Iranian Journal of Oil & Gas Science and Technology, Vol. 5 (2016), No. 2, pp. 21-44
http://ijogst.put.ac.ir

Evaluation of a Naturally-derived Deflocculant (Terminalia Chebula) in Bentonite Dispersions

Jalal Neshat[1] and Seyed Reza Shadizadeh*[2]

[1]M.S. Student, Department of Petroleum Engineering, Petroleum University of Technology, Ahwaz, Iran

[2]Professor, Department of Petroleum Engineering, Petroleum University of Technology, Ahwaz, Iran

Received: July 17, 2015; *revised:* November 02, 2015; *accepted:* December 05, 2015

Abstract

The unwanted addition of salt to drilling causes flocculation which has an adverse effect on mud rheological properties. To treat the flocculated mud chemical, deflocculants are commonly used; however, their disadvantages such as negative environmental effects, lower tolerance to contamination, and toxicity motivated scientists to search for effective additives. Using plant derived additives instead of commercial additives could help resolve the mentioned weaknesses, because they are nontoxic, cheap, easily accessible, and act multi-functional. In this paper the effect of black myrobalan rheological properties of flocculated bentonite mud was investigated and its performance was compared with chrome lignosulfonate (CLS). Rheological and filtration tests were conducted and properties such as plastic viscosity, yield point, gel strength, thixotropy, and apparent viscosity were calculated. It was perceived that by increasing black myrobalan concentration to 0.6 wt.%, rheological parameters and filtration loss decreased by 50% and 66.3% respectively, but they increased at higher concentrations, which indicated that black myrobalan acted as a deflocculant up to 0.6 wt.%. The deflocculation behavior of black myrobalan at low concentrations is attributed to ellagitannic acid and tannic acid. The comparison of the enactment of black myrobalan with chrome lignosulfonate showed that black myrobalan had a stronger decreasing effect on the rheological parameters and filtration compared to CLS.

Keywords: Flocculation, Black Myrobalan, Bentonite Mud, Deflocculant, Chrome Lignosulfonate.

1. Introduction

Drilling operation is the most significant stage in the development of oilfields and drilling fluid is the essential constituent of rotary drilling. It performs functions such as carrying cuttings from the hole and permit their separation at the surface, cooling and lubricating the drill bit and drill string, balancing formation pressure to prevent blowout, stabilizing the wellbore, and forming a thin, low-permeable filter cake (Benyounes et al., 2010; El-Sukkary et al., 2014). Drilling fluids typically are classified into three major groups, namely pneumatic, oil-based, and water-based. Oil-based drilling fluids have excellent properties such as stability, lubricity, and temperature stability. Though, the excessive use of oil-based drilling fluids harms the environment and it is important to develop more

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* Corresponding Author:
Email: shadizadeh@put.ac.ir

environmentally-friendly drilling fluids; hence water-based drilling fluids are more acceptable than other types (Meng et al., 2012). They do not damage the environment, and they are easy to construct, cost-effective to maintain, and proficient to overcome most drilling problems (Amoco Drilling Fluids Manual, 1994).

The simplest water-based mud is a mixture of water and bentonite, which is called bentonite–treated mud. It is usually utilized in drilling trouble-free shallow wells. Bentonite is added in order to provide viscosity to improve cutting carrying capability, suspension, lubricity, and reduce filtration loss. Viscosity or resistance to flow is provided by the large flat shape of the sheets, but it is the electrostatic charges on the sheets, which make bentonite unique (AVA Drilling Fluids Manual, 2004; Azar, 2007).

One of the major problems related to bentonite mud is flocculation. Flocculation occurs when clay platelets are electrically attracted to each other. One of main sources of flocculation is salt, which may originate from make-up water, salt stringers, massive salt sections, salt water flows, and commercial sources (Baker Hughes, 2006). When the Na^+ ion content is raised to 1%, the water becomes more positively charged than the ionized covering cloud that protects the clay platelet. The positive Al^{3+} edge joins the oxygen face, and the drift of edge to face is speeded up. The viscosity rises dramatically, and water loss becomes intense, when the clay flocculates edge to face in a "House of Cards structure," (Lyons, 2010). A common way to overcome this problem is to use deflocculants or thinners. Some of known chemicals are lignosulfonate, tannin, and lignite (Skalle, 2010).

There are good reasons, including economics, environmental protection, toxicity, etc. to improve drilling fluid performance and management. Drilling mud may represent 5% to 15% of drilling costs, but may cause 100% of drilling problems. Furthermore, increasing environmental concerns have limited the use of some of the most effective drilling fluids and additives and the industry is dedicated to replace them to low toxic, less harmful and less pure mud additives which are acceptable according to current environmental norms (Amanullah, 2007; Bloys et al., 1994).

Therefore, many researchers have tried to improve mud properties through replacing different commercial additives by environmentally-friendly additives. Yousif et al. (2011) assessed typical properties of lignite from Lakhra coal mines in Pakistan as a drilling mud thinner. They found that it was capable of increasing plastic viscosity, reduced yield point and gel strength, and satisfied its effectiveness as mud thinner and mud weight tolerance. Meng et al. (2012) studied the influence of carbon ash on the properties of bentonite dispersion and realized that by the addition of carbon ash, filtration loss and filter cake thickness increased dramatically, whereas the density decreased slightly. They also observed that increasing carbon ash concentration could result in high yield point and the ratio of yield point to plastic viscosity but low variation in viscosity. First time Narayana (2013) investigated the ability of black myrobalan in reducing the viscosity of the bentonite dispersion. He observed that among all the tannin-bearing materials, Myrobalan powder caused the greatest reduction in viscosity. Mehta and Jatkar (2013) showed that the addition of myrobalan powder changed the pH of the bentonite mud in a manner similar to its effect on viscosity. El-Sukkary et al. (2014) assessed several vanillin-modified polyoxyethylene surfactants in water-based drilling fluid and perceived that AV and YP increased, and hence they can be used as rheological modifiers. Muds formulated with some of these surfactants showed lower filtrate loss than blank mud, while these classes of additives were not stable against temperature. Moslemizadeh and Shadizadeh (2015) investigated the effect of Henna extract as a new additive on swelling of sodium bentonite in aqueous solution. They found that Henna extract had deflocculating characteristics at low concentrations up to 0.2 wt.% and indicated good inhibition properties to sodium bentonite swelling at about 3 wt.%. Shirmardi and Shadizadeh (2015) studied the effect of Sedr leaf extract (Ziziphys Spina Christi) on drilling fluid properties and observed that Sedr extract is capable of reducing shale and sodium bentonite swelling at 3% in WBM. It acted as a filtration control agent without affecting the rheological properties of drilling fluid. They proposed that inhibition characteristics of Sedr extract could be due to the wettability alteration of clay platelets.

This paper describes the results from the evaluation of black myrobalan as a new non-toxic and biodegradable additive in water-based mud for the development of environmentally-friendly and efficient water-based mud.

2. Materials and Methods

2.1. Materials

a. Bentonite

Bentonite is a type of clay consisting mainly of a hydrous silicate of aluminum. It is formed by weathering volcanic tuff and ash and consists mainly of montmorillonite [$(Al, Mg)_2(Si,Al)_4O_{10}Ca_x$ on H_2O] and contains varying amounts of other minerals like quartz (SiO_2) and calcium and sodium feldspar [$(CaAl_2Si_2O_8)$,$(NaAl_3Si_2O_8)$]. Bentonite is categorized into two types: Na-bentonite, which has a high swelling capacity, and Ca-bentonite, which is a non-swelling clay and forms colloidal very quickly in water (Abu-Jdayil, 2011). By comparing the specifications of bentonite X with API standard (Table 1), it was found that bentonite used in this study is in agreement with API standard. The particle size distribution of bentonite X was measured by Zetasizer model Zen 3600 (Malvern Instruments Ltd, UK) and the result is represented in Figure 1. According to this figure, bentonite X has a normal particle size distribution around 1500 nm. X-ray diffraction (XRD) was performed (by normal scan between 5° and 80° with a phase and time step of 0.02° and 0.5 second respectively) by Panalytical diffractometer model PW-1710 in order to characterize the chemical composition of bentonite X. The XRD pattern of bentonite X in Figure 2 shows that it mostly contains montmorillonite and quartz. The physical and chemical analyses of this bentonite are given in Table 2.

Table 1

Specifications of bentonite X (Amoco production company, 1994).

Test for Specification (22.5 lb./bbl. bentonite)	Bentonite X	API standard
θ_{600}	41	>30
YP/PV ($lb_f/100\ ft^2.cp$)	3.1	≤3
V_f (cc/30 min)	10.9	≤15
Yield (bbl./ton)	100	>91
Moisture (wt.%)	10	≤10
+200 mesh (wt.%)	0.3	≤4

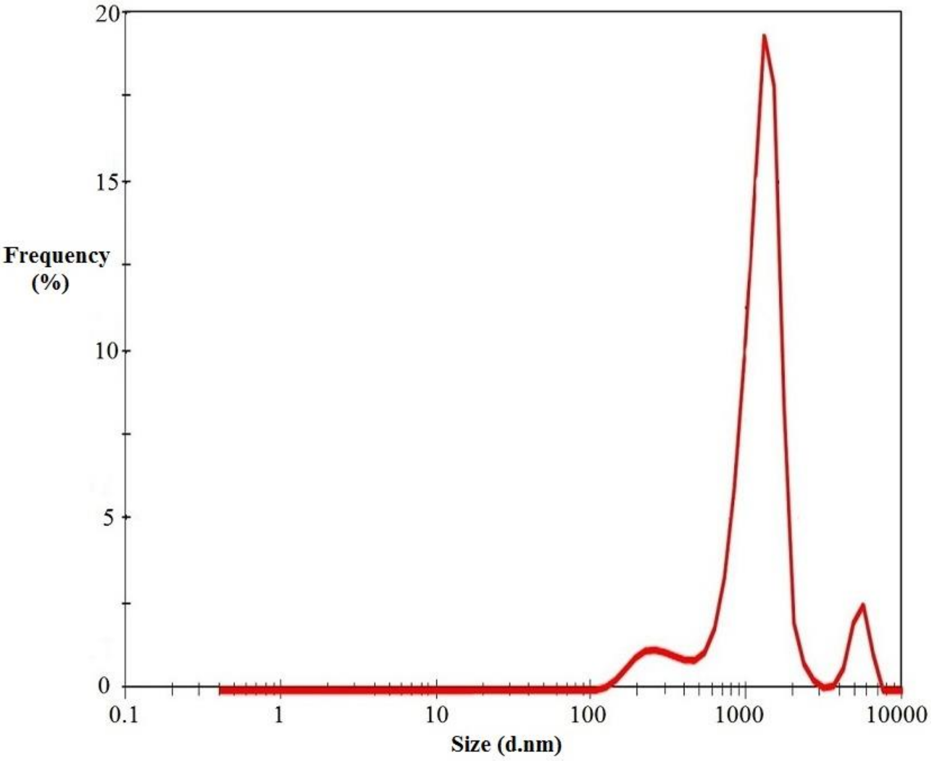


Figure 1
Particle size distribution of API bentonite (Z-Average=672.2 d.nm).

Table 2
Chemical and physical analyses of bentonite X.

Component	Mass percentage (wt.%)	Standard Error
SiO_2	69.3	0.2
Al_2O_3	15.4	0.2
Na_2O	3.9	0.09
MgO	2.8	0.08
CaO	2.4	0.07
Fe_2O_3	2.4	0.07
K_2O	1.2	0.05
P_2O_5	0.1	0.04
Density (gr/cc)	4.2	--------
Swelling volume (cc)	12	--------
pH	9	--------

The bentonite used in this study was supplied by Doreen Kashan (D.K., Tehran, Iran) company.

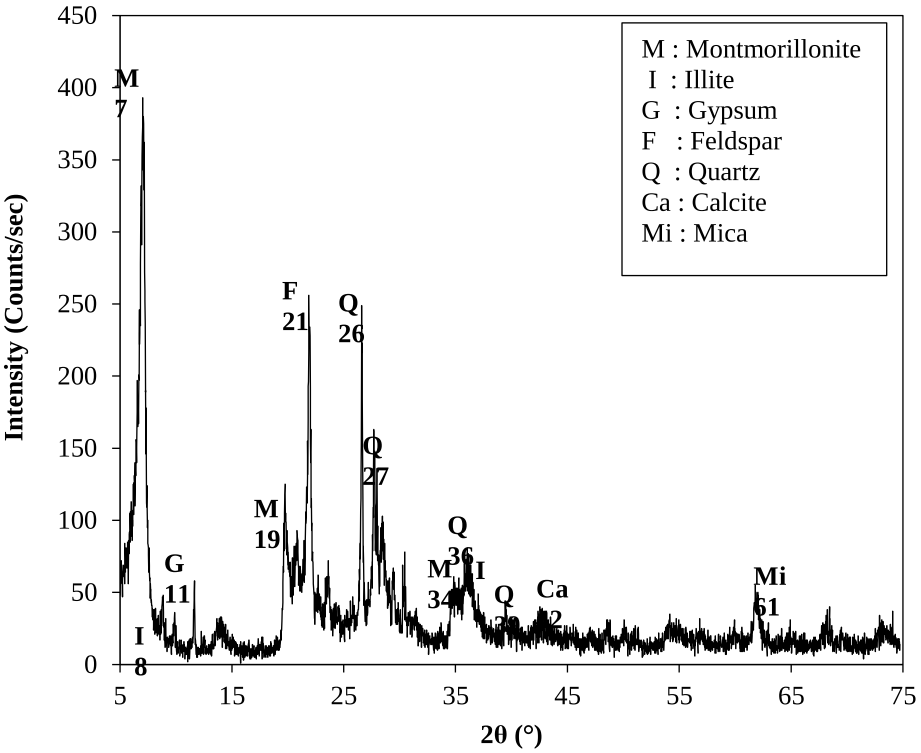


Figure 2

XRD pattern of API bentonite representing different minerals (number show phasing angles for minerals).

b. Black myrobalan (Terminalia Chebula)

Black myrobalan or Terminalia Chebula is a moderate tree which belongs to the family combretaceae. Its drupe is about 1 inch to 2 inch in size and has five lines or ribs on the outer shell (Figure 3). It grows in India, Myanmar, Bangladesh, Egypt, Turkey, and China. It is used in tanning the leather, in dyeing industry, in the basic aniline dyes as a mordant. In addition to the main uses, it has been used for centuries in the manufacture of writing inks (Mehta and Jatkar, 2013). It is also extensively used in traditional medicine as antimicrobial and is also employed to cure infectious diseases. Black myrobalan contains the triterpenes arjun glucoside 1, arjungenin, and the chebulisides 1&2. Other constituents are tannins up to 30%, chebulic acid 3% to 5%, chebulinic acid 30%, tannic acid 20% to 40% (Surya Prakash et al., 2012; Bag et al., 2013). Gas chromatography-mass spectrometry (GC-MS) analysis was performed by Perkin-Elmer GC Clarus 500 system interfaced to mass spectrometer equipped with an Elite-5MS (30mx250µm) composed of 5% Phenyl and 95% dimethylpolysiloxane. (Figure 4) revealed 1,2,3 Benzentriol, Levonoglucosenone, and n-Hexadecanoic acid (palmitic acid) as the frequent components in black myrobalan composition (Amala and Jeyaraj, 2014). Physical properties of black myrobalan are summarized in Table 3.

Figure 3
Terminalia chebula leaf, fruit, and extract (Rathinamoorthy and Thilagavathi, 2014).

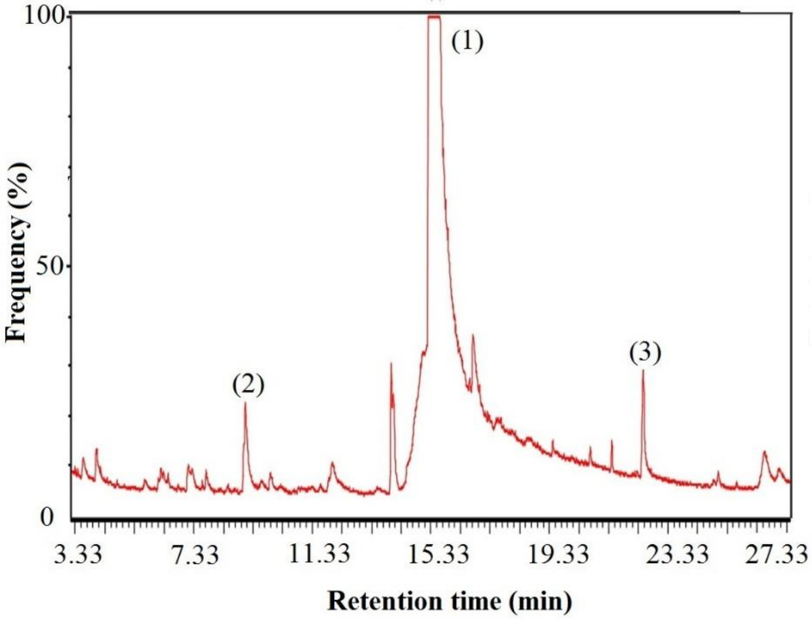
Table 3
Black myrobalan extract properties.

Property	Description
Type of Extract	Hydroalcoholic
Appearance	Brown
CMC	1.2 wt.%
Major component	1,2,3 Benzentriol
Nature	Acidic

The hydroalcoholic extract of black myrobalan in this study was purchased from Adonis Gol Daru (A.G.D.) Pharmacy Company (Tehran, Iran). Properties of black myrobalan are given in Table 3.

c. Chrome lignosulfonate (CLS)

Lignosulfonates are the strongly anionic by-product of the sulfite process. They are used in water based drilling fluids to deflocculate a clay dispersion which results in a cost effective reduction in fluid loss and cake thickness (AVA Drilling Fluids Manual, 2004). Chrome is added to lignosulfonate to assist with rheology stabilization (ASME Shale Shaker Committee, 2005). This additive was provided by Pars Drilling Fluid (PDF) Company (Tehran, Iran).



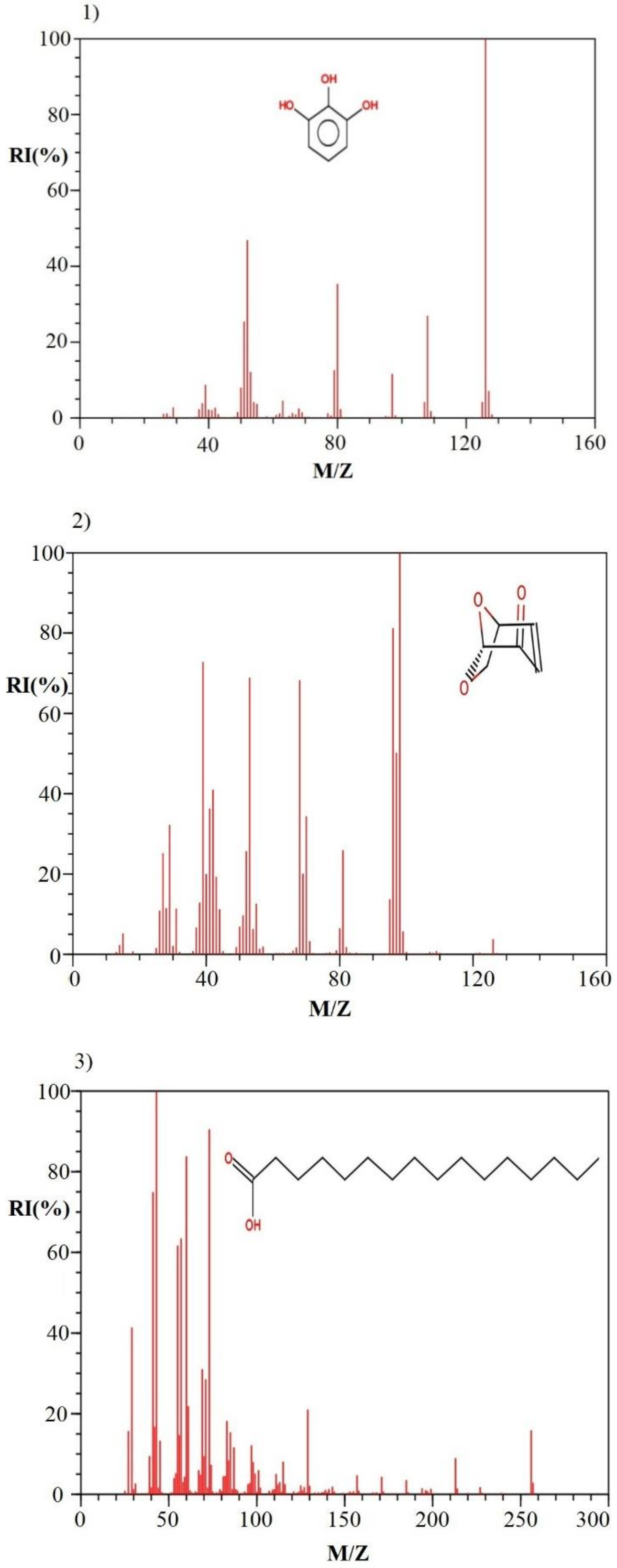


Figure 4
Structure and mass spectrum of major phytochemicals identified by GC-MS in Terminalia Chebula extract. (1). 1, 2, 3 Benzentriol; (2). Levonoglucosenone; (3). n-Hexadecanoic acid (palmitic acid)(Amala and Jeyaraj, 2014)

2.1. Experimental methods

a. Preparation of flocculated bentonite sample

A standard bentonite dispersion was prepared by the addition of 20 lb./bbl. (5.4 wt.%) bentonite to 350 cc distilled water and stirring at 6000 rpm. After 5 minutes, the mixing cup was removed from the mixer and the sides were scraped to dislodge any bentonite adhering to the cup. The mixer was replaced and stirring continued for an additional 10 min (for a total mixing time of 15 min). Then, the bentonite sample aged for 24 hours to hydrate fully (API RP-13I, 2009). Prior to flocculating the mud, it was stirred at 6000 rpm for 5 minutes to be homogenized. In order to increase additives solubility and clay stabilization, the pH of bentonite dispersion was increased to 9.5 by the addition of 1M KOH solution. Then, a 2 wt.% NaCl solution was added to the dispersed bentonite sample and mixed for 10 minutes. Predetermined contents (0.2, 0.6, 1.2, 2, and 3 wt.%) of black myrobalan or CLS were then added to the solution and mixed at 6000 rpm for an extra 10 minutes.

b. Characterization of bentonite mud

Fann V-G meter model 35 was used to measure the rheological properties of mud at high shear rates. Brookfield DVII$^+$ viscometer was also utilized to quantify the rheological behavior at low shear rates and at room temperature. API filtration loss of mud was determined by Baroid API filter press (Houston, Texas) at 100 psi (689.28 kPa) and 64 °F (17.8 °C). The pH value was measured by Kent EIL7045/46 digital pH meter. All the tests were carried out based on API-RP 13I. All the tests were carried out based on API-RP13I standard.

3. A review on rheological models

Rheological models are mathematical equations used to predict fluid behavior across a wide range of shear rates (Rabia, 2002). Three models used in this paper are explained as below:

3.1. Bingham plastic model

In the early 1900's, E.C. Bingham recognized that some fluids exhibit a plastic behavior and are distinguished from Newtonian fluids in a way that they require a yield stress to start flowing. These fluids are called to show a Bingham plastic behavior and are characterized by (Baker Hughes, 2006):

$$\tau = \tau_0 + \mu_p.\dot{\gamma} \qquad (1)$$

where τ_0 (lb$_f$/100 ft^2) is yield point; μ_p (cP) is plastic viscosity and γ (1/s) stands for shear rate.

According to this model, the rheological parameters are calculated by Equations 2-6:

$$AV = 0.5\theta_{600}(cP) \qquad (2)$$

$$PV = \theta_{600} - \theta_{300} \ (cP) \qquad (3)$$

$$YP = \theta_{300} - PV \left(\frac{lbf}{100ft^2}\right) \qquad (4)$$

$$RYP = \frac{YP}{PV} \quad (\frac{lbf}{100ft^2.cP}) \qquad (5)$$

$$LSRV = 2\theta_3 - \theta_6 \ (cP) \qquad (6)$$

where, θ_3, θ_6, θ_{600}, and θ_{300} are dial readings at 3, 6, 600, and 300 rpm respectively (Meng et al., 2012).
The primary problem of the Bingham plastic model is inability to accurately describe the rheological behavior of drilling fluids at low shear rates and yield point (Meng et al., 2012; Baker Hughes, 2006).

3.2. Herschel-Bulkely model

The Herschel-Bulkely (Yield-Power Law) is another model describes the rheological behavior of drilling muds more accurately than other models (Rabia, 2002).

This model is described by:

$$\tau = \tau_0 + K.\dot{\gamma}^n \tag{7}$$

where τ ($lb_f/100ft^2$) is the shear stress and τ_y ($lb_f/100ft^2$) represents the yield stress of the fluid; K $lb_f.s^n/100ft^2$) is the flow consistency index (and n represents the flow behavior index. Herschel-Bulkely (YPL) model reduces to the Bingham plastic model when n=1 and reduces to the Power Law model when τ_0=0 (Hassiba and Amani, 2012; Rabia, 2002).

3.3. Robertson-Stiff model

Robertson and Stiff (1976) proposed a three-parameter model to describe the rheological behavior of drilling fluids and cement slurries. Like the Herschel Bulkely model, it has been found to provide very close approximations for pressure losses in the circulating system in most drilling situations (Kök et al., 2000; Baker Hughes, 2006). In this model, shear stress is related to shear rate as given below:

$$\tau = A(\gamma + C)^B; (\tau > AC^B) \tag{8}$$

$$\acute{\gamma} = 0; (\tau \leq AC^B) \tag{9}$$

where, A, B, and C are constants determined by regression analysis. This model includes the Newtonian (C=0 and B=1), power law (C=0 and $B\leq$1), and Bingham plastic ($C\geq$0 and B=0) models as specials cases (Kök et al., 2000). Rheograms of all of the models which were explained before are shown in Figure 5.

Shear rate and shear stress values (for V-G with a bob radius of 1.7245 cm) are also calculated according to:

$$SS = 1.0678\,.N\,(\frac{lbf}{100\,ft^2}) \tag{10}$$

$$SR = 1.7034\,.\theta\ \ (s^{-1}) \tag{11}$$

$$SS = 1.7034\,.\theta\ \ (s^{-1})$$

4. Results & discussion

4.1. Rheological characterization of bentonite dispersion

a. High shear rates

The full flow curves of the flocculated bentonite mud in the presence of different concentrations of black myrobalan are represented in Figure 6. As could be seen in this figure, the addition of 2 wt.% NaCl solution increases the shear stress. By increasing black myrobalan concentration to 0.2 wt.%, shear stress decreases, and the further addition of black myrobalan extract up to 3 wt.% causes a slight increase in resistance to flow. According to Figure 7, some of active constituents of black myrobalan such as chebulic acid and 1-6 pentagalloyl glucose deprotonate in alkaline mud and become negatively charged. These deprotonated groups are physically adsorbed on the positively charged edges of clay platelets and neutralize them. The large size of these active constituents leads to establishing a repulsive force between the negative charges on clay surfaces and deprotonated groups. Thus a lower shear stress is needed to generate a known shear rate. A slight increase in shear stress at higher concentrations is due to the aggregation of black myrobalan molecules, which increases the solid concentration in the mud and consequently improves the flow resistance of mud.

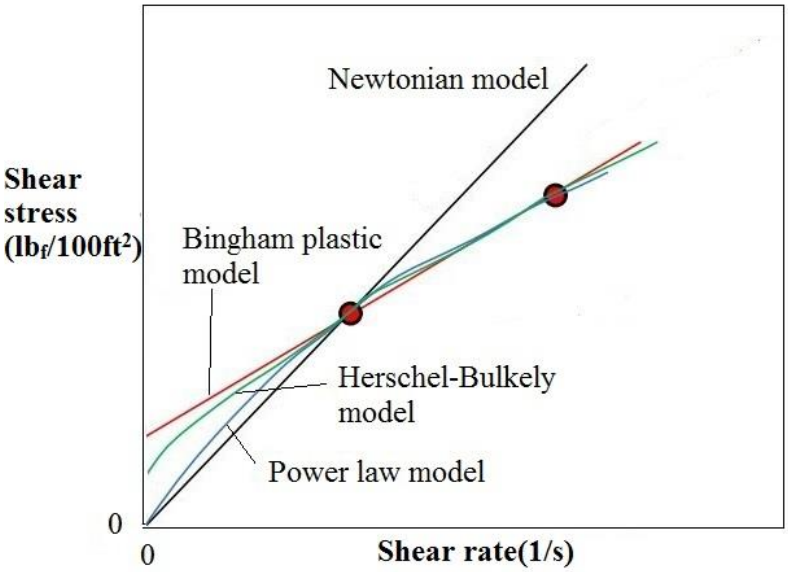


Figure 5

Rheograms of different rheological models (Skalle, 2010).

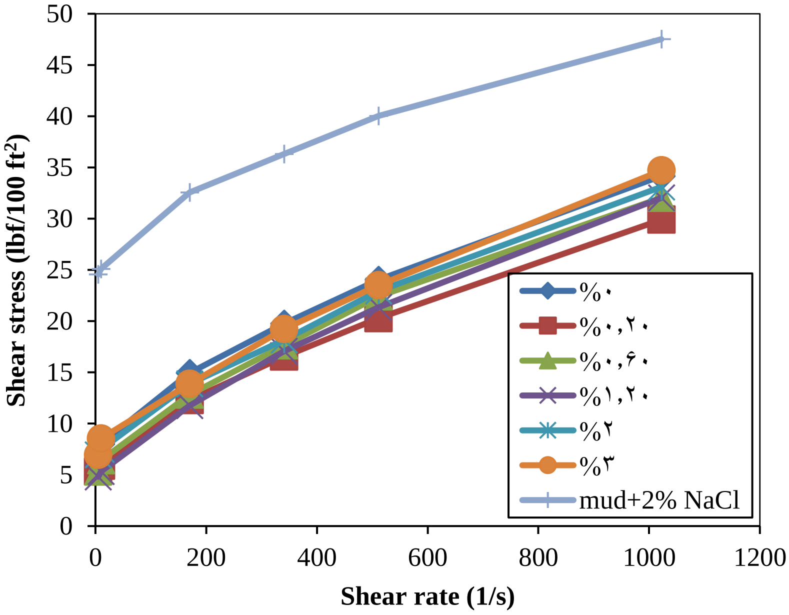


Figure 6

Rheological behavior of bentonite mud containing different concentration of Black Myrobalan.

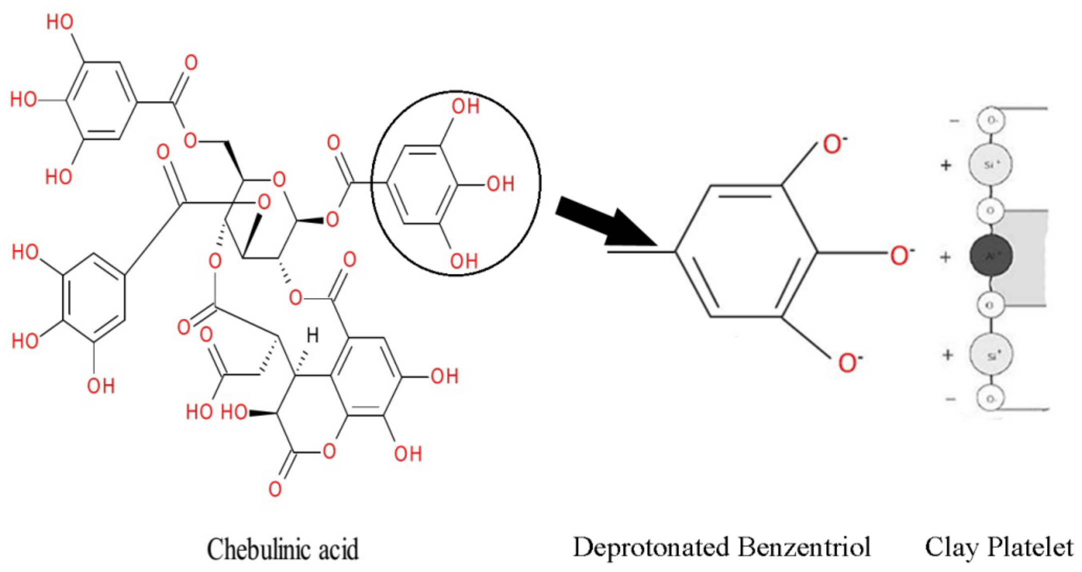
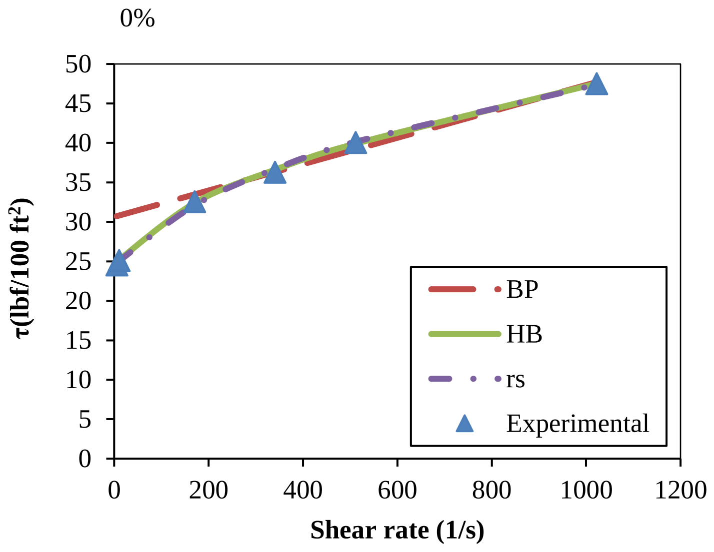


Figure 7
Mechanism of clay deflocculation by Black Myrobalan.

Figure 8 displays the effect of different concentrations of black myrobalan on fitting the rheograms of the experimental data with different rheological models. As illustrated in this figure, Herschel-Bulkely and Robertson-Stiff models are more accurate than Bingham plastic in describing bentonite dispersion including black myrobalan. By increasing black myrobalan extract to 0.6 wt.% rheograms tend to match Herschel-Bulkely model with a lower yield stress and higher slope compared to blank mud.

Parameters resulted from the regression analysis of different rheological models are given in Table 4.



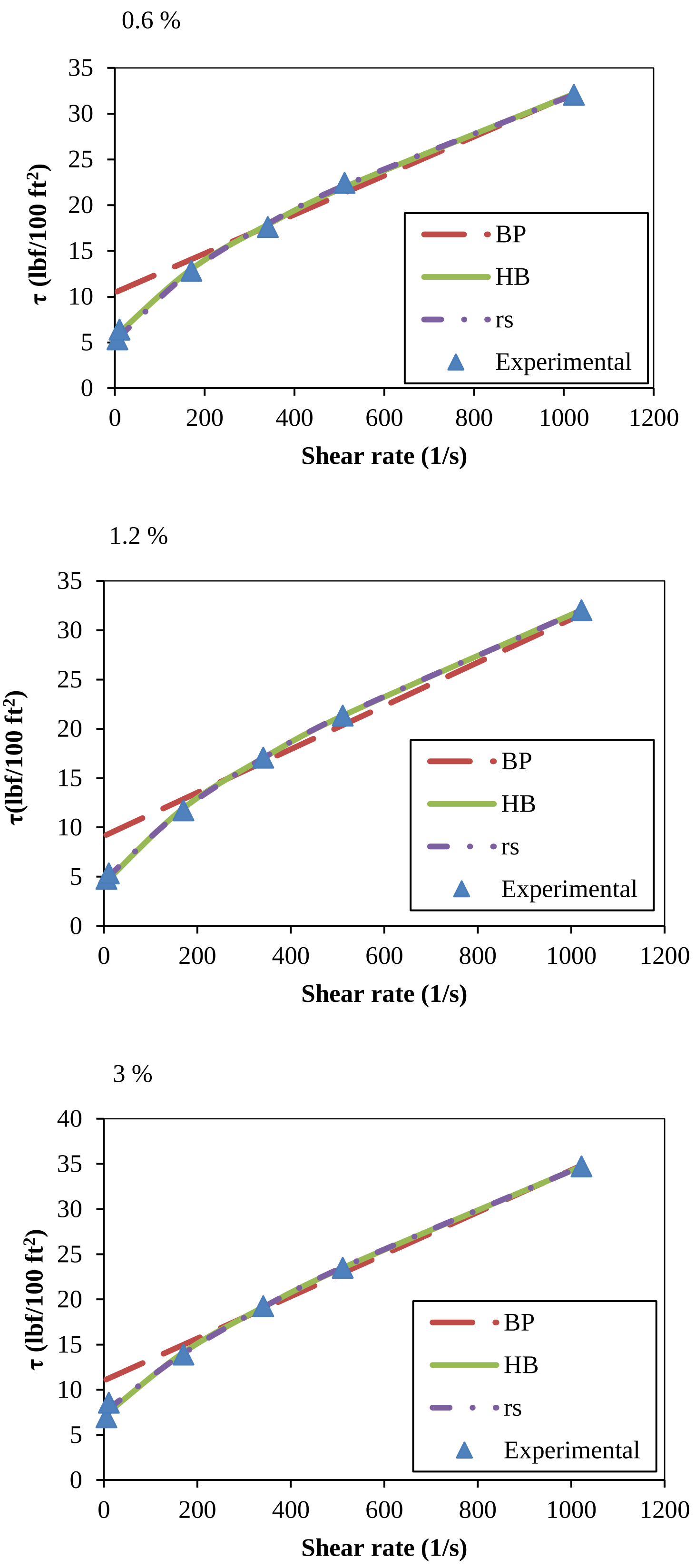


Figure 8
Fitting experimental data to different rheological models for different concentrations of Black Myrobalan extract.

Table 4

Parameters of rheological models for bentonite mud containing different concentrations of black myrobalan (HB: Herschel-Bulkely, RS: Robertson-Stiff, BP: Bingham plastic).

Black myrobalan concentration (wt.%)	**Rheological model**	**Parameters**	R^2	**RMSE**
0	HB	$K=0.5749$ $n=0.5408$ $\tau_y=23.14$	0.9999	1.389
	RS	$A=7.025$ $C=96.8$ $B=0.2719$	0.9989	0.3793
	BP	$\tau_0=30.63$ $\mu_p=0.0167$	0.9827	16.54
0.2	HB	$K=0.1808$ $n=0.7092$ $\tau_y=5.253$	0.9999	1.306
	RS	$A=0.3859$ $C=86.51$ $B=0.6204$	0.9998	2.705
	BP	$\tau_0=9.577$ $\mu_p=0.02$	0.9946	10.99
0.6	HB	$K=2584$ $n=0.6727$ $\tau_y=4.817$	0.999	0.4146
	BP	$\tau_0=10.45$ $\mu_p=0.0213$	0.9879	17.6
	RS	$A=0.6368$ $C=37.7$ $B=0.5626$	0.9991	5.435
1.2	HB	$K=0.2581$ $n=0.6791$ $\tau_y=3.492$	0.9999	2.131
	BP	$\tau_0=9.126$ $\mu_p=0.022$	0.9929	14.31
	RS	$A=0.4231$ $C=44.29$ $B=0.6206$	1	1.14
2	HB	$K=0.1813$ $n=0.7206$ $\tau_y=6.426$	0.9995	0.3002
	BP	$\tau_0=10.93$ $\mu_p=0.0218$	0.993	13.67
	RS	$A=0.4968$ $C=79.42$ $B=0.5995$	0.9996	0.3313
3	HB	$K=0.175$ $n=0.7333$ $\tau_y=6.539$	0.9998	2.786
	BP	$\tau_0=11.01$ $\mu_p=0.0233$	0.9953	12.02
	RS	$A=0.3903$ $C=100.6$ $B=0.639$	0.9993	0.3313

AV reveals the ability of drilling fluids to flow and is related to the rate of penetration. PV is caused by the friction between the suspended particles and influenced by the viscosity of the base liquid (Meng et al., 2012).

Figure 9 shows the effect of black myrobalan on the rheological parameters. As it is shown in Figure 9, by increasing black myrobalan concentration to 0.6 wt.%, AV decreases from 22.5 cP to 14 cP, and it then increases to 16.25 cP at 3 wt.%. Plastic viscosity increases slightly from 7 cP to 10.5 cP at 3 wt.%. Yield point decreases from 13 lb_f/100 ft^2 to 9 lb_f/100 ft^2 at 0.6 wt.%, and it increases again to 12 lbf/100 ft^2 at 2 wt.%; lastly, it decreases slightly to 11.5 lb_f/100 ft^2 at 3 wt.%. LSRV varies with the same trend as yield point and decreases from 22.5 cP to 4 cP at 1.2%; it increases to 6 lbf/100 ft^2 at 2% with slightly decreasing to 5 lbf/100 ft^2 at 3%.

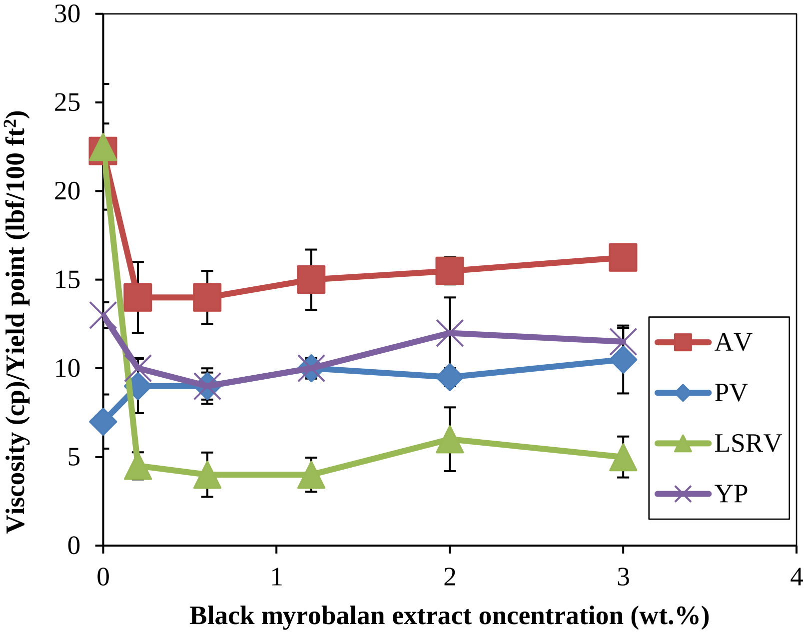


Figure 9

Rheological parameters of bentonite mud containing different concentrations of black myrobalan.

The gel strength is a measurement of the shear stress necessary to initiate the flow of a fluid which has been quiescent for a period of time (Annis & Smith, 1996). Thixotropy of the mud is the difference between the low readings after 10 sec and 10 min (El-Sukkary et al., 2013). The effect of black myrobalan on 10 sec and 10 min gel strength and the thixotropy of bentonite mud are illustrated in Figure 10. As can be seen, by increasing black myrobalan concentration to 1.2 wt.%, 10 min gel strength decreases by 44%, and 10 sec gel strength decreases by 70%, while thixotropy increases by 83%; however, their variation at higher concentrations of black myrobalan are trivial. According to Figures 9 and 10, it can be understood that increasing black myrobalan concentration to 0.6 wt.% decreases the rheological parameters of flocculated bentonite mud to minimum but increases them at higher concentrations. On the other hand, black myrobalan acts as a deflocculant in the range of 0.2 wt.% to 1.2 wt.%.

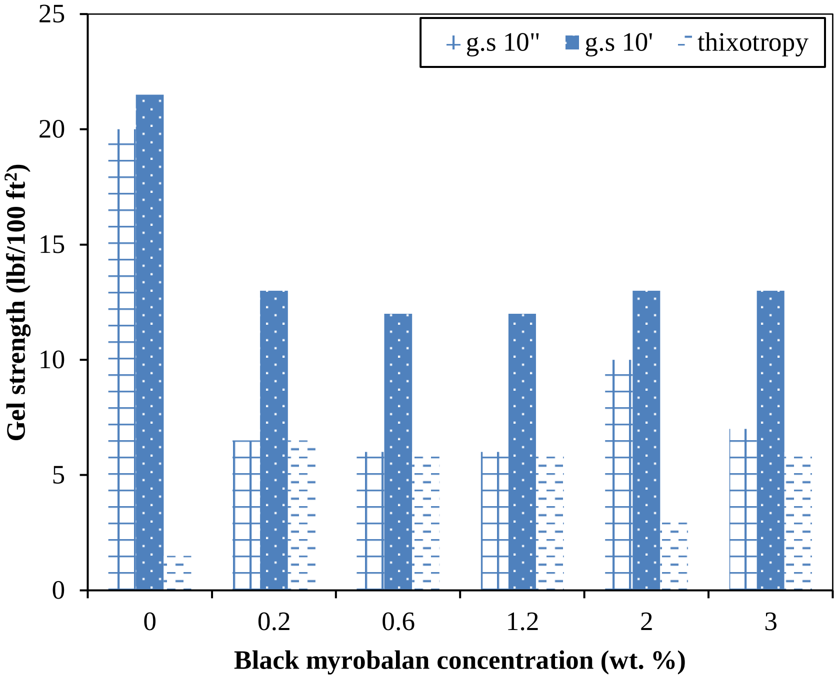


Figure 10
Variation of gel strength and thixotropy of bentonite mud containing different concentrations of black myrobalan.

The effect of black myrobalan on the rheological parameters of flocculated mud is compared with CLS in Figure 11. As can be observed, bentonite muds including 0.2 wt.% to 0.4 wt.% black myrobalan were found to have rheological parameters lower than the mud with the same content of CLS.

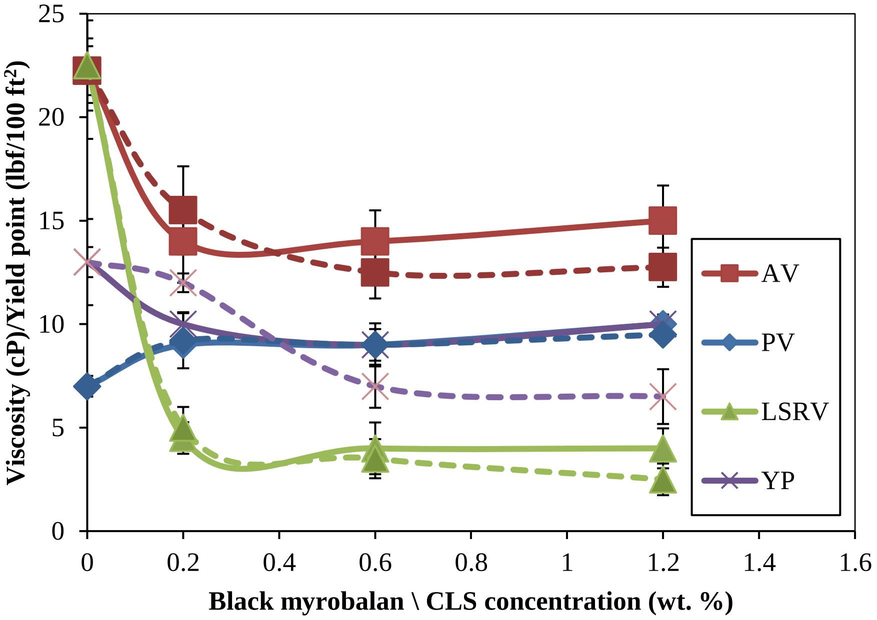


Figure 11
Comparison of rheological parameters of bentonite suspension comprising black myrobalan and CLS.

The gel strength of mud containing black myrobalan is compared with CLS in Figure 12. It is clear that gel strength and thixotropy values for CLS and black myrobalan are almost identical for a given concentration. The summary of the variation of rheological parameters of bentonite mud is given in Table 5.

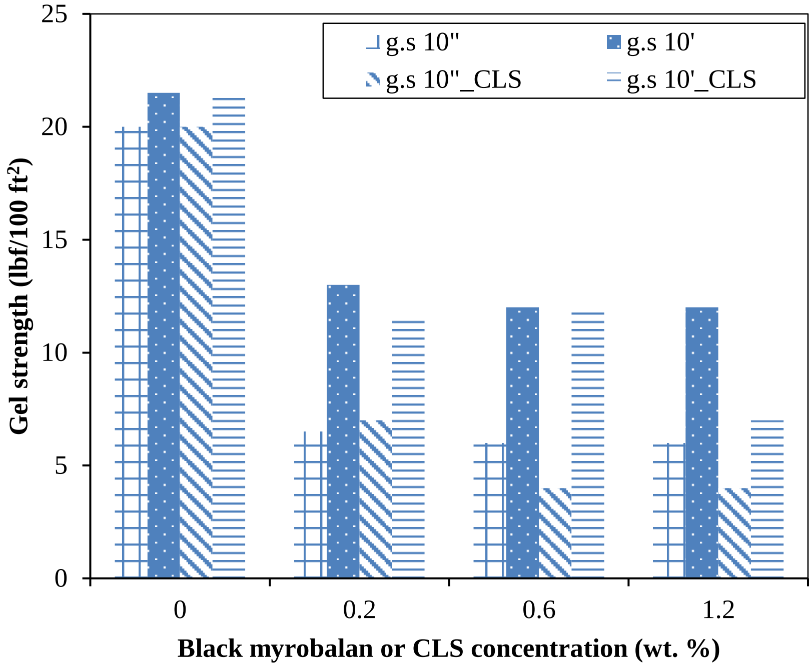


Figure 12
Comparison of gel strength values of bentonite mud containing different concentrations of black myrobalan by CLS.

Table 5
Rheological parameters data for bentonite mud containing different concentrations of black myrobalan or chrome lignosulfonate.

Concentration (wt.%)	AV(cP)		PV (cP)		G.S. 10” (lb$_f$/100 ft^2)		G.S. 10’(lb$_f$/100 ft^2)		YP (lb$_f$/100 ft^2)	
	B	C	B	C	B	C	B	C	B	C
0	22.25	22.25	7	7	20	20	21.5	21.5	13	13
0.2	14	15.5	9	9.2	6.5	7	13	11.5	10	12
0.6	14	12.5	9	9	6	4	12	11.5	9	7
1.2	15	12.75	10	9.5	6	4	12	7	10	6.5

*B: black myrobalan and C: Chrome Lignosulfonate.

Parameters of Herschel-Bulkely model for bentonite mud including black myrobalan and CLS are represented in Figure 13. As indicated in Figure 13-a, increasing black myrobalan and CLS concentration to 3 wt.% causes yield stress to decrease from 23.14 lb$_f$/100 ft^2 to 3.5 lb$_f$/100 ft^2 and 2.85 lb$_f$/100ft^2 respectively, and the value of yield stress for CLS is somewhat higher. According to Figure13-b, increasing black myrobalan or CLS concentration causes flow behavior index (*n*) to increase but decreases consistency index (*K*); however, they become steady at concentrations more than 0.6 wt.%. It is also clear that the values of K and n of black myrobalan are respectively lower and higher than those of CLS at concentrations lower than 0.5 wt.%, which shows that the degree of deflocculation for the mud containing black myrobalan is higher than the one having CLS.

RYP shows the capability of drilling fluids to carry the cuttings (Meng et al., 2012). A high RYP indicates that it is a shear thinning mud which is desirable for suspending the cuttings when circulation is stopped, and it breaks up quickly to a thin fluid when it is agitated by the continuation of drilling (Mahto and Jain, 2013).

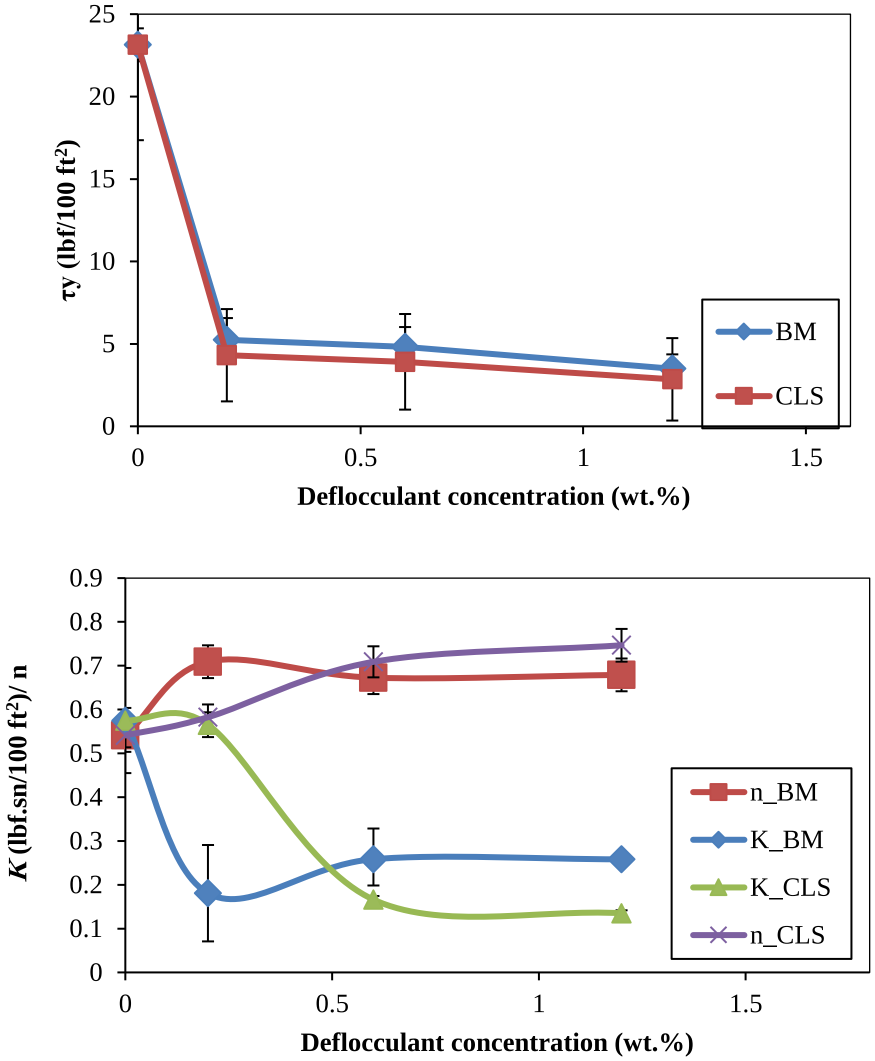


Figure 13

Parameters of Herschel-Bulkely model for bentonite mud including different concentrations of black myrobalan and CLS. (upper) yield stress (lower) consistency index and flow behavior index.

Figure 14 displays the variation of the RYP of bentonite mud against black myrobalan and CLS concentration. As their concentrations increase to 1.2%, RYP decreases gradually from 1.85lb$_f$/100 ft^2.cP to 1lb$_f$/100 ft^2.cP and 0.68 lb$_f$/100 ft^2.cP respectively; thus the drilling fluid capacity of bentonite mud to carry drilling cuttings is reduced, while the value of RYP of black myrobalan is higher than that of CLS, which is an indication of higher capacity of bentonite containing black myrobalan to carry the cuttings.

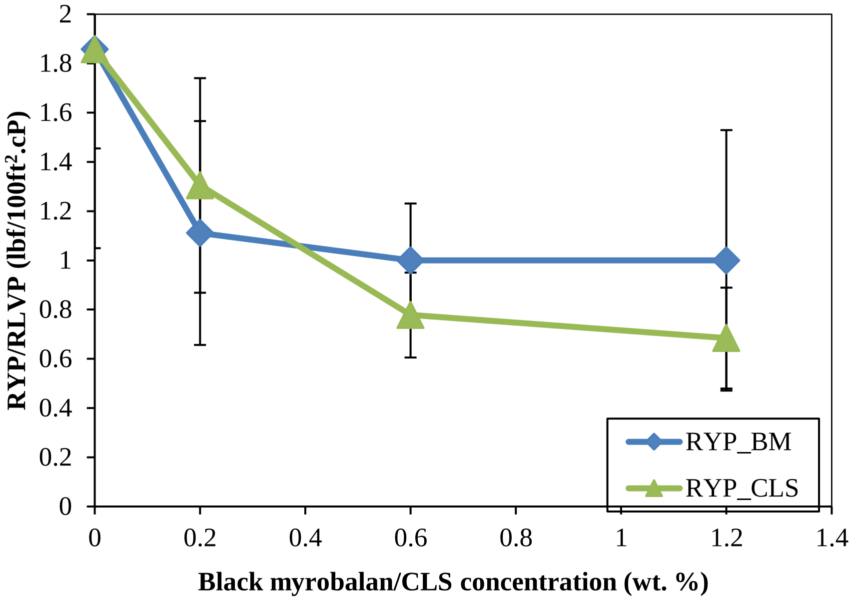


Figure 14
Effect of black myrobalan and CLS on RYP and RLVP of bentonite mud.

b. Low shear rates

Apparent viscosity of bentonite mud containing different concentrations of black myrobalan extract is shown in Figure 15. By increasing black myrobalan concentration to 3 wt.%, apparent viscosity decreases. The flow curve of bentonite mud comprising different concentrations of black myrobalan at low shear rates is also illustrated in Figure 16. It is obvious that increasing black myrobalan concentration to 0.2 wt.% causes shear rate to decrease suddenly, and by the further addition of black myrobalan shear stress changes insignificantly.

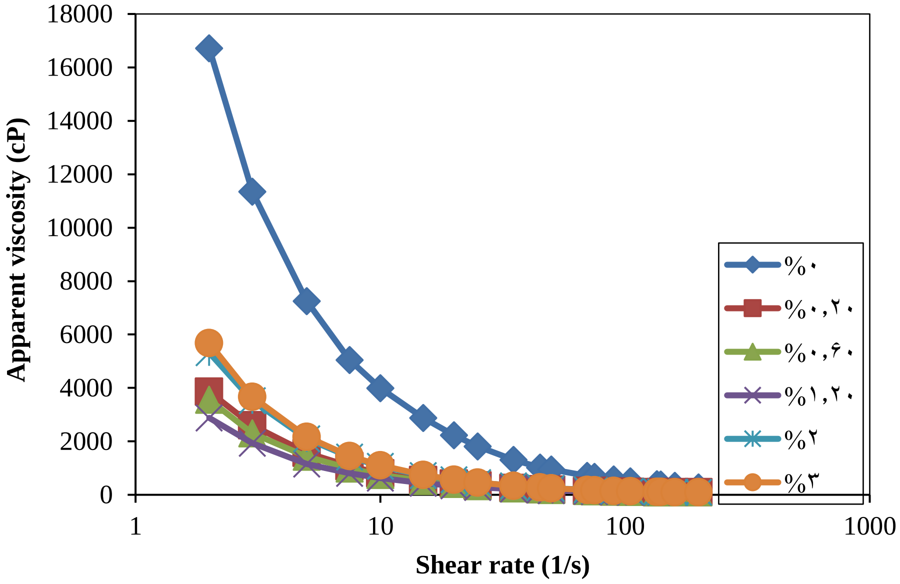


Figure 15
Apparent viscosity of bentonite suspension containing different concentrations of black myrobalan extract.

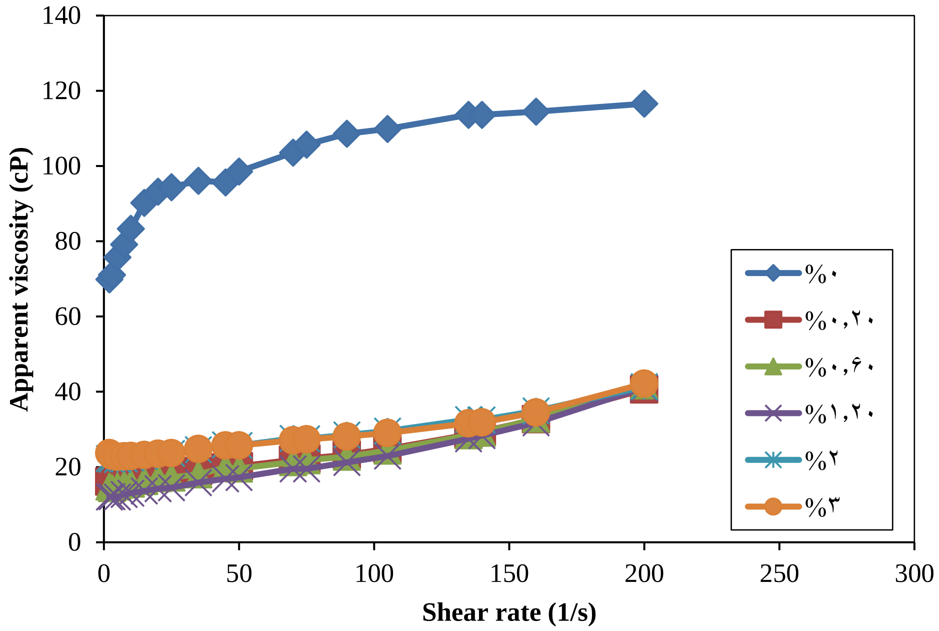


Figure 16
Rheological behavior of bentonite suspension comprising different concentrations of black myrobalan extract at low shear rates.

c. Filtration characteristics of bentonite mud

The profile of filtration volume of bentonite mud containing different concentrations of black myrobalan against time is shown in Figure 17. As expected, filtration volume is proportional to square root of time; by increasing black myrobalan concentration to 0.2 wt.%, filtration profile drops meaningfully and becomes unchanged at higher concentrations.

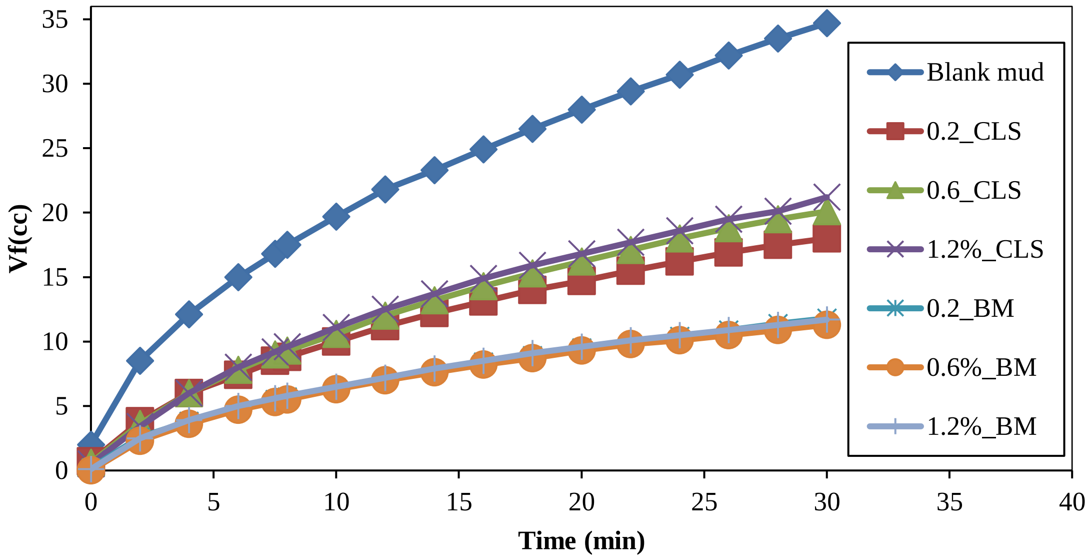


Figure 17
Comparing filtration profile of bentonite mud containing black myrobalan by CLS.

The filtration profile of mud containing black myrobalan is compared with CLS in Figure 18. The results show that mud containing black myrobalan has lower filtration than the one having CLS, which indicates that, in bentonite mud containing black myrobalan, clay platelets are dispersed highly and decrease the filtration volume more than CLS.

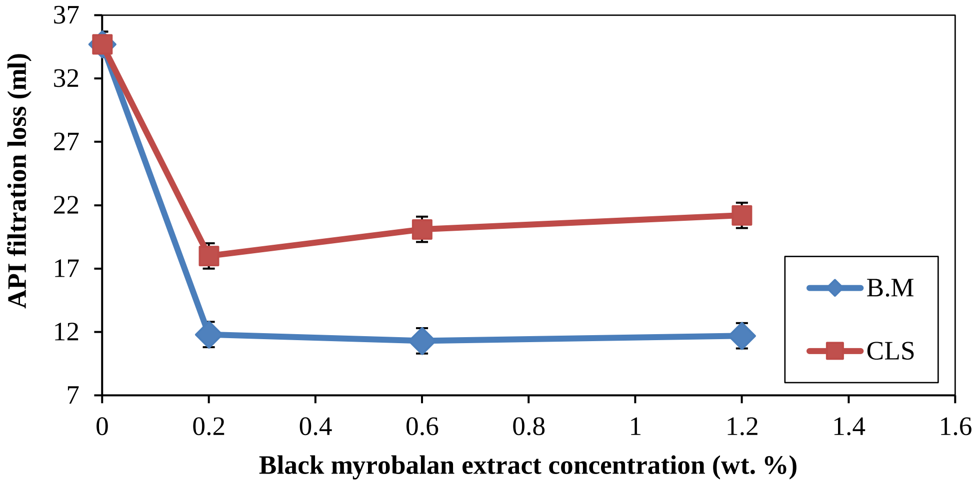


Figure 18
Filtration loss of bentonite dispersion comprising with different concentrations of black myrobalan/CLS.

4.2. Effect of black myrobalan on pH of bentonite mud

Figure 19 illustrates the pH of black myrobalan solution and bentonite dispersion at different concentrations of black myrobalan. As can be seen, the pH of bentonite dispersion decreases from 9 to 6.7 by increasing black myrobalan concentration to 3 wt.%. The pH of black myrobalan solution decreases to 4.4 at 1.2 wt.% and becomes steady for higher concentrations. The acidic pH of black myrobalan solution is due to the presence of components such as tannic acid, gallic acid, ellagic acid, etc. in its composition.

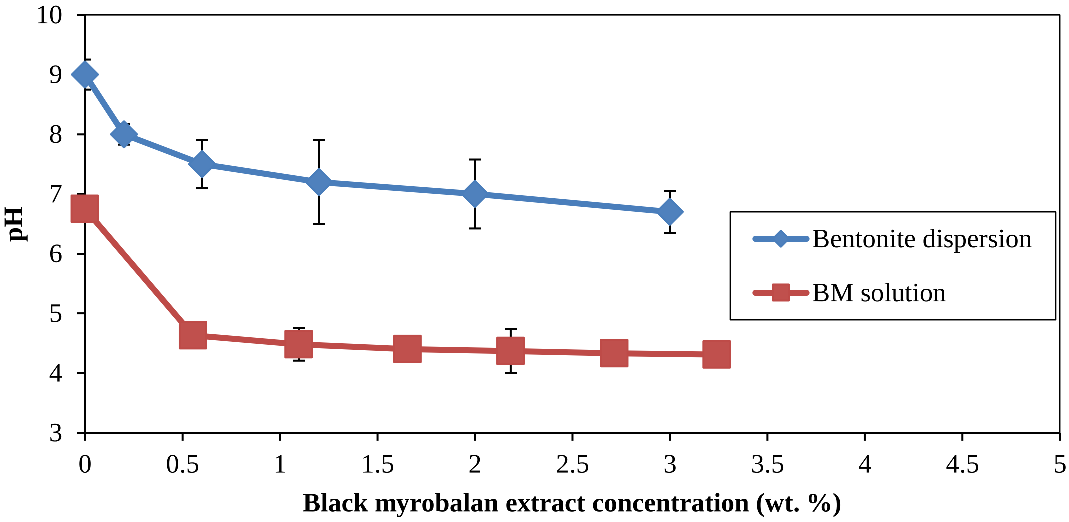


Figure 19
Variation of pH of bentonite dispersion by addition of black myrobalan

4.3. Effect of black myrobalan on the electrical conductivity of bentonite dispersion

Figure 20 demonstrates the electrical conductivity of bentonite dispersion containing different concentrations of black myrobalan. As shown, increasing black myrobalan concentration to 1.2 wt.% causes conductivity to increase to 319 µs/m, and it then becomes stable near this value for higher concentrations.

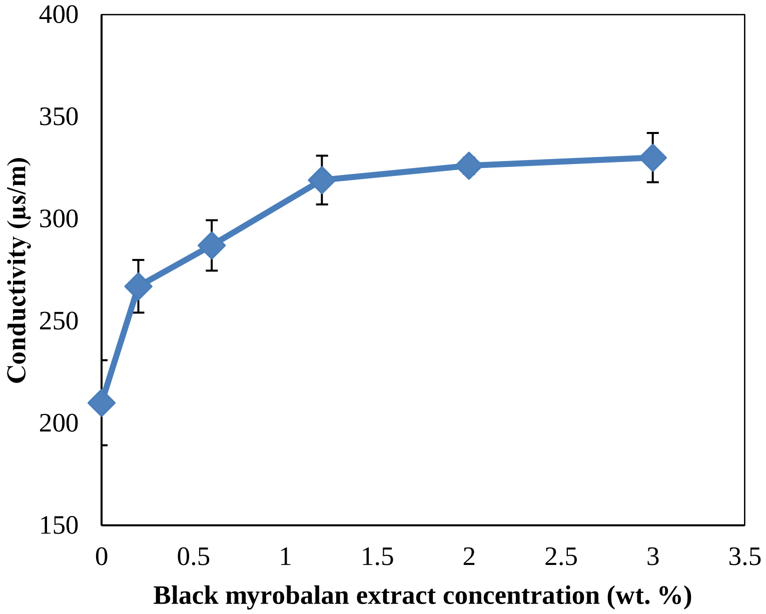


Figure 20
Effect of black myrobalan on electrical conductivity of bentonite dispersion.

5. Economic and environmental justification of utilization of black myrobalan

In present time, countries like India and China are the main sources of black myrobalan (Terminalia Chebula) and export it to Iran with an average cost of $2.6 per kilogram, while purchasing chrome lignosulfonate costs $5.5-6 per kilogram. According to geographical condition and climate of Iran, local planting of the black myrobalan rather than importing causes black myrobalan to be more available than chrome lignosulfonate. Other advantages such as non-toxicity, high solubility in water, and anti-corrosion characteristics suggest it for a potential replacement for chrome lignosulfonate. This extract is totally biodegradable and does not have any environmental problems, which makes it applicable in offshore drilling.

6. Conclusions

1. The addition of NaCl to bentonite mud enhances shear stress in the rheogram of mud significantly, which is due to the flocculation of clay platelets.
2. Herschel-Bulkely and Robertson-Stiff models predict the rheology of bentonite mud comprising black myrobalan with high accuracy.
3. The addition of black myrobalan to flocculated bentonite mud up to 0.6 wt.% decreases the AV; 10 sec and 10 min gel strength; LSRV; and RYP, but it increases PV.
4. Rheological parameters of mud comprising black myrobalan are lower than those of the mud having CLS at the same concentration up to 0.4 wt.%. Values of 10 sec and 10 min gel strength are the same for both black myrobalan and CLS in this range of concentration. It

could be realized that the deflocculation effect of black myrobalan is due to the neutralization of positive charges on the edges of clay platelets by deprotonated hydroxyl groups. Since the edge surface area of clay platelet is relatively a small proportion of the total area, this effects occurs at low concentrations of black myrobalan.

5. The gel strengths of mud for both black myrobalan and CLS are almost identical at the same concentration
6. The addition of black myrobalan and CLS to flocculated bentonite mud caused consistency index to decrease, but increased flow behavior index, which is an indication of the deflocculation of mud. At concentrations lower than 0.5 wt.%, consistency index and flow index of the mud having black myrobalan are lower and higher respectively compared to those of the mud having CLS. They also decrease the yield stress of Herschel-Bulkely model.
7. Increasing black myrobalan concentration to 0.6 wt.% decreases the apparent viscosity of mud at low shear rates; however, its effect at higher concentrations is insignificant.
8. Aging mud including black myrobalan for 24 hours caused the apparent viscosity and shear stress of bentonite mud to unexpectedly decrease.
9. Black myrobalan decreases the filtration volume of flocculated mud at concentrations lower than 1.2 wt.%, which is because of the deflocculation of clay platelets and plugging the pores by these platelets. Comparing the filtration of black myrobalan with that of CLS showed that black myrobalan decreases the filtration volume of mud more than CLS at the same concentration.
10. A comprehensive study of the performance of black myrobalan showed that black myrobalan is a more efficient deflocculant compared to CLS at concentrations less than 0.4 wt.%.
11. The addition of black myrobalan increases electrical conductivity up to 1.2 wt.%, and it then levels off.
12. The addition of black myrobalan decreases the pH of mud due to the presence of components such as tannic acid, gallic acid, ellagitannic acid, etc.
13. CLS is harmful to the environment because of the existence of chrome in its composition and is prohibited by environmental protection organizations, while black myrobalan is a plant-based and nontoxic material, which makes it a potential replacement for CLS.

Acknowledgments

The authors would like to thanks the laboratory technicians of Petroleum University of Technology for providing equipment and materials.

Nomenclature

AV	: Average viscosity
BM	: Black myrobalan
CLS	: Chrome lignosulfonate
G.S.	: Gel strength
K	: Consistency index [lbf.sn/100ft^2]
LSRV	: Low shear rate viscosity
PV	: Plastic viscosity
RLVP	: Ratio of low shear rate viscosity and plastic viscosity

RYP : Ratio of yield point and plastic viscosity

SR : Shear rate

SS : Shear stress

V_f : Filtration volume [cc]

YP : Yield point

YPL : Yield Power law

Greeks

$\dot{\gamma}$: Shear rate [s^{-1}]

μ_p : Plastic Viscosity [cP]

τ : Shear stress [lbf/100 ft^2]

τ_0 : Minimum shear stress [lbf/100ft^2]

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