STUDY ON MICROSTRUCTURE WAX EVOLUTION BEHAVIOR OF WAXY CRUDE OIL DURING COOLING

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The microcosmic characteristics of wax evolution of waxy crude oil directly affect the heat transfer process of crude oil. A microscopic experiment was carried out on the wax deposit process of waxy crude oil by polarized light microscopy. The phase change characteristics of wax crystals during wax deposit process were analyzed, and the process of wax crystal precipitation was quantitatively analyzed by the image analysis software ImageProPlus. The results show that the wax deposit process mainly includes wax crystal nucleation, growth and bonding, which together lead to the formation of wax crystal network. The sample oil particle number and the wax crystal area fraction curves have inflection points at 41℃ and 38℃, respectively, which are caused by different carbon number alkane content and wax crystal bonding. By analyzing the particle size and particle area curves, it is known that nucleation precipitation and growth are alternately dominant in the early stage of wax crystal growth. Through the wax roundness study, it is found that the growth of wax crystals in the early stage is mainly in the longitudinal direction, and the growth in the late stage is not directional. According to the phase change process of wax crystal, it is proposed that the temperature of the crude oil with porous media grid structure is coalescence point. By using the position point tracking of wax crystal and analysis of the variation characteristics of microscopic parameters, the coalescence point temperature of the sample crude oil in this experiment is 38℃.

Key words: *wax crystal, microscopic properties, polarized light microscopy, quantitative analysis, heat transfer process*

1. Introduction

The shutdown of hot oil pipeline is a common engineering problem in the field of storage and transportation. Especially for the long-distance crude oil pipeline, how to run more economically and more safely is very important. How to reduce energy consumption when the pipeline is in normal operation; When the pipeline is shut down, how to determine the safe shutdown time and restart pressure needs to fundamentally study the microscopic characteristics of paraffin crystallization in crude oil that can affect the low-temperature fluidity of waxy crude oil. After the shutdown, when the temperature of wax-containing crude oil is low to the wax extraction point, wax crystals continue to precipitate [1-5].

As the temperature continues to drop, the wax precipitated forms a microscopic network structure, resulting in the decline of crude oil mobility. In addition, the change of the phase state of the wax crystal during the cooling process also directly affects the transfer of heat transfer. When a small amount of wax crystals precipitate, the heat transfer mode is the same as that of liquid oil, and natural convection is the main heat transfer mode. With the increase of wax crystal precipitation, the crude oil presents the characteristics of porous medium, and its heat transfer and flow mode are different from that of liquid oil. Thereafter, when the crude oil solidifies, its heat transfer mode can be regarded as heat conduction. The physical process of phase change of crude oil is directly related to the precipitation of wax, so it is of great significance to study the phase change heat transfer of crude oil by carrying out the microscopic characteristics of wax-containing crude oil and defining the behavior characteristics of wax crystal and the change characteristics of phase state [6,7].During the temperature drop of crude oil, the amount of wax crystals increases continuously, and the phase state change process of wax crystals is accompanied by the transformation of crude oil from Newtonian fluid to non-Newtonian fluid. The existing studies on rheological properties mostly adopt microscopic microscope, rheological test instrument and differential scanning method [8]. He [9], Norman *et al.* [10] tested the rheological properties of waxy crude oil and analyzed the influence of nano-composite pour point depressant on the flow properties of waxy crude oil. Oliveira [11] characterized the density, water content, wax fraction (1-D and 2-D chromatography), pour point, yield stress and appearance temperature of 9 crude oil samples.

At present, there are a variety of techniques and means for observation and analysis of wax crystallization, such as polarized light microscopy, X-ray diffraction, microscopic infrared analysis, differential scanning calorimetry (DSC), etc., to analyze wax crystals from multiple perspectives, providing help for in-depth study of the microscopic characteristics of wax crystals [12-16].Moussa K. *et al.* [17,18] used electron microscopy to conduct experiments on cooling wax evolution and obtained morphology differences of wax crystals under different cooling conditions. When the amount of wax evolution was small, wax crystals were isolated from each other, and with the increase of the amount of wax evolution, flakes or layers of wax crystals were cross-linked and aggregated .Taheri-Shaikbj *et al.* [19,20] used scanning electron microscopy to obtain the behavior characteristics of wax crystals, and confirmed through experimental observation that the molecular structure, crude oil composition and asphaltene content of wax crystals would directly affect the shape and size of wax crystals.

In addition, the crystallization behavior of paraffins was studied by differential scanning calorimetry (DSC), X-ray diffractometry (XRD), micrography and scanning electron microscopy (SEM) by Emad A., *et al.* [21]. Létoffé J. M. *et al.* [22] found through thermal microscopy that the crystal size of the mixture of pure paraffin and crude oil matrix was small and depended on the length of the paraffin chain at the pour point. Srivastava, S.P. *et al.* [23] studied the crystallization behavior of n-alkanes in distillate wax from Bombay high crude oil by X-ray diffraction and micrography. The effects of solvents and pour point depressant additives on the lattice properties of paraffin wax and the mechanism of gel formation and additives were studied. The relationship between the crystal structure and morphology of wax was discussed. Li, H.Y. *et al.* [24] used digital image technology to carry out quantitative characterization of the morphology and size of wax crystals, adopted the width coefficient of particle size distribution to describe the width of particle size distribution, and used the fractal dimension to characterize the irregularity of particle size distribution. Jiang, C. *et al.* [25] used terahertz time domain spectroscopy (THZ-TDS) and microscopic techniques to study the model oil obtained by dissolving real mixtures of long-chain n-octadecane in diesel oil, so as to understand the clusters composed of asphaltene and wax with the increase of wax content. The fractal dimension was used to quantitatively characterize the morphology and structure of the model oil clusters. Xu, J. *et al.* [26] determined the effects of copolymers (MACs) with different polar carboxyl/non-polar octadecyl ratios on the size and shape of paraffin crystals by using polarizing microscope, rheology, differential scanning calorimetry and X-ray diffraction observation methods.

In terms of the study of wax evolution characteristics of crude oil, some scholars described the growth characteristics of wax crystal particles at different times of wax evolution, but the existing studies have not combined with the phase change heat transfer process, and there were no reports on the influence of the change of wax crystal phase state in the wax evolution process on the heat transfer mode transformation and the physical characteristics of the phase change process. In this paper, the microscopic characteristics of wax evolution process of waxy crude oil were studied by using polarizing microscope. The parameters such as wax crystal picture, particle number, area fraction, average particle area, particle size and roundness were quantitatively characterized. The wax evolution process of crude oil was investigated from the microscopic point of view, the corresponding relationship between wax evolution and temperature was revealed, and the characteristic temperature when the wax crystal formed the physical characteristics of porous media grid structure was determined. The research results can provide theoretical support for the study of phase change heat transfer of waxy crude oil.

2. Microscopic experiment on cooling of waxy crude oil

2.1. Physical properties of crude oil and experimental instruments

The waxy crude oil used in the experiment was taken from Daqing Oilfield, and its basic physical property parameters are shown in the Tab. 1.

Table 1. Physical parameters of waxy crude oil

Parameter	Value
Density at 50℃ (kg/m^3)	867
Density at 40℃ (kg/m^3)	872
The solidifying point (℃)	32.5
Wax precipitation point (℃)	44
Wax content (%)	15.5
Pectin content (wt%)	23.13
Asphaltene content (wt%)	0.7
The highest alkane	$C_{23}H_{48}$
The average carbon number	C27
N-alkanes content	55.5%
Isomeric alkane and impurity content	44.5%

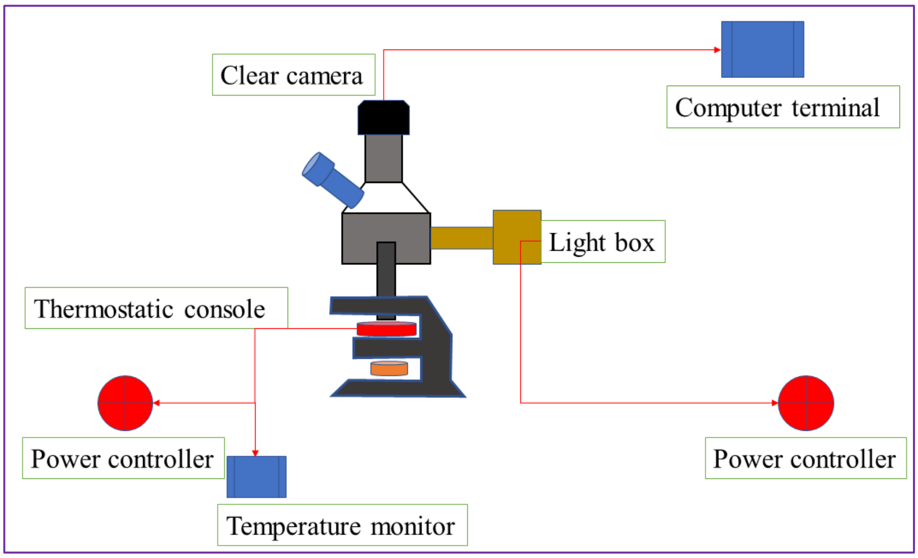


Figure 1. Experimental equipment diagram

The polarizing microscope used in the experiment is produced by Chongqing Aotter Optical Instrument Co., Ltd. The model is TP510, with a maximum magnification of 600 times. The microscope body was connected with the matching special digital camera to shoot the wax crystal image and transmit it to the computer for preservation. The temperature of the crude oil sample was controlled by BK-RDY thermostatic console with a special microscope. Other laboratory equipment included rotating heating table, cell scraper, smear needle and beaker. The experimental device is shown in the Fig. 1.

2.2. The experimental method

Before the experiment, the crude oil sample was heated and stirred in a constant temperature water bath at 75℃ for 1 hour to eliminate the influence of thermal history and ensure the uniform distribution of crude oil components. Then, the cell scraper, the smear needle, the slide and the cover glass were heated at 75℃ for 5min on the polarized heat platform. After preheating, take appropriate oil sample and smear it on the slide with the cell scraper, the thickness of which is about 5 microns. Then spread it evenly with the smear needle, cover the cover slide, and observe whether the oil film thickness is appropriate with the microscope transmission light. The finished sample was placed on the polarized heat table for isothermal cooling, and polarized light observation was carried out with a microscope. Take a sample photo every 0.5℃, and save the wax crystal picture to the computer.

The output wax crystal images were processed by Oppo image rating analysis software and ImageProPlus image analysis software. Firstly, the image was scaled, followed by threshold segmentation and image binarization. The characteristic parameters of wax crystal were statistically analyzed by using software measurement tools, including the particle number, area fraction, average particle area, particle size and roundness of wax crystal.

3. Wax crystal image feature extraction

The extraction principle of main characteristic parameters is as follows:

3.1. Number of wax grains

The target object needed to be measured was found after threshold segmentation, and then the number of particles was calculated statistically. The number of wax grains only reflects the quantitative characteristics of wax grains.

3.2. Wax grain diameter D_i and average diameter $\overline{D}$

The particle size of wax crystal can represent the change characteristics of its size, and its calculation formula is as follows:

$$D_i = \sqrt{\frac{4S}{\pi}} \tag{1}$$

$$\overline{D} = \frac{\sum_{i=1}^{k} n_i D_i}{\sum_{i=1}^{k} n_i} \tag{2}$$

Where： D_i-- Diameter of wax crystal particles, μm;

S-- The surface area of a wax crystal particle, μm^2;

n_i-- The number of wax grains within a certain diameter range, number;

$\overline{D}$--A range of diameters, μm.

3.3. Wax roundness R_0

Wax roundness R_0 reflects the complexity of the shape of the measured target. The more complex the shape of the measured target is, the greater the value of R_0 is. On the contrary, the closer the shape of the measured target is to a circle, the smaller the value of R_0 will be, approaching 1. Therefore, the value of R_0 can be used as a measure of the complexity of the target shape.

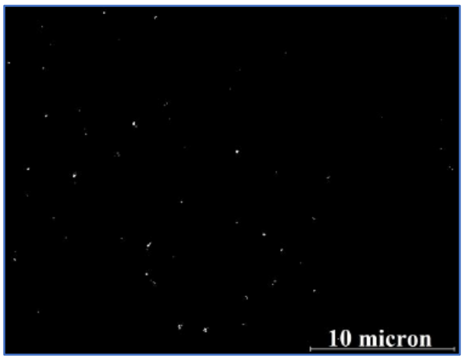
$$R_0 = \frac{L^2}{4\pi S} \tag{3}$$

Where:L-- The circumference of a wax grain, μm.

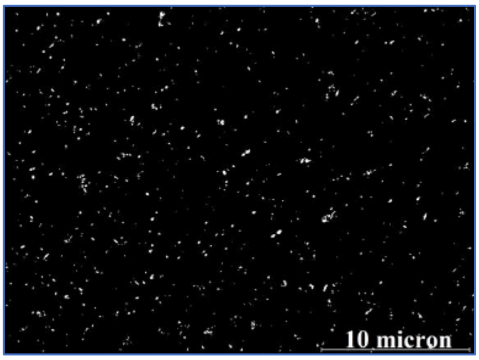
4. Results and analysis

4.1. Microscopic process of wax crystal precipitation

The wax crystal images at different typical temperatures are selected. It can be seen from the figure that the wax crystal images at different temperatures are very different. When the crude oil temperature is lower than the wax evolution point (44℃), wax crystals appear in the visual field, as shown in the Fig. 2(a). When the temperature decreases further, the amount of wax crystals precipitated gradually increases, covering the whole field of vision, as shown in the Fig. 2(b). As the temperature continues to decrease, wax crystals begin to grow and connect with each other at the same time of precipitation, as shown in the Fig. 2(c). A dense wax crystal network is eventually formed, as shown in the Fig. 2(d).



(a)



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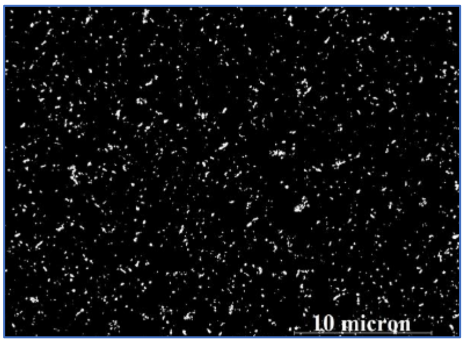
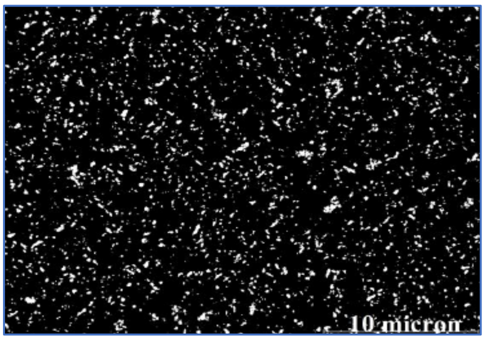


(c)

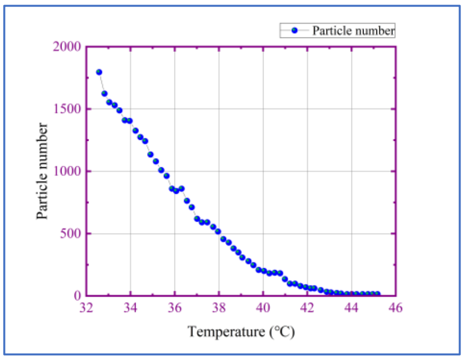


(d)

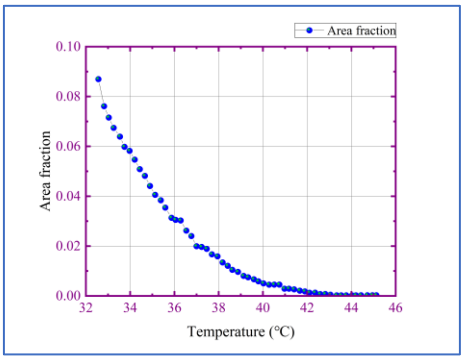
Figure 2. Binarization images of wax crystals at different temperatures (white parts are wax crystals) :(a) Image at 44℃; (b) Image at 40℃; (c) Image at 38℃; (d) Image at 33℃.

4.2. Quantitative analysis of wax crystal images

In order to ensure the correctness of the experiment, the average data of particle related parameters were obtained after repeated the experiment for three times. Particle number and area fraction are overall parameters, which can reflect the variation of wax crystals within a certain range. The curves are shown in the Fig. 3.



(a) Particle number



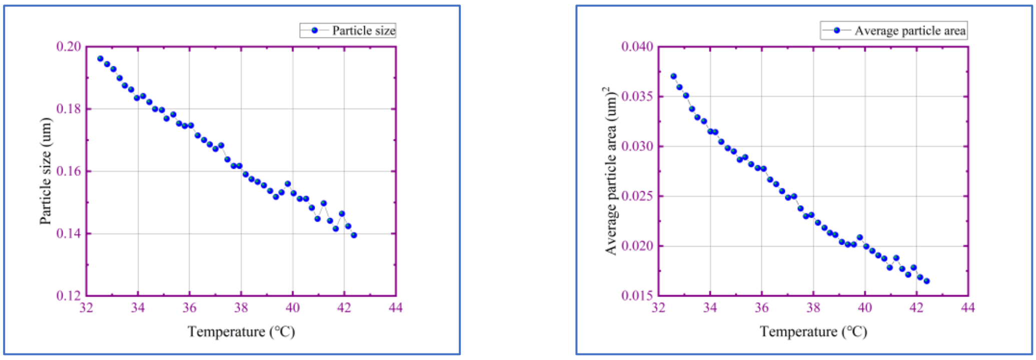
(b) Area fraction

Figure 3. The variation curves of Particle number and Area fraction with Temperature

According to the Tab. 1, the wax content of crude oil is 15.5%. When the temperature of crude oil dropped to the freezing point, the wax area fraction was about 9%, which was much lower than the wax content, mainly because the wax precipitated from crude oil had supersaturation. Until the temperature of crude oil dropped to the freezing point, there was still a large amount of dissolved wax in the liquid oil, which would continue to precipitate during isothermal rest or further temperature drop.

According to the two figures in the Fig. 3, wax crystals began to precipitate at about 44℃ during the whole temperature drop process. Each curve can be divided into two stages according to the slope change. The first stage is from 44℃ to 40℃. With the decrease of temperature, the number and area ratio of wax crystals precipitated in this stage increase slowly. When the temperature dropped to 40 ℃, wax crystals rapidly precipitated and grew, and their number and area ratio increased rapidly. As can be seen from the Fig. 3(a), the growth rate of particle number had two slow stages, which were 41℃-40℃ and 38℃-37℃ respectively. The slow increase of particle number may be due to, on the one hand, the small proportion of saturated alkanes in the temperature range, or the nucleation stage of wax crystal; on the other hand, it may be due to the connection of dispersed particles, which leads to the decrease of particle number. Combined with the DSC curve, it can be seen that the heat flow at 41℃ appeared the first peak due to nucleation, and the variation trend of the number of wax particles and the number of area fraction is consistent.

Therefore, it can be concluded that the flat segment at this time was caused by nucleation factors. However, when the temperature was 38℃-37℃, the increase rate of particle number significantly slowed down, while the increase rate of wax area did not change. Therefore, the reason for the decrease of particle number can be preliminarily confirmed as the connection between wax crystals. The rate of new wax crystals precipitated was similar to that of existing wax crystals, so the overall number of wax crystals remained basically unchanged, but the area of wax precipitation still increases steadily. Particle size and particle area are average parameters, which can reflect the variation of wax crystal individuals in the process of temperature drop. The curves were shown in the Fig. 4.



(a) Particle size **(b) Average particle area**

Figure 4. Variation curves of Particle size and Average particle area with Temperature

As can be seen from the Fig. 4, in the cooling process, the average particle size and average particle area fluctuated briefly at the initial stage, and then increased steadily. It was because of the process of temperature dropped, the precipitated wax crystal would continue to grow, and at the same time would also be accompanied by the generation of new crystal nucleus, the two were synchronous. Because the growth rate of wax crystal and the formation rate of crystal nucleus were not stable, when the formation rate of crystal nucleus was higher than the growth rate of crystal, the average diameter of wax crystal particle would decrease. When the formation rate of crystal nucleus was lower than the growth rate of crystal, the average diameter of wax crystal particles would increase, resulted in small fluctuations in the growth of average particle diameter and average particle area in the initial cooling stage. Since then, the number of wax crystals was increasing. Under the action of molecular forces such as electrostatic attraction and van der Waals force, the edges, angles and faces of wax crystals would be connected with each other. When the crystal surfaces were connected, the smaller crystals that were originally independent of each other would become a larger crystal, and the newly precipitated wax crystals would also adhere to the surface of the existing wax crystals and continue to grow, resulted in a steady increase in the average diameter and average area of the wax crystal particles.

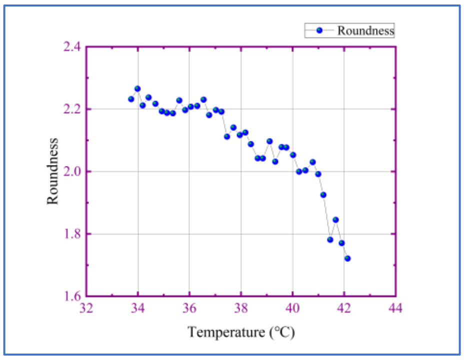


Figure 5. Corresponding relation between wax Roundness and Temperature

The average roundness of wax crystal is an important microscopic characteristic parameter of wax crystal, which reflects the degree of nearness of wax crystal. See the Fig. 5 for the roundness curve of wax crystal. The smaller its value is, the more nearness of wax crystal is (the roundness of perfect circle is 0).

As can be seen from the Fig. 5, the roundness of wax crystal increases with the decrease of temperature. After the rapid increase in the initial stage, the roundness increases gently, and the value mainly concentrates between 2 and 2.3. At the early stage of wax crystal precipitation, wax crystals grow rapidly and roundness increases rapidly. When the temperature reaches about 37℃, wax crystals are connected to each other, and some connected particles cannot be separated by threshold segmentation, so they are regarded as longer particles in calculation, the roundness increased slightly. When reaching a certain size and density, wax crystals begin to connect with each other to form larger particles and wax crystal network. At this time, wax crystals themselves no longer grow in the longitudinal direction, and roundness grows slowly or even basically does not change.

4.3. Determination of temperature when forming porous medium structure

The aggregation behavior of wax-producing particles and the phase state change characteristics of crude oil are closely related to temperature. Combined with the microscopic pictures, it can be seen that the cooling process of crude oil is accompanied by wax crystal formation, wax crystal growth, wax crystal particle connection and other processes. The diagram of the phase state change of wax crystal is shown in the Fig. 6.

In the process of crude oil cooling, when the temperature is lower than the wax evolution point, wax crystals will be precipitated, accompanied by the release of latent heat of wax evolution. Due to the uneven distribution and large spacing of wax crystals at the beginning of precipitation, with the decrease of temperature, the newly precipitated wax crystals will grow on the basis of the already formed wax crystals, and tend to gather locally, and eventually form a network structure similar to porous media with large pores. When the temperature continues to decrease, the number of wax crystals continues to increase. Due to van der Waals force, the wax crystals close to each other gather and continue to grow on the network structure that have been formed. The network structure will gradually become clear and the structure will become more and more dense.

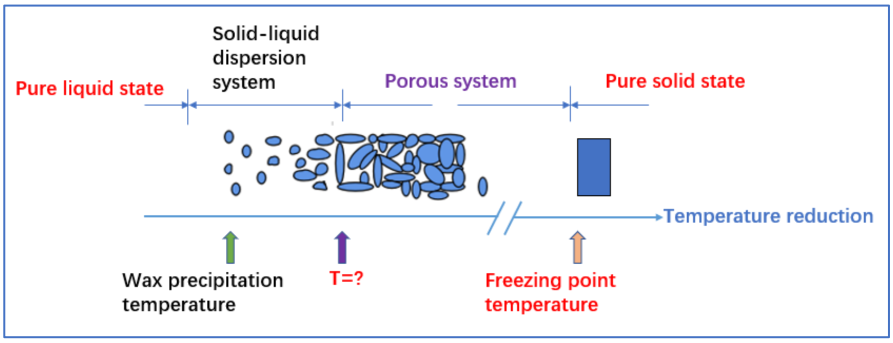


Figure 6. Wax crystal phase transformation during cold evolution of crude oil

In the process of crude oil phase change heat transfer, the formation of wax crystal grid structure is of great significance to the transformation of heat transfer mode. It changes the phase state characteristics of crude oil and make it show the characteristics of porous medium, leading to the change of heat transfer mode. Therefore, according to the changing characteristics of the phase state of wax crystal in the wax precipitation process of stationary crude oil, the temperature at which the mesh structure is formed between the wax crystals in the wax precipitation process is called " coalescence point " in this paper.

The determination of coalescence point should be based on the characteristics of microscopic parameters. According to the characteristic analysis of the relevant parameters such as the number of wax grains, particle area and average particle size in the process of wax precipitation, it can be preliminarily concluded that the coalescence point is 38℃. In addition, according to the study in literature [26-28], only 2% solid wax precipitated out of the stationary crude oil can make the crude oil gelling, and the cooling gelling is caused by the formation of three-dimensional grid structure between wax crystals [29]. In this study, according to the variation law of the number of wax grains and the area of wax grains, the temperature at which the mesh structure begins to form between wax grains is about 38℃, and the proportion of the area of wax grains at this time is about 2%, which is consistent with the quantitative analysis of the formation of the mesh structure of wax grains in the existing studies.

Through the comparison of dynamic microscopic images, it can be seen that the suspended wax crystal particles will keep moving during the cooling and phase transformation process of waxy crude oil. With the formation of spatial grid structure, wax crystal movement slows down or even disappears. In this paper, a typical position point 5 was selected for pixel positioning, as shown in the Fig. 7, and then the wax crystal dynamic tracking was carried out. A single pixel point was taken as the unit, and the overall pixel of the micro picture was 2592×1944. The pixel coordinates of each monitoring point at different temperatures were determined.

Comparing the displacement changes of wax crystal at each monitoring point above and below the coalescence point in the process of wax precipitation, the results were shown in the Tab. 2. As shown in the Tab. 2, when the temperature drops from 39℃ to 38℃, the wax crystals at each monitoring point shift significantly, and the one-way displacement of points 1, 2, 4 and 5 is as high as 11 to 14 pixels. When the temperature drops from 38℃ to 37℃, the displacement of each point is about 1-2 pixels, and each wax crystal point has no significant movement. Except that the displacement of 3 points is always small, the displacement of other points decreases by about 6-9 times.

Table 2. Location of monitoring points at different temperatures

	Point 1	Point 2	Point 3	Point 4	Point 5
39℃	1839×945	821×553	793×1499	1373×1133	213×1493
38℃	1839×933	807×555	795×1499	1385×1136	224×1495
37℃	1841×931	805×557	797×1499	1386×1136	225×1495
36℃	1841×931	805×559	797×1499	1386×1136	225×1495
35℃	1841×931	805×559	797×1499	1386×1136	225×1495

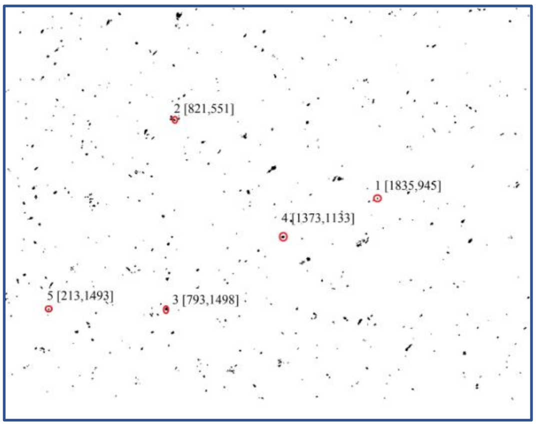


Figure 7. The positioning chart of wax crystal points monitored at 40℃

When the temperature reaches 37℃, the wax crystal at each monitoring point does not move. The decrease of the displacement of the wax crystal is due to the restriction of the movement of the wax crystal after the formation of the wax lattice structure. Therefore, it can be further proved that the coalescence point is 38℃.

5. Conclusion

In this paper, polarizing microscope was used to observe and photograph the microscopic change process of wax-containing crude oil's cooling and paraffin evolution, and image processing software such as ImageProPlus was used to carry out statistical and quantitative analysis on the wax crystal images. According to the results of this study, the phase change process of crude oil can be divided into liquid and solid liquid mixed porous system and the solid phase, including microscopy analysis is used to determine the temperature of nodes, grid structure is formed between the crystal-clear wax temperature, and precisely divide different phase partition, based on this study, to carry out the subsequent partitioning heat transfer of phase change heat transfer model is described. The conclusions are as follows:

1.The change of crude wax crystal with temperature mainly includes three stages: precipitation, growth and interconnection. In the early stage, wax crystals mainly precipitate and grow, and in the late stage, wax crystals mainly grow and join, and finally form a dense network of wax crystals.

2. Analyzing the wax crystal from the Angle of particle number and area fraction can reflect the

overall change of the wax crystal within the range. When wax crystals precipitate with decreasing temperature, there are obvious inflexion points at 41℃ and 38℃, in which the inflexion point at 41℃ is mainly caused by the difference of alkane content, while the inflexion point at 37℃ is caused by the interconnection between wax crystals.

3. According to the analysis of wax crystal individuals by particle size, particle area and roundness, it can be found that the wax crystal alternates between precipitation and growth in the early stage, and then gradually becomes stable, and the precipitation and growth reach a balance. At the same time, the wax crystals mainly grow longitudinally in the early stage, but no directional tendency in the later stage.

4. The concept of " coalescence point " was put forward, and the temperature of the crude oil forming porous media grid structure in this experiment was determined to be 38℃ by using the position point tracking of wax crystal combined with the analysis of the characteristics of microscopic parameter variation.

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