



## Investigation of the effect of polyelectrolyte structure and type on the electrokinetics and flocculation behavior of bentonite dispersions

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### HIGHLIGHTS

- Electrokinetics behavior of bentonite-polyelectrolyte dispersions is investigated.
- PAM structure has a great influence on the dispersion turbidity and zeta potential.
- Larger flocs are produced using cationic PAMs than anionic or amphoteric PAMs.

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### ABSTRACT

The influence of surface charge type and density, and molecular weight of cationic, anionic and amphoteric polyacrylamide (PAM) on the electrokinetics, adsorption and flocculation behavior of highly stable bentonite dispersions is investigated. The results showed that PAM structure has a great influence on the dispersion turbidity and zeta potential. The reduction in the dispersion turbidity and negative zeta potential is considerably greater in the presence of cationic PAMs than anionic and amphoteric PAMs, due to an increased adsorbed layer thickness of cationic PAMs. In addition, the adsorption studies showed a higher bentonite surface affinity for cationic PAMs than anionic and amphoteric PAMs. Amphoteric PAMs exhibit both cationic and anionic behaviors and their strength and relative number of sites varies depending on surface charge density. Hence, the amphoteric PAMs adsorption isotherms were located in a region between cationic and anionic PAMs. Furthermore, the floc size and adsorption results suggested strong dependence on PAM surface charge density, type and molecular weight. Generally, at the optimum concentration, larger flocs are produced using cationic PAMs than anionic or amphoteric PAMs. It is also observed that the floc sizes increased with increasing cationic surface charge density but decreased with increasing anionic surface charge density. Molecular weight was also an essential factor influencing floc sizes. According to the floc size results, increasing the molecular weight of anionic and cationic PAMs increased the floc size. In conclusion, cationic PAM FO 4800 SH (very high charge density, high molecular weight) was determined as the best flocculant among the investigated PAMs. Overall, this work has been very successful in establishing the relationship between electrokinetics behavior and the produced flocs for the flocculated bentonite dispersions.

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### 1. Introduction

Wastewater treatment in minerals and metal industries is a core area of research considering the challenges faced in separating a myriad of contaminants from water. These contaminants mainly comprise of organic compounds, heavy metals and clay minerals [1–3]. The treatment involved is often characterized by low economic viability and is less environment-friendly. One significant example is the separation of clay minerals from wastewater.

Commonly found minerals include kaolinite and bentonite, industrial minerals that are often incorporated as functional fillers in several industrial applications [4,5].

The commercial significance of bentonite as a raw material is high due to its versatility in applications in different industries including, cosmetics and medical products, paint, dyes, water treatment [6–8], pharmaceuticals [9,10], dyes [11] and papermaking industry [12]. Its specific applications are primarily determined by its mineral composition, mainly smectite, and exchangeable cations based on the quality and quantity of each. Bentonite may be utilized in its natural form or pretreated through physico-chemical processes like acid activation, soda activation, ion

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exchange and heating treatment, depending upon the field of application [13]. Bentonite exhibits outstanding properties of adsorption and ion exchange. The negatively charged structure allows for various types of cations from the surrounding environment to electrically balance the structure. It offers remarkable adsorption capacity arising from the large surface area and high rate of ion exchange facilitating more active adsorption of organic pollutants.

Bentonite is known chemically as a hydrated aluminum silicate of composition  $(\text{Na})_{0.7}(\text{Al}_{3.3}\text{Mg}_{0.7})\text{Si}_8\text{O}_{20}(\text{OH})_4 \cdot n\text{H}_2\text{O}$  which occurs in the form of thin, roughly hexagonal platelets [14]. Its structure (see Fig. 3) comprises of three layers – two outer tetrahedral layers of silica ( $\text{SiO}_2$ ) surrounding a central octahedral layer of alumina ( $\text{Al}_2\text{O}_3$ ). The octahedral  $\text{Al}^{3+}$  ions in the structure are prone to isomorphous substitution with metal ions like  $\text{Fe}^{2+}$  and  $\text{Mg}^{2+}$  possessing lower valence. Similarly, the tetrahedral  $\text{Si}^{4+}$  ions can also be substituted by  $\text{Al}^{3+}$  ions. Such substitutions result in a net negative charge over the structure that are balanced usually by exchangeable cations like  $\text{Na}^+$  and  $\text{Ca}^{2+}$  along with water molecules that may surround the bentonite structure. In most bentonites,  $\text{Ca}^{2+}$  is found to be the abundant exchangeable ion. Few carry ions like  $\text{Na}^+$ ,  $\text{H}^+$  and  $\text{K}^+$  [15].

Bentonite plates have amphoteric properties and the faces or surfaces carry a permanent negative charge and depending on the pH, there is a net positive charge on the edges [15]. The structure of bentonite and therefore the rheology of the suspension are strongly affected by a change in the electrolyte and medium pH. When plate-like clay particles such as bentonite flocculate, three different modes of particle association may occur: face-to-face (FF), edge-to-face (EF) and edge-to-edge (EE). FF association leads to thicker and larger flakes, and EF and EE association lead to three-dimensional voluminous 'house-of-cards' structures [15,16]. The easy formation of the different types of association depends on the balance of electrostatic interactions (attractive or repulsive), which are controlled by the chemistry of the suspension, and the attractive van der Waals forces between the particles [16]. There are other theories describing the modes of interaction between plates-like clay particles have been proposed. Including the above mentioned, they are [16–19]:

- i. Mutual repulsion of the particles is a result of the interactions between their double layers.
- ii. Edge-to-edge association to form cross-linked ribbons is due to the high repulsive potential between basal surfaces.
- iii. Parallel associations of plates are held together by water molecules.

Tackling the treatment of clay dispersions is a key concern in mineral processing industries. Commonly occurring as contaminants, clay minerals like bentonite, kaolinite and montmorillonite are often washed from the mineral deposits into tailings using large quantities of water rendering highly stable clay-water suspensions. This feature has posed serious problems with regard to dewatering and handling problems in several industries. Direct discharge of wastewater into water bodies significantly increases water turbidity arising from persistently suspended clay particles. This has strained the feeding habits of several marine species and considerably reduced the population and the number of fish species. Therefore, treatment of effluents is essential before discharge into water bodies to prevent highly stable clay suspensions to hamper the health of aquatic species [1,20].

Coagulation/flocculation can be an optimum technique for destabilizing such stable colloidal dispersions in terms of feasibility, efficiency as well as cost-effectiveness. Formerly, the use of inorganic coagulant metal salts as coagulants was common with acceptable efficiencies achieved. However, several drawbacks such as high dosages, sensitivity to changes in pH, temperature and characteristics of the suspension, handling vast quantities of sludge formed subsequently and coagulant specific flocculation mechanism (charge neutralization with small bridges) that produced flocs with low shear resistance limited their use [21]. Consequently, there has been a shift in focus towards flocculation using high molecular weight polymeric flocculants (or polyelectrolytes) that are biodegradable, cheap, efficient, versatile and pose minimum disadvantages [20,22,23].

Flocculation of bentonite particles can take place through several mechanisms: polymer bridging, neutralization of surface charge, complex formation between polymer and suspension particle on particle surface and depletion flocculation [24]. The polyelectrolyte is mixed with bentonite particles followed by adsorption of polymer on particle surface. The adsorbed polymer chains undergo rearrangement in their configuration on the surface. Collisions among particles induce aggregation subsequently forming large flocs that settle down. The flocs may even deform in cases of intense agitation or depletion flocculation [25].

At present, a number of researches have been conducted to characterize bentonite structure and its flocculation behavior with different flocculants. The application of natural polymers such as chitosan and polysaccharide flocculants in bentonite dispersions have been investigated for exploring more economical and environment-friendly reagents [26]. Moreover, several studies discuss the flocculation process for dispersions like kaolinite analyzed in terms of zeta potential and turbidity measurements only. Further, a few papers discuss bentonite removal by coagulation-flocculation process using cationic polymers (like polyacrylamide) and inorganic salts [22].

In brief, for the first time, the effect of high molecular weight polyacrylamides with various surface charge types and densities has been examined on the flocculation of simulated bentonite suspensions. Unlike natural flocculants, such as chitosan, high molecular weight PAMs successfully destabilized stable bentonite suspensions using low concentrations and without the need for inorganic salts as coagulant aids. The floc structures have also been analyzed by means of particle size distribution.

Hence, the objective of this study involves optimizing flocculation behavior of fine bentonite suspensions by analyzing the effect

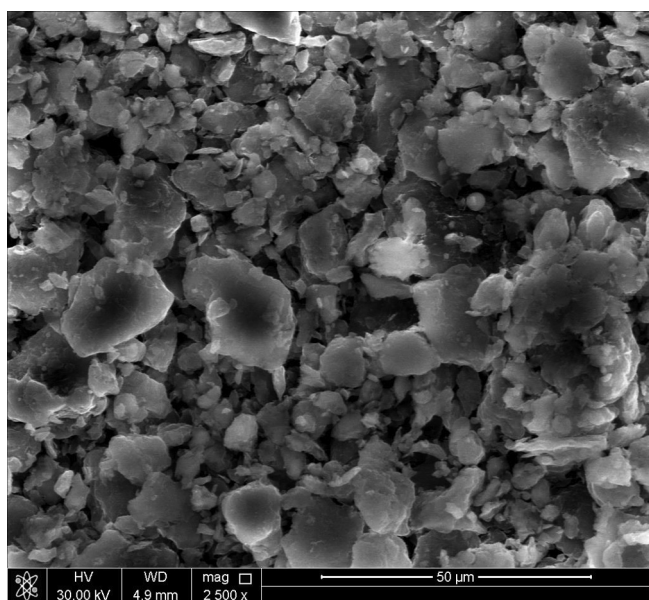


Fig. 1. SEM pattern for pure bentonite.

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