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Intercalation of ionic liquids into bentonite: Swelling and rheological behaviors

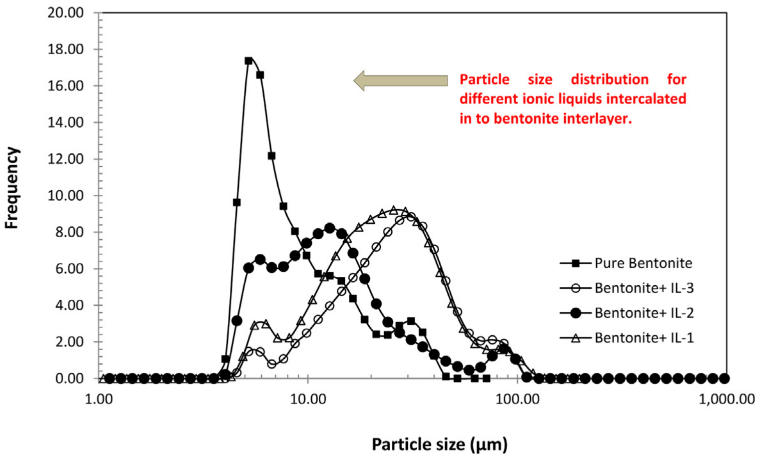
M.S. Nasser[a,*], Sagheer A. Onaizi[b,c], I.A. Hussein[a], M.A. Saad[d], M.J. Al-Marri[a], A. Benamor[a]

[a] Gas Processing Center, College of Engineering, Qatar University, P.O. Box 2713, Doha, Qatar
[b] School of Chemical Engineering and Advanced Materials, Newcastle University, 535 Clementi Road, Blk 35 #02-01, Singapore, Singapore
[c] School of Chemical Engineering and Advanced Materials, Newcastle University, Newcastle upon Tyne, NE1 7RU, United Kingdom
[d] Department of Chemical Engineering, College of Engineering, Qatar University, P.O. Box 2713, Doha, Qatar

HIGHLIGHTS

- Intercalation of different ionic liquids into bentonite interlayer is investigated.
- All the studied samples showed elastic rather than viscous rheological behavior.
- The swelling of bentonite reached equilibrium at 3 mM concentration of tested ILs.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, direct intercalation of three different green ionic liquids (ILs) having different salt and cation sizes into bentonite interlayer is investigated. The ILs covered in this study are: 1-hexyl-3-methylimidazolium chloride [IL-1], 1-butyl-3-methylimidazolium octyl-sulfate [IL-2], and 1-butyl-3-methylimidazolium bromide [IL-3]. The aim of this IL modification of bentonite is to enhance both rheological and thermal stability of the modified bentonite. This objective is achieved by increasing the swelling of the bentonite interlayer, which is essential for many applications. The basal d-spacing of the ILs intercalated bentonite in comparison with dried bentonite powder as detected by XRD results showed that the cations of the three ILs are successfully intercalated into the interlayer of the bentonite platelets by a cation exchange mechanism and the swelling is likely influenced by the cation size, type and concentration of the ILs. The measurements of the equilibrium concentrations of the ILs in the solution suggested that adsorption is taking place on the external surfaces. This assumption is validated by Zeta potential measurements. The results from particle sizes and Zeta potential measurements have shown that bentonite intercalated with IL-1 and IL-2 are the most effective in decreasing the overall repulsive

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* Corresponding author.
E-mail address: m.nasser@qu.edu.qa (M.S. Nasser).

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forces and increasing the attractive forces on the bentonite surface. Consequently, larger bentonite aggregates are produced by the chloride based IL intercalated bentonite rather than those intercalated with the bromide based IL or pure bentonite. In a similar manner, rheological measurements showed that the bentonite intercalated with IL-1 and IL-2 produced strong bentonite aggregates with higher G′, G″, τ_c and η^* than those intercalated with IL-3 and the untreated bentonite samples. Rheological, Zeta Potential and XRD measurements have confirmed that green ionic liquids can be used in different applications to enhance bentonite swelling.

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1. Introduction

Bentonite consists mainly of montmorillonite [$(Al,Mg)_2(OH)_2(Si,Al)_4O_{10}(Ca)_x$ on H_2O] and different amounts of other minerals such as quartz (SiO_2), calcium and sodium feldspar [($CaAl_2Si_2O_8$),($NaAl_3Si_2O_8$)] [1]. Bentonite is used in a wide range of applications including, drilling fluids [2–4], pharmaceuticals [5,6], dyes [7–10], papermaking industry [11–13], and in many other applications. The structure of bentonite is principally composed of layers including silica and alumina sheets linked together and arranged on top of each other. Bentonite is an aggregate of lamellar platelets, packed together by electrochemical forces and containing interposition water. Each platelet consists of three sandwich-arranged layers: a central octahedral alumina (Al_2O_3) layer, and two tetrahedral silica (SiO_2) layers (Fig. 1). Bentonite surface carries negative charges on their faces due to isomorphic substitutions which are Al^{3+} for Si^{4+} substitution in tetrahedral sites and Mg^{2+} for Al^{3+} substitution in octahedral sites [14]. The negative charge is balanced by cations intercalated between the structural platelets/sheets and these cations may be alkaline earth ions (Ca^{2+} and Mg^{2+}) or the sodium ion (Na^+).

When water is added to the bentonite surface, it will be absorbed in the interlayer region of the clay and this initiates swelling. Swelling is the moving apart of the clay particles until they reach their maximum equilibrium separation [15–18]. Because of this unique behavior, the clay intercalations have been widely studied by many researchers [19–24]. The main objective of the clay intercalation was to produce compounds that could have exceptional properties such as improved mechanical strength, thermal stability, photoluminescence catalysis, etc. [25]. Ionic liquid (IL) modified clays have recently attracted the attention of many researchers and this is due to the IL unique properties such as negligible vapor pressure, non-flammability, high electrical conductivity and thermal stability [26–28]. Ionic liquids (ILs) are molten salts, which remain liquids (non-volatile) at room temperature. ILs can be used in many important applications such as catalysis, electrochemistry, and extraction [26].

Bentonite-based clays are the main component of drilling fluids. Because of their exceptional rheological properties, bentonites are used to formulate the body of drilling fluids, and they work as a viscosifier additive and thus reduce water losses [29–31]. In drilling fluids, the favourable bentonite properties are high swelling capacity, zero free water, controlled fluid loss, and good rheological behaviours [2,16]. Attempts have been made to upgrade bentonite by treating them with Na_2CO_3 [31,32], with H_2SO_4 [33], and with MgO [32] in order to obtain a bentonite with improved swelling capacity and viscosity. In this study, ILs modification of bentonite is proposed to enhance both rheological and thermal stability of the modified bentonite by increasing the swelling of the bentonite interlayer. ILs are known to be green materials, which are a major advantage over other chemical modifiers.

This is the first attempt to examine the intercalations of different ILs into the bentonite platelets and explore its swelling, flocculation and rheological behavior. The progress of the intercalation process was observed in terms of the bentonite swelling using particle size distribution and XRD tests. Rheological techniques are used to explore the impact of ILs on the degree of bentonite dispersion and rheology of suspensions. Zeta potential measurements are conducted to evaluate the stability of IL-bentonite colloidal dispersions.

2. Theory

2.1. Rheology of colloidal dispersions

The rheological measurements of colloidal dispersions are important tool to investigate the stability or flocculation of colloidal dispersions and their flow and deformation behaviours [34,35]. Drilling fluids, for example, are time-dependent and show viscoelastic behavior (i.e., they show both viscous and elastic behaviours under an applied deformation force). The viscoelasticity of the drilling fluid is related to the important task of pressure peak, static barite sag, cuttings transport, weighting particles suspended in the drilling fluid, and hydraulic modelling and optimization [36]. The viscoelastic tests are also crucial to evaluate low shear viscosity, gel structure formation and breakage [37–39]. This gel structure keeps heavy particles suspended, and prevents cuttings from precipitating and settling at the wellbore wall during the drilling operations [36,39]. The results from the steady and dynamic rheological tests are expected to provide useful insight into colloidal dispersions structure, stability, and time-dependent behavior.

The viscoelastic properties of the materials are mainly defined in terms of storage modulus (G'), loss modulus (G''), critical stress (τ_c) and complex dynamic viscosity (η^*). The values of G' and G'' are usually determined from oscillatory tests by applying a sinusoidal input strain [$\gamma = \gamma_0 \sin(\omega t)$] and recording the subsequent sinusoidal shear stress $\tau = \tau_0 \sin(\omega t + \delta)$ in a strain controlled rheometer. The opposite takes place in a stress controlled rheometer. In the previous formulas, γ_0 is the maximum amplitude of the strain while τ_0 is the maximum stress displaying the amplitude and δ is the phase angle.

At low stress amplitudes, G' and G'' are not strain or stress dependent. The linear region where the rheological properties are not strain or stress dependant is called the linear viscoelastic region (LVR). The corresponding critical stress (τ_c) is defined as the stress where G' falls to 90% of the plateau value.

2.2. Particle size distribution (PSD)

Number average or first moment (D_n) and weight average or second moment (D_w) will be used to evaluate the effect of IL type and concentration on bentonite swelling behavior. These parameters are defined in Eqs. (1) and (2):

$$D_n = \sum \left(\frac{N_i \times D_i}{N_i} \right) \tag{1}$$

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