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The Modification of Waste Polystyrene and Its Application as a Heavy Oil Flow Improver

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Abstract: The widespread use of polystyrene has brought great convenience to people's lives, but inappropriate recycling practices can also have a significant negative impact on the environment and public health. In this work, waste polystyrene was refined using maleic acid anhydride to produce flow improvers. The effect of the modified polystyrene perception of viscosity reduction and pour point depression of Henan oil was evaluated. The results show that modified polystyrene reduces the viscosity of Henan oil by 96.5% at most and depresses the pour point by 6.1 °C at most. Fourier transform infrared (FT-IR) and proton nuclear magnetic resonance (^{1}HH NMR) were used to investigate the modified polystyrene. The mechanism of modified polystyrene that improves the fluidity of heavy oil was proposed and analyzed through the study of wax crystal morphology (at 20 °C below the pour point).

Keywords: heavy oil; pour point; viscosity reduction; modified polystyrene

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1. Introduction

With the accelerated logistics development in various countries, a large number of plastic packaging materials are used in the process of article packaging, among which polystyrene is one of the main compositions. While the biodegradation of polystyrene does occur, the rate of degradation in the natural environment is very slow, so polystyrene persists as solid waste [1]. The disposal of waste polystyrene by incineration and landfill will cause pollution to the ecological and natural environment. Therefore, it is necessary to treat it properly to solve environmental pollution problems. On the other hand, with the increase in oil demands, the extraction and viscosity reduction technology of heavy oil reservoirs has attracted wide publicity. Reducing the viscosity of heavy oil is the primary problem to solve in the development and utilization of heavy oil [2]. Heavy oil and bitumen reserves are estimated to be 70% of the world's total recoverable oil [3,4]. It is generally believed that the high viscosity of heavy oils is directly attributable to a high resin content, asphaltene components, and the high polarity of the monomer molecules [5,6]. Multiple methods have been developed to improve the mobility of heavy oil [7]. Polymers such as homopolymers and copolymers (containing waxy kinds of paraffin and polar components) have been used as heavy oil flow improvers in the oilfield [8,9]. These polymers can slow down or change the crystallization of paraffin components to reduce the viscosity of heavy oil [10,11]. Polystyrene is structurally similar to some heavy oil flow improvers, but its solubility in heavy oil is limited, so it is necessary to modify polystyrene to further enhance its solubility in heavy oil [12,13]. In addition to what has been stated, the malleated modification of waste polystyrene is a convenient process that can be easily applied to comprehensive applications (such as in the preparation of emulsion-type adhesives, which are also widely used). Therefore, we designed a flow improver of heavy oil from waste polystyrene by maleylation modification and evaluated the properties of modified polystyrene. The modified polystyrene can disperse the aggregates of colloid and asphaltene and disrupt the composition of waxy crystals, which leads to a subsequent reduction in the viscosity of crude oil.

The versatile chemical structure of maleic anhydride makes it incredibly versatile, with low toxicity and easy handling for a variety of applications [14]. Polymers such as maleic acid polymers and major polyolefins have been used to achieve coupling in composites with polymeric matrices of polyethylene and polypropylene, which have been extensively reported in the literature. Combination-form polymers such as copolymers of maleic anhydride with styrene or α-olifins or vinyl esters are also evaluated as PPDs [15]. Thus, it is the target of the currently available studies to further improve efficacy [16]. In this research, we surveyed the structure of comb-type polymers via the Malay acylation of maleic anhydride with polystyrene. Maleic anhydride is a monomer that accepts electrons, and its properties are derived from active double bonds and active anhydride groups. The obvious benefits of using these comb copolymers as flow improvers for heavy oil are clear: they incorporate both non-polar side alkyl chains and polar groups [17]. At the same time, these copolymers can modify shapes so that wax crystals cannot interlock with each other, prevent asphaltenes from aggregation, and hinder the flow of the crude in lines.

2. Materials and Methods

2.1. Materials

Maleic anhydride was purchased from China, Nanjing, Nanjing Chemical Reagent Co., Ltd. (at AR reagent grade), pyridine was purchased from China, Beijing, Pharmaceutical Group Co., Ltd Sinopharm Chemical Reagent Co., Ltd. (at AR reagent grade), and 0# diesel oil was purchased from Sinopec. The waste polystyrene plastic is derived from household waste. The heavy oil was obtained from the Henan Oilfield. Its properties are shown in Table 1.

Table 1. The main physical parameters of the heavy oil from Henan Oilfield.

Pour Point, °C	Saturated HC, %	Aromatic HC, %	Resin, %	Asphaltene, %
24.2	26.98	28.47	34.12	10.43

2.2. Maleylation Modification of Polystyrene

The synthesis scheme is shown in Figure 1. A certain amount of waste polystyrene was dissolved in an amount of pyridine in a flask, and $n_{AlCl3} = 0.02\%$ was added. The ratio of the phenyl of polystyrene to maleic anhydride was set as 1:1, 2:1, 1:2, and 1:5, respectively. The reaction was conducted at 115°C for 4 h. After the completion of the reaction, the precipitate was filtered out, then the product was mixed with diesel oil in different proportions. Five flow improvers were selected according to the solubility of diesel. The abbreviation of five flow improvers is shown in Table 2, in which modified polystyrene is abbreviated as MPS.

Table 2. Abbreviation of five flow improvers with different conditions.

Abbreviation Name	$n_{styrene}:n_{maleic\ anhydride}$	$v_{modified\ polystyrene}:v_{diesel}$
MPS1	1:1	1:1
MPS2	2:1	1:1
MPS3	2:1	1:2
MPS4	1:2	1:1
MPS5	1:5	1:1

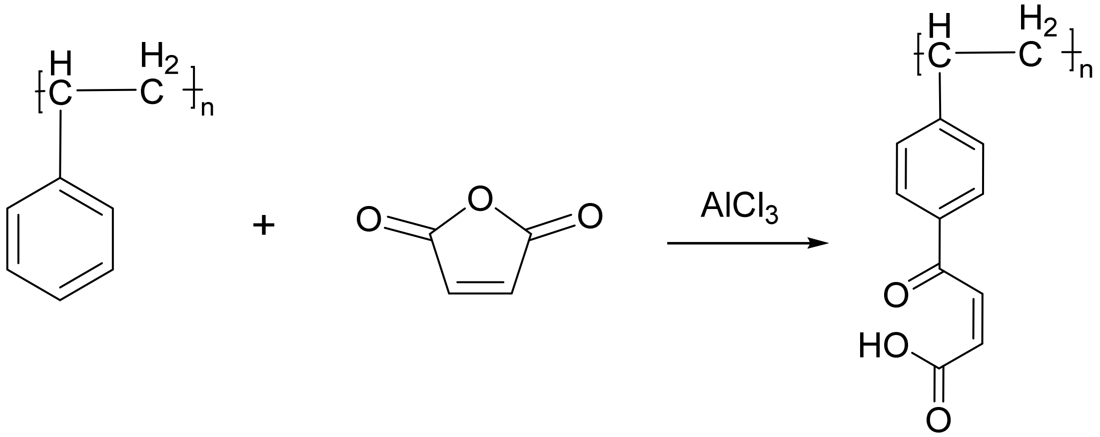


Figure 1. Modification of polystyrene by maleylation.

2.3. FT-IR Spectra Analysis

The modified polystyrene was characterized by the Fourier transform infrared spectroscopy (FT-IR) liquid film method (the sample was applied as a slim, thin liquid film that was placed in the light circuit for examination) [18].

2.4. ^{1}H NMR Spectroscopy

The ^{1}H NMR analyses of the copolymers were realized using a BRUKER 400 model NMR spectrometer. The samples were dissolved in $CDCl_3$ prior to the analysis.

2.5. Pour Point Evluation

Pour points of heavy oil samples with different concentrations of flow improvers [19] were determined according to SY/T0541-2009 (Crude oil pour point determination method).

2.6. Viscosity Evaluation

In total, a 30 g Henan oil sample was poured into a measuring cup, sealed with a plastic film, heated to 60 °C, maintained at a constant temperature at the water bath for 1 h, placed in a thermostatic temperature water bath at the temperature to be measured, and kept there for 30 min. The viscosity of the blank oil sample and the viscosity of the oil sample after adding different concentrations of the flow improver (100–1000 mg/L) were measured separately with a rotational viscometer. When the viscosity value of the rotary viscometer was stable, a record was kept of the oil samples' viscosity at different temperatures.

2.7. Optical Microscopy

According to the SY/T5119-2008 (analysis of soluble organic matter in rocks and group components of crude oil), saturated hydrocarbon components were isolated from heavy oil. We studied the wax crystal morphology using the OPTPro-3000 polarization microscope. The saturated hydrocarbon components were isolated from the oil sample and then put in a steady-temperature water bath at 20 °C below the pour point for 1 h. The saturated hydrocarbon components were placed on a glass sheet, and the morphology of the wax crystal was visualized by a polarized light microscope.

3. Results and Discussion

3.1. IR Spectrum Analysis

The functional groups of the modified polystyrene ($n_{\text{styrene}}/n_{\text{maleic anhydride}} = 1{:}1$) were characterized by IR spectrometry. The obtained spectra are clearly shown in Figure 2, and an absorption band corresponding to the vibrations of C=C is present at 1439 and 1491 cm^{-1}. The symmetrical vibration of 1714 cm^{-1} can be observed, and antisymmetric stretching vibration peaks appeared at 1754 cm^{-1} of the C=O groups within maleic anhydride. In addition, 698 cm^{-1} is the monosubstituted peak of a benzene ring; 3059 and 2924 cm^{-1} represent the characteristic peaks of the CH group and CH_2 groups, correspondingly. These characteristic peaks are in fundamental concordance with the standard spectra of maleic anhydride-modified polystyrene. Compared with the IR spectrum of polystyrene, the IR spectra of modified polystyrene showed a new peak at 1754 cm^{-1}, indicating the success of the maleic anhydride modification of polystyrene.

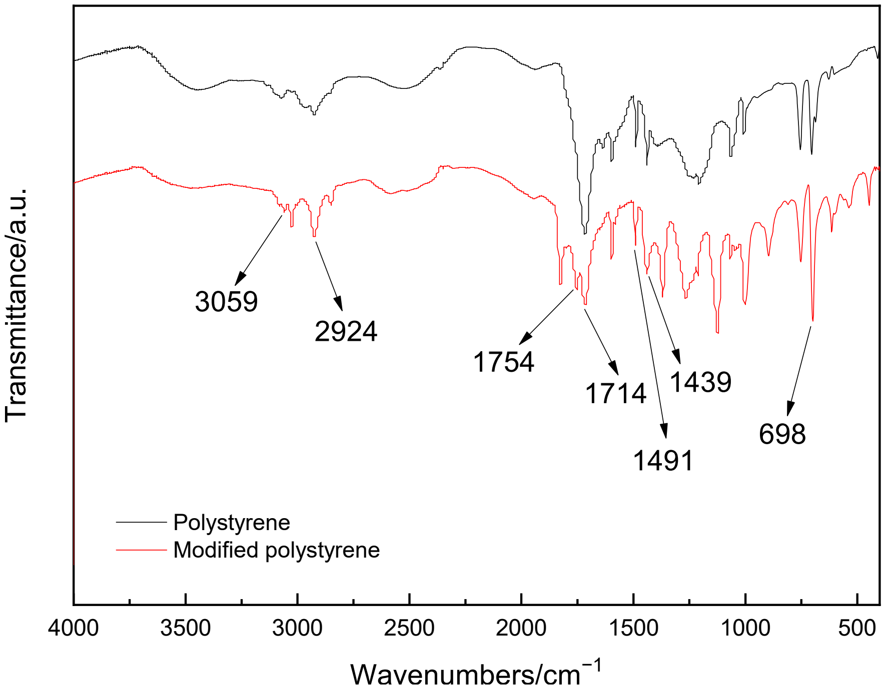


Figure 2. The IR spectra of the modified polystyrene.

3.2. ^{1}H NMR Spectroscopy

Figure 3 showcases the ^{1}H1H NMR spectra of modified polystyrene (MPS) derived from waste polystyrene foam, which inherently contains varying ratios of saturated hydrocarbon-blowing agents and impurities, such as polypropylene. This composition gives rise to pronounced low-field signals that obscure certain spectral regions, notably the peaks at h and i in the MPS spectrum. Nonetheless, the presence of distinct peaks at specific chemical shifts provides compelling evidence of the successful modification process. The symmetry of the benzene ring in the MPS structure is reflected in the clustering of hydrogen signal peaks near δ 7.09, 7.04, 6.50, and 6.45 ppm, indicating the preservation of the aromatic backbone during modification. Additionally, the presence of maleic anhydride, the modifying agent, was confirmed by the emergence of hydrogen signal peaks at δ 7.34 and 7.32 ppm, corresponding to the e position on the maleic anhydride moiety. While the expected signal peaks at f may be obscured by the intense signals from the benzene ring, the combined analysis with infrared spectroscopy further solidifies the conclusion that the modification of polystyrene with maleic anhydride was achieved. This comprehensive spectroscopic analysis not only verifies the successful incorporation of maleic anhydride into the polystyrene structure but also underscores the complexity of working with recycled materials, necessitating the careful interpretation of spectral data to accurately assess the structural changes.

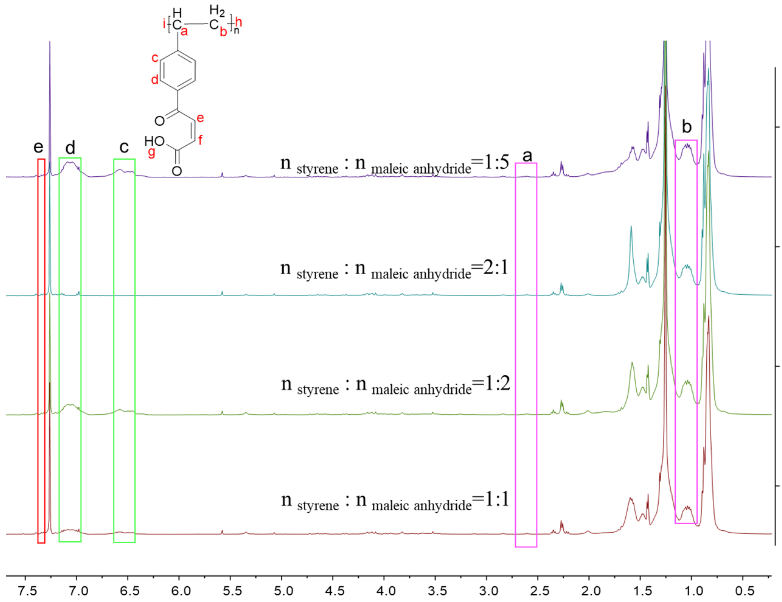


Figure 3. MPS ^{1}H NMR spectra with different molar ratios.

3.3. Pour Point

The investigation into the impact of modified polystyrene (MPS) on the pour point of heavy oil yielded intriguing results, as summarized in Table 3. Notably, the introduction of MPS into the oil samples effectively mitigated the pour point (−20 °C~30 °C), which is a critical parameter influencing the flowability of heavy oil at low temperatures. The baseline pour point of the untreated oil sample was recorded at 23.5 °C, highlighting the challenge posed by wax crystallization at lower temperatures. Upon the addition of MPS at varying volume ratios, a discernible trend emerged: the pour point decreased progressively with an increase in the MPS concentration. Among the tested formulations, MPS1 demonstrated the most pronounced effect, achieving a maximum reduction of 6.1 °C in the pour point of the Henan oil sample. This remarkable performance can be attributed to the unique ability of MPS to disrupt and disperse wax crystal aggregates, which are primarily responsible for the elevation of the pour point. By preventing the formation of large, interconnected wax crystal networks, MPS facilitates the flow of oil even at temperatures below its original pour point, thereby enhancing the operational flexibility and efficiency of heavy oil processing and transportation.

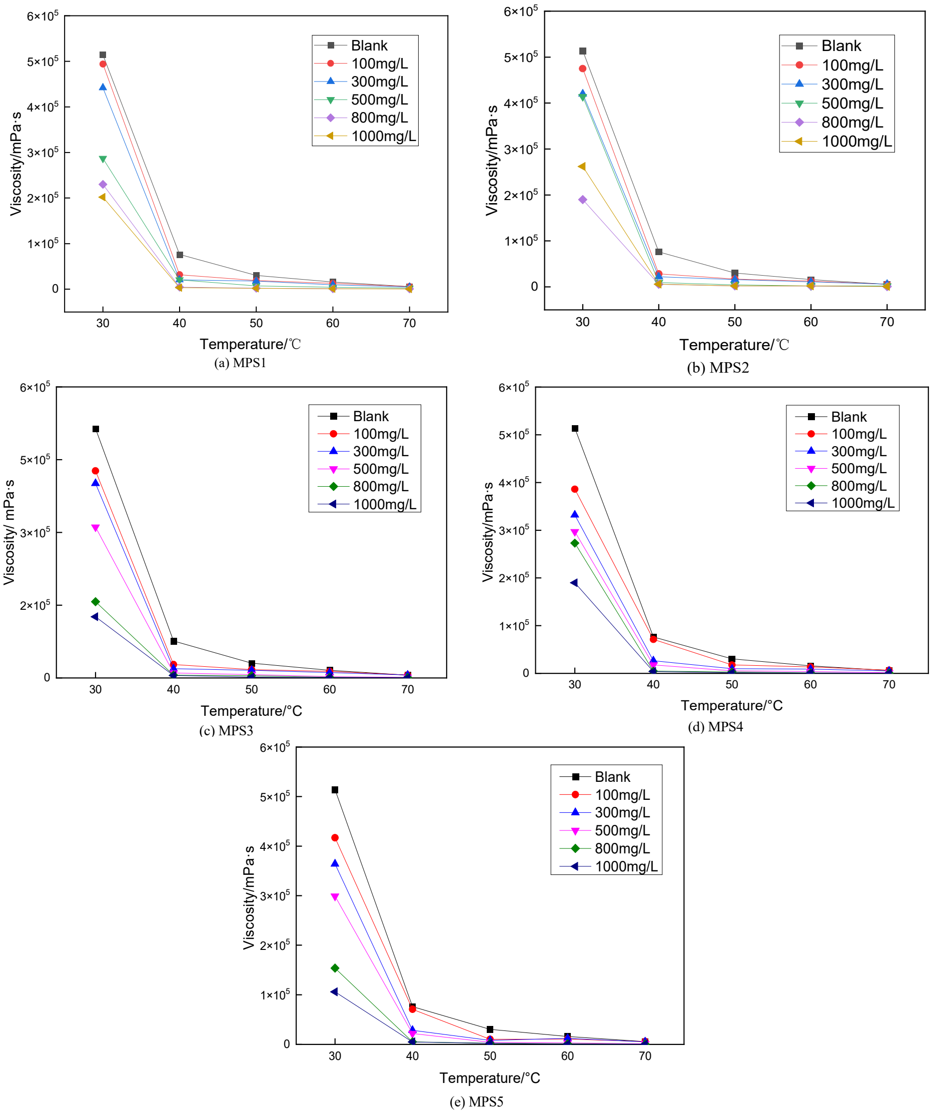
Table 3. Pour point of Henan crude oil.

Concentration, mg/L	**Pour Point, °C**				
	MPS1	**MPS2**	**MPS3**	**MPS4**	**MPS5**
0	23.5	23.5	23.5	23.5	23.5
100	23.0	23.1	22.8	23.3	23.2
300	22.3	21.7	21.5	23.1	22.9
500	22.0	21.1	22.0	21.9	20.7
800	21.4	20.5	19.1	19.8	20.1
1000	17.4	20.9	18.3	18.7	18.5

3.4. Viscosity

The viscosity modulation of the oil sample under investigation is intricately intertwined with the interplay of five distinct flow improvers and varying temperature conditions. Notably, when temperatures dip below 40 °C, the viscosity of the oil is significantly influenced not only by the concentration of viscosity reducers but also by the ambient temperature itself, exhibiting a more pronounced sensitivity. Conversely, as temperatures surpass the 40 °C threshold, a remarkable transformation occurs, with the viscosity of the crude oil undergoing a substantial decrease. Under higher temperature conditions of 50–70 °C, the viscosity profiles for various concentrations of flow improvers converge, indicating a diminished sensitivity to concentration changes.

Figure 4 provides a compelling visualization of this phenomenon, highlighting the steep escalation in viscosity reduction efficiency as the concentration of crude oil flow improvers increases [20]. This trend underscores the potency of these additives in enhancing the flow properties of the oil. Specifically, the study reveals that the optimal performance, marked by a staggering 96.5% reduction in viscosity, is achieved at a temperature of 50 °C and with a concentration of 1000 ppm of MPS3, demonstrating the synergy between temperature, concentration, and the specific flow improver in achieving maximum viscosity modulation. These findings have significant implications for the oil and gas industry, where optimizing the flow properties of crude oil can significantly enhance transportation efficiency and reduce operational costs. From the results in Sections 3.3 and 3.4, it can be seen that MPS2 and MPS3 have different functional efficiencies despite having the same chemical structure. This may be due to the different solubility and dispersibility of polymers with different concentrations in solutions mixed with diesel in different proportions, resulting in significant differences in the experimental results. In addition, adding different amounts of diesel to crude oil can also result in variations. Dilution and viscosity reduction effects may occur and may have different synergistic effects with polymer viscosity reducers.



(a) MPS1

(b) MPS2

(c) MPS3

(d) MPS4

(e) MPS5

Figure 4. Effects of the flow improver with different concentrations on oil viscosity.

3.5. Paraffin Crystal Morphology

An optical microscope was used to explore the impact of modified polystyrene on the crystal morphology of wax derived from saturated hydrocarbons. It was observed via polarizing light microscopy, as shown in Figure 5a; due to the existence of modified polystyrene, paraffin crystals have a flocculent form, and wax crystals are easy to cross-connect to form a network of structures to improve the viscosity or reduce the fluidity of crude oil [21]. From Figure 5b, not only were shorter and smaller paraffin crystals as modified polystyrene added, but the morphology of the particles also changed to a needle-like form, which indicate that modified polystyrene ha the ability to decrease the propensity for paraffinic crystallization to form three-dimensional networks [22,23], thus decreasing the viscosity of the poorly fluid thick oil caused by the wax component [24].

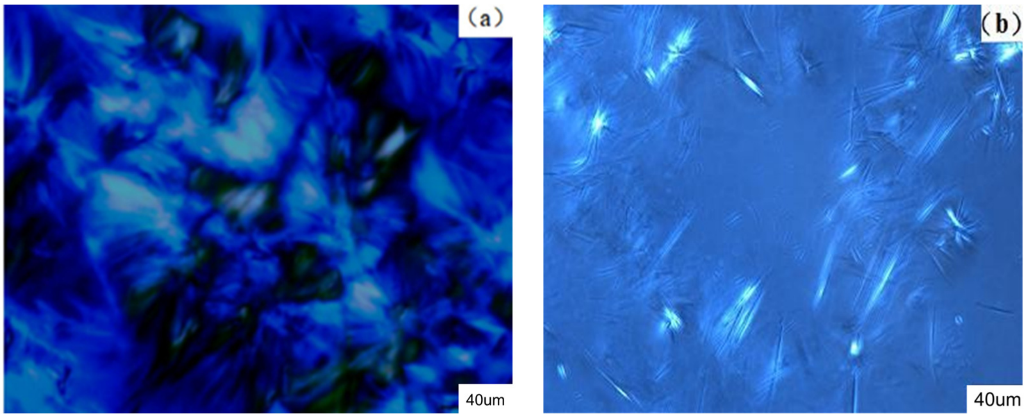


Figure 5. Wax crystal structure before (**a**) and after (**b**) modified polystyrene was added.

3.6. Mechanism

Figure 6 presents an insightful illustration of the underlying mechanism by which flow improvers exert their beneficial effects on oil viscosity. The intricate process of paraffin crystallization, which often contributes to the increase in viscosity in crude oil, encompasses multiple mechanisms, including co-crystallization, nucleation, and adsorption [25,26]. In this context, flow improvers play a pivotal role in adhering to the crystal surfaces of waxes, effectively hindering their growth and consequently minimizing the likelihood of the formation of intricate three-dimensional networks that can impede oil flow. Furthermore, these flow improvers demonstrate a dual-pronged approach to viscosity reduction, particularly in heavy oils. By interacting with asphalt molecules, they facilitate the dispersion or even breakage of aggregated particles, thereby reducing the overall viscosity. Specifically, modified polystyrene, a type of flow improver, leverages its polar groups to establish robust hydrogen bonds with asphalt, disrupting its native hydrogen bond network and subsequently dispersing the asphalt aggregates throughout the crude oil. This disintegration of large, viscous aggregates into smaller, more manageable particles significantly enhanced the fluidity of heavy oil, making it easier to transport and process [27–29]. This comprehensive understanding of the mechanism of action of flow improvers not only provides valuable insights into their functioning but also paves the way for the development of more effective additives tailored to specific oil compositions and operational conditions.

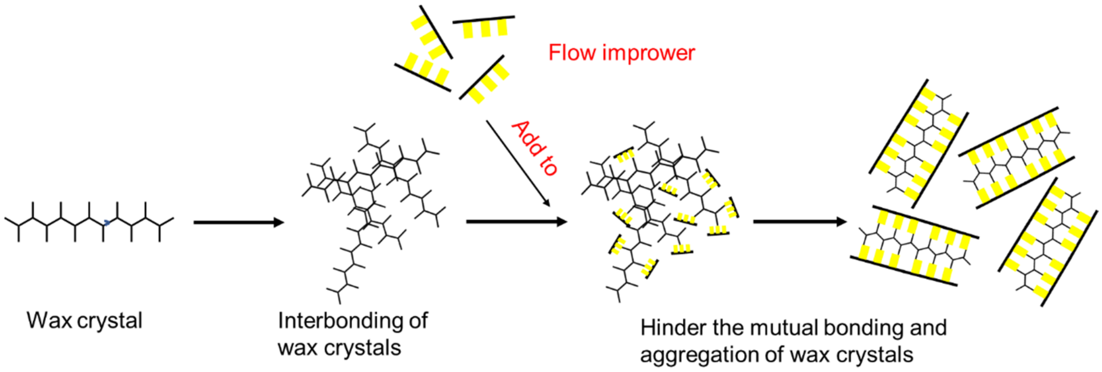


Figure 6. Interaction of the flow improver along with wax crystals.

4. Conclusions

The efficiency of the modified polystyrene as a flow improver was studied in crude oil samples. The results show that modified polystyrene MPS3 can reduce the viscosity by 96.5% at 30 °C, and MPS1 can depress the pour point by 6.1 °C at most. The mechanism of improving fluidity through the interaction between modified polystyrene and crude oil components was elucidated. The co-crystallization of modified polystyrene and saturated hydrocarbons is potentially possible according to an examination of the wax crystal's shape, which can alter the crystal morphology, limit the aggregation of waxy crystals, and decrease the pour point of the oil sample. To enhance the fluidity of heavy oil, modified polystyrene interacts with resin and asphaltene polymers to form a strong hydrogen bond, destroys its original hydrogen bond, and disperses the asphaltene aggregates in crude oil. This work provides a solution for the collection and transportation of crude oil by recycling waste polystyrene.

Author Contributions: Conceptualization, T.W. and G.C.; methodology, T.W.; software, Y.Z.; validation, T.W., C.W. and M.W.; formal analysis, T.W.; investigation, G.C.; resources, J.W.; data curation, M.W.; writing—original draft preparation, T.W.; writing—review and editing, T.W.; visualization, J.W.; supervision, J.W.; project administration, G.C.; funding acquisition, G.C. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

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Conflicts of Interest: Authors Wang Chunhui and Wang Meng were employed by the company Changqing Oilfield Company. Author WYunlei Zang was employed by the company Lanzhou Petrochemical Company. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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