



# Ultrasonic oscillations induced property development of water-bentonite suspension containing sulfonated wood coal

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## Abstract

During drilling fluid preparation, ultrasonic oscillations were introduced into water-bentonite suspension incorporating sulfonated wood coal (SMC) by a specially designed device. The influences of ultrasonic oscillations on fluid loss and rheological performances of the drilling fluid as well as mechanism of ultrasonic action were investigated. The experimental results showed that the filtrate volume decreased with the increase of ultrasonic time till a certain extent and then leveled off. In the presence of ultrasound, shorter time of 15 min and mild intensity of 250 W could lead to a satisfactory result in fluid loss properties, including the reasonable filtrate volume and thin and compact filter cakes. With increasing ultrasonic intensity, the fluid loss properties changed relatively little but various rheological data of the drilling fluids always increased. Adsorption tests through total organic carbon, infrared spectrum and thermogravimetric analyses as well as clay particle size analysis confirmed that as compared with the conventional agitation, ultrasound-assisted mud preparation could not only increase adsorbed amount of SMC on bentonite but also decrease average clay particle diameter attributed to acoustic cavitation. A plausible mechanism based on sonochemical thermodynamics is proposed to explain the improvement of the colloidal structure and performances of drilling fluid.

**Keywords** Ultrasound · Bentonite · Water-based drilling fluid · Filtration · Adsorption · Dispersion

## Introduction

As a clean and efficient oscillation technology, high-intensity ultrasound has been universally applied in these areas of cleaning, plastic welding, machining, biology, medicine, material and chemistry (Guo et al. 2009). Of all the applications, sonochemistry captures the most attention of scientists and engineers in recent years (Chatel et al. 2016). Cavitation (Guo and Peng 2007) includes the birth, growth and drastic breakdown of gas micro-bubbles in solutions as a consequence of pressure changes with ultrasonic waves passing through the liquid medium. It can repeatedly create frictional force, shock wave and some hot spots with locally high temperature (up to 5000 K) and high pressure (up to 100 MPa), thus causing the concentrated release of much of energy at short intervals of time (Didenko et al. 2000). The acoustic cavitation in liquids is responsible for nearly

all the phenomena of sonochemistry, for example, synthetic chemistry, electrochemistry, increased reaction kinetic, degradation, emulsification effect, precipitation, extraction, etc. (Chatel et al. 2016).

Clays with abundant resources on the earth include some layered minerals such as bentonite, illite, chlorite, vermiculite, palygorskite and sepiolite, etc. They are environmentally friendly and widely used in different fields according to various molecular structures and properties. Bentonite is mainly comprised of smectite type clay (Darvishi and Morsali 2011), whose chemical formula is typically defined as  $(Al_{3.34}Mg_{0.66})(Si_{7.0}Al_{1.0})O_{20}(OH)_4$ . The layered structure is two tetrahedral sheets of  $Si^{4+}$  oxides sandwiching an octahedral sheet of  $Al^{3+}$  oxides. The layers relative to each other are likely to slip, thus causing the further swelling and dispersion of the bentonite particles (Chatel et al. 2016). Heterovalent substitution of metal atoms in the tetrahedral and octahedral sheets endows a negative charge to the crystal lattice of clays (Anderson et al. 2010). The charge can be balanced by interlayer exchangeable cations such as  $Na^+$ . Further, the simple inorganic cations may trade with other more complex organic cations. In addition to these

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characteristics, bentonite possesses the abundant active reaction points (silanol, hydroxyl, etc.) on the surface (Novikova et al. 2014). These provide a possibility of uses of bentonite suspension in conjunction with ultrasonic oscillations. The relative research works mainly focus on following categories (Chatel et al. 2016):

- (1) More than half of the uses are concentrated on the ultrasonic development of colloidal solution stability, including the increase of zeta potential and the reduction of bentonite particles size in suspension (Darvishi and Morsali 2011; Mekhamer 2010; Nguyen et al. 2013). These belong to theoretical research.
- (2) Ultrasonic oscillations can induce the intercalation of cationic, anionic or nonionic surface active agents into montmorillonites (Chaudhary and Liu 2013; Onder et al. 2013; Xi et al. 2005; Zhang et al. 2010). The resulting materials are very promising in the two areas of preparation of polymer nanocomposites and treatment of wastewater.
- (3) In the presence of ultrasonic oscillations, some contaminant adsorption from the liquor to montmorillonite is obviously increased (Ealias and Saravanakumar 2019; Taghizadeh and Seifi-Aghjekohal 2015).
- (4) Natural and modified montmorillonites are high performance catalysts for some organic reactions. Ultrasonic oscillations can provoke the better accessibility of the reactants to catalysts and consequently reduce the reaction time and elevate the yield (Dar et al. 2013; Mohammadpoor-Baltork et al. 2010).

It is well known that water-bentonite suspension can also be used as the base mud of drilling fluid, due their excellent colloidal performances. A primary function of the water-based drilling fluid is to creating the thin and impermeable mud cake on well walls for preventing the loss of water from drilling fluids into geological formations. The increase of filtrate volume may produce a series of drilling accidents such as shrink of the borehole dimension, excessive torque and formation collapse. To avoid this, filtrate reducers consisting mainly of some modified natural products and synthetic organic substances have been prepared and applied in the water-bentonite suspension. There is a general consensus among researchers with regard to mechanism of action of the filtrate reducers. It has been described previously by us (Guo and Peng 2012; Peng et al. 2010) that the filtrate reducers are firstly adsorbed on the bentonite surface by physical or chemical linkage manners. As a result, the negative charge density and hydration membrane thickness of clay particles get increased, along with the strengthening of particle multiplex dispersion and colloidal structure stability by virtue of electrostatic repulsion between clay particles. Finally, the solid composition of drilling fluids is precipitated to form

highquality filter cakes minimizing fluid loss. Developing high performance filtrate reducers has been the subject of considerable research activity (Ahmad et al. 2018a, b; Chang et al. 2019; Chu and Lin 2019; Li et al. 2015). In addition to abovementioned traditional filtrate reducers, many kinds of inorganic nanoparticles and hybrid (inorganic–organic) nanocomposite materials have been recently applied to improve filtration performances (Aftab et al. 2017; Rafati et al. 2018). However, the further occurrence of new additives for optimizing filtration properties will become more and more difficult, because a limitation to practical implementation is the lack of adequate chemical preparation methods. So, it is worthy of be expected that the ultrasound-induced adsorption and dispersion of bentonite technology, combined with existing filtrate reducers, provides an innovative route to enhance fluid loss properties of water-based drilling fluids. So far, no correlative work has been reported.

Sulfonated wood coal (SMC) is commonly synthesized from the reaction of wood coal, formaldehyde and sulfonating agent (e.g., sodium bisulphite, sodium sulphite and sodium pyrosulphite) at alkaline medium. SMC can be resistant to the action of elevated temperature in excess of 180 °C but no tolerance to salt (Kelessidis et al. 2007). As a conventional filtrate reducer with a certain rheology-adjusting function, it has been successfully and widely used in deep well drilling as early as the 1970s. Humic acid is the major constituent of wood coal. It belongs to the natural admixture of various polycyclic aromatic hydrocarbons with active functional groups of benzene ring, carboxy and hydroxyl (Kelessidis et al. 2009). Researchers have attributed the filtration control behavior of SMC primarily to watersoluble humic acid and its derivatives (Kelessidis et al. 2007). In the article, ultrasound was introduced into water-bentonite suspension incorporating SMC by a specially designed device in our lab. The influences of ultrasonic oscillations on fluid loss and rheological performances of the drilling fluid as well as mechanism of ultrasonic action were investigated. The objectives of the article are to explore the possibility of filtration property development using ultrasonic oscillations during mud preparation and to provide the theoretical guidance for field use of water-based drilling fluids with the assist of ultrasonic technique.

## Experimental

### Materials and equipment

Na<sub>2</sub>CO<sub>3</sub> was chemically pure and purchased from Kelong Reagent Company of China. Sodium bentonite and SMC were commercial grades supplied by Renzhi Oilfield Company of China. SMC was used with further purification to remove insoluble carbon residue.

The sample preparation was performed in a self-designed device described in Fig. 1. An ultrasonic probe with a diameter of 1.5 cm was vertically inserted into a cuboid vessel, and gap between the probe tip and the centre of vessel bottom was kept at 2 cm. The constant frequency and variable energy output of the probe were 20 kHz and 200 to 1000 W, supplied by the piezoelectric transducer. For continuous running at room temperature, a valid cooling unit was equipped on the ultrasonic device.

### SMC purification

The purification procedure involved dissolving in pure water, filtrating to remove insoluble residue and freeze drying of the filtrate.

### Sample preparation

According to the Chinese standard of SY/T 5092-2017, a bentonite stock suspension was prepared through dispersing 21 g of bentonite and 0.74 g of  $\text{Na}_2\text{CO}_3$  into 350 mL of pure water and agitating for 15 min at high speed. The resulting bentonite suspension was placed for 24 h at room temperature before use. The drilling fluid was obtained with an addition of 10.5 g of refined SMC into the bentonite suspension and stirring for 15 min at high speed.

Subsequently, the drilling fluids were entirely fed into the ultrasonic reactor. After ultrasonic oscillations of a certain

time and power, the drilling fluids were removed to take some measurements.

### Thermal aging experiment

The aging test of drilling fluids sealed in pressure jars was conducted in a BGRL-7 roller oven (Tongchun Company of China) by the normal 16 h thermal rolling at 180 °C.

### Filtration property test

The filtration testing was performed using the SD6A medium-pressure filter press equipped with filter paper (Tongchun Company of China) in accordance with typical American Petroleum Institute (API) specifications. After the drilling fluids were loading into the filter press cup, the filtration test started run at room temperature and pressure difference of 0.69 MPa. The filtrate was collected in a graduated flask, and total filtrate volume within 30 min was denoted as  $FL_{API}$ . The reported data were the average value taken from four tests. After each experiment, the residual fluid in filter press cup was taken away to gain intact filter cakes for the coming analyses.

### Permeability of filter cake and scanning electron microscope (SEM) analysis

According to Darcy's Law, permeability ( $K$ ) of fresh filter cakes could be calculated by the following equation:

$$K = \frac{\mu L dV}{PA dt} = \frac{\mu L Q}{PA} \quad (1)$$

where  $\mu$  is the viscosity of filtrate obtained through Ostwald viscosimeter at room temperature (mPa.s);  $L$  and  $A$  are the thickness (cm) and cross section area ( $45.8 \text{ cm}^2$ ) of filter cakes, respectively;  $P$  is the drop of pressure across filter cakes (0.69 MPa); and  $Q = (dV/dt)$  is the filtration speed (mL/s), which is obtained using the method of water flowing through already formed filter cake (Li et al. 2015).

Surface features of dry filter cakes were observed with the SU8010 SEM (Hitachi Company of Japan). Before observation, the surfaces were coated with gold sputter.

### Rheological property test

Rheological data of the drilling fluids were acquired through using a ZNN-SD12 rotational viscosimeter (Tongchun Company of China) at room temperature. The speeds of rotation were 3, 6, 100, 200, 300, 600 rpm corresponding to the shearing rates ( $\dot{\gamma}$ ) of 5.11, 10.22, 170.3, 340.7, 511, 1022  $\text{s}^{-1}$ , respectively. The shearing stress ( $\tau$ ) and apparent viscosity ( $\eta$ ) were calculated as follows:

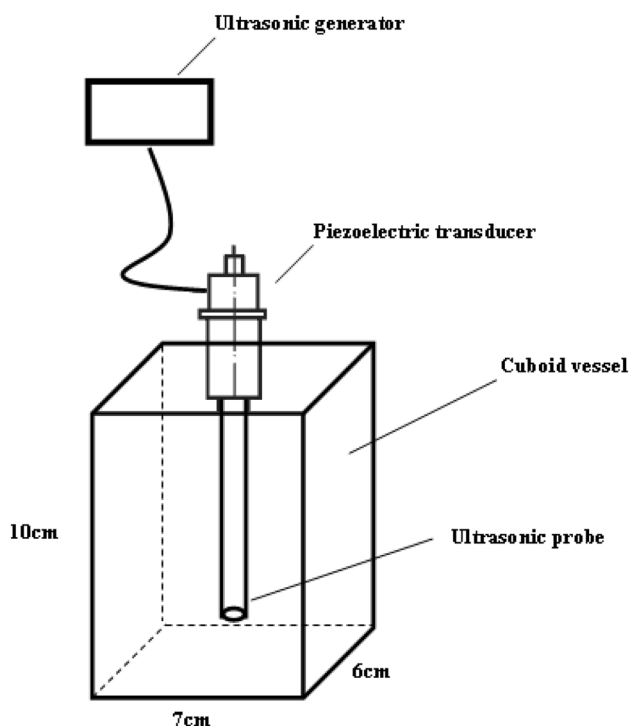


Fig. 1 Schematic diagram of ultrasonic reactor

$$\tau = 0.511\theta \text{ (Pa)} \quad (2)$$

$$\eta = \frac{\tau}{\dot{\gamma}} \times 10^3 \text{ (mPa s)} \quad (3)$$

where  $\theta$  is the reading of instrument panel at a designated speed of rotation. The reported data were the mean value from four tests.

### Adsorption measurement

The adsorption quantity was calculated according to the difference between SMC concentrations in solutions before and after adsorption equilibrium. To test the initial concentration of SMC before adsorption equilibrium, an accurately weighed amount (10.5 g) of refined SMC was mixed with a known volume (350 mL) of pure water, where the solution concentration was circa 2.9%. The accurate concentrations of SMC in the solution and the filtrate from  $FL_{API}$  tests were finally determined with TOC-L type total organic carbon (TOC) analyzer (Shimadzu Company of Japan).

The dry filter cake was again dispersed in pure water and then centrifugated to eliminate unadsorbed SMC. The process was repeated four times. The final sediments were dried to constant weight under the vacuum condition of 80 °C and later used for the test of Fourier transform infrared spectrum (FTIR) and thermogravimetric analysis (TGA). FTIR measurements were performed on 170X FTIR spectrometer (Nicolet Company of USA) with 32 scans and the resolution of 4  $\text{cm}^{-1}$ . Before measurement, the samples were prepared with KBr through compression. TGA was performed by using TG209 thermal analysis instrument (Netzsch Company of Germany) from room temperature to 700 °C in a flowing  $\text{N}_2$  atmosphere. The rate of temperature rise was 20 °C/min.

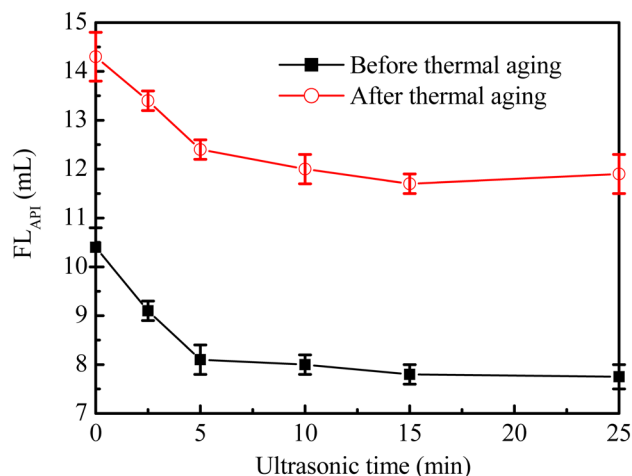
### Particle size measurement

The size of bentonite particle in drilling fluid was determined using a JL-1166 laser particle analyzer (Jingxin Company of China).

## Results and discussion

### Fluid loss

Figure 2 illuminates the effect of ultrasonic time on the  $FL_{API}$  of the drilling fluids at fixed oscillation intensity of 500 W, to decide the threshold time for the sonication treatment. It can be discovered that the  $FL_{API}$  values before and after thermal aging depend strongly on ultrasonic reaction time. As expected, elevated temperature deteriorates the

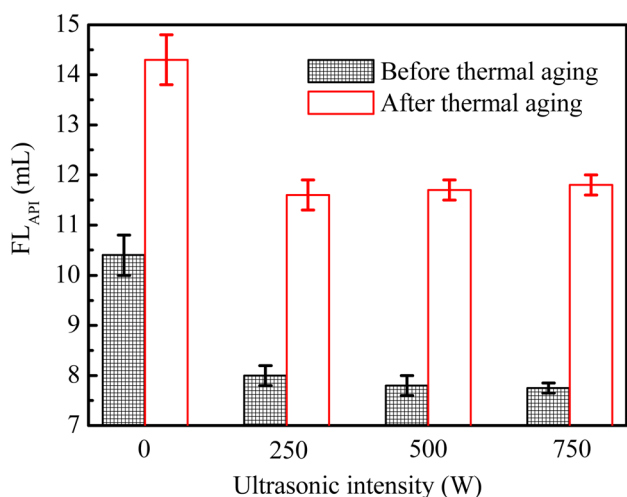


**Fig. 2** Effect of ultrasonic time on  $FL_{API}$  of drilling fluids at oscillation intensity of 500 W

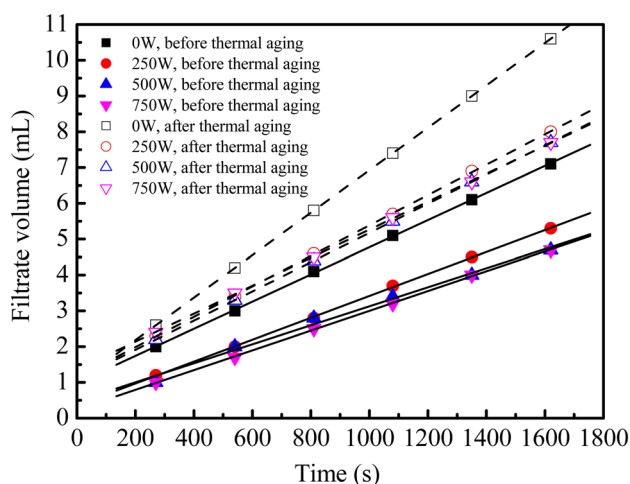
fluid loss properties. The hot aging leads to the flocculation among bentonite flakes and the dehydration of bentonite surface, which can increase the fluid loss. At initial 5 min of ultrasonic oscillations, the  $FL_{API}$  values show a rapid reduction irrespective of thermal aging of the drilling fluids. After that, the decrease continues, but the extent becomes weaker. When ultrasonic reaction time reaches 15 min, the  $FL_{API}$  values appear to approach a limiting value. Before and after thermal aging, ultrasonic oscillations can minimize the  $FL_{API}$  values by approximate 25% and 18%, respectively. Such short ultrasonic reaction time has considerably practical significance in mud preparation, because very long time of ultrasonic oscillations is generally needed for other treatments of clays (often more than 50 h (Chatel et al. 2016)). Accordingly, ultrasonic oscillations time is set at 15 min in the latter experiments. Subsequently, various  $FL_{API}$  values are displayed in Fig. 3, as a function of the ultrasonic intensity between 0 and 1000 W in steps of 250 W. In our experiment, drilling fluids could not endure a harsh ultrasonic oscillation of 1000 W as a result of the excessive acoustic cavitation and thus boiled continuously and overflowed from the reactor, making the filtrate volume of the samples unobtainable. Except 1000 W, the ultrasonic intensities have little influence on the  $FL_{API}$  values before and after thermal aging. It indicates that a relatively mild oscillation intensity of 250 W can also improve the filtration properties.

### Filter cake permeability and SEM analysis

Besides the lower filtrate volume, satisfactory filtration properties of drilling fluids should relate to the formation of thin and dense filter cakes on borehole walls. Accordingly, the permeability and microtexture of filter cakes are further investigated. Figure 4 displays the variation of filtrate



**Fig. 3** Effect of ultrasonic intensity on FL<sub>API</sub> of drilling fluids at oscillation time of 15 min



**Fig. 4** Filtrate volume versus time for the already formed filter cake from drilling fluids at different ultrasonic intensities and fixed oscillation time of 15 min

volume flowing through already generated filter cakes from the drilling fluids at different ultrasonic intensities with filtration time. Straight lines were gained by plot the volume of

collected filtrate vs. filtration time. Their slopes are equivalent to Q values. By substituting the  $\mu$ , L and Q values into Eq. 1, the K values were obtained. All the data are summarized in Table 1. It is found that there is nearly no change in all the  $\mu$  values, indicating that the viscosity of filtrate is not a decisive factor for the filtration control. Before and after hot aging of drilling fluids, the L values of ultrasonically treated samples decrease approximate 57.6% and 39.3%, respectively, compared to those of untreated samples, whereas the K values of ultrasonically treated samples decrease approximate 65% and 59%, respectively, compared to those of untreated samples. In the presence of ultrasonic oscillations, L and K values remain constant, respectively, regardless of the magnitude of ultrasonic intensity. The changing pattern of characteristics of filter cakes as a function of ultrasonic intensity is the same with that observed in the filtration property test, indicating that the filtration properties are basically decided by the features of mud cakes and ultrasonic oscillations have an important effect on them. The conclusions are also confirmed in SEM micrographs. Figure 5 shows the SEM images of dried filter cakes from ultrasonically treated and untreated drilling fluids, when time and intensity of ultrasonic oscillations are fixed at 15 min and 500 W. In the absence of ultrasonic oscillations, a great number of wrinkle and agglomerate appears on the filter cake surface (Fig. 5a). The wrinkle should be perpendicularly layer by layer stacked, indicating that there may be lots of cracks helpful to accelerate filtration. The rough and loose surface structure is more remarkable after thermal aging (Fig. 5c). Ultrasonic oscillations not only help to shrink the cracks but also alleviate the particle coalescence, thus generating a relatively smooth and dense microstructure. The phenomena are clearly observed in Figs. 5b and d.

**Rheological properties**

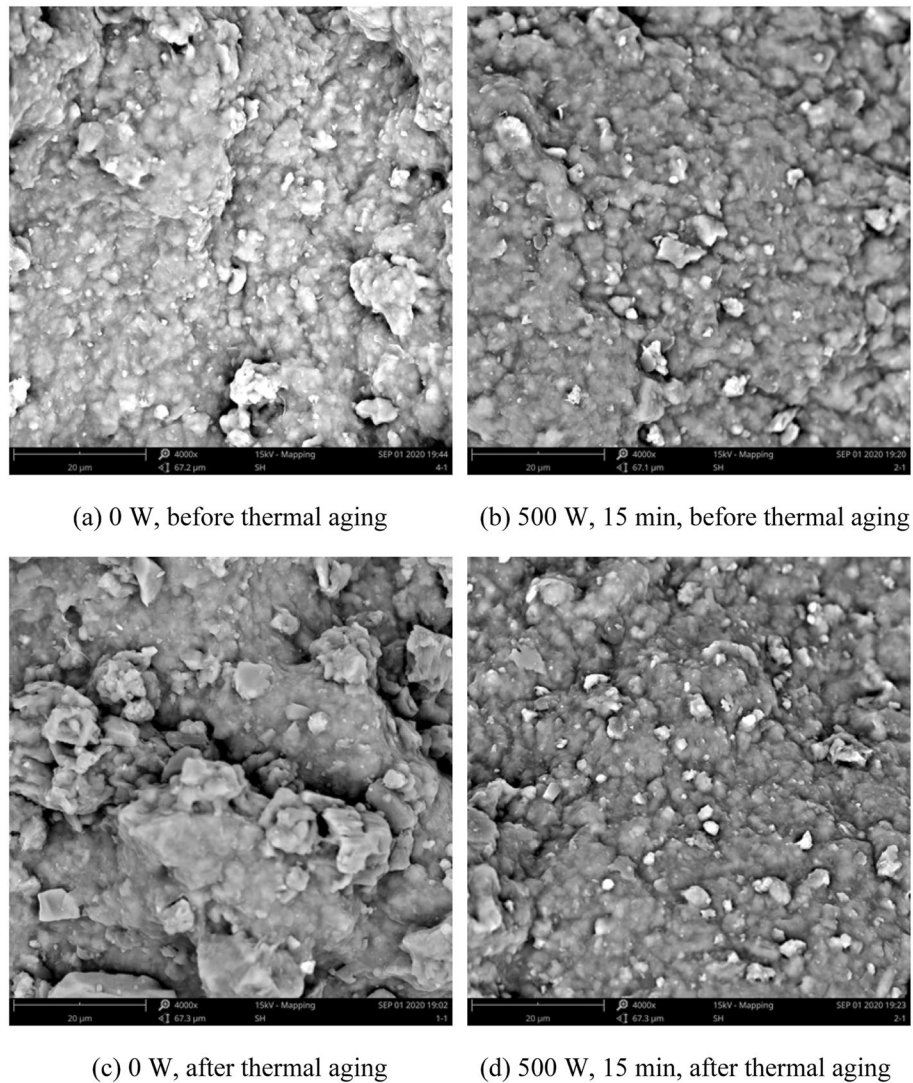
The effects of ultrasonic intensity on the shear stress and apparent viscosity of the drilling fluids before and after hot aging are studied. Figure 6a and b show shear stress and apparent viscosity against shear rate of the drilling fluids, respectively. All the samples produce shear thinning behaviors. After thermal aging, the increase in shear stress and

**Table 1** Viscosity ( $\mu$ ) of filtrate and thickness (L), filtration speed (Q), permeability (K) of filter cake from drilling fluids at different ultrasonic intensities and oscillation time of 15 min

Ultrasonic intensity (W)	Before thermal aging				After thermal aging			
	$\mu$ (mPa s)	L (cm)	$Q \times 10^3$ (mL/s)	$K \times 10^3$ (mD)	$\mu$ (mPa s)	L (cm)	$Q \times 10^3$ (mL/s)	$K \times 10^3$ (mD)
0	0.900	0.033	3.79	0.36	0.890	0.056	5.93	0.95
250	0.925	0.014	3.06	0.13	0.890	0.034	4.24	0.41
500	0.972	0.014	2.66	0.12	0.890	0.034	4.07	0.39
750	0.960	0.014	2.76	0.12	0.900	0.034	3.90	0.38



**Fig. 5** SEM micrographs of dried filter cakes



apparent viscosity should be ascribed to thermal induced swelling and flocculation of bentonite flakes (Ahmad et al. 2018a, b). The resembling results have been also found in the causticized lignite mud (Zhang et al. 2016). It can be seen from Fig. 6 that introduction of ultrasonic oscillations of various intensities into drilling fluids increases the magnitude of shear stress and apparent viscosity. The larger the ultrasonic intensity is, the more evident the increases are especially before thermal aging, showing an obvious difference compared with the influence of ultrasonic intensity on the filtration control. The extent of improvement in filtration properties is independent of the ultrasonic intensity, as already described in front. It is well known that in drilling fluid formulation design, diverse viscosifiers are added to enhance the rheological performances up to a desired extent needed for hole cleaning and suspending and carrying cuttings. Nevertheless, the performances can't exceed beyond specific levels. Inadequate increase may bring some negative

influences during mud circulation in borehole, for instance, excessive frictions resulting in drilling tool damage. Hence, it is concluded that ultrasonic oscillations of various intensities can be utilized to tailor rheological behaviors of drilling fluids and the filtration properties keep optimum all the while. Adsorption measurements by TOC, FTIR and TGA as well as clay particle size analysis will be conducted to investigate the reason behind ultrasound-induced improvement of filtration and rheological performances in the water-based drilling fluids.

### Adsorption properties

Figure 7 depicts the variation of adsorbed amount of SMC on bentonite versus ultrasonic intensity, before and after thermal aging. In the adsorption process, it is usually accepted that hydroxyl group of humic acid in SMC produces hydrogen and coordination bonding with oxygen atom

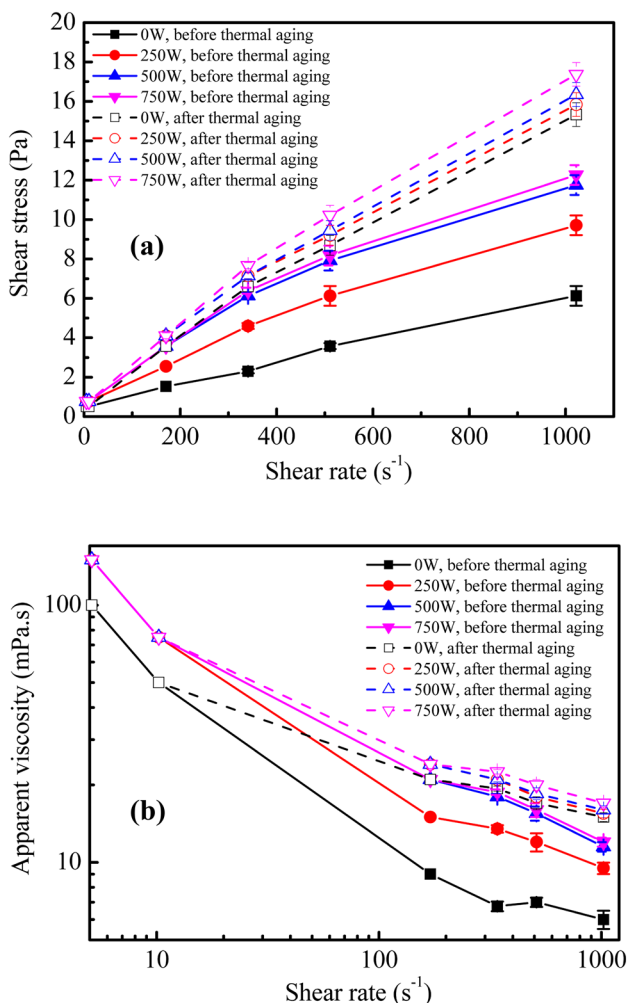


Fig. 6 Rheograms of drilling fluids at different ultrasonic intensities and fixed oscillation time of 15 min

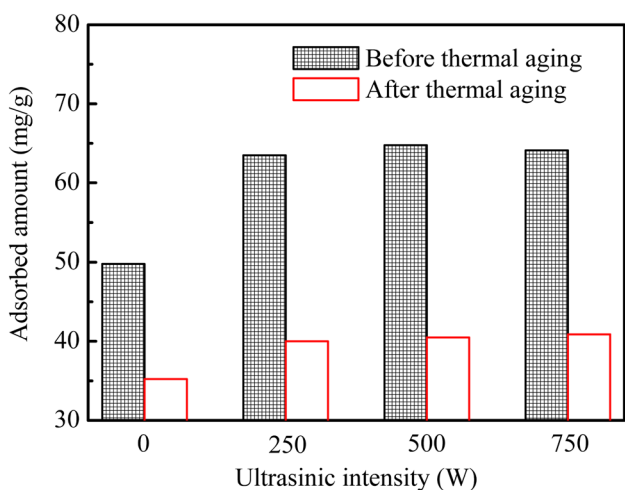
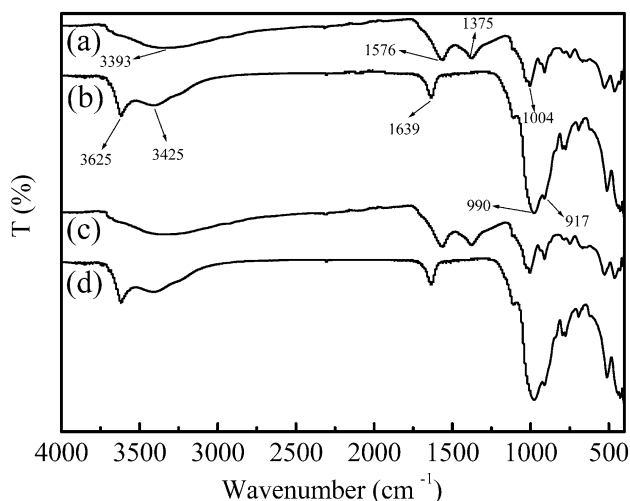


Fig. 7 Adsorbed amount of SMC on bentonite at different ultrasonic intensities and constant oscillation time of 15 min

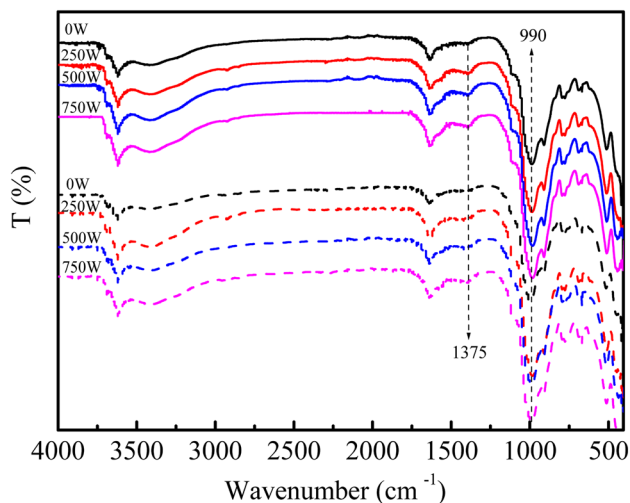
and aluminium atom of clay flakes, respectively. With the increase of temperature, the equilibrium between adsorption and desorption shift toward the desorption direction due to the exothermic characteristics of adsorption reaction. As a result, the adsorbed amount decreases after aging at 180 °C. In addition, the mud preparations under silent and ultrasonic conditions exhibit a remarkable difference in adsorptive capacity of SMC on bentonite. The adsorbed amount is obviously increased under ultrasonic oscillations in comparison with the conventional agitation but no obvious effect of intensity on the adsorption properties is observed. The adsorption process, in which the mass transfer occurs, may be limited by convection–diffusion in the reaction system. A drop in resistance to mass transfer can achieve through strengthening the convection in the circumstance. Ultrasonication has the ability to produce convection in colloidal solution. Ultrasound induced cavitation nearby the bentonite adsorbent can effectively decrease the boundary layer thickness and enhance the porosity on the bentonite surface in favor of mass transfer. At the same time, the high specific surface area and abundant active adsorption site of bentonite are obtained due to particle size reduction in the presence of ultrasonic oscillations (described in more detail later). The factors together lead to an increase of the adsorbed amount (Ealias and Saravanakumar 2019). On the other hand, only a slight dependence of the adsorbed amount on ultrasonic intensity should be ascribed to the following two mechanisms. One is that the extreme ultrasonication is likely to produce the desorption that the SMC molecules leave the surface of bentonite particles (Hassani et al. 2016; Taghizadeh and Seifi-Aghjekohal 2015). The other reason is that parts of ultrasonic energy may be scatter to the container walls instead of being absorbed in the liquid when the ultrasonic intensity exceeds certain criteria and consequently the efficacy of ultrasound utilization decreases (Su et al. 2012).

To obtain the ultrasonically treated samples of SMC and bentonite, ultrasonic oscillations were introduced into 2.9% aqueous solution of SMC and the bentonite stock suspension, respectively. Figure 8 gives the FTIR spectra of ultrasonically treated and untreated dry SMC and bentonite, when time and intensity of ultrasonic oscillations are fixed at 15 min and 500 W. In the SMC spectrogram, the wide band maximized at 3393 cm<sup>-1</sup>, and the absorption peak at 1576 and 1375 cm<sup>-1</sup> can be distributed to the stretching vibrations of OH group, aromatic C = C bond and C-O bond of phenolic OH, respectively (Zhang et al. 2016). The peak of 1004 cm<sup>-1</sup> represents the stretching vibration of S = O bond in sulfonic acid group (Chang et al. 2019; Zhang et al. 2016). The FTIR patterns of bentonite exhibit the representative character of smectite group. The main absorption peaks are described as follows (Mekhamer 2010; Xuan et al. 2015): the stretching band of structural hydroxyl group (3625 cm<sup>-1</sup>), the wide



**Fig. 8** FTIR spectra of **a** virgin SMC, **b** virgin bentonite, **c** ultrasonically treated SMC and **d** ultrasonically treated bentonite

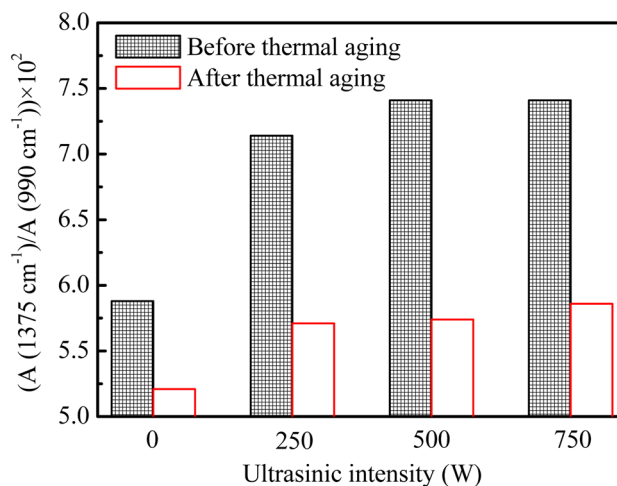
stretching band of bound water ( $3425\text{ cm}^{-1}$ ), the deformation band of water ( $1639\text{ cm}^{-1}$ ) and the bands of Si–O stretching ( $990\text{ cm}^{-1}$ ) and Al–Al–OH bending ( $917\text{ cm}^{-1}$ ). No obvious difference in FTIR spectra is detected between ultrasonically treated and untreated samples, meaning that SMC and bentonite with ultrasonic oscillations don't undergo any structural change. FTIR analysis of the SMC-bentonite composites at different ultrasonic intensities is carried out and showed in Fig. 9. As compared with SMC and bentonite, in the FTIR spectra of all SMC-bentonite composites, there are no new absorption bands and only show a band overlapping of both components. It also



**Fig. 9** FTIR spectra of ultrasonically treated and untreated SMC-bentonite composites at different ultrasonic intensities and oscillation time of 15 min (solid line: before hot aging; broken line: after hot aging)

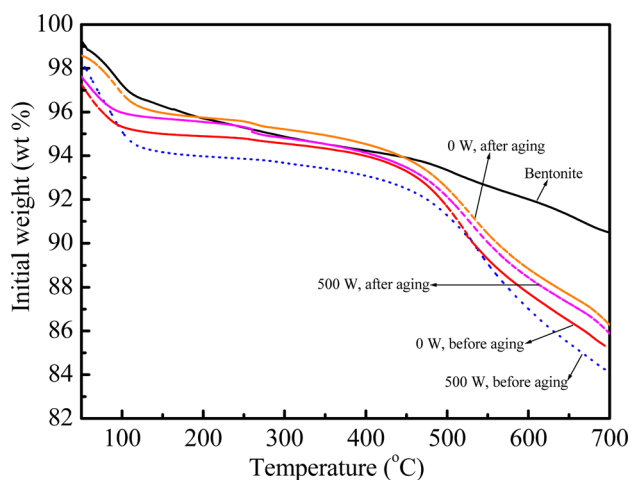
confirms the effective adsorption of SMC on bentonite, irrespective of ultrasonic oscillations.  $A(1375\text{ cm}^{-1})/A(990\text{ cm}^{-1})$  values of various SMC-bentonite composites are shown in Fig. 10, where  $A(1375\text{ cm}^{-1})$ , and  $A(990\text{ cm}^{-1})$  are the area of absorption band at  $1375$  and  $990\text{ cm}^{-1}$ , respectively. The higher the value is, the more abundant the C–O content relative to Si–O group in composites is, indicating the increasing of the adsorbed amount of SMC on bentonite. The results of FTIR are well consistent with those of TOC tests.

When time and intensity of ultrasonic oscillations are fixed at 15 min and 500 W, ultrasonically treated and untreated SMC-bentonite composites are chosen to assess the thermal decomposition behaviors through thermogravimetric techniques. TGA patterns of natural bentonite and the composites in the temperature range of room temperature– $700\text{ }^{\circ}\text{C}$  are shown in Fig. 11. The thermal decomposition curve is separated into two parts. The first part of quality loss observed below  $200\text{ }^{\circ}\text{C}$  is attributed to the expulsion of bound water on bentonite and hydrophilic SMC molecule (Chang et al. 2019; Zhong et al. 2016). The larger mass loss may mean that the sample tends to take up more water through sulfonic group and carboxyl group on SMC molecule and thus form a thicker hydrated shell on the surface of bentonite. It is beneficial in improving filtration properties. Besides dehydroxylation of bentonite, subsequent mass loss is mainly induced by the thermolysis of organic substance (Peng et al. 2013). The larger mass loss in the second step implies abundant adsorbed amount of SMC on bentonite. Here, the effects of ultrasonic oscillations and thermal aging on the absorbability get confirmed again.

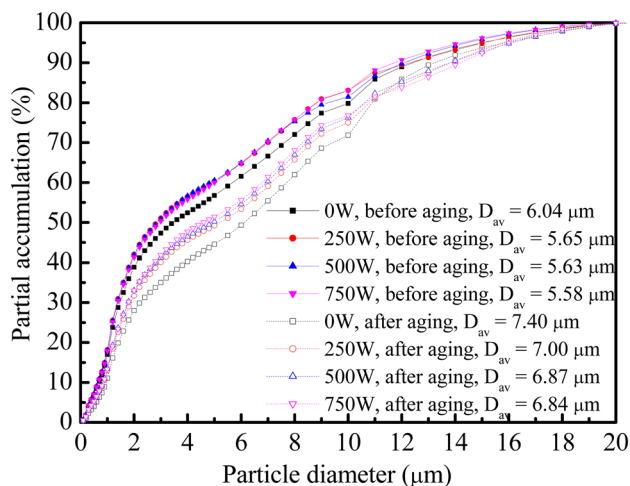


**Fig. 10** Variation of  $A(1375\text{ cm}^{-1})/A(990\text{ cm}^{-1})$  values of various SMC-bentonite composites





**Fig. 11** TGA curves of bentonite and ultrasonically treated and untreated SMC-bentonite composites at different ultrasonic intensities and fixed oscillation time of 15 min



**Fig. 12** Particle size accumulation curves of ultrasonically treated and untreated drilling fluids at different ultrasonic intensities and fixed oscillation time of 15 min

### Bentonite particle size analysis

Figure 12 presents the effects of ultrasonic oscillations and thermal aging on the clay particle size of the drilling fluids. It can be found that the particle size accumulation curves move to the right region in the graph, and average particle diameter ( $D_{av}$ ) is increased after thermal aging test, indicating that bentonite particles tend to aggregate with increasing temperature as a consequence of chemical degradation and desorption of SMC and elevated mutual collision of clay particles (in accordance with Sun (Chang et al. 2019) and Zhu (Ma et al. 2017)). Introduction of ultrasonic oscillations of 250 W into drilling fluids shifts the particle size

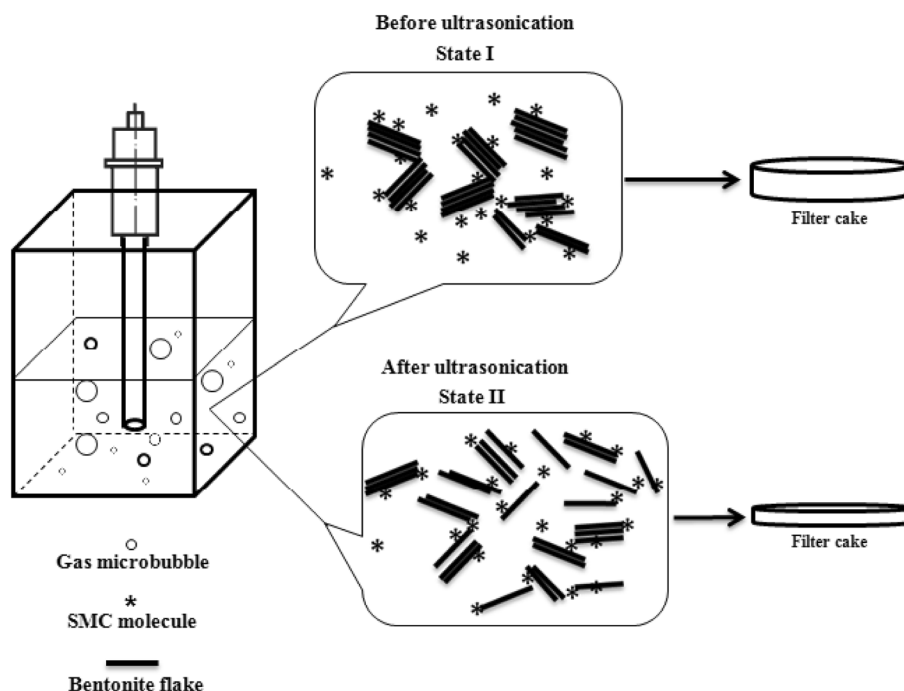
accumulation curves to left, corresponding to the decrease of  $D_{av}$ . However, the curves and  $D_{av}$  change relatively little with the further increase of ultrasonic intensity. It should be ascribed to the following two factors. On one hand, ultrasonic cavitation leads to microjet and shock-wave impact on the surface of clay particles and interparticle collisions, directly delaminating the larger particles (Mekhamer 2010). On the other hand, increased adsorbed amount of SMC on bentonite with the introduction of ultrasonic oscillations is more beneficial to make and maintain fine particles in the system (Ma et al. 2017), due to improved hydration dispersion effects on clay surface (which has been confirmed by TGA). Combined with the abovementioned analyses about TOC data, it can be concluded that the ultrasound-induced adsorption and dispersion assist each other and boost each other.

### Mechanism for improvement of filtration property

The classical action mechanism of traditional filtrate reducer in water-based drilling fluids has been already mentioned in front INTRODUCTION parts. On the basis of the theory and our works in the paper, a schematic illustration shown in Fig. 13 is put forward to explain the development of filtration performances of ultrasonically treated drilling fluids. All the proofs point to the outcomes that ultrasound-assisted adsorption of SMC on bentonite and dispersion of bentonite clay particle are responsible for the improvement. Introduction of ultrasonic oscillations during mud preparation in nature settle the question how bentonite and additive efficiently react with each other and maximally convert into corresponding composites. Unfortunately, the present routes for solving the problems still rest on the excessive addition of conventional drilling fluid additives.

In our experiments, ultrasonic oscillations can break the thermodynamic equilibrium of colloidal stability and impel the equilibrium state to shift from I toward II. The degree of the shift is heavily dependent on some ultrasonic parameters such as time and power. Finally, a new thermodynamic equilibrium state is achieved and kept forever even if ultrasonic oscillations don't continue. Excellent adsorptive and dispersive capacity derived from ultrasonication are the precondition of improving filtration properties, whereas generating of thicker hydration shell on the clay surface and more stable gel structure is a foregone conclusion of the process. Further, the ultrasonically treated SMC-clay composites under hydrostatic and formation pressure difference can layer by layer pile up to yield thin and compact filter cakes, acting as valid barrier layers to alleviate the fluid loss. Simultaneously, the improved dispersion state (high specific surface area of clay particles) can strengthen the resistance to flow, meaning the increase of viscosity at macroscopic level (Ahmad et al. 2018a, b; Ma et al. 2017; Rafati et al. 2018), since viscosity

**Fig. 13** Schematic diagram of effects of ultrasonic oscillations on interaction of bentonite clay and SMC molecule and formation of filter cake



is considered as internal friction between two layers of a liquid under shearing strength. In addition, we think that the thermodynamic state of mud hydrosol directly affect the rheological properties and the formation of filter cake. Characteristics of filter cake further determine the filtrate volume. In other words, the filtrate volume is indirectly affected by the state of the adsorption and dispersion. As a result, rheological properties of drilling fluid are more susceptible to ultrasonic intensity compared to the filtrate volume.

## Conclusion

Under our experimental conditions, in-situ improvement of filtration properties in water-bentonite suspension containing SMC could be realized by introduction of ultrasonic oscillations during drilling fluid preparation. As compared with the conventional agitation, the addition of ultrasonic oscillations created thin, dense, and low permeability mud cakes, leading to lower loss of fluid. The API filtrate volume decreased with increasing ultrasonic time, and it tended to reach to a critical value. The lower ultrasonic energy output of 250 W could bring a rather satisfactory result in filtration properties, above which the filtration properties are almost unchanged. However, it was interesting to note that shearing strength and apparent viscosity of the drilling fluids were increased with introduction of ultrasonic oscillations, and continue the increase with increasing ultrasonic intensity. It indicated that we may use ultrasonic oscillations of various intensities to regulate and control the rheological behavior of drilling

fluids according to the actual demand and at the same time obtain the optimal filtration properties. Ultrasound-induced adsorption of SMC on bentonite and ultrasound-induced dispersion of bentonite clay particle are responsible for the improvement. Adsorption experiments by TOC, FTIR and TGA as well as particle size analysis displayed that adsorption quantity of SMC on clay increased with introduction of ultrasonic oscillations due to acoustic cavitation, along with the decrease of average clay particle diameter.

## Suggestions for future work

The findings in this paper are only limited to a simply water-based drilling fluid system. Before the field application of the ultrasound-assisted mud preparation technology, some research works still need to be done, including the adjustment of more ultrasonic parameters such as frequency and energy distribution, other ultrasonic oscillations of water-bentonite suspension containing one even several additives, and verification of ultrasonic improvement of other drilling fluid properties (lubrication, inhibition and plugging).

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## Declarations

**Conflict of interests** On behalf of all the co-authors, the corresponding author states that there is no conflict of interest.

**Human Participants and/or Animals** No research involving Human Participants and/or Animals.

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