

Experimental study on the effect of ions on the surface hydration characteristics of fine quartz

Chunfu Liu^{1,2}, Fanfei Min², Lingyun Liu², Jun Chen², Bao Ren², Kai Lv², Yujiao Tan²

¹ State Key Laboratory of Mining Response and Disaster Prevention and Control in Deep Coal Mine

² Anhui University of Science and Technology

Corresponding author: ffmin@aust.edu.cn (Fanfei Min), chfuliu@aust.edu.cn (Chunfu Liu)

Abstract: The fine quartz particle hydration and effects of metal ions on the hydration characteristics of fine quartz surface are investigated using the rheological experiment. Several important factors affecting hydration factors, such as particle sphericity, solution pH, ion species, ion concentration were investigated. The results show that viscosity and hydration factor of fine quartz suspension increase with the increase of solution pH. wherein quartz particles have more negative charges on the surface in alkaline environment and strong hydration repulsion; The introduction of metal ions enhances the hydration strength of fine quartz surface to a certain extent. In contrast, high valence and high concentration will increase the viscosity of fine quartz suspension, and the hydration factors of particle surface also increase. At the same ion concentration, the order of influence on the hydration factors of fine quartz particles is $Mg^{2+} > Ca^{2+} > Na^{+} > K^{+}$. This finding has been attributed to the combination of metal ion hydration and its adsorption on the mineral surface. This study will provide the theoretical guiding significance for the refractory coal slime water and other mineral processing wastewater containing quartz particles.

Keywords: quartz, hydration cation, particle sphericity, hydration factor, viscosity

1. Introduction

Quartz is one of the most common minerals on earth, often associated with many high-value minerals. Quartz surface hydration plays a crucial role in many industrial processes, such as sewage sludge treatment (Świerczek et al., 2018), sands deposition (Huang et al. 2019), petroleum (Pei et al. 2018; Moslemizadeh et al. 2018), mineral processing and wastewater treatment (Li et al. 2020; Min, Peng, and Liu 2015). For example, regarding slime water treatment, when hydrated particles approach, repulsive force prevents them from getting closer owing to the overlap of hydration layers. Combined with electrostatic and Van der Waals interactions between particles, a stable colloidal system is formed, which leads to difficulties in sedimentation and clarification of coal slime water. For this reason, it is recognized that mineral surface hydration characteristics are the key aspect of solving the issue of difficult settlement of coal slurry.

Hydration phenomenon and characteristics of mineral have been researched using the simulation and experimental methods. Density functional theory (DFT) and Molecular dynamics (MD) simulation are widely used in the study of study of mineral hydration and interface reactions. Characteristics of electronic structure, charge transfers, dynamic and thermodynamics can be simulated. And the interaction mechanism revealed from the atomic level (Luo et al. 2022; Peng et al. 2016). Water molecules interact with the α -quartz (001) surface via two hydrogen bonds between the surface active atoms and water molecules, generating the hydration layer, which is consist of three monolayers of water molecules (Liu et al. 2019; Liu et al. 2022). Reagent interactions with quartz surfaces are also described, for example, the effects of various nitrogen-containing compounds on the hydroxylated α -quartz (001) and (100) surfaces. Wherein, 2,4,6-trinitrotoluene (TNT) and 2,4-dinitrotoluene (DNT) are preferably adsorbed on the α -quartz(001) surface, while 3-nitro-1,2,4-triazole-5-one (NTO) is found on both α -quartz surfaces (Tsendra et al. 2019). When encountering macromolecules or large systems simulation,

the dynamic statistical properties of mineral-water molecules (Li et al. 2018; Yu et al. 2021; Wang, Ren, and Yan 2021) interface interactions can be simulated and analyzed by the MD method. For example, the adsorption of water molecules and ions on the quartz surface (Quezada, Rozas, and Toledo 2017; Zambrano, Walther, and Jaffe 2014; Dewan et al. 2014).

The hydration force has been studied by SFA (Schrader et al. 2015), AFM (Butt 1991), 3D-SFM (Miyazawa et al. 2016), 3D-AFM (Martin-Jimenez et al. 2016) and QCM-D (Chen et al. 2016) techniques from the first observation of periodic hydration forces between mica surfaces in dilute electrolyte solutions (Pashley 1981a, 1981b; Pashley and Israelachvili 1984). Sun studied the origin of hydration force, its date from the hydrophobic interactions and how it is shaped by the mineral surface (Sun 2017). Drost-Hansen (Drost-Hansen 1977) proposed a typical three-layer structure hydration model for polar and non-polar surfaces, wherein there is a great difference in how water molecules are arranged on hydrophobic or hydrophilic surfaces. Based on the viscosity of Einstein's equations, Song (Song et al. 2005; Song and Peng 2005) and Min (Min, Peng, and Liu 2015) modified Einstein's viscometry in order to study the surface hydration characteristics of nano-silica, kaolinite and montmorillonite. Zhao et al. (2017) (Zhao and Yi 2017) proposed combining AFM and viscometry to investigate the thickness of the hydrated layer on the nano-montmorillonite surface. Firstly, the thickness of nano-montmorillonite and its aspect ratio were measured by AFM, and then the hydration factor on the surface of montmorillonite was calculated by viscometry. Subsequently, the thickness of the hydrated layer on this surface was calculated to be 1.6 nm.

At present, the research on quartz hydration mainly focuses on the theoretical simulation of hydration mechanism, but the cognition of the actual hydration on the quartz surface and the microscopic effect mechanism of inevitable solution metal cation is still not well understood. Samples mentioned in previous viscosity experiment are flake clay minerals, and the nano silica takes the form of spherical particles, which differ from the objective of the irregular flakes quartz and cannot be completely applicable. Therefore, based on the real particle shape to modify Einstein's viscosity equation to determine the different solution chemistry environment, it was possible to characterize the hydration factor of quartz surface particles and metal ion/hydrated cation of quartz grain surface hydration characteristics. Clarifying the influence of metal ions on the surface hydration of fine quartz particles has theoretical guiding significance for the refractory coal slime water and other mineral processing wastewater containing quartz particles.

2. Materials and methods

2.1. Sample and reagents

The quartz sample with the density of 2.569 g/cm³ test by the pycnometer. The X-ray diffraction (XRD) and atomic fluorescence spectroscopy (XRF) test results of the samples are shown in Fig. 1 and Table 1.

XRD analysis results showed there were no crystalline mineral impurities in the quartz samples. Also indicated here is that the quartz sample has good crystallinity and the strongest diffraction peak appears at 28.10, which is very consistent with the standard pattern of α -quartz. According to Table 1, the SiO₂ content in the quartz sample is 97.764%, the total proportion of various impurities in the quartz sample is less than 3%, and the loss of combustion is 0.198%. It means that the quartz sample is characterized by high purity and meets the requirements of test.

Reagents used in this study, namely NaCl, KCl, MgCl₂, CaCl₂, hydrochloric acid, and sodium hydroxide were obtained from China National Reagent Corporation Ltd. The grade is that of analytical reagents with a purity of 99.9%. The water is deionized water which was filtered through a resin layer with a pore size of 0.2 μ m

Table 1. Chemical components atomic fluorescence spectra analysis of quartz

Composition	SiO ₂	Al ₂ O ₃	MgO	P ₂ O ₅	SO ₃	K ₂ O	CaO	TiO ₂	Fe ₂ O ₃	ZrO ₂
content (%)	97.764	1.533	0.048	0.018	0.011	0.299	0.019	0.034	0.068	0.008

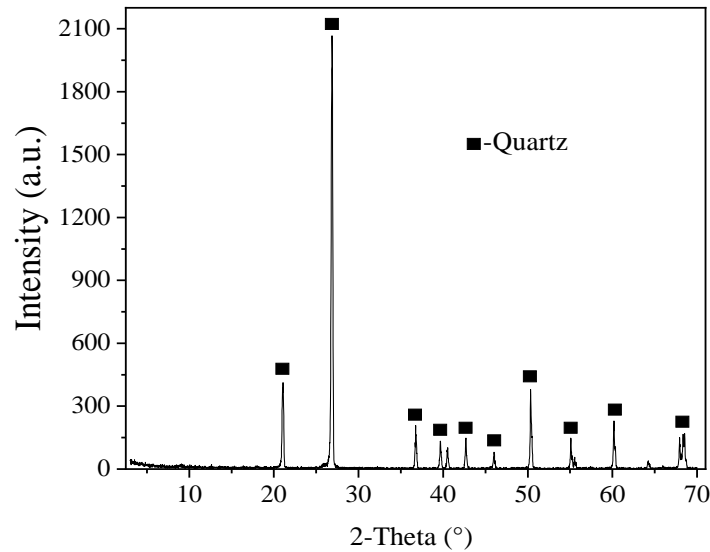


Fig. 1. The X-ray diffraction pattern of the quartz sample

2.2. Experimental method

In the dispersion system with very low concentration, Einstein viscosity equation can well describe the relationship between the volume fraction of rigid spherical particles and the viscosity of the dispersion system (Mardles 1940) (Eq.1).

$$\eta/\eta_0 = 1 + 2.5\phi_{sp} \quad (1)$$

where the η/η_0 is the relative viscosity, η is the viscosity of the dispersion system, η_0 is the viscosity of the solvent and ϕ_{sp} is the volume fraction of solid particles.

However, when the solid particles are hydrophilic particles, the surface hydration will affect the water molecules at the particle interface. The viscosity of the particles in the water suspension and the effective density of the dispersed layer increase with the increase of the volume of the particles in the water suspension. The difference between the effective volume of particles and the volume of particles is the volume of the hydration layer on the surface of particles. Song (Song et al. 2005) characterized the surface hydration of hydrophilic particles by modifying Einstein viscosity equation, defining hydration factor. And referring the derivation process of relative viscosity characterization hydration namely the relationship between the relative viscosity increments of suspension and the change of effective volume fraction.

Based on the Einstein's viscosity equations, Song et al. (2005), Song and Peng (2005) and Min et al., (2015) modified Einstein's viscometry in order to study the surface hydration characteristics of nano-silica, kaolinite and montmorillonite. The parameter of 2.5 in Einstein's viscosity equation corresponds to the viscosity shape factor of rigid spherical particles, while in practice, quartz is in flakes, and its real viscosity shape factor needs to be corrected. In order to modify the particle shape in the viscosity equation, batch statistical analysis was employed to analyze a certain amount of particle morphology and size, and the actual sphericity or roundness of the particles was obtained by the proportional relationship with spherical particles. Firstly, the surface morphology of sample particles is observed, and then the particle size is calibrated under the standard of SEM scale. The calibration rule is to select a relatively flat surface, measure its length and width, obtain the length width ratio of the particle, and do a statistical comparative analysis on a large number of particles. SEM photos and size calibration pictures of particles are shown in Fig. 3. It can be seen from Fig. 3 that quartz comprises spherical block particles, 75% of the quartz particles are below 9.862 μm , the aspect ratio of particles is counted in combination with SEM, and the results are shown in Fig. 4. According to the statistical analysis concerning the aspect ratio of particles, it is distributed between 1 ~ 3, and particles are spherical with the aspect ratio of 1. The larger the aspect ratio of particles, the worse the sphericity will be. The average aspect ratio of the particle group is 1.723, with 76% of the particles having an aspect ratio less than the particle swarm's average.

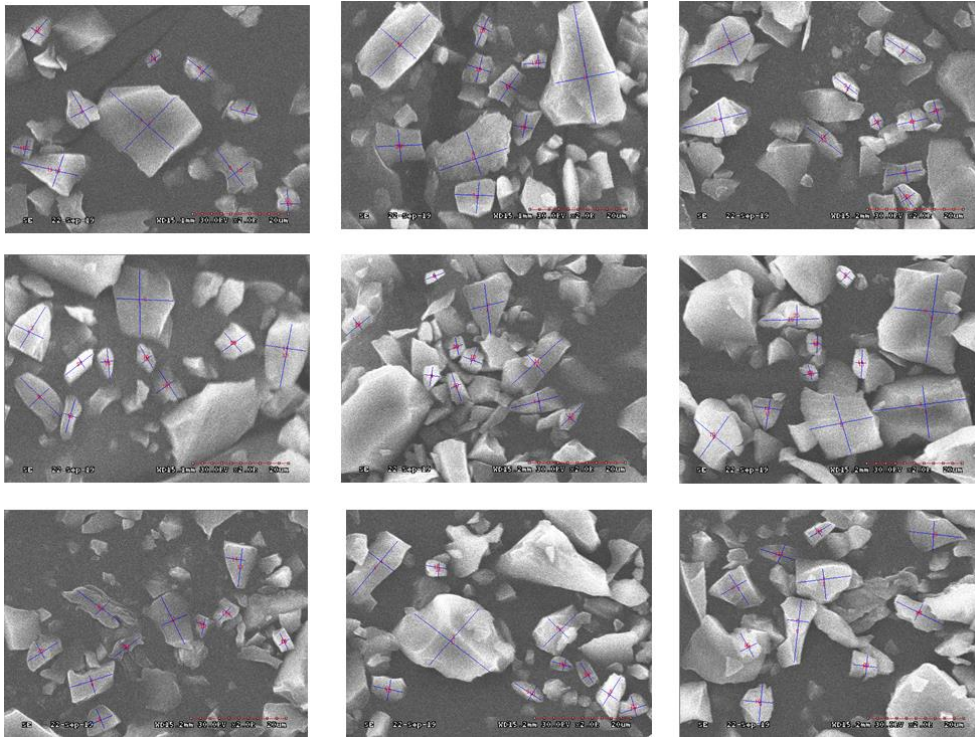


Fig. 3. SEM image of quartz particles and particle size calibration

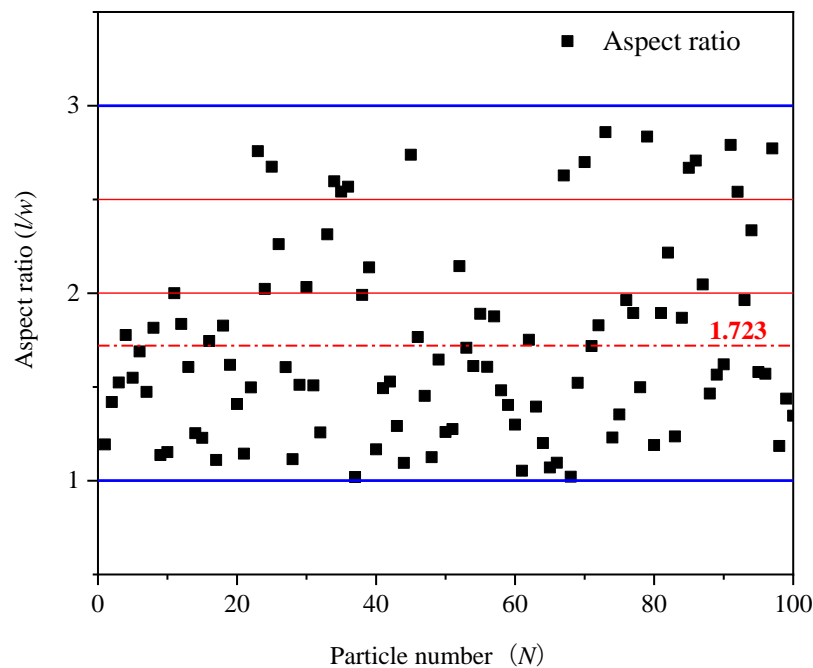


Fig. 4. Statistical analysis of particle aspect ratio

After the statistical analysis of the aspect ratio of particles, the difference between particles and spherical particles needs to be considered, namely, the actual sphericity of particles needs to be known. The method of analyzing the projection area of particles is adopted. Since the research's quartz sample comprises flake particles, its projection should be oval. The particle sphericity (s) is the ratio of the circumference of the equivalent projected area of the particle to the actual circumference of the projected contour (Lv et al., 2021):

$$s = \frac{C_s}{C_t} \quad (2)$$

where C_e is the circumference of the circle equivalent to the projected area, and C_t is the circumference of the actual projection of the particles selected in the test. For non-spherical particles, the closer the s value of the particle is to 1, the closer the projected area of the particle is to a circle and the closer the particle morphology is to a sphere. The analysis shows that the sphericity of the particle size quartz amounts to approximately 0.899. This particle sphericity can help to modify the shape factor of the real fine quartz particles, so that the viscosity equation is modified and made suitable for the spherical like fine quartz particles in this study. The relationship between the hydration factor (f) and the relative viscosity of the suspension and the slope k of the particle volume fraction curve can be expressed as follows:

$$f = 0.4k/s - 1 \quad (3)$$

A series of metal ion (Na^+ , K^+ , Mg^{2+} and Ca^{2+}) suspensions containing fine quartz particles with volume fractions of 0.2%, 0.4%, 0.6%, 0.8% and 1.0% are prepared, respectively. In order to make sample particles and aqueous solution fully mixed and dispersed, the prepared suspensions are placed on a magnetic stirrer and stirred evenly at a rotational speed of 300 r/min for 24 hours.

AR-G2 rheometer (TA Instruments Ltd, United States) measured the viscosity of the quartz suspension. The test was carried out after checking the instrument's condition and using standard liquid to measure the standard curve. The fixed shear rate was 150 s^{-1} and each test was repeated 20 times.

3. Results and discussion

3.1. Effect of pH on the hydration of quartz surface

Solution pH will affect the charging characteristics of the particle surface, and also have a certain impact on the hydration of the particle surface, thus affecting the diffusion of the suspension and the viscosity of the suspension. Fig. 5 shows the effect of solution pH on the surface hydration of fine quartz particles.

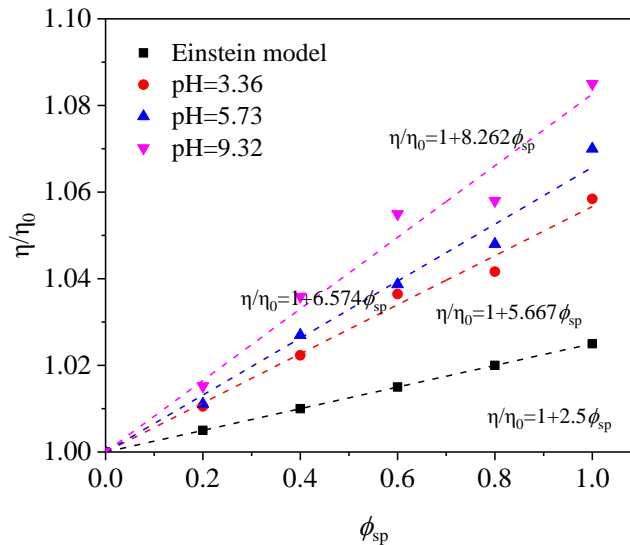


Fig. 5. Effect of solution pH on surface hydration of quartz particles

Due to the surface hydration of quartz particles, the viscosity of fine quartz suspension is greater than that predicted by Einstein viscosity equation. By fitting and analyzing the viscosity curves under different volume fractions and solution pH, the relationship equations (4), (5) and (6) are obtained.

$$\eta/\eta_0 = 1 + 5.667\phi_{sp} \quad (4)$$

$$\eta/\eta_0 = 1 + 6.574\phi_{sp} \quad (5)$$

$$\eta/\eta_0 = 1 + 8.262\phi_{sp} \quad (6)$$

According to equation 3, the hydration factors are related to the slope of the fitting curve. The linear fitting parameters under different pH conditions and the corresponding hydration factors of quartz particles are shown in Table 2.

Table 2 Hydration degree of quartz particles under different pH conditions

pH	Slope (<i>k</i>)	hydration factors (<i>f</i>)
pH=3.36	5.667	1.038
pH=5.73	6.574	1.365
pH=9.32	8.262	1.972

It can be seen from the results in Table 2 that the hydration factors on the surface of quartz particles increase with the increase of solution pH, which means that the viscosity of fine quartz particle suspension increases with the increase of solution pH; The viscosity of quartz suspension in alkaline solution is higher than that in acidic solution. From acidic to alkaline solution, the hydration factor of particle surface increase nearly twice. The reason is that the solution pH affects the dissociation condition on the surface of quartz particles (Liu et al. 2018). The zeta potential on the quartz particle surface increases with the increase of solution pH. Under alkaline environmental conditions, there is more charge on the surface of quartz particles, resulting in the suspension of fine quartz particles attributable to electrostatic repulsion. The dipole effect between more charge and water molecules is also enhanced, with strong hydration and large thickness of hydration film, The hydration repulsion interaction between particles is strong. On the contrary, the smaller the pH value of the suspension, the weaker the hydration of its surface.

3.2. Effect of ions on viscosity and hydration factor of quartz surface

The coagulation and diffusion characteristics of the suspension will certainly change after the ions interact with the fine particles in the suspension due to their charging and hydration characteristics. In order to investigate the effects of ion types and ion concentrations on the hydration characteristics of the particle surface, the effects of Na⁺, K⁺, Mg²⁺ and Ca²⁺ on the viscosity of the fine quartz particle suspension under different concentrations were found, and the specific results are shown in Fig. 6. The relative viscosity and volume fraction of fine quartz suspension under varied ion concentrations and different ion types are fitted, and the fitting curve is shown in the equation indicated in the Fig.. The hydration factor results of fine quartz particles under different conditions are shown in Table 5.

Table 5 Effect of ions species and concentrations on the hydration factor

Ion concentration	hydration factor (<i>f</i>)			
	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺
0.001mol/L	2.173	2.180	3.295	2.713
0.005mol/L	3.137	2.882	4.251	3.870
0.01mol/L	3.554	3.151	5.422	5.096
0.05mol/L	4.576	4.093	6.707	6.268

Under the same salt concentration, the hydration factors of fine quartz particles are in the order of Mg²⁺ > Ca²⁺ > Na⁺ > K⁺. With the introduction of ions, these metal ions will be adsorbed to the surface of quartz particles through electrostatic interaction or specific adsorption. Quartz dissolution was found to increase up to 100 times in the presence of small amounts of some alkali metal (M⁺) and alkaline earth (M²⁺) cations (Allen et al., 2017), more active sites are exposed on quartz surface. In addition, metal ions on the mineral surface and metal ions dispersed in aqueous solution interact with polar water molecules to form hydrate cation. Hydrate cation forms the first hydration layer through dipole interaction. The hydrogen bond between water molecules promotes the formation of its outer hydration layer, so as to enhance the order of water structure and the interaction between water molecules and quartz surface, which is closely related to the hydration ability of ions, The hydration capacity of Mg²⁺ and Ca²⁺ in these salt solutions is significantly stronger than that of Na⁺ and K⁺, nearly 4-5 times and the hydration capacity of Mg²⁺ is the strongest (Yin et al., 2017). Which is consistent with the hydration test results of different ions. Therefore, in the presence of Mg²⁺, the surface hydration factor *f* of fine quartz is the largest, followed by Ca²⁺, and in the presence of K⁺, the surface hydration factor of fine quartz particles is the smallest.

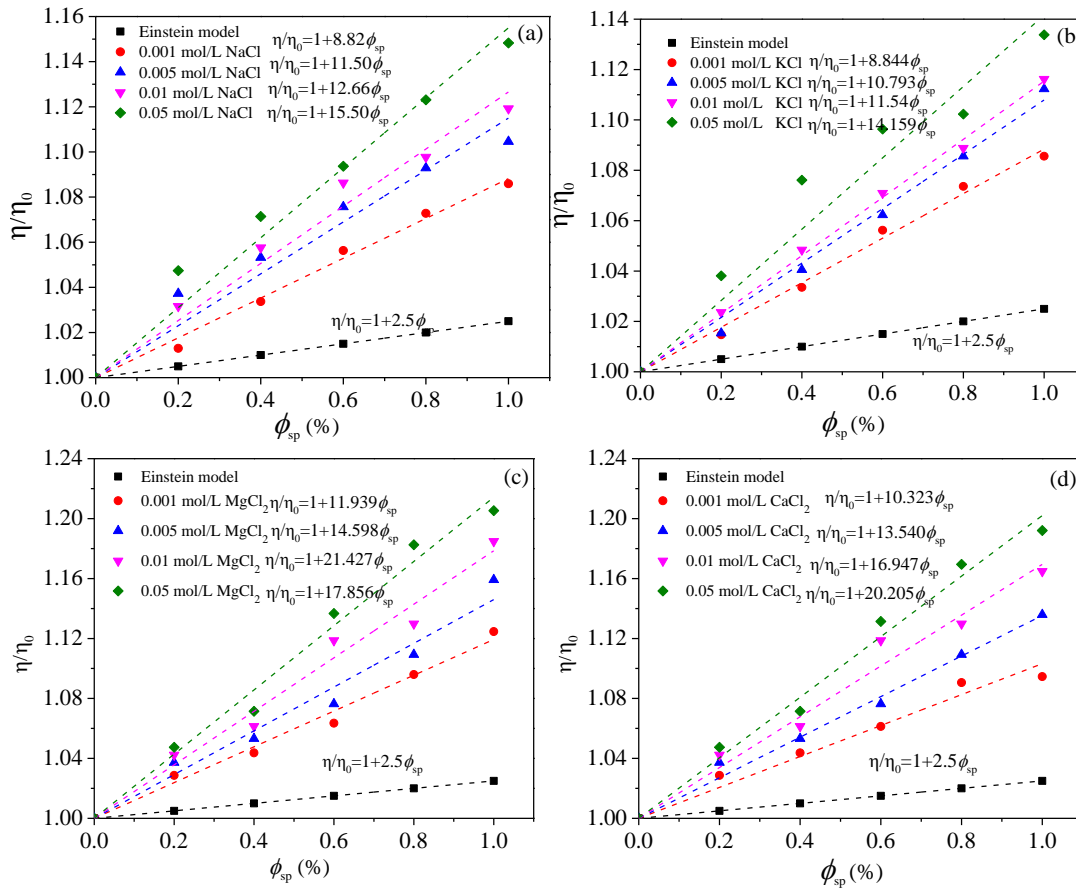


Fig. 6 Effect of ionic concentrations on the relative viscosity (a) NaCl; (b) KCl; (c) MgCl₂; (d) CaCl₂

4. Conclusions

The suspension viscosity of fine quartz particles increases with the increase of solution pH. The relatively strong hydration on the quartz surface appears in the alkaline environment. At this time, the quartz surface has more negative charge, strong hydration, strong hydration repulsion between particles, good particle dispersion in the suspension, large viscosity of the suspension and large hydration factors on the particle surface.

Compared with pure water, with the introduction of ions, metal ions have dipole interaction with polar water molecules in aqueous solution to form hydrate cation. Hydrate cation enhances the order of water structure due to its interaction with water molecules. When hydrate cation is adsorbed to the surface of fine particles, it will also enhance the hydration strength of particle surface. The relative viscosity of fine quartz suspension increases, and the hydration factors of particle surface also increase. Under the same ion concentration, the order of influence on the hydration factors of fine quartz particles is Mg²⁺ > Ca²⁺ > Na⁺ > K⁺.

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