



Research article

Resource utilization of expired progesterone medicines as flow improver for waxy crude oils

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ABSTRACT

At present, the treatment of expired medicines mainly involves burning, which means waste of resources and carbon dioxide emissions, and it does not comply with the concept of resource recycling. In this study, in order to explore the resource utilization pathways of expired medicines, progesterone drugs were evaluated as crude oil flow improvers as an example. The results shows that progesterone injection (PI) and progesterone capsule (PC) both act as viscosity reducer and pour point depressant in different crude oil, and 500 ppm PI and 300 ppm PC are the best dosage respectively. 500 ppm PI can reduce the viscosity of HN oil sample by 60.40%, and depress the pour point by 8.5 °C. 300 ppm PC can reduce the viscosity of HN oil by 54.7%, and depress the pour point by 10.9 °C. Furthermore, through DSC and wax crystal morphology analysis, the possible mechanism of this expired medicine as crude oil flow improver was further discussed. Finally, considering the costs of concentration, transportation, treatment, processing and other links, the possible cost of crude oil flow improver was summed up, and its market feasibility was analyzed. This study provides a reference case for the resource utilization of expired medicines.

1. Introduction

In recent years, with the continuous enhancement of public health awareness, more and more people are accustomed to storing medicines at home, and the production of expired medicines is naturally more. According to the Appendix "Exemption Management List for Hazardous Wastes" of the National Catalogue of Hazardous Wastes (2021 Edition) in China, expired medicines generated in household daily life or activities providing services for daily life are classified as hazardous waste in household waste. In recent years, some local government departments, pharmaceutical companies, and retail pharmacies have often carried out centralized recycling activities for expired medicines in households. It is recommended that families with conditions can send expired medicines to these formal recycling points for recycling and unified harmless treatment. The expired medicines collected centrally require harmless treatment (GB 39707-2020, 2021). At present, the treatment methods for expired medicines include incineration, landfill and mixed with domestic garbage treatment. There are four common treatments, incineration treatment, chemical system treatment, alkaline treatment,

and biological treatment (high temperature biodegradation). Comparing the treatment methods, it is clear that they possess different advantages and disadvantages. Of these, high operating costs and energy consumption emerge as the most prominent challenges to achieving environmentally sound treatment. According to data, China's annual production of expired medicines is about 15,000 tons. If the medicine is not handled properly (Toh and Chew, 2017), its toxicity and corrosiveness will not only endanger the health and life safety of the public but also pollute water sources, land, and destroy the biological chain. Furthermore, the large amount of fuel consumed during the treatment process will also aggravate carbon emissions and bring immeasurable threats to society (Zhang et al., 2022). According to the United Nations Intergovernmental Panel on Climate Change (IPCC), the global surface temperature from 2011 to 2020 increased by 1.09 °C. Compared with the industrial revolution, the highest level in about 125,000 years, and it is the greenhouse gas emissions caused by the burning of fossil fuels such as coal and natural gas. Therefore, the resource utilization of expired medicines is a green and efficient way to deal with such problems. From the perspective of global resource management, the cost of recovery,

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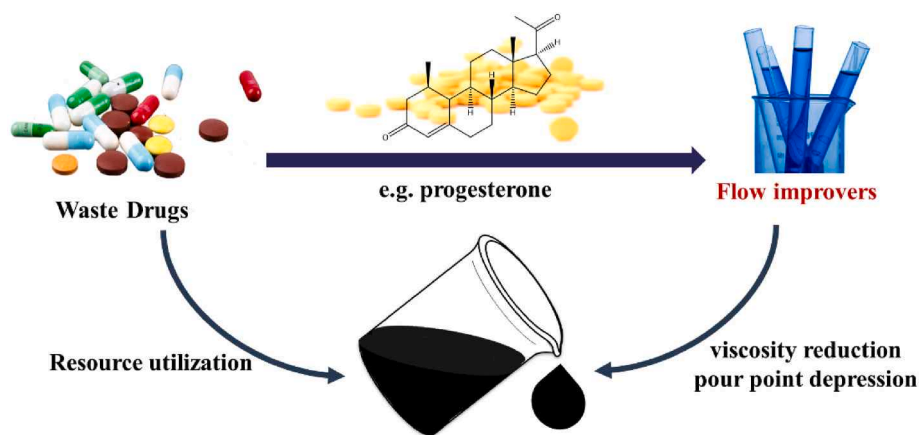


Fig. 1. Resource utilization scheme for expired progesterone medicines.

treatment, and utilization of expired medicines is immeasurable. Various countries have different recycling methods, and even some underdeveloped countries have not established medicine recovery systems. The most direct approach to reducing expired emissions into the environment is to decrease the production quantity of pharmaceuticals. Nevertheless, achieving this reduction directly is challenging. Therefore, considering the cost of medicine recovery and treatment, we should focus on maximizing the value of pharmaceuticals even after expiration (Abdel-Shafy and Mansour, 2018; Alexandridou et al., 2018).

The gradual increase in global energy demand and the depletion of low-viscosity crude oil have compelled numerous enterprises to invest in heavy crude oil production. However, the characteristics of heavy oil, high viscosity and poor fluidity, present significant challenges in subsequent exploitation and transportation. Therefore, during petroleum production, various crude oil liquidity improvers are employed to enhance the fluidity (Kumar et al., 2017). The most extensively used flow improvers for crude oils are ethylene–vinyl acetate copolymer (Machado André et al., 2001; Taraneh et al., 2008), alkyl ester of unsaturated carboxylic acid–olefin copolymer (Soldi et al., 2006), and the maleic anhydride alkyl ester of unsaturated carboxylic acid copolymer (Gu et al., 2020a; Song et al., 2005). These additives functioned by one or more of several postulated mechanisms, viz, nucleation, adsorption, cocrystallization, and improved wax solubility, which results in the formation of smaller wax crystals with more-regular shapes (Lv et al., 2020).

Progesterone is a first-line drug for clinical prevention and treatment of miscarriage, and the structure of its active ingredients is shown in Fig. 1. Progesterone contains non polar cycloalkyl structures and carbonyl groups similar to comb like copolymers used as crude oil flow improvers. Previous studies have shown that comb like copolymer crude oil flow improvers can inhibit the crystallization of paraffin components in crude oil, or weaken the network structure strength of wax crystals, thereby reducing the viscosity of crude oil. At the same time, these copolymers can change the shape of wax crystals that cannot be interlocked with each other, prevent asphaltene aggregation, and improve the flow performance of crude oil in pipelines. Based on the understanding of the structure of the two and the concept of waste resource utilization, In this study, progesterone drugs were evaluated as crude oil flow improvers as an example as shown in Fig. 1.

2. Experimental

2.1. Experimental reagents and materials

The names of crude oil samples were abbreviations of different oil fields or its block. HN, YL, GC oil samples from Henan oilfield, XJ oil samples from Xinjiang oilfield special zone. Diesel is purchased from

Table 1

Progesterone Injection and Progesterone Capsules list of ingredients.

Oil sample	Density/(g·cm ⁻³ , 25 °C)	Saturates /wt%	Aromatics /wt%	Asphaltenes /wt%	Resins /wt%
HN	0.944	37.78	30.21	14.25	17.76
YL	0.943	45.63	33.24	8.31	12.82
XJ	0.940	40.82	26.53	14.29	18.46
GC	0.931	31.96	29.9	17.44	20.7

Sinopec, and expired progesterone medicines are recycled from pharmacies. The physicochemical properties of the different oil samples are shown in Table 1. The progesterone injection solution with a dosage of 1 mL:20 mg is composed of the following weight percentages of components: progesterone at a range of 0.5–2%, injection oil 15–35%, emulsifier 1–12%, co-emulsifier 0.1–1%, stabilizer 0.2–2%, antioxidant 0.01–2.5%, and water as the remaining quantity. The 0.1 g progesterone capsule is composed of the following weight percentages: 40% progesterone, 59.6% rapeseed oil, and 0.4% soy lecithin.

2.2. Preparation of flow improver

Progesterone injection (hereinafter referred to as PI): each 1 mL PI contains 20 mg of progesterone, the excipient is injection oil, and 100, 200, 300, 400, 500 ppm flow improvers can be directly prepared to add oil samples. Progesterone capsule (hereinafter referred to as PC): After removing the capsule shell, 100, 200, 300, 400, and 500 ppm flow improver was prepared with diesel as the solvent and added to the oil sample.

2.3. Pour point evaluation

The pour point test was measured using the SY/T0520-2008 (SY/T0520-2008, 2008), The oil sample undoped or doped with PI/PC was placed in a 25 mL measuring cup, stood in a constant temperature cold bath for 30 min, then taken out, cooled at room temperature and tilted at 45 °C to observe the fluidity of the oil sample. The pour point of each sample was tested three times and the average value was defined as the final pour point.

2.4. Viscosity evaluation

According to SY/T0541-2009(SY/T0541-2009, 2009), the viscosity of oil samples at different temperatures was determined. Take a certain quality of oil sample and put it in a water bath at 60 °C, keep the temperature for 30min, take 25 g and put it in a measuring cup, put the measuring cup in the water bath at the temperature to be measured, and

Table 2
The effect of PI on depression of pour point.

Concentration (ppm)	HN	GC	XJ	YL
Blank	28.5	24.0	16.0	36.0
100	20.0	22.5	16.0	33.0
200	22.5	21.0	11.0	27.0
300	22.5	21.5	13.0	30.0
400	20.0	18.0	11.0	31.5
500	20.0	22.5	10.0	32.0

Table 3
The effect of PC on depression of pour point.

Concentration (ppm)	HN	GC	XJ	YL
Blank	28.5	24.0	16.0	36.0
100	23.7	22.1	10.3	28.5
200	25.0	22.8	10.3	28.1
300	17.6	22.5	9.0	28.6
400	18.0	21.7	5.1	27.7
500	20.2	22.9	5.5	28.5

use NDJ-8S rotary viscometer to determine the viscosity at the corresponding temperature.

2.5. Differential scanning calorimetry analysis

The differential scanning calorimetry (DSC) analysis of crude oil (Ahmadi et al., 2019), PC, and PI treated crude oil were performed using

a Mettler-Toledo DSC822e DSC apparatus. The chemicals, suppliers, and purities used for the DSC calibration were: *n*-C₇ (Scharlau, 99%), *n*-C₈ (Panreac, 99%), *n*-C₁₂ (Alfa-Aesar, >99%), *n*-C₁₆ (Alfa-Aesar, >99%), *n*-C₁₈ (Fluka, P99.8%) and Indium (Mettler Toledo). The temperature profile follows two steps: (1) Previous step: sample was heated from room temperature to 80 °C at 11 °C/min; (2) Cooling step: each sample was cooled down from 80 °C to -25 °C at 8 °C/min.

2.6. Optical microscopy

The saturated hydrocarbon component was separated from crude oil according to the method of SY/T 5119-2008(SY/T5119-2008, 2008) (Analysis of soluble organic matter in rocks and group components of crude oil). Wax crystal morphologies were studied using an OPTPro-3000 polarizing microscope. The heavy oil sample was separated from the saturated hydrocarbon components and then placed in a 20 °C constant temperature water bath for 1 h. The saturated hydrocarbon components were placed on a glass sheet, and a polarized light microscope was used to examine the morphology of wax crystal.

3. Results and discussion

3.1. Pour point of the treated crude oil

Pour point reflects the ease with which wax molecules form network structures. The effect of flow improvers on pour point of crude oil is presented in Table 2 and Table 3. Poly (benzyl oleate-maleic anhydride) pour point depressants is a popular flow improver (Elkatory et al.,

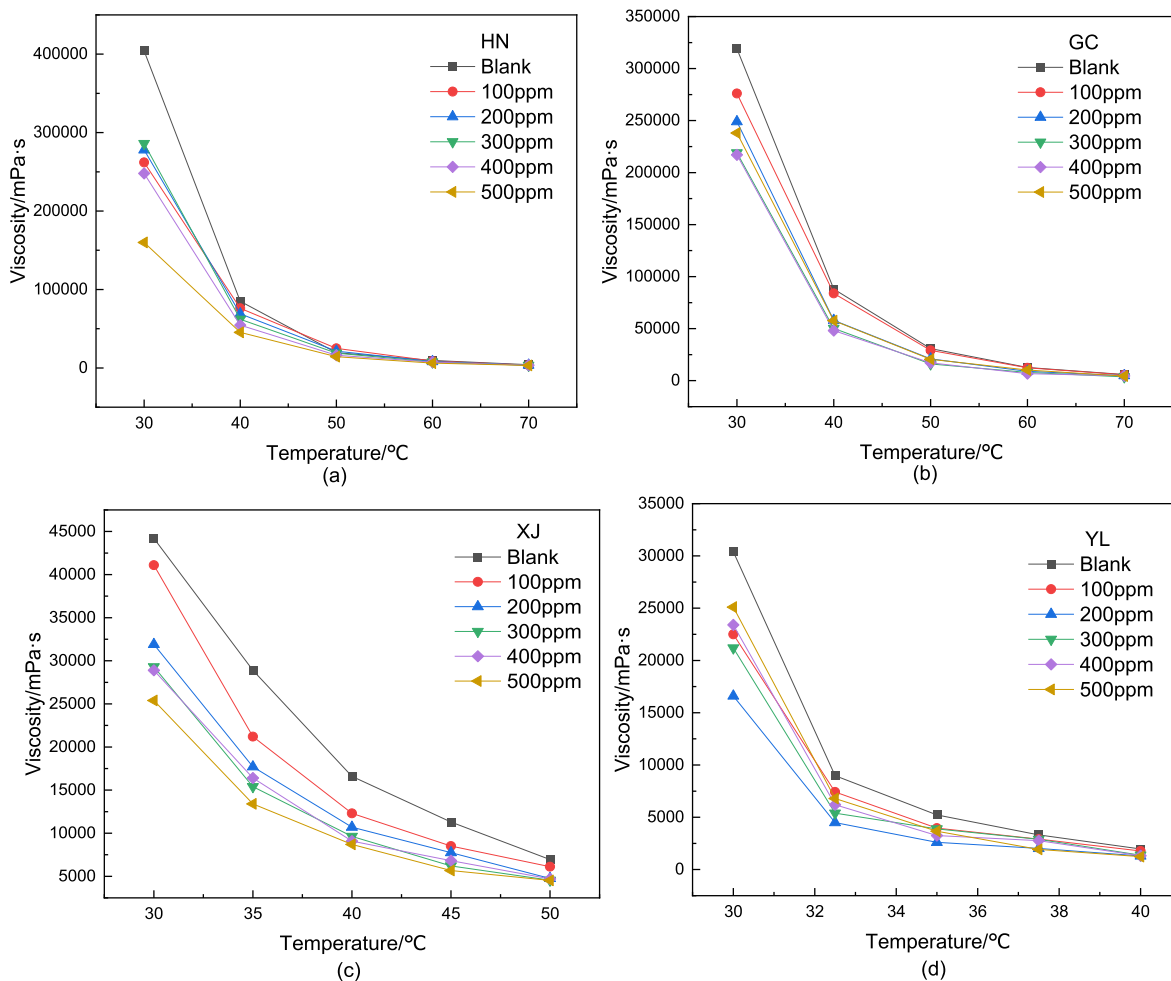


Fig. 2. The viscosity reduction effect of PI (a–d) and PC (e–h) on oil samples with different concentrations

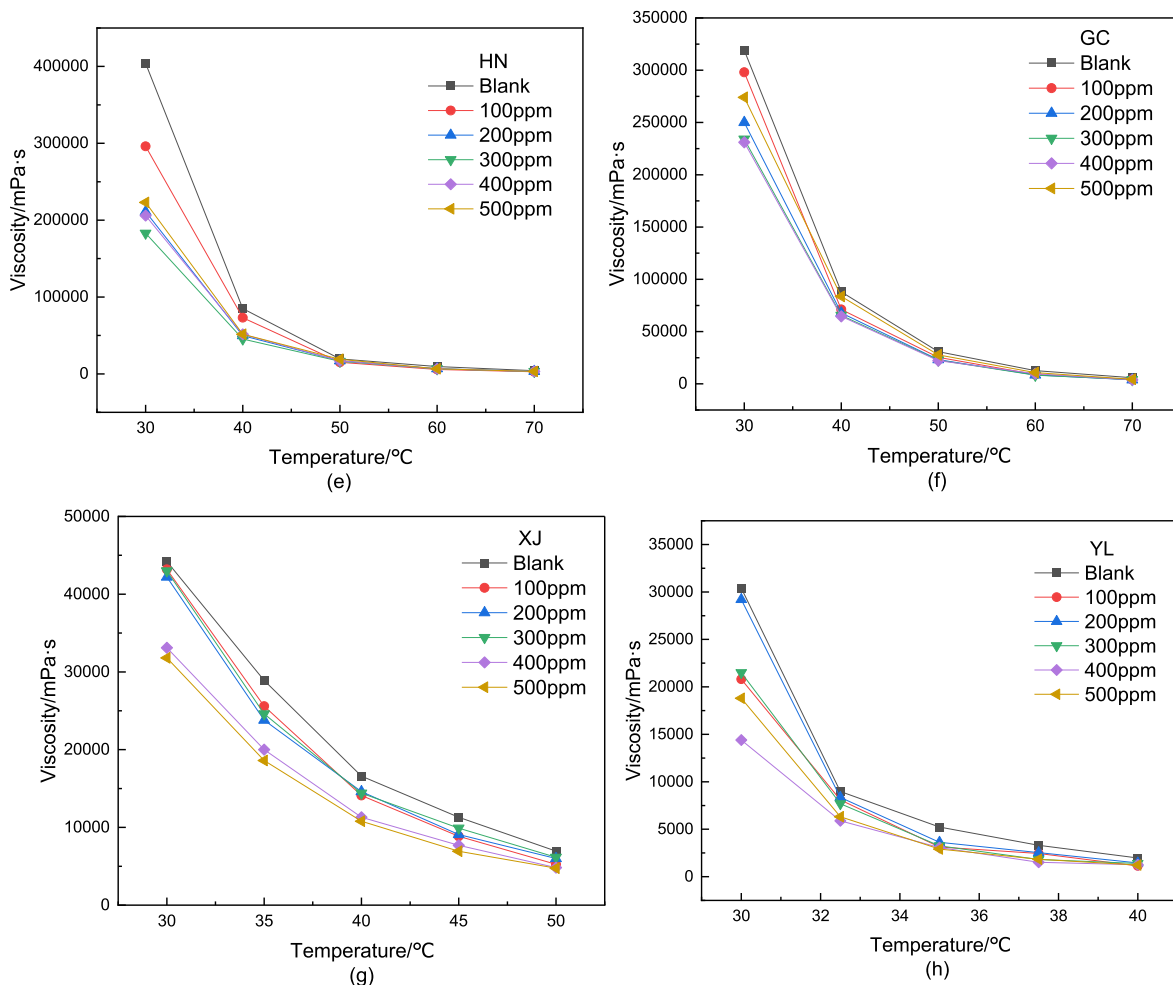


Fig. 2. (continued).

2023), which depresses the pour point of wax crude oil by 21 °C at a dose of 2000 ppm. It was shown that the polymer additive could hinder the formation of wax crystals and retard the growth of paraffin crystals. In this experiments, the concentration of PI flow improver has different anticoagulation effects on different oil samples, compared with PI flow improver, the effect of PC flow improver is more pronounced (Gao et al., 2019). With 300 ppm PC the pour point of XJ oil is depressed by 10.9 °C, the pour point of HN, GC and YL oil is depressed by 9.5 °C, 2.3 °C, 8.3, respectively, the coagulation effect of the flow improves increases with the increase of concentration, and after reaching the optimal concentration, the pour point increases slightly when the concentration is exceeded. This phenomenon is because the molecules in the flow improver co-crystallize with the wax molecules at the optimal concentration (Li et al., 2021), destroying the crystalline directionality of the wax crystals and not aggregating with each other, thereby increasing the fluidity of crude oil. Excessive flow improvers are embedded in wax molecules, which are not conducive to the eutectic properties of flow improvers and wax molecules (Zhou et al., 2022). These results are similar to those reported work (Elkatory et al., 2023).

3.2. Rheological properties of the treated crude oil

The effects of PI and PC flow improvers on different oil sample viscosity in a certain temperature range are shown in Fig. 2(a–d) and (e–h), respectively. As shown in Fig. 2, PI and PC flow improvers have different adhesive effects on different oil samples. At 30 °C, the viscosity reduction rate of 500 ppm PI to Henan oil samples reached 60.40%, compared

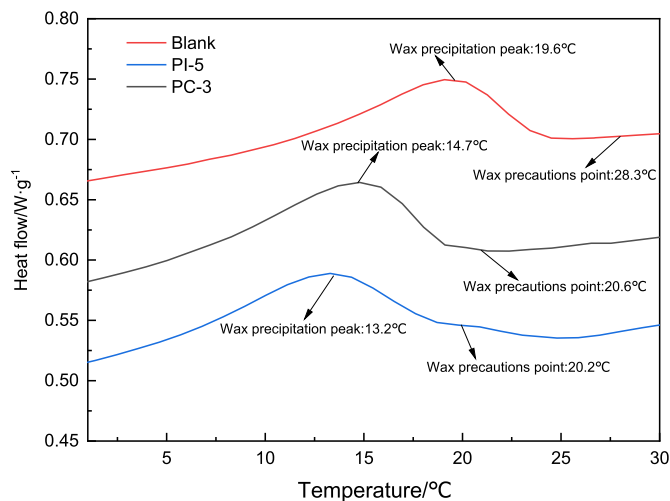


Fig. 3. The viscosity reduction effect of PC on oil samples with different dosage.

to PI, PC flow improver is more effective in reducing viscosity, at 30 °C, the viscosity reduction rate of 300 ppm PC has reached 54.70%. Obviously, 300 ppm PC can reduce the viscosity while saving the cost of the flow improvers.

In terms of the overall viscosity-temperature change trend, with the

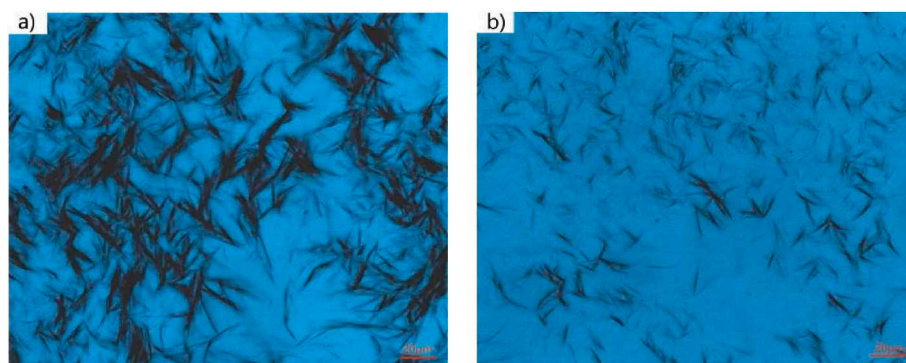


Fig. 4. Wax crystal in saturated hydrocarbon (a) without PI; (b) with PI.

temperature increases, the viscosity of various oil samples decreased significantly at different concentrations (Fang et al., 2012; Zhu et al., 2018), HN, GC, and XJ oil samples are between 30 and 40 °C and YL oil samples are between 30 and 32.5 °C. When the temperature is higher, the viscosity of crude oil with and without flow improver added is lower and almost the same. This indicates that the addition of flow improvers can significantly improve the cryogenic liquidity of crude oil (Tang et al., 2022).

3.3. DSC analysis

500 ppm PI and 300 ppm PC were added into the HN oil sample for DSC analysis respectively. As shown in Fig. 3, wax precautions point and wax precipitation peak in the control sample were 28.3 °C and 19.6 °C, respectively. With the addition of 300 ppm PC, HN oil sample's wax precautions point and wax precipitation peak drops to 20.6 °C and 14.7 °C, respectively. With the addition of 500 ppm PI, HN oil sample wax precautions point and wax precipitation peak drop to 20.2 °C and 13.2 °C, respectively. After adding 500 ppm PI and 300 ppm PC, the wax precautions point and wax precipitation peak of the oil sample shift to the lower temperature significantly. This indicates that the addition of PI and PC as flow improvers inhibits the wax precipitation, lowers the precipitation temperature of paraffin in the waxy crude oil, and reduces the pour point of the crude oil, resulting in improving oil fluidity even at lower temperature. It may be attributed to the modification of the flow

improver on the wax crystals, which reduces the tendency of initial wax crystals to aggregate into larger ones or even reticular structures, thereby reducing the viscosity of the crude oil with poor fluidity due to wax component at low temperature. (Elarbe et al., 2022).

3.4. Wax crystal morphology analysis

In order to study the microscopic changes of wax crystals with doped/undoped 500 ppm PI, microscopic observations were used to record their morphology (Gu et al., 2019). Fig. 4a shows that without PI the morphology of wax crystals is feathery, the wax crystals are arranged compactly and regularly but not dispersed, and the wax crystals have been aggregated into a dense wax crystal lattice to form the 3-D view. However, the addition of 500 ppm PI results in a transformation in the morphology and structure of the wax crystals, as illustrated in Fig. 4b. The mesh-like structure of the wax crystals became smaller, with refined needle-like morphology, increased dispersion, and reduced aggregation. This can be attributed to the influence of the crude oil flow improver on the crystallization behavior of the wax component. It alters the crystallization orientation and distribution pattern of wax crystals in crude oil, inhibits their aggregation and cross-linking growth, and disperses the wax component within the waxy crude oil. These macroscopic effects are manifested as a viscosity reduction at relative low temperature (Ren et al., 2022; Sun et al., 2018). Correspondingly, the pour point of crude oil will also decrease, which is consistent with the previous

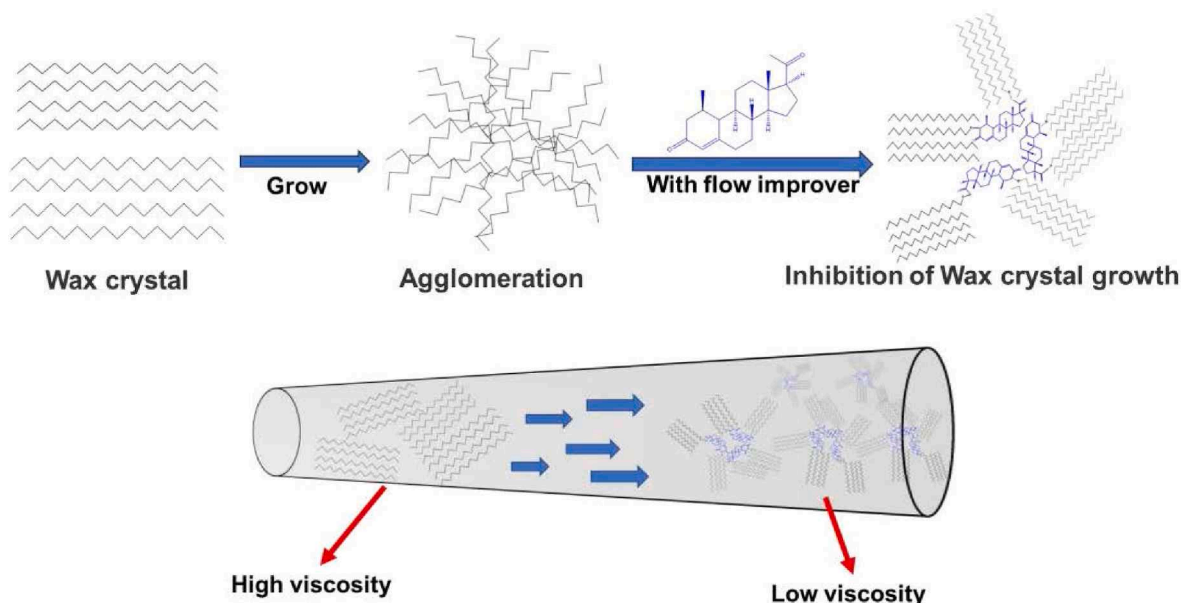


Fig. 5. The co-crystallization and dispersion of flow improver on wax crystals.

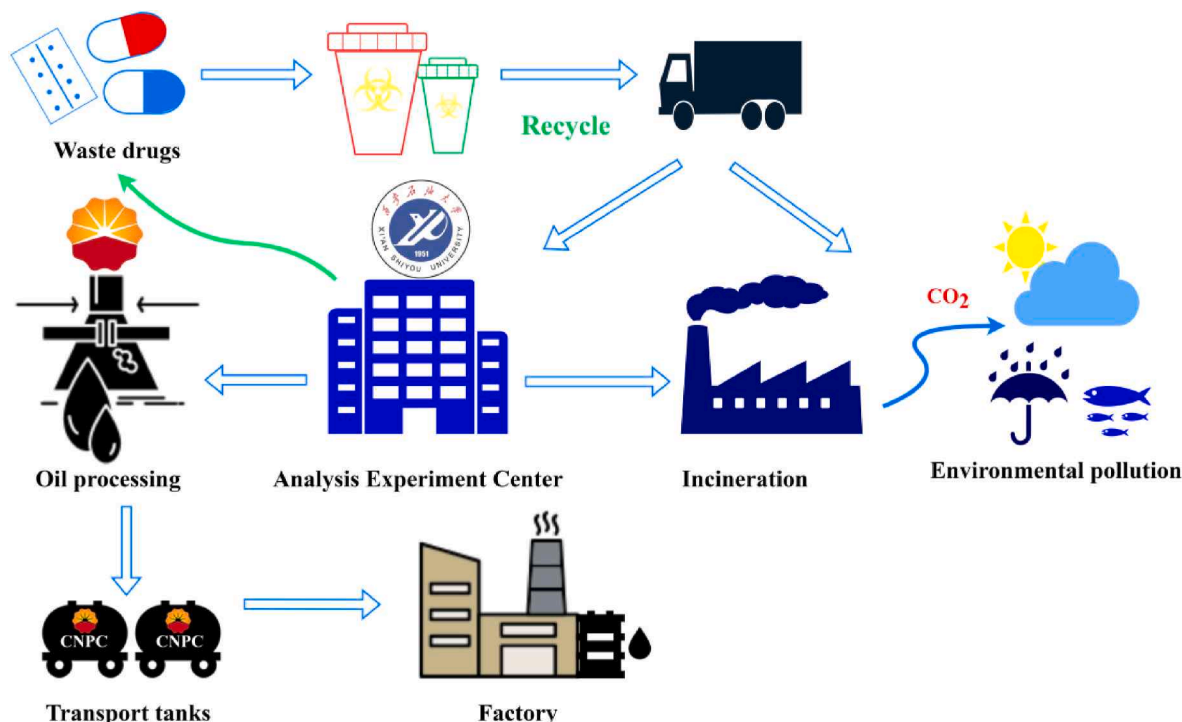


Fig. 6. Resource utilization flow chart of expired medicines.

analysis results (Gu et al., 2020a,b).

3.5. Proposed mechanism

As shown in Fig. 5, a hypothesis about the mechanism of flow improvers is proposed. Due to the interaction between the hydrocarbon chain of the flow improver and the saturated hydrocarbon, molecules can adsorb to the surface of the wax crystallization when the wax is formed (Zhang et al., 2018; Ma et al., 2023). Because the flow improver adsorbs-eutectic with wax crystals through the unique molecular configuration, the crystalline orientation of wax crystals is changed. The spatial resistance of the flow improver molecule itself inhibits the growth of wax crystals, which leads to the increase of wax crystals dispersion, which improves the influence of wax crystals on crude oil fluidity (Chen et al., 2021; Yu et al., 2022). In addition, flow improver can also act on the hydrogen bond of recombinant components, destroy and loosen the accumulation layer, and to some extent disperse the resin and asphalt components, thereby increasing the liquidity of crude oil (Zhang et al., 2021; Eke et al., 2021). Progesterone contains non polar cycloalkyl structures and carbonyl groups similar to comb like copolymers used as crude oil flow improvers. When the temperature decreases, these aromatic hydrocarbons precipitate first, providing a large number of nuclei for the precipitation of paraffin wax, which affects the arrangement of wax crystals. Besides, the polar groups contained in the progesterone molecule will distort and deform the formed nuclei and hinder the growth of wax crystals, thus improving the fluidity of crude oil (Elkatory et al., 2022).

3.6. Cost and feasibility analysis

In this study, the problem of high demand and high cost of reducing additives in traditional oilfields is compensated by converting expired medicines into crude oil flow improvers that can adapt to the field of the oil field (Wang et al., 2022), while also addressing the environmental hazards associated with the traditional disposal of expired medicines. The resource utilization of expired medicines is shown in the following Fig. 6. The widely used flow improvers in Changqing Oilfield, such as the

XG series flow improvers, have a market price of \$3172 per ton, while the K-12 oil flow improver has a price of \$2317 per ton. In contrast, the cost of using expired medicines as flow improvers is only US\$1062 per ton - US\$1241 per ton, which includes the cost of transportation, handling, processing, and packaging of finished products of expired medicines, etc. The recycling of expired medicines comes from domestic expired and fixed-point collection, which does not generate additional costs, compared with conventional incineration and landfill methods, the transportation cost of expired medicines is US\$27 per truck - US\$69 per truck, and the finished products transport, processing and packaging costs are generated by the large machine purchased in the early stage, the cost is US\$965, and the maintenance cost at the later stage is US\$69 - US\$100. It is obvious that using expired medicines as crude oil flow improvers not only increases the fluidity of crude oil, but also reduces the cost by about 50%. More importantly, conventional incineration disposal increases CO₂ emissions by about 15,000ton/year, and realizing the resource utilization of used pharmaceuticals (Cui et al., 2022; Yakubu and Zhou, 2019) can trade these carbon emissions with the remaining plants or enterprises, compensating for the cost of crude oil flow improvers while contributing to a certain extent to the early achievement of peak carbon and carbon neutrality targets in China. The following is the cost calculation equation (1)

$$C = M_p C_p + M_d C_d + C_o \quad (1)$$

where C is the total cost of the flow improver in ¥/ton; M_p is the ratio of PI or PC [14] per ton of flow improver, 0.30 tons/ton and 0.50 tons/ton respectively, C_p is the total cost per ton of PI or PC, estimated at M_d is the proportion of diesel fuel per ton of flow modifier containing PI, 0.65 ton/ton; C_d is the cost per ton of diesel, now US\$1014 per tonne; C_o is the total cost of transportation, packaging, and processing, estimated at US\$96 per tonne - US\$179 per tonne.

4. Conclusions

In this study, expired progesterone medicines were used as crude oil flow improvers, and were pour point and viscosity reduction effects

were evaluated due to their similar structure to traditional crude oil flow improvers. The results show that both PI and PC can reduce the viscosity and depress the pour point of crude oil at a certain extent. At 30 °C, the viscosity of HN oil were reduced by 60.40% and 54.70%, respectively, and the pour points of HN oil were depressed by 8.5 °C and 10.9 °C, respectively. Further research has shown that these two medicines also have a certain effect on improving the fluidity of crude oil from other sources. The mechanism was studied by DSC analysis and crystal morphology observation. PI and PC flow improvers may inhibit the growth of wax crystalline structures, loosen aggregates of resin and bitumen molecules, and make crude oil easier to flow. In addition, the total cost to use expired progesterone medicines as crude oil flow improver is estimated to be around US\$1062 per ton - US\$1241 per ton, which is far lower than the price of crude oil flow improvers currently used in oil fields. This work not only provides methods for the resource utilization of expired medicines and low-cost crude oil flow improvers, but also contributes to promoting carbon peaking and carbon neutrality goals.

CRedit authorship contribution statement

Yunlei Zang: Investigation, Data curation, Writing – original draft. **Guibin Liu:** Data curation, Investigation, Formal analysis. **Wenyu Ji:** Data curation, Investigation. **Yongfei Li:** Validation, Conceptualization, Supervision. **Gang Chen:** Methodology, Project administration, Funding acquisition, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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