Application of Tea Polyphenols as a Biodegradable Fluid Loss Additive and Study of the Filtration Mechanism

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**ABSTRACT:** Drilling fluids with poor filtration property are disadvantageous for well drilling, easily causing wellbore instability and formation collapse. This work reports the novel utilization of tea polyphenols (TPs) as a fluid loss additive in the bentonite–water-based drilling fluids (BT-WDFs). The influence of TP concentration and temperature on the filtration property of the fluids was described. The results showed that an increase in the TP concentration contributed to a decrease in fluid loss. Especially BT-WDFs added with 3.0 wt % TP exhibited a low fluid loss (less than 10 mL) at room temperature and high temperatures (∼150 °C), displaying better filtration property and temperature resistance than common fluid loss agents. Through the investigations on the viscosity, the particle size of TP/BT-WDFs, and micromorphology of filter cakes, the dispersion effect of TP was considered as the dominant factor for the filtration property of TP/BT-WDFs. TP molecules, containing many functional groups, could attach to the surface of bentonite platelets, improve the hydration of bentonite particles, and promote the dispersion of bentonite particles. At room temperature, TP facilitated the dispersion of hydrated bentonite. The existing “house-of-cards” structure was weakened, decreasing the particle size and viscosity of TP/BT-WDFs. At high temperature, bentonite dehydrated and aggregated, thereby increasing the particle size of bentonite particles, decreasing the viscosity of bentonite dispersion, and resulting in a high fluid loss. The addition of TP dispersed bentonite from face-to-face (FF) attraction to edge-to-face (EF) attraction, recovered the house-of-cards structure, and increased the viscosity of TP/BT-WDFs. Under the dispersion effect of TP, an appropriate grain composition of bentonite particles was formed and the pore throats were plugged to prevent the penetration of water. Finally, a compact and thin filter cake was built and the fluid loss was greatly reduced. The TP/BT-WDFs exhibited good filtration property. TP is a prospective candidate to be a high-performance and biodegradable fluid loss additive in well-drilling applications.

1. INTRODUCTION

Drilling fluid is an important functional fluid in the well-drilling operation. The success of drilling operations greatly depends on the performance of drilling fluids. In the drilling process, drilling fluids are pumped from the surface into subsurface reservoirs through the drilling string. They perform important roles such as cleaning the rock fragments, balancing formation pressure, stabilizing the borehole wall, and cooling and lubricating drilling tools.1,2 Due to the cost and environmental requirements, water-based drilling fluids (WDFs) are more commonly used. The WDFs are mainly composed of water, bentonite (BT), and various chemical agents (fluid loss additive, tackifier, shale inhibitor, etc.).3 The filtration property is one of the most important properties for WDFs. In the case of a poor filtration property, too much water will penetrate into the formation and form a thick filter cake. This may lead to formation damage, borehole collapse, pipe sticking, and eventually an increase in drilling costs.4 Therefore, over the past decades, a great number of additives have been employed to reduce the filtration volume of WDFs, for example, starch,5,6 polyanionic cellulose (PAC),7 copolymers,8–10 humic acid,11 phenolic resin,12 graphene,13 and nanosilica.14–16

Tea polyphenols (TPs), as one of the most extensive plant polyphenols worldwide, have received much attention from the daily chemical, pharmaceutical, and food industries owing to their extraordinary antioxidative and antibacterial characteristics.17,18 The major polyphenols in TPs are flavonols, and the major components in flavonols are catechins. In detail, there are four major components in catechins, including (−)-epigallocatechin gallate (EGCG), (−)-epicatechin gallate (ECG), (−)-epicatechin (EC), and (−)-epicatechin gallate (EGC). EGCG accounts for 50−70% of catechins and is the major

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catechin in TPs. TPs are water-soluble and quite stable with good temperature-resistant property and oxidation-resistant property. Based on these special characteristics, TPs have been widely applied in the engineering field and demonstrated good thermal stability, antioxidative property, and emulsion stabilization ability. In fact, some plant-based phenolic materials have been employed as chemical additives in petroleum engineering due to their distinctive characteristics (biodegradability, water-solubility, surface activity, adsorption dispersibility, etc.). As a kind of phenolic materials, the utilization of TPs as more effective fluid loss additives in the WDFs is worth researching.

The filtration property of BT-WDFs is greatly influenced by the filter cake quality and filtrate viscosity. Various fluid loss additives have been used to reduce fluid loss by increasing the filtrate viscosity and improving the filter cake quality. Nanoparticles could plug the pores in filter cakes as scaling agents to decrease the filter cake permeability. Some copolymers, such as acrylamide-based copolymers, can be adsorbed onto clay particles and form a hydration shell, preventing the aggregation of clay particles. Downhole, the conditions are pretty complex, especially at high temperatures. Drilling fluid additives must possess high-temperature resistance. Therefore, it is necessary to evaluate the filtration property when adding TPs at high temperatures.

This work aims to study the promising application of TPs as biodegradable fluid loss additives in BT-WDFs and uncover the underlying mechanism. The characteristics (e.g., solubility of the TP solution, \( \zeta \)-potential, morphology, and thermogravimetric property) of TPs were studied. The lower fluid loss was observed with increase in the concentration of TPs. Through the investigations on the viscosity, the particle size distribution of TP/WDFs, and micromorphology of filter cakes, the filtration mechanism of TPs was proposed.

2. MATERIALS AND METHODS

2.1. Materials. TP (purity \( \geq 98\% \)) extracted from green tea leaves was purchased from Qingdao Youshuo Chemical Technology (China). Bentonite was obtained from Huaiian Tengtei Development Co., Ltd. (China). Carboxymethyl starch (CMS) was purchased from Aladdin Industrial Corporation (China). Polyanionic cellulose (PAC) was obtained from the China National Offshore Oil Corporation (CNOOC). All of the chemicals were used without further purification.

2.2. Characterization of TP. 2.2.1. Proton Nuclear Magnetic Resonance (\(^1\)H NMR). The \(^1\)H NMR spectrum of TP was recorded with a Bruker AVANCE III 600M NMR apparatus. Dimethyl sulfoxide (DMSO) was used as the solvent.

2.2.2. TP Solution Stability. The TP solutions at different pH values were prepared to observe their solution forms and stability.

2.2.3. \( \zeta \)-Potential. The \( \zeta \)-potential of TP solutions at different pH values was measured by a Malvern Zetasizer Nano series. The concentration of all of the samples in this test was about 1.0 g L\(^{-1}\).

2.2.4. Thermogravimetric Analysis (TGA). The TGA and derivative thermogravimetry curves of TPs were measured by PE Pyris 1 in the temperature range of 50–500 °C at a heating rate of 10 °C min\(^{-1}\) and under an N\(_2\) gas flow.

2.2.5. Biodegradability Test. The biodegradability of the TP was evaluated through the BOD\(_5\)/COD\(_m\) method. In the petroleum industry, the values of BOD\(_5\)/COD\(_m\) in the range of <1.0, 1.0–5.0, and >5.0% represent hardly degradable, nondegradable, and biodegradable, respectively.

2.3. Preparation of BT-WDFs and TP/BT-WDFs. Four percent BT-WDFs were prepared by adding 4% (w/v) bentonite in distilled water and stirring at 1500 rpm for 12 h, followed by static standing for 24 h. Then, 1, 2, and 3% (w/v) TPs were dissolved in 4% BT-WDFs with stirring at 5000 rpm for 30 min and the pH of the TP/BT-WDFs was adjusted to 9 by adding NaOH aqueous solution.

2.4. Filtration Tests of BT-WDFs and TP/BT-WDFs. Filtration tests were carried out by the American Petroleum Institute (API) guidelines. About 250 mL of the fluids (BT-WDFs or TP/BT-WDFs) were poured into a standard filter press (Fann Instrument Company) equipped with reinforced filter paper (9 mm in diameter and 2.7 \( \mu m \) in pore size). The filtration tests were conducted under a pressure of 100 psi using \( N_2 \) gas chargers for 30 min. The volume of fluid loss was recorded at 0.5, 1.5, 3.0, 7.5, 15, and 30 min after each test had begun.

2.5. Rheology Analysis of BT-WDFs and TP/BT-WDFs. The rheological properties of BT-WDFs and TP/BT-WDFs were measured through a Haake Mars rheometer (Thermo Electron Corporation, Waltham, MA). The apparent viscosity was measured under a shear rate from 1000 to 1 s\(^{-1}\). All of the tests were conducted at 25 °C. Before the rheological measurement, the dispersions were stirred energetically for 30 min.

2.6. Microstructural Analysis. The exterior morphologies of BT-WDFs and TP/BT-WDFs dispersions with different TP concentrations were observed by a Leica DM4 M microscope with a total magnification of 50\( \times \).

3. RESULTS AND DISCUSSION

3.1. Characteristics of TP. The molecular structure of the TP was first characterized. As mentioned above, the major polyphenols in TP are catechins, which account for 60–80% of the total TPs. In detail, there are four major tea catechins: EGCG, EGC, ECG, and EC (as shown in Figure 1), among which EGCG was reported as the most abundant ingredient. Meanwhile, the \(^1\)H NMR spectroscopy was also used to characterize the structure of the TP. As shown in Figure 2, typical chemical shift values in the TP were preliminary presented: \( \delta = 2.6–2.9 \) ppm mainly for EGCG and EGC, \( \delta = 4.6, 6.6, 6.8 \) ppm for EGC, \( \delta = 4.7 \) ppm for EC, \( \delta = 5.0 \) ppm for ECG, \( \delta = 4.9, 5.3, 5.8 \) ppm for mainly EGC, EGCG, and \( \delta = 6.4 \) ppm for EGCG. The TP exhibited various polyphenol structures.

The \( \zeta \)-potential of TP versus pH was first measured to evaluate its charged nature (Figure 3). TP was almost negatively charged over the whole pH range. There are many
phenolic hydroxyls in TP molecules. These functional groups can ionize and produce H⁺, thereby resulting in the negative charge of TP. Due to the ionizing effect, the TP solution was acidic and its natural pH was 2.51. A variety of external parameters can affect the oxidation state of the TP solution, of which pH is the most important one. The oxidation rate will improve with increase in the pH value, which can be evident by the gradual deepening color of the TP solution (Figure 3).

The TGA results were also analyzed (Figure 4). The slight mass loss for TP from 50 to 150 °C corresponded to the evaporation of free water and crystal water in the TP. After the temperature was subsequently increased to 500 °C, the remaining weight of TP was still 58.28%. The TP mass started to dramatically decrease at 210 °C, showing a high thermal decomposition temperature. The results indicated that TP had the potential to be used in high-temperature conditions.

Environmental protection property is pretty important for drilling fluid additives due to strict environmental regulations. According to the previous literature, the TP was nontoxic and good for people’s health. However, as a drilling fluid additive, there was no direct proof to show its degradability. Thus, a biodegradability test was conducted to further evaluate the environmental performance of the TP. As shown in Table 1, the value of the BOD₅/COD₅ was 26.97%. The result proved that the TP was environmentally friendly and biodegradable as a drilling fluid additive.

3.2. Filtration Properties of TP in BT-WDFs. A series of API filtration tests for TP were conducted in 4% BT-WDFs at different TP concentrations and temperatures (Figure 5a). The fluid loss of pure BT-WDFs was as high as 23 mL within 30 min at room temperature. When 1.0, 2.0, and 3.0% TPs were added into BT-WDFs, 43.5, 58.3, and 59.1% reductions in the fluid loss volume were achieved, respectively, displaying an improved filtration property.

### Table 1. Biodegradability of the TP

| Sample | COD₅ (mg/L) | BOD₅ (mg/L) | BOD₅/COD₅ (%) | Biodegradability grade |
|--------|-------------|-------------|---------------|------------------------|
| TP     | 3.70 × 10⁴  | 9.98 × 10³  | 26.97         | Biodegradable          |

Figure 4. TGA analysis of TP.

Figure 5. Plots of fluid loss versus time for BT-WDFs at different TP concentrations after aging at room temperature (a), 120 °C (b) and 150 °C (c). The fluid loss volume (mL/30 min) of TP/BT-WDFs with different TP contents under different temperatures (d).
The downhole temperature rises gradually with an increase in the drilling depth, which might limit effective filtration control. Therefore, we also measured the fluid loss variation of BT-WDFs and TP/BT-WDFs after aging at 120 and 150 °C (Figure 5b,c). For pure BT-WDFs, the fluid loss increased dramatically with increase in the aging temperature, displaying a poor filtration property at high temperatures. As expected, TP/BT-WDFs still maintained remarkable filtration property. The fluid loss volume was still further decreased with increase in the TP concentration (as shown in Figure 5d). For example, when the aging temperature was 120 °C, the addition of 1.0, 2.0, and 3.0% TPs greatly decreased the fluid loss from approximately 30 to 19.5, 13.4, and 9.5 mL, respectively. Moreover, the fluid loss curve of fluid with the addition of 3.0% TP reached a plateau after 15 min. Similar phenomenon was also observed at 150 °C. The fluid loss was reduced to 10.2 mL when 3.0% TP was added, where as that of pure BT-WDFs was about 50 mL at 150 °C. The improvement in filtration property was beneficial for maintaining the stability of borehole and avoiding the formation of collapse. These results demonstrated that TP could be used as an effective fluid loss additive in BT-WDFs.

Subsequently, the filter cakes obtained in the filtration tests were taken into account. Figure 6 shows the photos of fresh filter cakes formed by TP/BT-WDFs at different TP concentrations and temperatures. For pure BT-WDFs, bentonite particles will imbibe water and hydrate immediately when dispersed in water, forming diffused electric double layers. This will cause separation of individual bentonite layers and the formation of typical “house-of-cards” structure, producing a desired high viscosity of the bentonite dispersion. Finally, bentonite settled to build a relatively thin and compact filter cake (Figure 6a), forming a barrier layer to prevent the penetration of water. That was the typical filtration mechanism for pure BT-WDFs. However, the filter cakes of BT-WDFs became greatly thicker after aging at 120 and 150 °C (Figure 6a1,a2), demonstrating a deteriorated filtration property. For TP/BT-WDFs, improved appearances of filter cakes were seen by adding TPs with different concentrations (Figure 6b−d,b1−d1,b2−d2). In detail, with the addition of 1.0% TP, the filter cake became thinner and denser. This positive effect became stronger as the TP concentration increased to 3.0%. Especially, the filter cake was almost transparent after the addition of 3.0% TP at room temperature. In addition, very thin, clear, and smooth filter cakes of TP/BT-WDFs were also observed after aging at 120 and 150 °C, showing a better filtration property. In addition, the thickness of the filter cakes was also measured to show a quantitative description. As shown in Table 2, the thickness of the filter cakes from pure BT-WDFs increased with an increase in the temperature. After TP was reacted with BT-WDFs, the thickness of the filter cakes was sharply reduced. For example, after 3% TP was added at 150 °C, the thickness of the filter cake was 1.02 mm, which was slightly thinner than that of pure BT-WDFs at room temperature. The filtration property of BT-WDFs is greatly determined by the filter cake quality. A thinner filter cake usually shows better filtration property. TP could improve the filter cake quality and reduce the filter loss of BT-WDFs at temperatures ranging from room temperature to 150 °C, showing the potential of TP as an effective fluid loss additive.

To further evaluate the filtration property of TP, some common fluid loss additives were also tested at 150 °C to compare with TP. Diverse fluid losses were obtained when different additives were added at a fixed concentration of 3.0% (Figure 7a). Compared with CMS and PAC, the addition of TP in BT-WDFs resulted in the lowest fluid loss. Moreover, the filter cake of TP/BT-WDFs also performed the thinnest and the most compact appearance (Figure 7b). CMS and PAC were common fluid loss additives in drilling engineering, but these materials exhibited poor filtration property under high temperature. The comparative tests further proved that TP displayed better filtration property.

### Table 2. Thickness of the Filter Cakes

| Temperature   | 0%  | 1%  | 2%  | 3%  |
|---------------|-----|-----|-----|-----|
| room temperature | 1.04 | 0.76 | 0.58 | 0.56 |
| 120 °C         | 2.08 | 1.14 | 0.90 | 0.72 |
| 150 °C         | 3.12 | 1.22 | 1.08 | 1.02 |

**Figure 6.** Images of fresh filter cakes from TP/BT-WDFs at TP concentrations of (a) 0 wt %, (b) 1.0 wt %, (c) 2.0 wt %, and (d) 3.0 wt % at room temperature. Filter cakes (a1)−(d1) and (a2)−(d2) were those of filter cakes (a)−(d) after aging at 120 and 150 °C.

**Figure 7.** Fluid loss (a) and filter cakes (b) of base fluids added with different fluid loss additives after aging at 150 °C.
loss. Therefore, if the viscosity of drilling fluids is the dominant factor for the filtration mechanism, drilling fluids with higher viscosity should show lower fluid loss volumes. Therefore, the plots of viscosity versus shear rate for TP/BT-WDFs at different TP concentrations and temperatures were first evaluated (Figure 8). Distinctive trends of viscosity were observed by incorporating TP at different temperatures. At room temperature, pure BT-WDFs displayed typical shear thinning behavior and showed higher viscosity than all of the TP/BT-WDFs samples. However, the viscosity of TP/BT-WDFs presented a downward trend with the increase of TP concentration. Meanwhile, with the increase in the temperature from room temperature to 150 °C, the viscosity of pure BT-WDFs decreased gradually. On the contrary, the viscosity of TP/BT-WDFs became higher. Moreover, the viscosity of TP/BT-WDFs at 150 °C was higher than that of pure BT-WDFs at 150 °C but still almost equal or less than that of pure BT-WDFs measured at room temperature. Besides, at the same temperature, the viscosity of TP/BT-WDFs always showed a downward trend with increase in the TP concentration. Filtration experimental results showed that the fluid loss decreased with an increase in the TP concentration at every temperature. The above results were contrary to the conception that drilling fluids with a higher viscosity should have a lower fluid loss. Therefore, the viscosity was not the primary factor for the filtration property of TP/BT-WDFs.

Subsequently, the particle size of bentonite was also taken into account for understanding the role of TP in the filtration process. The particle size distribution of BT-WDFs and TP/BT-WDFs at different temperatures is shown in Figure 9. Distinct particle size distribution was obtained after adding TP at different TP concentrations. However, the viscosity of TP/BT-WDFs became higher. Moreover, the viscosity of TP/BT-WDFs at 150 °C was higher than that of pure BT-WDFs at 150 °C but still almost equal or less than that of pure BT-WDFs measured at room temperature. The above results were contrary to the conception that drilling fluids with a higher viscosity should have a lower fluid loss. Therefore, the viscosity was not the primary factor for the filtration property of TP/BT-WDFs.

Figure 8. Plots of viscosity versus shear rate for BT-WDFs at different TP concentrations after aging at room temperature (a), 120 °C (b), and 150 °C (c).

Figure 9. Particle size distribution of BT-WDFs at different TP concentrations after aging at room temperature (a), 120 °C (b), and 150 °C (c).

Figure 10. D_10 and D_50 of BT-WDFs at different TP concentrations after aging at different temperatures.

WDFs increased dramatically after aging at high temperatures, correlating to a high filter loss. On the contrary, after TP with different concentrations was added, the D_10 and D_50 of TP/BT-WDFs decreased gradually, whereas the filtration property was improved.

In fact, the filtration property of BT-WDFs is greatly influenced by the particle size distribution of the solid phase in BT-WDFs. Generally, the particles with an appropriately small size can have more access to the pores and pore throats. Many solid particles, such as nanosilica, ultrathin calcium carbonate (CaCO_3), laponite nanoparticles, and some fibrous materials, have been developed and used as sealing agents. Satisfactory filtration property can be achieved by choosing the appropriate nanoparticles that match the size of pores. After adding TP, the particle size of bentonite became smaller and exhibited a wide distribution range. Namely, TP could promote the dispersion of bentonite particles, even at high temperatures. The combination of nanosized and micronized clay particles contributed to plugging the pore throats and forming an effective barrier layer to prevent the penetration of water, thereby reducing the fluid loss.

3.4. Microstructural Analysis of TP/BT-WDFs. The micromorphology of bentonite particles was observed by microscopy (Figure 11). Pure bentonite dispersion displayed quite homogeneously distributed particles at room temperature (Figure 11a). With an increase in the aging temperature, bentonite particles aggregated and the particle size obviously increased. The addition of TP as a filtration agent restored the bentonite to a smaller size, which was consistent with the particle size measurement results. These results further proved that TP could promote the dispersion of bentonite and induce
smaller particles, which was beneficial for reducing fluid loss. The changing pattern of particle size of bentonite as a function of TP concentration was identical to that observed for the fluid loss. This demonstrated that the size change of bentonite particles might be the predominant reason for the improved filtration property of TP/BT-WDFs.

SEM micrographs have been usually applied to examine the influence of fluid loss additives on the microstructure of dry filter cakes, further elaborately accounting for filtration mechanism. The SEM micrographs of dried filter cakes from TP/BT-WDFs are shown in Figure 12. For pure BT-WDFs, at room temperature, bentonite could form a relatively compact filter cake (Figure 12a). However, at high temperatures, the bentonite aggregated and generated rough morphology, especially at 150 °C. A large number of wrinkles appeared on the surface of the filter cakes, and the surface was very rough (Figure 12a1,a2). These wrinkles seemed to be stacked in layers, indicating that there might be many channels available for fluid penetration. For TP/BT-WDFs, with the addition of TP, such stacked wrinkles gradually disappeared. A more smooth and compact microstructure was clearly observed (Figure 12d−d2). Obviously, the addition of TP improved the filter cake quality. The results were also in accordance with that of filtration tests.

### 3.5. Filtration Mechanism Analysis

Generally, the bentonite platelets are negatively charged on the flat surface, while the edges can be negative, neutral, or positive depending on the pH value. Bentonite particles hydrate, swell, and disperse in water. The ionic attraction between the negatively charged flat surface and positively charged edge (edge-to-face (EF) attractions) forms a special house-of-cards structure, which is responsible for the viscosity of bentonite dispersion. These special features enable bentonite to be an ideal rheological and filtration material in BT-WDFs. However, bentonite is sensitive to temperature. Under high-temperature conditions, the adsorption of clay particles to water molecules becomes weak and clay particles dehydrate and aggregate (Figures 9 and 11), thereby resulting in thick filter cake and poor filtration property (Figures 5 and 6).

TP molecules contained many functional groups, such as phenolic hydroxyls, carbonyls, and carboxyls. TP could be attached to the surface of bentonite platelets via hydrogen bond between carbonyl/hydroxyl groups of TP and hydroxyl groups of bentonite platelets. The negative hydration groups in TP increased the electronegativity of bentonite, improved the hydration of bentonite particles, and promoted the dispersion of bentonite particles. At room temperature, bentonite fully hydrated and dispersed in pure BT-WDFs. A house-of-cards structure was formed by EF attraction (Figure 13a).
bentonite dispersion displayed quite a high viscosity, showing typical shear-thinning non-Newtonian characteristics (Figure 8a). After the addition of TP, the adsorption of TP further accelerated the dispersion of bentonite and broke the house-of-cards structure (Figure 13b). Thus, the particle size and viscosity of TP/BT-WDFs decreased with increase in the dosage of TP (Figures 8a and 9a). At high temperature, as shown in Figure 13c, the bentonite in pure BT-WDFs dehydrated, aggregated, and settled through face-to-face (FF) attraction (Figure 14c). The viscosity of pure BT-WDFs decreased and fluid loss increased sharply. As exhibited in Figure 13d, the addition of TP dispersed bentonite from FF attraction to EF attraction (Figure 14d). The house-of-cards structure was recovered and the particle size of bentonite increased and the viscosity of bentonite dispersion decreased sharply, leading to a high fluid loss. The addition of TP dispersed bentonite from FF attraction to EF attraction. Then, the house-of-cards structure was recovered and the viscosity of TP/BT-WDFs was increased. As a result, a compact and thin filtration cake was formed and the fluid loss was greatly reduced.

3.6. Cost Implication. The prices of different fluid loss additives were investigated and compared (as shown in Table 3). Among these additives, CMS had the lowest price, while TP was the most expensive. However, in the above study, TP showed better filtration property than CMS and PAC at the same concentration. The sulfonated phenolic resin (SMP) was also a common fluid loss additive in the oilfield. However, this material was restricted in use because of its toxicity. From the investigation, although TP had good filtration property and was environmentally friendly, it still had the problem of high cost.

4. CONCLUSIONS

In this work, TP was utilized as an effective and environmentally friendly fluid loss additive in BT-WDFs. The addition of TP could effectively decrease the fluid loss at room temperature and high temperature (~150 °C), displaying better filtration property than common fluid loss agents. Moreover, based on the investigations on the viscosity, the particle size distribution of TP/BT-WDFs, and micromorphology of filter cakes, the filtration mechanism of TP was also proposed. The dispersion effect of TP was predominant for the filtration property of TP/BT-WDFs. At room temperature, the addition of TP further promoted the dispersion of bentonite, decreased the particle size of bentonite, and increased the particle size distribution range. The house-of-cards structure of bentonite was weakened, leading to a decrease of the viscosity. At high temperature, pure bentonite dehydrated and aggregated through FF attraction. The particle size of bentonite increased and the viscosity of bentonite dispersion decreased sharply, leading to a high fluid loss. The addition of TP dispersed bentonite from FF attraction to EF attraction. Then, the house-of-cards structure was recovered and the viscosity of TP/BT-WDFs was increased. As a result, a compact and thin filtration cake was formed and the fluid loss was reduced. Therefore, TP was expected to be used as an effective and biodegradable fluid loss reducer in well-drilling applications. Meanwhile, it still has some deficiencies. TP is more expensive than some conventional fluid loss control materials, such as PAC and CMS. Besides, TP may decay in drilling fluids and affect the durability of its effect. Considering these concerns, it may be better to use TP in combination with other fluid loss agents and fungicides.

Table 3. Prices of Different Fluid Loss Additives

| additive | CMS | PAC | SMP | TP |
|----------|-----|-----|-----|----|
| price (yuan/t) | 8500 | 15000 | 12000 | 45000 |

Figure 13. Schematic illustrations for the formation of the filter cakes from BT-WDFs and TP/BT-WDFs at room temperature and high temperature.

Figure 14. TEM micrographs of pure BT-WDFs (a) and TP/BT-WDFs (b) at room temperature and pure BT-WDFs (c) and TP/BT-WDFs (d) at 150 °C. The concentration of TP was 3.0%.
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Notes
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