

THE DISPERSIVE EFFECT OF SODIUM HEXAMETAPHOSPHATE ON KAOLINITE IN SALINE WATER

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Abstract—Freshwater has become increasingly scarce in many countries. To reduce the consumption of freshwater, the use of saline water resources in industry could provide an opportunity to meet the challenge of water-supply sustainability. However, the presence of electrolytes in saline water causes the coagulation of kaolinite, the colloid stability of which plays a key role in the processing of a number of minerals. Therefore, the dispersion of kaolinite in saline water was studied here. Electrophoretic mobility and colloid stability studies were conducted on a sodium hexametaphosphate-kaolinite system in the presence of NaCl, KCl, CaCl₂, and MgCl₂, the major electrolytes in saline water resources. The effect of each electrolyte on kaolinite dispersion was studied. Based on the studies of individual electrolytes, a method was developed to disperse kaolinite in 1:1 diluted synthetic seawater with distilled water, which may potentially reduce the consumption of freshwater by 50% when applied in industry.

Key Words—Kaolinite, Sodium Hexametaphosphate, Dispersion, Coagulation, Seawater.

INTRODUCTION

Freshwater resources have faced a crisis worldwide in recent decades (Zeid, 1998) and freshwater is scarce in virtually all countries (Radif, 1999). To reduce the consumption of freshwater, the use of saline water resources in industry provides an opportunity to meet the challenge of water-supply sustainability (Hamoda, 2001). The current trend in mineral-processing plants is already towards maximum recycle of process water, in order to reduce the consumption of freshwater (Haran *et al.*, 1996). Continued water recycling from tailing ponds results in the build-up of various metal ions, however, resembling the characteristics of saline water.

Kaolinite is of very fine particle size and its colloid stability plays a key role in the processing of a number of minerals such as iron ore, oil sands, kaolin, *etc.* (Mathur, 2002; Li *et al.*, 2005; Ma *et al.*, 2009; Ma and Bruckard, 2010). The dispersion mechanism of a number of dispersants involves electrostatic forces, which decrease with increasing ionic strength because of the compression of the electrical double layer. The tolerance of electrostatic stabilization to high ionic strength has received very limited attention. Through AFM measurements, Veeramasuneni *et al.* (1998) reported that, even in saturated salt solutions, surface charge still affects interparticle interactions, but whether such effects are strong enough to affect the settling behavior of particles has not been studied to date.

Process water and saline water contain a variety of metal ions, the effects of which on kaolinite coagulation

vary widely. The hydrolyzed species of alkaline earth ions are more strongly adsorbed than their free aquo ions at solid–liquid interfaces and play a key role in mineral processing (Fuerstenau and Palmer, 1976; Mpofu *et al.*, 2003). The concentration of Ca and Mg is often found to be of the order of 10⁻³ M in process water (Iwasaki *et al.*, 1980), which causes quick coagulation of kaolinite (Ma, 2010, 2011a). In addition to the difference between alkali and alkaline earth ions, the water-structure making/breaking capability of alkali ions further complicates the coagulation behavior of mineral particles (Ma and Pawlik, 2005). The effective dispersion of kaolinite in the presence of these metal ions is, therefore, of critical importance for the maximum recycle of process water and the use of saline water resources in mineral processing.

In the present study, the dispersive effect of sodium hexametaphosphate (HMP), a widely used dispersant of clays (Castellini *et al.*, 2005), on kaolinite in saline water containing Na, K, Ca, and Mg was investigated.

EXPERIMENTAL METHODS

Materials

Kaolinite from Georgia, USA, was provided by Ward's Natural Science Establishment. Quantitative X-ray diffraction (XRD) analysis showed that the sample contained 93% kaolinite and 7% illite. The average particle size (d_{50}) of the sample was determined to be 4.2 μm using a Malvern Mastersizer 2000 (Malvern, Worcestershire, UK). The BET (Brunauer, Emmett, Teller) specific surface area of the sample, determined using a Micromeritics Tristar 3000 instrument (Gosford, NSW, Australia), was 10.7 m²/g.

Sodium hexametaphosphate (96% pure) was supplied by Sigma. CaCl₂, MgCl₂, KCl, NaCl, and NaOH are

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ACS-certified chemicals from Ajax Finechem (Sydney, NSW, Australia). The synthetic seawater was prepared according to the recipe of Culberson *et al.* (1978), using 0.48523 M NaCl, 0.01058 M KCl, 0.01068 M CaCl₂, and 0.05518 M MgCl₂, similar to the recipes used by Dickson (1990) and Roy *et al.* (1993).

Methods

The turbidity method is a convenient tool for studying the coagulation process (Gregory, 2009). As the coagulated particles grow in size, their settling rates increase. Strongly coagulated suspensions produce clear supernatants of low turbidities, while dispersed systems give supernatants of high turbidities. The colloidal stability of kaolinite was investigated through turbidity measurements using a Hach 2100AN turbidimeter (Loveland, Colorado, USA). Turbidity values were expressed in nephelometric turbidity units (NTU). In turbidity tests, 0.5 g of kaolinite was first conditioned with 50 mL of salt solution or distilled water for 20 min. Then 50 mL of a sodium hexametaphosphate solution with known concentration was added and the entire mixture was further conditioned for 30 min. After stirring, 30 mL of the sample was collected for turbidity measurements. The experimental method was detailed in the author's previous work (Ma, 2011a, 2011b).

Zeta (ζ) potential measurements were performed with the use of a Zetacompact Z8000 model (CAD Instrumentation, France). An electric field of 80 V/cm was applied to kaolinite suspensions. Results were based on an automated video analysis of particles. Zeta potentials were calculated from electrophoretic mobilities using the Smoluchowski equation. 0.001 M NaCl was used as a background electrolyte in the tests. The standard deviation in these experiments was 2–3%.

RESULTS

Mineral suspensions containing kaolinite are usually dispersed in alkaline media in the mineral-processing industry (Li *et al.*, 2005; Ma *et al.*, 2009) and, thus, all the tests were conducted at pH 10 in the present study. The PZC of the edges of the kaolinite sample was determined to be between pH 7 and 8 (Ma, 2011b). Hence, the surfaces of kaolinite are free of positive charge in this work. The effect of HMP on the ζ potential of kaolinite in the presence of NaCl, KCl, CaCl₂, and MgCl₂ was studied here (Figure 1). In the absence of HMP, the magnitude of the ζ potentials was found to decrease in the order NaCl > KCl > CaCl₂ > MgCl₂. In the presence of HMP, the dispersant effectively rendered the surfaces of kaolinite more negatively charged despite the type of electrolytes. Such an effect is more pronounced for CaCl₂ and MgCl₂ than for NaCl and KCl.

In distilled water, only 10⁻⁵ M HMP was needed to obtain the optimum dispersive effect (Figure 2). In the presence of NaCl and KCl, a greater concentration of

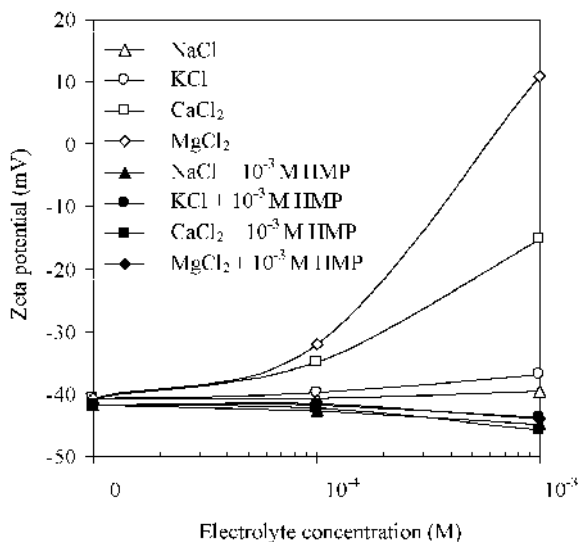


Figure 1. Effect of HMP (10⁻³ M) on the ζ potential of kaolinite in the presence of NaCl, KCl, CaCl₂, and MgCl₂ at pH 10. 10⁻³ M NaCl was used as a background solution.

HMP was needed to disperse kaolinite. K⁺ was generally more detrimental to the dispersion of kaolinite than Na⁺. When the ionic strength increased to 0.4 M, the dispersive effect of HMP disappeared completely.

In 10⁻⁴ M CaCl₂ and MgCl₂ solutions, the degree of dispersion of kaolinite increased slowly with HMP concentration (Figure 3). In contrast, in 10⁻³ M CaCl₂ and MgCl₂ solutions, when kaolinite particles were

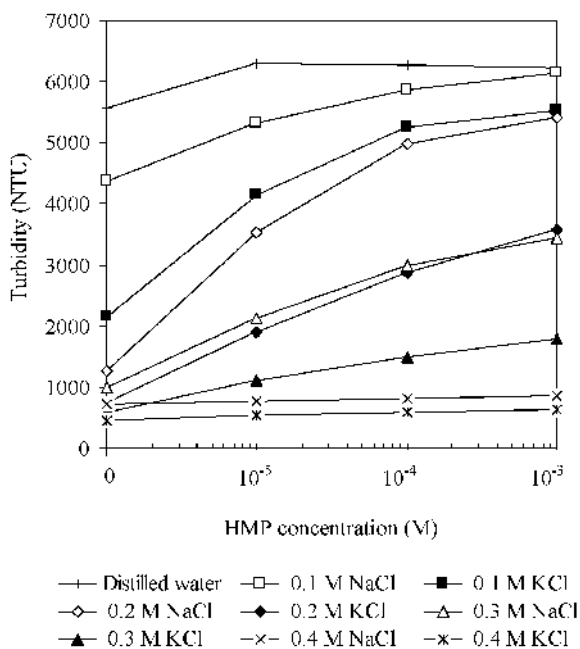


Figure 2. Effect of HMP concentration on the colloidal stability of kaolinite in the presence of NaCl and KCl at pH 10.

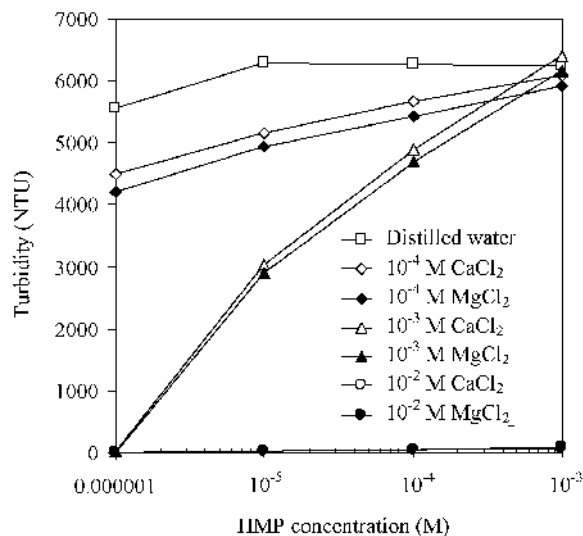


Figure 3. Effect of HMP concentration on the colloid stability of kaolinite in the presence of CaCl₂ and MgCl₂ at pH 10.

strongly coagulated, the degree of dispersion of kaolinite increased quickly with HMP concentration. In 10⁻² M CaCl₂ and MgCl₂ solutions, no dispersive effect of HMP was observed.

Without HMP, kaolinite particles coagulated when the concentrations of NaCl and KCl were ≥0.2 and 0.1 M, respectively (Figure 4). In contrast, 10⁻³ M CaCl₂ or MgCl₂ was sufficient to destabilize kaolinite.

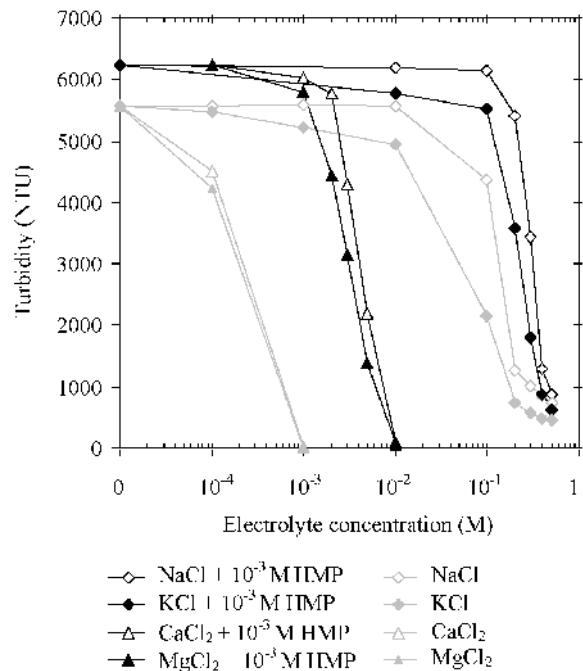


Figure 4. Effect of NaCl, KCl, CaCl₂, and MgCl₂ concentration on the colloid stability of kaolinite in the presence and absence of HMP at pH 10

The presence of HMP significantly enhanced the stability of kaolinite with improved tolerance to salts. The dispersive effect of HMP (10⁻³ M) on kaolinite in synthetic seawater and softened synthetic seawater (without Ca and Mg), diluted with distilled water, was also studied (Figure 5). In synthetic seawater, even after a 5:1 (distilled water:synthetic seawater ratio) dilution, fast coagulation of kaolinite still occurred. After softening, however, 1:1 dilution of synthetic seawater was sufficient to form a stable dispersion of kaolinite.

DISCUSSION

Effect of high ionic strength on electrostatic stabilization

The electrostatic interaction of approaching particles is due to the overlap of their diffuse layers. As two particles approach each other, the overlapping of diffuse layers leads to long-range repulsive forces due to entropic effects (Israelachvili, 1997). The profile of the interaction depends on the Debye length (Quemada and Berli, 2002). Debye length is the reciprocal of the effective thickness of the diffuse layer (κ^{-2}). The value of κ^2 is given by:

$$\kappa^2 = \frac{2e^2 N_A \sum C_i z_i}{\epsilon k_B T} \tag{1}$$

where ϵ , e , N_A , k_B , T , z_i , and C_i denote the permittivity of the electrolyte solution (F/m), the electronic charge (C), the Avogadro constant (mol⁻¹), the Boltzmann constant (J/K), the absolute temperature (K), the valency, and the concentration of ions (mol/L), respectively.

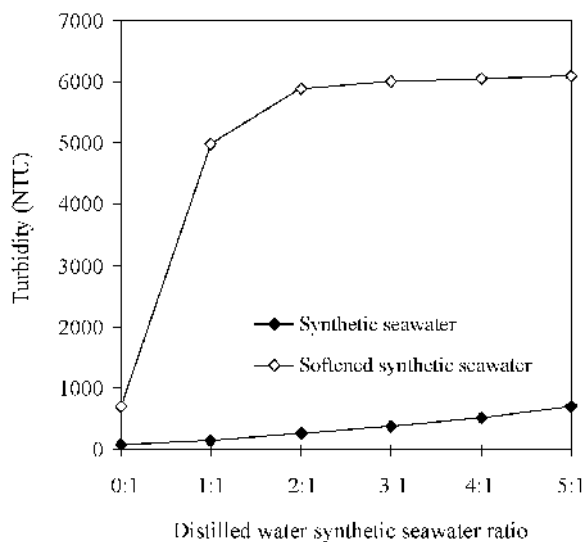


Figure 5. The dispersive effect of HMP (10⁻³ M) on kaolinite in synthetic seawater and softened synthetic seawater, diluted with distilled water, at pH 10.

Using equation 1, the Debye length in 0.1 and 0.2 M NaCl solutions was calculated to be 0.96 and 0.68 nm, respectively. According to the estimates by Nightingale (1959) and Osakai *et al.* (1997), the diameters of a hydrated Na^+ ion are 0.72 and 0.61 nm. Although the estimates differ somewhat, the Debye length in 0.1 M NaCl solution is larger than any of the estimated sizes of a hydrated Na^+ ion. Likewise, the turbidity of kaolinite in 0.1 M NaCl solution (without HMP) maintained a relatively high level (4362 NTU), indicating the still significant electrostatic repulsion between the clay particles (Figure 4). As the ionic strength increased to 0.2 M, the Debye length was reduced to 0.68 nm, which falls in the range of the estimated diameters of a hydrated Na^+ ion from 0.61 to 0.72 nm. At this point, the diffuse layer was compressed to such a level that only a single layer of Na^+ could be accommodated in it and electrostatic repulsion between kaolinite particles was reduced drastically. Consequently, the turbidity of kaolinite in 0.2 M NaCl solution (without HMP) decreased sharply to 1275 NTU, from 4362 NTU in 0.1 M NaCl solution (Figure 4).

In the presence of HMP, kaolinite dispersion remained stable up to 0.2 M NaCl. Even in 0.3 M NaCl solution, HMP still demonstrated a moderate dispersive effect on kaolinite. Because the Debye length was compressed drastically at such high ionic strength, the dispersive effect of HMP may not be attributed entirely to electrostatic stabilization. According to Lagaly (1989), HMP induces steric stabilization by forming a protective layer on clay particles which impedes the direct contact between particles. The results in Figure 4 demonstrate that such steric stabilization induced by HMP was also affected by ionic strength. Although still significant in 0.3 M NaCl solution, the steric forces diminished quickly at greater ionic strength.

Effect of Na^+ and K^+

Alkali ions form a sequence known as the Hofmeister series, *i.e.* Cs^+ , K^+ , Na^+ , Li^+ , which orders the ions from the least hydrated to the most hydrated. The least hydrated ions are also water-structure breakers (Cs^+ , K^+), while strongly hydrated Na^+ and Li^+ are water-structure makers. The adsorption behavior of these ions on mineral surfaces varies. According to Dumont *et al.* (1990), minerals with a high heat of immersion preferentially adsorb water-structure makers while surfaces with a low heat of immersion preferentially adsorb water-structure breakers. The heat of immersion is related to the PZC of the minerals (Healy and Fuerstenau, 1965). Generally, high-PZC minerals preferentially adsorb water-structure makers, while low-PZC minerals preferentially adsorb water-structure breakers. This concept explains satisfactorily the adsorption sequences observed on a number of oxides (Dumont *et al.*, 1990).

The ζ potential of kaolinite in the presence of K^+ is less negative in comparison to Na^+ , indicating that the

clay surfaces preferentially adsorb K^+ (Figure 1). The greater affinity of K^+ toward kaolinite surfaces resulted in stronger compression of the diffuse layer and faster coagulation of kaolinite particles, in comparison to Na^+ . For example, 0.1 M K^+ reduced the turbidity of kaolinite to 2160 NTU, while 0.1 M Na^+ only decreased it to 4362 NTU (Figure 4). In the presence of HMP, K^+ was found to be more detrimental to the dispersive effect of HMP than Na^+ . For example, in the presence of 10^{-3} M HMP, the effects of 0.1 M K^+ and 0.2 M Na^+ on kaolinite coagulation were practically the same (Figure 2).

At high concentrations of Na^+ and K^+ , where electrostatic stabilization diminished and steric stabilization induced by HMP dominated as discussed above, the dispersive effect of HMP was less pronounced in the presence of KCl than in the presence of NaCl. The more detrimental effect of K^+ than Na^+ on steric stabilization was also observed in previous work on a guar gum-quartz system by Ma and Pawlik (2005).

Effect of calcium and magnesium

In contrast to alkali ions, a trace of Ca and Mg was adequate to destabilize kaolinite (Figures 3, 4). Clays coagulated by Ca and Mg are difficult to disperse (Lagaly, 1989; Manfredini *et al.*, 1990). Andreola *et al.* (2006) reported that, even in the presence of 2.12×10^{-2} M HMP, Ca still drastically reduces the electrophoretic mobility and colloidal stability of kaolinite. However, in the present study, at up to 2×10^{-3} M CaCl_2 , the HMP-kaolinite system was still stable (5782 NTU) (Figure 4). The Ca concentration studied by Andreola *et al.* (2006) was 5×10^{-3} M. In 5×10^{-3} M CaCl_2 solution, the turbidity of kaolinite decreased to 2201 NTU (Figure 4), which corresponds very well to the results reported by Andreola *et al.* (2006). Therefore, in a very dilute solution of alkaline earth ions, *e.g.* $\leq 2 \times 10^{-3}$ M CaCl_2 or $\leq 10^{-3}$ M MgCl_2 , kaolinite can form a stable dispersion in the presence of HMP.

The HMP disperses kaolinite by adsorbing on the edges (positively or negatively charged), which increases the overall negative surface charge (van Olphen, 1963; Lagaly, 1989). The adsorption of HMP on kaolinite was reported to be of chemical nature, with the formation of an inner sphere complex with edge aluminol groups (Lyons, 1964; Diz and Rand, 1990; Castellini *et al.*, 2005). In the present study, HMP interacted strongly with kaolinite irrespective of the type of electrolytes (Figure 1). In earlier work by Ma (2010), Ca and Mg ions adsorbed on kaolinite in alkaline medium was found to increase significantly the adsorption density of starch by providing extra adsorption sites for the polymer. For HMP, however, alkaline earth cations adsorbed on kaolinite did not provide extra adsorption sites for the dispersant, because the ζ potentials of kaolinite coated with HMP are similar to each other in the presence of alkaline earth cations and alkali cations. This observation agrees very well with the

hypothesis of Andreola *et al.* (2006) that the Ca ions adsorbed in the electrical double layer of kaolinite do not affect HMP adsorption.

In addition to adsorbing on the edges of kaolinite, HMP can also disperse the clay particles by complexing the alkaline earth cations in kaolinite suspensions (Andreola *et al.*, 2004) because HMP forms a very stable complex with Ca (Kura and Ohashi, 1974).

Dispersion of kaolinite in synthetic seawater

The synthetic seawater was prepared according to the recipe of Culberson *et al.* (1978), using 0.48523 M NaCl, 0.01058 M KCl, 0.01068 M CaCl₂, and 0.05518 M MgCl₂, similar to the recipes used by Dickson (1990) and Roy *et al.* (1993). The minimum concentrations for NaCl, KCl, CaCl₂, and MgCl₂ to destabilize the HMP-kaolinite system were 0.3, 0.2, 0.003, and 0.002 M, respectively (Figure 4). The concentrations of all the electrolytes in seawater, except KCl, were greater than the levels required to destabilize the HMP-kaolinite system. Therefore, diluting seawater with freshwater was necessary to form a stable dispersion of kaolinite.

To reduce the amounts of NaCl, KCl, CaCl₂, and MgCl₂ in seawater to 0.3, 0.2, 0.003, and 0.002 M, respectively, 1:1 dilution with freshwater would be sufficient for NaCl, while even a 10:1 (freshwater:synthetic seawater ratio) dilution was inadequate to decrease the concentration of MgCl₂ to a satisfactory level (Figures 4, 5). Thus, Ca and Mg have to be removed by softening the seawater. In the absence of Ca and Mg, a 1:1 dilution with freshwater produced a very stable dispersion of kaolinite.

CONCLUSIONS

(1) In the absence of HMP, from 0 to 0.1 M NaCl, the Debye length was extensive and kaolinite formed a stable dispersion. At greater ionic strength (≥ 0.2 M), the Debye length was compressed to the size of a single hydrated Na⁺ ion, leading to the collapse of electrostatic stabilization. The presence of HMP improved significantly the clay's tolerance to high ionic strength, probably by inducing steric forces.

(2) The surface properties of kaolinite resemble that of low-PZC minerals, showing greater affinity toward a water-structure breaker (K⁺) in comparison to a water-structure maker (Na⁺). Consequently, K⁺ was more efficient at compressing the diffuse layer and caused quicker coagulation of kaolinite. K⁺ was also found to be significantly more detrimental to the dispersive effect of HMP than Na⁺.

(3) Alkali earth ions were more efficient at destabilizing kaolinite than alkali ions, but the quick coagulation of kaolinite caused by alkali earth ions can be dispersed effectively by HMP, when the concentrations of CaCl₂ and MgCl₂ were $\leq 2 \times 10^{-3}$ M and 10^{-3} M, respectively.

(4) Based on the effects of individual metal ions on kaolinite coagulation, 1:1 dilution of softened synthetic seawater with freshwater was found to be sufficient to form a stable dispersion of kaolinite in the presence of HMP.

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