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Article

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Research paper

Influence of shear on rheology of the crude oil treated by flow improver

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ABSTRACT

Because the influence of shear on the rheology of waxy crude oil must be considered according to the composition and structure of the waxy crude oil transported via pipeline when a new pipeline is constructed, the risks associated with changes in the waxy crude oil composition during the pipeline operation must be considered in advance. To provide a policy-making basis for designers of new pipelines and managers of existing pipelines, the wax appearance temperature, crude oil components, wax structure and wax crystal morphology were analyzed by differential scanning calorimetry (DSC), gas chromatography (GC), proton nuclear magnetic resonance (¹H-NMR) and polarization microscopy. The influence of shear on the rheology of waxy crude oil treated by a flow improver and the relationships among this influence and the waxy crude oil composition, the wax structure and the shear temperature were investigated. The results indicated that above the wax appearance temperature, shear does not affect the rheology of waxy crude oil treated by a flow improver, whereas below the wax appearance temperature, the influence of pipeline shear on the rheology of waxy crude oil is related to the composition of the waxy crude oil and wax structure, which can improve the rheology of certain crude oils but can also deteriorate the rheology of other crude oils. However, pump shear can deteriorate the rheology of crude oil treated by a flow improver. The main reasons for the abovementioned phenomena are that a large number of wax crystals in the crude oil and shear changes the structures of the larger wax crystals formed by eutectic interactions between the flow improver molecules and wax molecules below the wax appearance temperature. In summary, this study provides a reference value for flow improvement research, pipeline design and pipeline management.

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1. Introduction

The heating of transported crude oil is often performed during pipeline transportation, although the consumption of fuel oil and the transportation costs are high; therefore, waxy crude oil is modified by the addition a flow improver as an energy-saving and cost-reducing measure. Great progress has been made in flow improver research, and various types of flow improvers with various structures have been developed. The widely used types are linear polymers and comb-shaped polymers; the former's functional unit is a polymeric backbone, such as ethylene-vinyl acetate copolymer (Vieira et al., 2012) and ethylene-vinyl acetate-sodium allyl sulfonate terpolymer (Wang et al., 1989), while the latter's functional unit is appended to the backbone of the

polymer, such as polyoctadecyl methacrylate (Soldi et al., 2007), octadecyl acrylate-maleic anhydride copolymer (Du et al., 2012), acrylate-maleic anhydride copolymer and its derivatives (Nassar and Ahmed, 2010), alkyl linoleate-maleic anhydride copolymer and its derivatives (Soliman et al., 2018), octadecene-maleic anhydride copolymer (El-Ghazawy et al., 2014), styrene-maleic anhydride-octadecyl acrylate terpolymer and its derivatives (Liu et al., 2017; Li et al., 2016), polyesters with long alkyl chains (Deka et al., 2018; Quan et al., 2018).

In the process of waxy crude oil transport via a pipeline, the oil inevitably suffers from pipeline shear for an extended length of time as well as pump shear for short periods. The shear history has a large influence on the rheology of the waxy crude oil; therefore, during the process of researching and applying a flow improver, it is important for the flow improver to have a suitable static modification effect on the waxy crude oil; however, its shear-resisting performance is more important because this factor affects the stability of the modification effect on the waxy

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crude oil along the entire pipeline and is related to the safe operation of the pipeline.

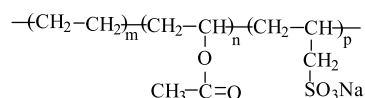
Previous studies investigated the effect of the shear history on the rheology of waxy crude oil treated by a flow improver. Yu et al. (2012) analyzed field test data and found that when the shear temperature is lower than the wax appearance temperature, pipeline shear and pump shear cause a rise in the pour point of Tuha crude oil. Zhang et al. (1996), performed an emulsion experiment and found that pipeline shear and pump shear caused a rise in the pour point and viscosity of No. 10 Xinjiang mixed crude oil and showed that the effect of pipeline shear exceeded that of pump shear. A study by Xia and Zhang (2001), indicated that pipeline shear deteriorated the rheology of a mixed crude from Qijuling and Lunku. A study by An and Qiao (2017) revealed that pump shear deteriorated the rheology of three Dingjing pipeline crude oils. However, the abovementioned works investigated the effect of shear on the rheological properties of only one crude oil and did not analyze the relationship between the shear effect and the crude oil composition. The influence mechanism of shear on the rheology of waxy crude oil has not been summarized.

One objective of this study is to provide a policy-making basis for new pipeline construction. When a new pipeline is designed, it is necessary to study the influence (especially the deterioration action) of shear action on the crude oil rheology based on an analysis of the crude oil composition and structure. Another objective is to provide a policy-making basis for managers of existing pipelines. Since the composition of the crude oil transported by a pipeline (different sources of imported crude oil) from port to inland usually changes, the risks of crude oil composition changes to the pipeline operation must be considered in advance. To achieve this goal, this study investigated the influence of shear on the rheology of four typical waxy crude oils and the relationships among this influence and the composition, wax structure and shear temperature. The rule summarized in this study is not absolute and exceptions may be identified with further research; however, this rule can still serve as a reference for policy-makers.

2. Experiment

2.1. Flow improver and waxy crude oil

The structural formula of the flow improver used in the experiments is as follows:



where $m = 550\text{--}700$, $n = 280\text{--}450$ and $p = 150\text{--}350$.

The flow improver is a linear polymer, and its functional unit provides nucleation sites, and cocrystallization with wax occurs in the polymeric backbone. The ester carbonyl and sulfonic groups are dissimilar to the wax crystals and block the extensive growth of wax crystals with a net-like structure.

The four types of waxy crude oils used in the experiment are obtained from the Luning pipeline, Xinjiang-out pipeline, Kushan pipeline and Lunku pipeline.

2.2. Analysis of the waxy crude oil composition

The composition of the waxy crude oil was determined by the Chinese Petroleum and Natural Gas Industry standard SY/T7550-2012. The results are shown in Fig. 1.

Fig. 1 shows that the wax content of the waxy crude oil from the Luning pipeline is higher, while those of the other waxy crude oils are lower. The content ratios of resins-asphaltenes to wax in the waxy crude oils from the Luning pipeline and Lunku pipeline are relatively high and those of the other crude oils are lower.

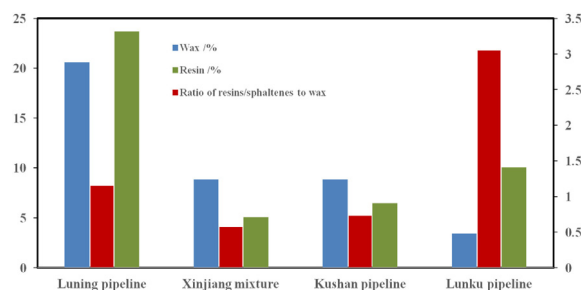


Fig. 1. Crude oil composition.

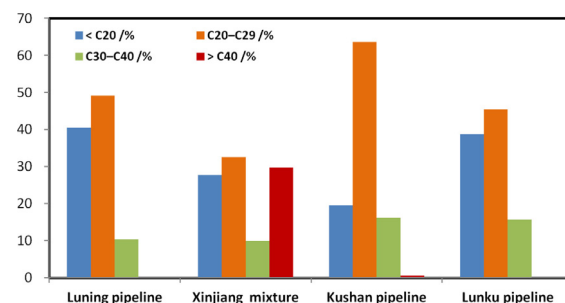


Fig. 2. Carbon number distribution of the wax.

Table 1
Branched-chain degree of the wax in the four crude oils.

| Wax | Luning pipeline | Xinjiang-out pipeline | Kushan pipeline | Lunlu pipeline |
|-----------------------|-----------------|-----------------------|-----------------|----------------|
| Branched-chain degree | 0.45 | 0.09 | 0.31 | 0.36 |

2.3. Analysis of the wax carbon number

The carbon number distribution of wax in the waxy crude oil was determined by the Chinese Petroleum and Natural Gas Industry standard SY/T5779-2008. The results are shown in Fig. 2.

Fig. 2 shows that the wax carbon number distribution of the Kushan pipeline crude oil is relatively concentrated ($C_{20}\text{--}C_{29}$ wax accounts for more than 60% of the total wax amount), the Xinjiang-out pipeline crude oil contains more high-carbon wax (wax with $>C_{40}$ accounts for more than 30% of the total wax amount), while the wax carbon number distributions of the other three crude oils are not concentrated and the content of high-carbon wax is extremely low.

2.4. Analysis of the wax branched-chain degree

The branched-chain degree is the ratio of CH_3 to CH_2 in the wax molecule and was measured by proton magnetic resonance ($^1\text{H-NMR}$). The results are listed in Table 1.

2.5. Differential scanning calorimetry (DSC) analysis of the crude oil

The DSC spectral curves of the samples (Fig. 3) were obtained by a DSC thermal instrument made by NETZCH corporation in Germany, and the wax appearance temperature and wax appearance peak temperature were obtained by the Chinese Petroleum and Natural Gas Industry standard SY/T0545-1995. The results are listed in Table 2.

Table 2
Wax appearance temperature and wax appearance peak temperature.

| | Luning pipeline | Xinjiang-out pipeline | Kushan pipeline | Lunku pipeline |
|-------------------------------------|-----------------|-----------------------|-----------------|----------------|
| Wax appearance temperature /°C | 33.96 | 23.13 | 22.07 | 27.14 |
| Wax appearance peak temperature /°C | 23.22 | 8.11 | 9.49 | 7.23 |
| Average latent heat /J/g | 9.43 | 2.75 | 10.41 | 2.84 |

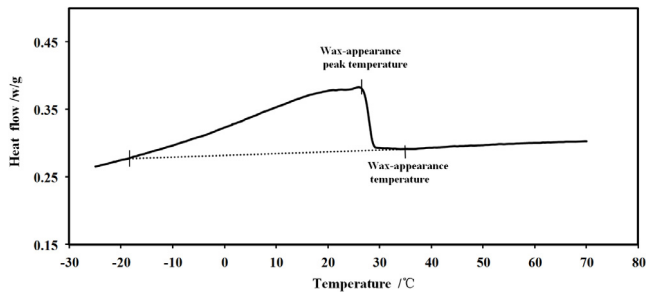


Fig. 3. DSC spectra of the crude oil.

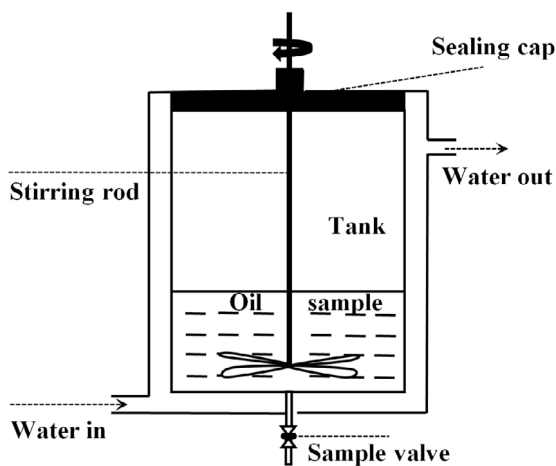


Fig. 4. Sealed tank schematic.

2.6. Emulsion experiment

Experimental installation. The experimental device (Fig. 4) consists of a well-sealed cylindrical tank with a water bath jacket (inner diameter of 20.2 cm, stainless steel), an IKA agitator (the agitator propeller has 4 blades, the blade width is 3.6 cm, the inclination angle is 45° and the diameter is 14 cm) and a Hakke programmed cooling water bath.

Emulation of pipeline shear. The pipeline shear rate is generally $10\text{--}70\text{ s}^{-1}$, and the computation of the stirring speed (80–120 rotations/min) is performed according to the method provided by Zhang et al. (2002, 2003a) to emulate pipeline shear ($20\text{--}40\text{ s}^{-1}$).

Emulation of pump shear. The pump shear rate is generally $2000\text{--}15000\text{ s}^{-1}$, and the computation of the stirring speed (6600–12000 rotations/min) is performed according to the method provided by Zhang et al. (2002, 2003a) to emulate pump shear ($3000\text{--}6500\text{ s}^{-1}$).

Emulation procedures (Fig. 5). First, crude oil was added to the well-sealed cylindrical tank. The height of the oil sample in the tank was 9–15 cm, and the blade was 1 cm from the bottom of the tank. The crude oil was maintained at the temperature of the crude oil at the first station for 30 min. Samples were collected, the pour point was determined, and wax crystals (virgin crude

Table 3
Emulsion experimental conditions.

| (a) Temperature of the crude oil at the transportation station | | | | |
|---|-----------------|-----------------------|-----------------|----------------|
| | Luning pipeline | Xinjiang-out pipeline | Kushan pipeline | Lunku pipeline |
| First station | 63 °C | 35 °C | 35 °C | 55 °C |
| Second station | 40 °C | 25 °C | 25 °C | 35 °C |
| Third station | 30 °C | 9 °C | 12 °C | 8 °C |
| (b) Flow time of the crude oil from one station to the next station | | | | |
| | Luning pipeline | Xinjiang-out pipeline | Kushan pipeline | Lunku pipeline |
| From the first station to the second station | 40 h | 20 h | 48 h | 50 h |
| From the second station to the third station | 46 h | 40 h | 72 h | 40 h |

oil) were observed. Then, a flow improver was added and stirred for 30 min to mix the flow improver fully with the crude oil; then, samples were collected, and the pour point was determined. Second, the cooling program of the Hakke programmed cooling water bath was compiled under the dynamic condition of pipeline shear emulsion, and the temperature of the crude oil was reduced from the temperature of the crude oil at the first station to the temperature of the crude oil at the second station for a certain time (flow time of the crude oil from the first station to the second station, Table 3); then, samples were collected, and the pour point was determined. Third, pump shear emulsion was used; samples were taken, and the pour point was determined. Fourth, the cooling program of the Hakke programmed cooling water bath was compiled under the dynamic condition of pipeline shear emulsion, and the temperature of the crude oil was reduced from the temperature of the crude oil at the second station to the temperature of the crude oil at the third station for a certain time (flow time of the crude oil from the second station to the third station, Table 3); then, samples were collected, the pour point was determined, and wax crystals were observed. Fifth, pump shear was emulsified, samples were collected, the pour point was determined, and wax crystals were observed.

The emulsion experimental conditions are listed in Table 3.

2.7. Measurement of the pour point

The evaluation indexes of the crude oil rheology include the pour point, viscosity and yield value. For a waxy crude oil, the pour point has a positive relationship with the viscosity and yield value and can represent the rheology of the waxy crude oil.

The pour point was determined according to the Chinese Petroleum and Natural Gas Industry standard SY/T0541-2009.

Figs. 6–9 show the pour points after adding 40 mg/kg, 100 mg/kg, 30 mg/kg or 40 mg/kg flow improvers to the crude oils from the Luning pipeline, Xinjiang-out pipeline, Kushan pipeline and Lunku pipeline, respectively, and the influences of pipeline shear and pump shear on the crude oil pour points (the inlet pour point at the first station is the pour point of the virgin crude oil).

In Fig. 6, the pour point of the Luning pipeline is obtained from field test data. The pour point before/after pipeline shear is exactly the pour point when the crude oil flows out of a station and flows into the next station, and the pour point before/after pump shear is exactly the pour point when the crude oil flows into/out of a station. The emulsion experimental data and field test data from the Luning pipeline are consistent (Zhang et al., 1999a), thus demonstrating that the emulsion experimental data are reliable.

In Figs. 7–9, the pour points of the three other crude oils are obtained from the emulsion experiments.

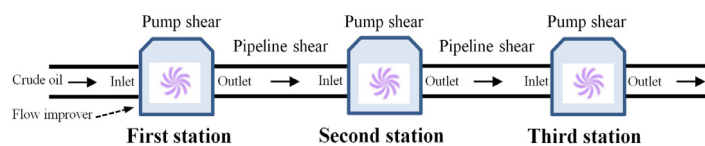


Fig. 5. Emulsion procedures.

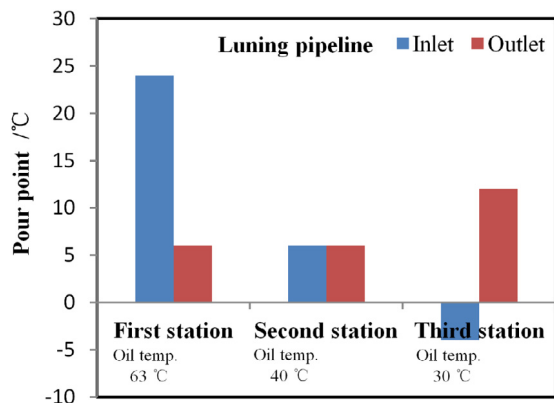


Fig. 6. Pour point of the crude oil at the inlet and outlet of the transportation station of the Luning pipeline.

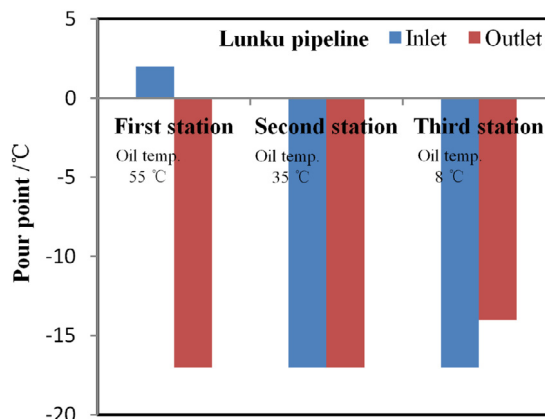


Fig. 9. Pour point of the crude oil at the inlet and outlet of the transportation station of the Lunku pipeline.

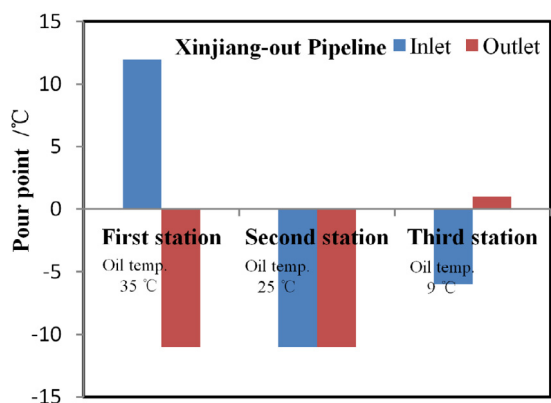


Fig. 7. Pour point of the crude oil at the inlet and outlet of the transportation station of the Xinjiang-out pipeline.

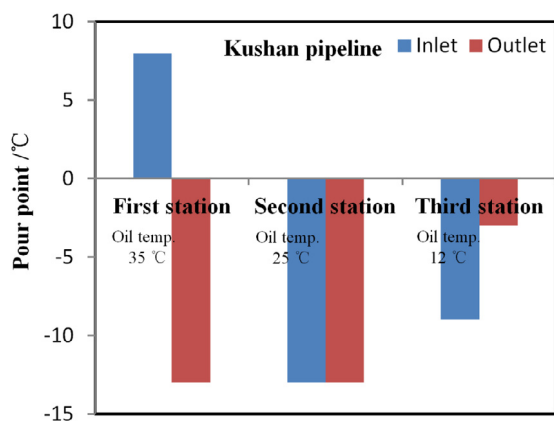
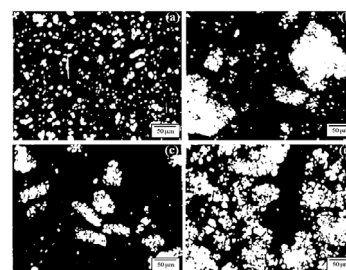
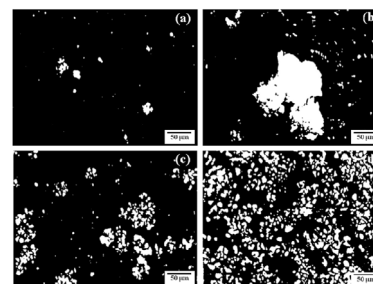


Fig. 8. Pour point of the crude oil at the inlet and outlet of the transportation station of the Kushan pipeline.



(A) Virgin crude oil; (B) treated crude oil (before shear); (C) treated crude oil (after shear); and (D) treated crude oil (recombination after shear)

Fig. 10. Microscopic images of the wax crystals in the Luning pipeline crude oil at 30°C (treated with 40 mg/kg flow improver).



(A) Virgin crude oil; (B) treated crude oil (before shear); (C) treated crude oil (after shear); and (D) treated crude oil (recombination after shear)

Fig. 11. Microscopic images of the wax crystals in the Xinjiang-out pipeline crude oil at 9°C (treated with 100 mg/kg flow improver).

2.8. Observation of wax crystals

Observation of the wax crystals could be performed by polarization microscopy, and photographs of the wax crystals could be taken. The results are shown in Figs. 10 and 11.

3. Results and discussion

3.1. Effects of resins-asphaltenes and carbon number distribution on rheology

Researchers worldwide have conducted insightful studies on the pour point reduction mechanism, and it is commonly believed that the eutectic interactions between the wax and flow improver play a main role in pour point reduction (Zhang et al., 2003b; Coto et al., 2014; Chen et al., 2010). The flow improver has a higher molecular weight than the wax, and eutectic interactions occur between the flow improver and wax, during which multiple crystal nuclei are formed on the molecular chain of the flow improver. With enlarged wax crystals, the quantity of wax crystals per unit volume of crude oil declines and the crude oil system stabilizes; therefore, the pour point of the crude oil decreases.

Resin and asphaltene chemically contains condensed aromatic rings, heteroatoms, often heavy metals, and aliphatic chains. Certain studies (Kriz and Andersen, 2005; Tinsley et al., 2009; Venkatesan et al., 2003) have reported that resins and asphaltenes act on wax crystals to change the wax crystal structure during crude oil wax crystallization; therefore, they are natural flow improvers and can significantly decrease the pour point, viscosity and yield stress. Furthermore, Long et al. (2012) thought that the agglomerates formed by resins, asphaltenes and flow improvers became the efficient nucleator of the wax crystal and were able to change the process of wax crystallization and greatly depress the pour point of the additive-treated crude oil; therefore, resins and asphaltenes can coordinate flow improvers to improve the rheology of a waxy crude oil.

Wax is high molecular weight n-paraffin, the solubility of which decreases as the temperature decreases. Wu et al. (2014) and Zhang et al. (1999b) thought that the carbon number distributions of wax in crude oils had a large influence on their pour points, and wax with a concentrated carbon number distribution could result in the concentrated precipitation of wax crystals and a poor response to the flow improver. García et al. (2001), García and Urbina (2003) found that the n-paraffin chain length was correlated with the crude oil pour point and indicated that a narrower molecular weight distribution of wax in the crude oil corresponded to a higher wax crystallization risk.

3.2. Influence and mechanism of pipeline shear on the rheology of crude oil

Figs. 6–9 show that above the wax appearance temperature (from the outlet of the first station to the inlet of the second station), pipeline shear has no effect on the rheology of the four kinds of crude oil because only a very small amount of wax crystals precipitates from the crude oil above the wax appearance temperature and pipeline shear has little effect on the wax crystal structure.

Fig. 6 shows that below the wax appearance temperature, pipeline shear can improve the rheology of the Luning pipeline crude oil. After 46 h of pipeline shear in the pipeline (from the outlet of the second station to the inlet of the third station, the temperature of the crude oil falls from 40 to 30 °C), the pour point of the Luning pipeline crude oil falls by 10 °C (from 6 to –4 °C). The main reasons why pipeline shear can improve the rheology of crude oil are as follows: the Luning pipeline crude oil contains no high-carbon wax, the carbon number distribution of wax is scattered (Fig. 2), and the contents of resins-asphaltenes as natural flow improvers are high. In this case, pipeline shear can promote the synergistic action of the resins-asphaltenes and flow improver to change the wax crystal structure (Kriz and Andersen, 2005; Tinsley et al., 2009; Venkatesan et al., 2003; Long et al., 2012), so that larger wax crystals are easily formed.

Fig. 7 shows that below the wax appearance temperature, pipeline shear deteriorates the rheology of the Xinjiang-out pipeline crude oil. After 40 h of pipeline shear (from the outlet of the second station to the inlet of the third station, the temperature of the crude oil falls from 25 to 9 °C), the pour point of the Xinjiang-out pipeline crude oil rises by 5 °C (from –11 to –6 °C). The reason for this increase is that the Xinjiang-out pipeline crude oil contains more high-carbon wax (Fig. 2), which has a low branched-chain degree (Table 1), and the crude oil is almost entirely composed of normal paraffin. Researchers have found that wax tends to form smaller needle-like wax crystals (Ganeeva et al., 2010; Zhang et al., 2014) that are stacked together to form large inclusions with a relatively low structural strength. Therefore, although the pipeline shear strength is sufficient to break the inclusions with needle-like wax crystals (the force between the needle-like wax crystals and their surroundings is relatively low) to form new small wax crystals with active centers (Yi and Zhang, 2011), the flow improver cannot affect the newly formed active centers and cannot alter the structure of the wax crystals. In addition, the contents of resins-asphaltenes that act as natural flow improvers are low and the synergistic action between the resins-asphaltenes and flow improver is low (Kriz and Andersen, 2005; Tinsley et al., 2009; Venkatesan et al., 2003; Long et al., 2012), which results in a higher pour point of the crude oil.

Fig. 8 shows that below the wax appearance temperature, pipeline shear deteriorates the rheology of the Kushan pipeline crude oil. After 96 h of pipeline shear (from the outlet of the second station to the inlet of the third station, the temperature of the crude oil falls from 25 to 12 °C), the pour point of the crude oil of the Kushan pipeline rises by 4 °C (from –13 to –9 °C). The reason for this increase is that the content ratio of resins-asphaltenes to wax in the crude oil of the Kushan pipeline is relatively low and the synergistic action between the resins-asphaltenes and flow improver is low (Kriz and Andersen, 2005; Tinsley et al., 2009; Venkatesan et al., 2003; Long et al., 2012). In particular, the carbon number distribution of wax is relatively concentrated (Fig. 2). When the temperature of the crude oil falls to below the wax appearance temperature, the supersaturation of wax molecules is high and the wax appearance rate is also high (Wu et al., 2014; Zhang et al., 1999b; García et al., 2001; García and Urbina, 2003). At this point, the pipeline shear action will promote the appearance of wax crystals and the formation of crystal nuclei but does not favor wax crystal growth. There is not enough time for the flow improver to interact with the rapidly generated wax crystals, resulting in the formation of a large number of small wax crystals. Small wax crystals easily interconnect, thereby forming a network structure with a higher structural strength, and the pour point of the crude oil greatly increases.

Fig. 9 shows that below the wax appearance temperature, pipeline shear has no effect on the rheology of the Lunku pipeline crude oil. The reason is that the wax content in the Lunku pipeline crude oil is relatively low (Fig. 1), and even below the wax appearance temperature, there are fewer wax crystals in the crude oil (Wu et al., 2014; Zhang et al., 1999b; García et al., 2001; García and Urbina, 2003).

3.3. Influence and mechanism of pump shear on the rheology of the crude oil

Figs. 6–9 show that above the wax appearance temperature (at the second station), pump shear has no effect on the rheology of the four kinds of crude oil. The reason is that only a very small amount of wax crystallization occurs in the crude oils above the wax appearance temperature, and pump shear has little effect on the wax crystal structure.

Table 4
Influence of shear on the viscosity of the flow improver diesel solution.

| Concentration of the flow improver /wt.% | 1 | 3 | 5 |
|---|------|------|-------|
| Solution viscosity before shear (35 °C/cst) | 3.89 | 6.86 | 12.19 |
| Solution viscosity after shear (35 °C/cst) | 3.89 | 6.86 | 12.19 |

Figs. 6–8 show that below the wax appearance temperature (at the third station), pump shear deteriorates the rheology of the Luning pipeline, Xinjiang-out pipeline and Kushan pipeline crude oils, and after pump shear, the pour points increase by 16 °C (from –4 to 12 °C), 7 °C (from –6 to 1 °C) and 6 °C (from –9 to –3 °C), respectively. The main reason for the significant increases in the pour points of the crude oils after pump shear is not that the molecular chain of the flow improver breaks under shear, which is based on the fact that the flow improver is a low-molecular-weight polymer and its molecular structure is not easily damaged under shear. The flow improver was prepared with a 5% diesel solution and loaded into an airtight container for 10 h of shear. The viscosity before/after shear was measured by a viscometer, and no change in the viscosity was found (Table 4).

For a long period of high-strength shear, the molecular chain of the flow improver is not broken, and the amount of flow improver added to the crude oil is only a few tens of mg/kg. Therefore, it is very unlikely that pump shear will result in chain breakage of the flow improver. The repeated heating of the oil sample after shear to the initial heat treatment temperature followed by restoration of the pour point to the level before shear indirectly indicate that pump shear cannot lead to breakage of the molecular chain of the flow improver.

After pump shear, the main reason for the sharp rise in the pour point of the crude oil is that below the wax appearance temperature, there are a large number of wax crystals in the crude oil and the larger wax crystals formed under the action of the flow improver can be broken (Figs. 10 and 11) to form small wax crystals with new profiles after shear (Yi and Zhang, 2011). The newly formed profile is that of the new active center, and the flow improver cannot be redistributed, which means that the flow improver is unable to react with the new active center to change the wax crystal structure. In this case, as the temperature decreases, wax molecules will continue to appear and adsorb on the active centers of wax crystals, and the wax crystals will continuously grow. At the same time, the wax crystals form an interconnected network with a structure, which encloses the light components of the crude oil (Yi and Zhang, 2011), so that the crude oil flows less easily at a higher temperature, thus resulting in a higher pour point of the crude oil.

Fig. 9 shows that below the wax appearance temperature (at the third station), the increasing magnitude (only 3 °C, from –17 to –14 °C) of the pour point of the Lunku pipeline crude oil is lower than those of the three other kinds of crude oil after pump shear. The reason for this finding is that the wax content in the Lunku pipeline crude oil is relatively low (Fig. 1), and even below the wax appearance temperature, there are fewer wax crystals in the crude oil (Wu et al., 2014; Zhang et al., 1999b; García et al., 2001; García and Urbina, 2003), which does not contain high-carbon wax (Fig. 2). In addition, the content ratio of resins-asphaltenes, which act as natural flow improvers, to wax in the crude oil is high (Fig. 1). The synergistic action between the resins-asphaltenes and flow improver can change the structures of the wax crystals and promote the formation of large crystal aggregates (Kriz and Andersen, 2005; Tinsley et al., 2009; Venkatesan et al., 2003; Long et al., 2012); therefore, pump shear has little effect on the rheology of the Lunku pipeline crude oil.

4. Conclusions

Above the wax appearance temperature, pipeline shear has no effect on the rheology of crude oil, whereas below the wax appearance temperature, the effect of pipeline shear on the rheology of crude oil is related to the composition of the crude oil and the wax structure. In other words, for crude oil with a high content ratio of resins-asphaltenes to wax, shear can improve the rheology of the crude oil, whereas for crude oil with high-carbon wax or wax with a concentrated carbon number distribution, shear can deteriorate the rheology of the crude oil.

Above the wax appearance temperature, pump shear has no effect on the rheology of the crude oil. Below the wax appearance temperature, pump shear can deteriorate the rheology of the crude oil and the influence degree is related to the crude oil composition and wax structure.

Data availability

The raw/processed data required to reproduce these findings cannot be shared at this time because the data also form part of an ongoing study.

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