Contents lists available at ScienceDirect

### Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

#### Research paper

# Experimental investigation of the effect of henna extract on the swelling of sodium bentonite in aqueous solution



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#### ARTICLE INFO

Article history: Received 31 August 2014 Received in revised form 18 December 2014 Accepted 19 December 2014 Available online 4 January 2015

Keywords: Henna extract Clay stabilizers Sodium bentonite Conductivity Swelling Adsorption

#### ABSTRACT

This paper reports for the first time the effect of Henna extract as a new, naturally occurring, and ecofriendly additive on swelling of sodium bentonite in aqueous solution. This is performed via a number of methods including dynamic linear swelling test in two distinct temperatures of 28 °C and 82 °C, sodium bentonite inhibition and sodium bentonite sedimentation tests. The results indicated that inhibition properties of Henna extract are a function of its concentration and are comparable with potassium chloride and polyamine as the two most common clay stabilizers. Inhibition mechanism was assessed using adsorption measurements obtained by conductivity technique, contact angle measurements via the sessile drop method, and scanning electron microscopy (SEM) analysis. Results of adsorption measurements indicated that Henna extract has a lower adsorption isotherm in alkaline medium (pH = 9) compared to natural medium (acidotic). Adsorption of Henna extract increases hydrophobicity of sodium bentonite particle surface; nevertheless, this adsorption mechanism is slightly weakened in alkaline medium due to the effect of caustic soda as a pH adjustment agent. SEM analysis demonstrated that sodium bentonite particles have an extended area when exposed to Henna extract solution instead of distilled water indicating sodium bentonite particle stability. Henna extract has deflocculating properties at low concentrations (especially up to 0.2 mass%); yet it indicates good inhibition properties to sodium bentonite swelling at a concentration several times higher than that of deflocculating concentration, about 3 mass%. Inhibition properties could be attributed to the hydrogen bonding between the hydroxyl group of Henna extract constituents and oxygen atoms available on silica groups of sodium bentonite, especially to the lawsone (2-hydroxy-1,4 napthaquinone) constituent.

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#### 1. Introduction

Wellbore instability is one of the important as well as threatening factors in raising the cost of drilling operations. According to the statistics (Zeynali, 2012) wellbore instabilities in oil and gas wells could result in a waste of U\$\$ 1 billion per year worldwide. Roots of wellbore instability can be divided as mechanical and physic-chemical effects (Tan et al., 1996; Osuji et al., 2008). Unlike mechanical effect, physico-chemical effects are time dependent (Manohar Lal and Amoco, 1999) and are a direct result of interaction between the rock (especially shale) and drilling fluids.

Shales can be defined as sedimentary rocks with laminated layered characteristics and high clay content (Diaz-Perez et al., 2007). These rocks are allocated for about 75% of drilled formations and source of 90% of wellbore stability problems (Steiger and Leung, 1992) and are known as troublesome formations for drilling (Chenevert and Amanullah, 2001; Akhtarmanesh et al., 2013). Non-clay minerals are

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also present in shale but they are generally considered inert making contributions. Therefore, swelling and dispersion properties of a particular shale are functions of the amount and types of clay minerals present (Steiger, 1982). Among all the clays, sodium saturated smectites have attracted a lot of interest, mainly owing to their high swelling potential and their occurrence frequencies during drilling operations (Anderson et al., 2010). Exposure of water sensitive shales, especially sodium saturated smectites, to the conventional water-based drilling fluids results in their hydration and swelling due to water adsorption which in effect leads to more drilling problems.

Clay swelling occurs under two consecutive regimes including the inner crystalline swelling and osmotic swelling (Madsen and Müller-Vonmoos, 1989). The swelling process can be defined as a phenomenon in which water molecules surround a clay crystal structure and position themselves to increase the structure's c-spacing, thus resulting in an increase in volume (Patel et al., 2007b). Although oil-based muds have a superior performance such as excellent shale inhibition, wellbore stability, lubricity, anti-accretion properties, contamination resistance and possibility of reuse (Chegny et al., 2008), the use of these fluids to control clay rich shales has been limited due to their high cost and environmental restrictions (Clark and Benaissa, 1993; Mody et al., 2002). Up to date,







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#### Nomenclature

Acronyms	
XRD	X-Ray Diffraction
XRF	X-Ray Fluorescence
SEM	Scanning Electron Microscopy
PHPA	Partially Hydrolyzed Polyacrylamide
LOI	Loss On Ignition
LSCA	Linear Swelling Cap Assembly
PPM	Part Per Million
Variable	S
q	Adsorption of Henna extract on sodium bentonite sam-
	ple particles, mg/g sodium bentonite sample
m <sub>tot.solution</sub> Total mass of solution in original bulk solutions, g	
c°	Henna extract concentration in initial solution before
	equilibrated with sodium bentonite sample, ppm
С	Henna extract concentration in aqueous solution after
	equilibrated with sodium bentonite sample, ppm
m <sub>clay</sub>	Total mass of sodium bentonite sample added,

various drilling fluid additives namely "clay stabilizers" that can inhibit clay swelling have been introduced and utilized to strengthen the clay rich shales compared to water-based drilling fluids. Potassium chloride known as a commonly clay stabilizer has been utilized from several past decades up to date which has a high performance in controlling the clay swelling owing to the cationic size and hydrational energy (Pruett, 1987). Presence of potassium ion in water-based drilling fluids at a concentration above 1 Wt%, fails these fluids according to the mysid shrimp bioassay test (Anderson et al., 2010). Therefore, it can be claimed with certainty that high concentration of potassium chloride required to inhibition could yield harmful effects as adversely affecting the environment including the high disposal costs, high fluid loss due to high clay flocculation (Zhong et al., 2011), and higher corrosion rates like any high salinity aqueous solution (Clark et al., 1976). Setting aside the aforementioned drawbacks, the use of potassium chloride in conjunction of partially hydrolyzed polyacrylamide (PHPA) has been recommended to control the water sensitive shales (Clark et al., 1976). Quaternary amine salts have a high inhibition property. However, a number of disadvantages including their toxicity (tetra methyl ammonium chloride and high molecular weight guaternary amine), flocculation of fluids with high solid concentration, and incompatibility with anionic drilling fluids additives are associated with these salts which restrict their applications (Patel et al., 2007a). The ability of polyetheramine components in suppressing the swelling potential has already been investigated (Qu et al., 2009; Wang et al., 2011; Zhong et al., 2011). Zhong et al. (2012) indicated the superior inhibitive properties of poly (oxypropylene)-amidoamine, which caused a significant reduction of hydration and swelling. Recent studies have revealed the high applicability of dopamine (Xuan et al., 2013) and bis (hexamethylene) triamine (Zhong et al., 2013) in clay swelling reduction.

According to previous discussions, defects of clay stabilizers are more related to their toxicity, high costs, and corrosion. Therefore, finding the appropriate clay stabilizers free from these problems is highly essential. The primary aim of this study is to implement of Henna extract, a natural dye, as new, naturally occurring, and ecofriendly additive to investigate its effect on swelling of sodium bentonite. Motivations behind this study were the some of the fascinating properties of Henna extract such as being a naturally occurring material and thus its environmentally friendly characteristics, acting as an anti-corrosion in various metallic mediums (Al-Sehaibani, 2000; Chetouani and Hammouti, 2003; El-Etre et al., 2005; Ostovari et al., 2009; Abdollahi and Shadizadeh, 2012), decreasing the hydrophilic properties of sandstone (Abdollahi, 2011; Abdollahi and Shadizadeh, 2013), its low cost and wide availability. In the present work, adsorption of Henna extract on non-prehydrated sodium bentonite was investigated by conductivity method. The effect of Henna extract on sodium bentonite swelling was also assessed through a combination of dynamic linear swelling, sodium bentonite inhibition, and sodium bentonite precipitation tests. Wettability of sodium bentonite film modified with Henna extract was investigated using contact angle measurement via sessile drop method. Finally, interaction between the Henna extract and sodium bentonite was observed by scanning electron microscopy (SEM) analysis.

#### 2. Experimental

#### 2.1. Materials

#### 2.1.1. Henna extract

Lawsonia inermis L, which is commonly referred to "Henna" is a shrub or small tree frequently cultivated in India, Pakistan, Egypt, Yemen, Iran, and Afghanistan (Ali et al., 2009). It has been reported that leaves of Henna contain lawsone, gallic acid, glucose, fats, resine, mucilage and traces of alkaloid (Chaudhary et al., 2010). In current study, Henna extract that is obtained from leaves of Henna was purchased from Ebnemasouyeh Company, Tehran, Iran. The main constitutes of Henna extract are lawsone (2-hydroxy-1,4 napthaquinone, C<sub>10</sub>H<sub>6</sub>O<sub>3</sub>), gallic acid (3,4,5-trihydroxybenzoic acid, C<sub>7</sub>H<sub>6</sub>O<sub>5</sub>), dextrose  $(\alpha - D - Glocose, C_6H_{12}O_6)$ , and tannic acid which were characterized using gas chromatography, mass spectrometry, and methanolysis by methanol and sulfuric acid (Fig. 1) (Ostovari et al., 2009). Humans have employed Henna containing lawsone (principle coloring matter) as hair dye, body paint, and tattoo dye for more than 4000 years (Kirkland and Marzin, 2003). Lawsone can be used as a sensitive cyanide and acetate sensor due to several specifications including capability of hydrogen bonding with anions by the hydroxyl of the enol in structure, containing conjugated ketone that is suitable conjugate or nucleophilic addition of strong nucleophiles such as cyanide, and containing naphthoquinone chromophore as a signaling unit (Hijji et al., 2012). Henna extract powder has brown color with special odor, is soluble in water (with special pH curve as shown in Fig. 2) and alcohol. Properties of Henna extract are summarized in Table 1.

#### 2.1.2. Sodium bentonite

Sodium bentonite with high montmorillonite content and cation exchange capacity of 66.5 meg/100 g (determined by methylene blue test) was obtained from Pars Drilling Fluid Company, Tehran, Iran, X-ray fluorescence (XRF) and X-ray diffraction (XRD) were implemented to characterize the chemical and semi-quantitative mineral compositions of sodium bentonite. The chemical and semi-quantitative mineral compositions of sodium bentonite are presented in Table 2. LOI demonstrates loss on ignition that is related to the volatile components.

#### 2.2. Methods

#### 2.2.1. Adsorption measurements

The adsorption isotherms of Henna extract were determined using conductivity method both in natural pH and adjusted pH of 9 (using caustic soda solution). The adsorption tests were carried out in a number of consecutive steps including: a) preparing different concentrations of Henna extract aqueous solution both in natural pH and adjusted pH of 9, b) measuring the conductivity of each series solutions by Sartorios PP-20 conductivity meter, c) plotting the conductivity versus concentration for each series solutions as a reference conductivity curve, d) adding 0.5 g of dried sodium bentonite powder (at 105 °C) to 100 ml of each solutions, e) stirring the dispersions by magnetic stirrer for 24 h to reach equilibrium, f) centrifuging the certain amount of

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