydrates on the surface PP and quartz particle at 4 h of hydration, respectively. As can be seen, both acicular and granular hydration products were grown, but the distribution on the surface of the particles was still sparse and there was no specific growth direction. Bar-shaped ettringite appeared on the surface of PP and quartz particles, as shown in Figure 9b,e. The hydration products seem to be different in Figure 9b₁,e₁. This is attributed to the fact that the granular products of Figure 9b₁ are identifiable substances attached to the surface of the particles, rather than the hydration products growing on the surface of the particles. Figure 9c,c₁,f,f₁ shows the surface hydrates on the surface PP and quartz particle at 7 h of hydration, respectively. As can be observed, an amount of slender needle-like hydration products was distributed on the surface of PP and quartz particles. Acicular hydration products were grown a lot after hydration for 7 h. The distribution of hydration products was still uneven and there was no uniform direction of growth, as shown in Figure 9c₁,f₁. Comparing the morphology of the two particles at different time, it is found that the nucleation and growth of hydrates on PP and quartz particles are very similar. Similar crystallization characteristics of quartz have been reported in relevant literature [13,22].

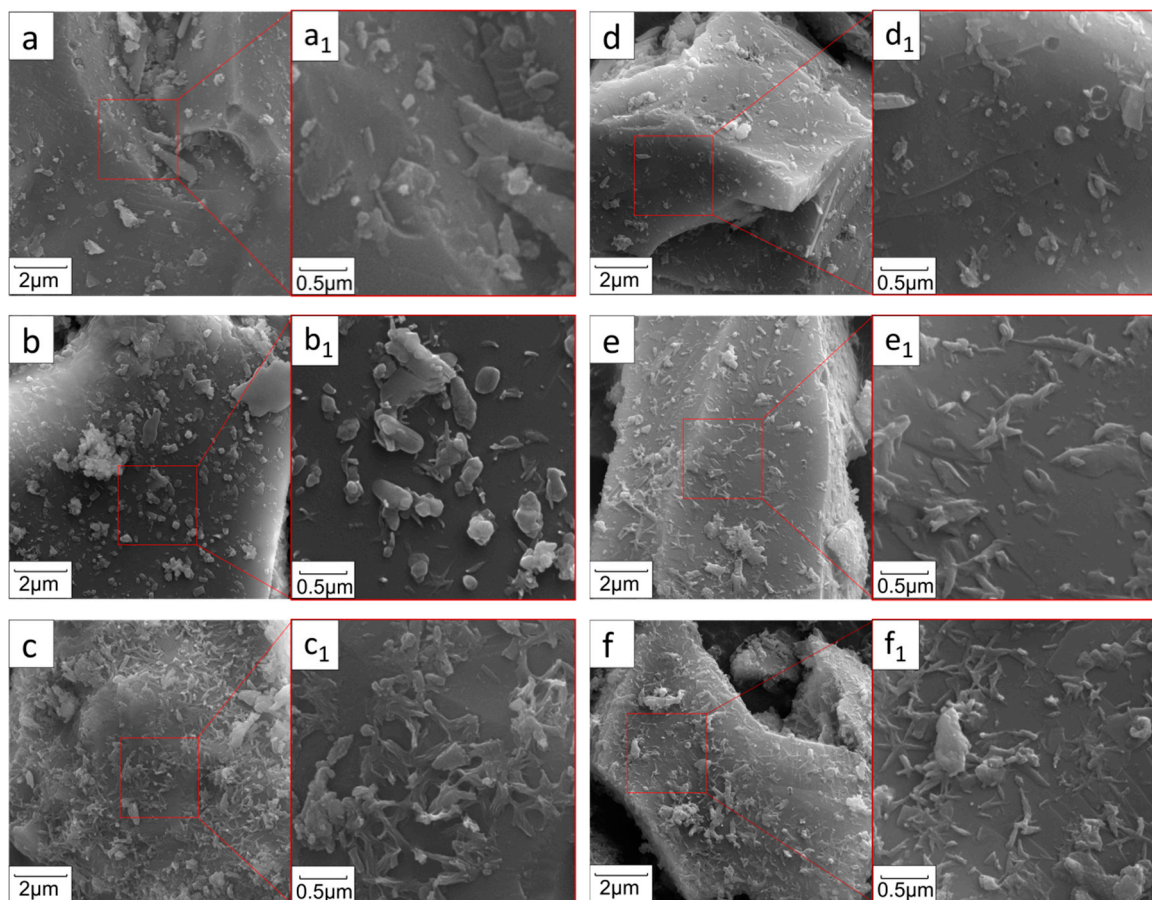


Figure 9. Morphology of hydration products on surface of PP particle at 1 h (a,a₁), 4 h (b,b₁), 7 h (c,c₁) and quartz particle at 1 h (d,d₁), 4 h (e,e₁), 7 h (f,f₁) (a–f are 25,000×, and a₁–f₁ is 90,000×).

3.4. Discussion

The nucleation and growth of hydration products on the surface of PP and quartz particles depends on its surface properties and the interactions between related ions and its surfaces. Previous research [13] has reported that cement has similar surface chemical properties to quartz, because their surfaces are rich in silicate phase, and therefore they show similar surface charge characteristics. The evolution of the zeta potential of PP seems to be a little more positive with rising ion concentrations. This may be attributed to the similarity of the surface composition of PP and quartz, after all, the surface compositions of both PP and quartz are mainly SiO_2 . The PP surface is also rich in silicate phase, and therefore it is assumed that it has similar surface charge characteristics as quartz particles and has similar C-S-H nucleation and growth mode. In Figures 4–8, it can be observed that the zeta potential of PP varies in a similar way as that of quartz in solutions of NaOH, $\text{Ca}(\text{OH})_2$, $\text{Ca}(\text{OH})_2 + \text{NaOH}$, $\text{Ca}(\text{OH})_2 + \text{KOH}$, and $\text{Ca}(\text{OH})_2 + \text{K}_2\text{SO}_4$. This preliminarily confirms that the surface chemical properties of PP and quartz powder have high similarity and the surfaces of both PP and quartz particle are rich in silicate phase.

The aluminum phase in PP seems to be a factor influencing the experimental results, but the zeta potential test shows the surface potential of PP is similar to quartz in the same solution. The aluminum atoms are less exposed to the surface of the compound and very difficult to dissolve in a relatively low-alkaline environment. Even a small amount of aluminum ions in the solution would not affect the experimental results. This is consistent with the results observed by SEM. Therefore, the aluminum phase in PP does not affect the formation of its early hydration products.

Ca^{2+} ion plays a main role in regulating the crystallization of C-S-H on the particle surface. Ca^{2+} ion is an important factor that determines the dynamics, morphology, and structural characteristics of C-S-H and also controls the nucleation characteristics in the heterogeneous C-S-H nucleation process [23]. Zeta potential confirmed that the interaction of Ca^{2+} ion with quartz and PP surfaces is similar. It is a relatively weak electrostatic interaction, which is not conducive to forming “stable nuclei” which is proven by the microscopic observation in morphology and distribution of hydration products, as shown in Figure 9. Therefore, in this study, it is considered that PP and quartz have similar surface properties and have the same effect on the nucleation and growth of hydrates in cement paste at a very early-age of hydration. Therefore, from the viewpoint of the effect on very early hydration, PP can be used in cement paste similar to quartz powder.

4. Conclusions

In this study, the effect of PP on the nucleation and growth of hydrates in cement paste at very early ages was investigated. Quartz was used as a reference. The interactions of their surface with various ions in cement paste solution, which has an important effect on the nucleation and growth of hydrates, were studied by using the zeta potential test. The morphology of the nucleus and crystal of hydrates was investigated by using SEM. The influence of the chemical properties of PP on the ion interaction and the very early hydration of blended cement were discussed. On the basis of the experimental results, the following conclusions can be drawn:

1. The zeta potential measurements show that the affinity of the surface of ceramic polishing powder and quartz to ions in the pore solution of cement paste is similar;
2. The SEM observation shows that there is also not much difference in the crystallization of hydrates on the surfaces of ceramic polishing powder and quartz;
3. Because both PP and quartz particle are rich in silicate phase, the adsorption of calcium ions onto their surfaces is due to the weak electrostatic interaction, resulting in uneven and unstable nuclei on their surface.

These evidences show that ceramic polishing powder has surface chemical properties similar to quartz. From the viewpoint of the effect on very early hydration, ceramic polishing powder can be used in cement paste similar to quartz powder.

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Conflicts of Interest: The authors declare no conflict of interest.

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