
Experimental research and simulation verification of recycling process of heat transfer oil

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Abstract: Heat transfer oil will inevitably deteriorate after long time use, leading to the gradual accumulation of material insoluble in acetone in hot medium oil that seriously exceeds the standard. Therefore, extending the service life of heat transfer oil is an important means to improve economic benefits. In this experiment, solvent extraction settlement method was used to remove material insoluble in acetone. By designing a set of extractive settlers, sedimentation experiments were carried out with different solvents, moreover, the acetone-insoluble content, kinematic viscosity and neutralisation value of the extracted heat transfer oil were detected. Computational fluid dynamics was used to verify the effect of asphalt settlement separation tank by numerical simulation. The experiments were validated by measuring the extraction effects of different solvents. The results have important implications for industrial production.

Keywords: heat transfer oil; acetone insoluble; extraction sedimentation; orthogonal test; numerical simulation.

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Biographical notes: Guorong Liu graduated from the Department of Mechanical Engineering, East China Petroleum University in July 1984, majoring in Chemical Machinery, and stayed to teach. He received his Master's in Chemical Process Machinery from University of petroleum in June 1992 and was promoted to Professor in 2005. His main research is direction liquid solid filtration theory and related engineering technology in heterogeneous separation technology. He is a member of the filtration and separation expert group of Sinopec.

1 Introduction

Heat transfer oil is the most commonly used heat carrier at present (Wang et al., 2015; Vignarooban et al., 2014; Gao et al., 2013). As a heat carrier, heat transfer oil transfers heat to other substances. It is a kind of heat medium with excellent performance used in indirect heat transfer system (Zeraouli et al., 2000; Hrifech et al., 2000). Heat transfer oil is mostly light yellow or brown, and it is widely used in petrochemical, food, textile, building materials and many other industries because of its high heat transfer efficiency, good thermal stability, low freezing point and viscosity, small environmental pollution advantages (Nicole et al., 2014; Zhao et al., 2021; Hekmatipour and Ji, 2019). In modern industrial production, heat transfer oil occupies a larger and larger market share, using heat transfer oil system has become the mainstream of indirect heating.

Although heat transfer oil has good thermal stability, it inevitably deteriorates in the process of heating and heat transfer due to long-term operation at high temperature (Law et al., 2018; Wright, 2014). If the system temperature is too high, the transfer oil will decompose when it is used for a long time at high temperature. Decomposition of the

product is more active and will react with other molecules to form a polymer. These reactions are interlinked. With the extension of time, the performance of heat transfer oil will change and deteriorate, and the deterioration rate will accelerate, which will pose a great threat to the operation of the system (Bellos et al., 2018). The asphalt will stick to the pipes of the heat transfer oil system and the inner wall of the equipment and then it will coke, which will affect the heat transfer efficiency and the normal operation of the device. At the same time, the existence of asphalt and other macromolecular groups will accelerate the deterioration of heat transfer oil, resulting in local overheating of furnace body and pipeline, which will damage equipment and endanger personal safety after a long time. The coking and deterioration of heat transfer oil is an urgent problem to be solved in the heating process. Among them, removing asphalt from oil and improving the performance of degraded heat transfer oil are the most key problems.

Through physical and chemical methods, the coke or gelatin attached in the heat transfer oil system is removed under the condition of stopping or switching the standby pipeline, including mechanical coke cleaning method and

chemical coke cleaning method (Kazak et al., 2014; Piepiorka-Stepuk et al., 2016). These methods need to be carried out when the system is fully or partially parked, then the heat transfer oil will be exited from the system. When the heat transfer oil temperature drops to the appropriate temperature it is returned to the heat transfer oil manufacturer in barrels or cans. These processes will cause long downtime and increase packaging and transportation costs and inject new heat transfer oil should be carried out in advance oil dewatering and the low boiling, which consumes time, wastes resources and affects effective production time. Because the treatment of waste heat transfer oil is usually cumbersome, online centrifugal settlement and online batch distillation have been proposed. These two methods can achieve online treatment, but there is no exact application yet.

In view of the problems existing in the above heat transfer oil treatment methods, the solvent deasphalting method (Long et al., 2012) was used for reference in this paper, and the extraction settlement method was adopted to remove the gum asphaltene, so as to realise the online regeneration of heat transfer oil. In this paper, the ideal extraction solvent was determined by the extraction sedimentation experiment, and the numerical simulation was combined to provide a strong basis for the design of online regeneration system. The research on the online regeneration of heat transfer oil has done a good interpretation of the concept of innovation and green development (Cui et al., 2020), which is of great significance to industrial production, energy saving and emission reduction. This paper is organised as follows. The second part introduces the selection process of process combination in the process of extraction sedimentation. In the third part, the extraction solvent is selected. In the fourth part, the factors influencing the extraction conditions are analysed. In the fifth part, the numerical simulation of asphalt settlement separation tank is carried out. The sixth part is the induction and analysis of the research content.

2 Selection of process combination

2.1 Component analysis of residual precipitation of heat transfer oil

The heat transfer oil used in this experiment contains a large amount of acetone-insoluble matter. Therefore, before carrying out the orthogonal test, one of the solvents was first used to conduct an extraction experiment on the heat transfer oil, and component analysis was conducted on the obtained precipitation to understand its composition.

The composition analysis of the precipitate mainly includes three aspects: saturated component, aromatic component and asphalt component. The analysis results are shown in Table 1.

As can be seen from Table 1, the main component of the residual precipitation of heat transfer oil solvent is colloidal asphaltene, accounting for 85.2% of the total. Therefore, acetone-insoluble substances, namely colloidal asphaltenes,

need to be targeted to select the appropriate solvent to extract and refine the heat transfer oil, and remove acetone-insoluble substances.

Table 1 Component analysis of the extracted sediment of HTF

Component	Saturated fraction	Aromatic fraction	Asphalt
Mass fraction (%)	6.2	8.6	85.2

2.2 Orthogonal experimental study on extraction subsidence of heat transfer oil

In this experiment, a total of eight extraction solvents were selected, numbered CN1–CN8 (in order of acetone, n-pentane, acetone, and butanone mixture 1:1, petroleum ether, furfural, NMP, n-heptane, toluene). In the orthogonal test, three factors affecting the extraction sedimentation effect in the extraction sedimentation experiment were mainly considered: solvent ratio, settling temperature, settling time, and each factor in three levels, using L9 (3⁴) orthogonal table, under normal circumstances the need for 27 times test, so the use of orthogonal table for tools, in the case of test times as much as possible to reduce, the results and the comprehensive test the results were obtained to determine the best parameter combination. In the orthogonal table, there are two very important factors, k and R , respectively. k represents the removal rate of acetone insoluble matter corresponding to each factor at a single level. The larger the value of k is, the higher the removal rate of acetone insoluble matter is. R is the range of the factor, reflecting the influence of the level change of the factor on the test results, and the value size reflects the importance of the factor. Orthogonal test should be carried out for the eight extraction solvents, and the optimal combination of conditions for each solvent was selected and then tested again, so as to screen the solvents.

The orthogonal test of CN1 solvent was taken as an example to analyse the results. It is assumed that the factor solvent ratio affecting the extraction sedimentation of heat transfer oil is A, the sedimentation temperature is B (°C), and the sedimentation time is C(h). The solvent ratio, settling temperature and settling time of each solvent were selected for practical industrial applications. Through orthogonal test, the result data of CN1 solvent was presented in the form of orthogonal table, and the analysis of orthogonal test results was shown in Table 2.

As can be seen from Table 2, the order of range is $RA > RB > RC$. It can be seen that the influence degree of various factors on the removal rate of acetone insoluble matter is the ratio of solvent > precipitation temperature > precipitation time, and the ratio of solvent is the most important influencing factor. Therefore, A should be controlled at the optimal level first, and the order of the three main effects of factor A is $A_3 > A_2 > A_1$. Therefore, the optimal level of A is A_3 . Similarly, the second important factor is B, the optimal level is B_2 , and the optimal level of the third factor

C is C1. Through the above analysis, the optimal process combination can be obtained as A3B2C1, that is, the solvent ratio is 2:1, the settling temperature is 100°C, and the settling time is 0.5h.

According to the above experimental and analytical steps, orthogonal experiments were conducted on CN1–CN8 respectively, and the experimental results were analysed to obtain the extraction and sedimentation effects of different extraction solvents in different solvent ratio, settling temperature and settling time, and to study the influence of various factors on the removal rate of acetone insoluble matter, to find the optimal process combination of each solvent.

Table 2 CN1 solvent analysis of results of orthogonal test

Factor level	A solvent ratio	B settlement temperature (°C)	C settlement time (h)	X removal rate of acetone insoluble
1	0.5:1	80	0.5	0.398
2	1:1	100	1	0.396
3	2:1	120	2	0.625
4	0.5:1	100	2	0.391
5	1:1	120	0.5	0.382
6	2:1	80	1	0.793
7	0.5:1	120	1	0.371
8	1:1	80	2	0.385
9	2:1	100	0.5	0.858
k1j	0.387	0.525	0.546	
k2j	0.388	0.548	0.520	
k3j	0.759	0.459	0.467	
R	0.372	0.089	0.078	
Better level	A3	B2	C1	
Factor primary and secondary	1	2	3	

3 Preliminary selection of extraction solvent

The optimal process parameters of each solvent were used to test again, and the extraction effect of each solvent was compared, so as to select the better solvent. The extraction effects of different solvents were mainly compared by the removal rate of acetone insoluble matter, and the changes of acid value and viscosity were also investigated. Under optimal process conditions, the properties of the extracted heat transfer oil obtained by eight solvents are shown in Table 3. Figures 1, 2 and 3 show the comparison between different solvents and related performance indicators.

Table 3 Eight solvents relevant properties data of extracted HTF under better technological conditions

Number	A	B	C	D	E	F
CN1	2:1	100	0.5	0.858	28.19	0.138
CN2	2:1	100	0.5	0.436	31.47	0.251
CN3	1:1	120	2	0.396	30.28	0.296
CN4	2:1	120	2	0.591	32.35	0.278
CN5	1:1	120	2	0.095	38.28	0.436
CN6	2:1	140	1	0.832	30.14	0.147
CN7	2:1	120	1	0.378	32.43	0.316
CN8	1:1	160	2	0.108	37.83	0.412

A Solvent ratio

B Settlement temperature (°C); C: time (h)

D Removal rate of acetone insoluble matter

E Viscosity (mm²/s)

F Neutralisation value (mg KOH/g).

Figure 1 Comparison of different solvent and acetone insoluble solids removal rate

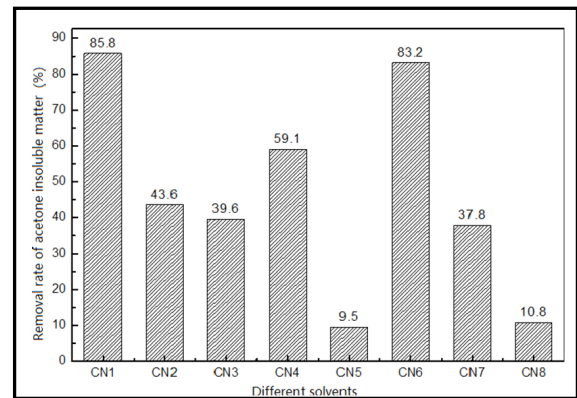
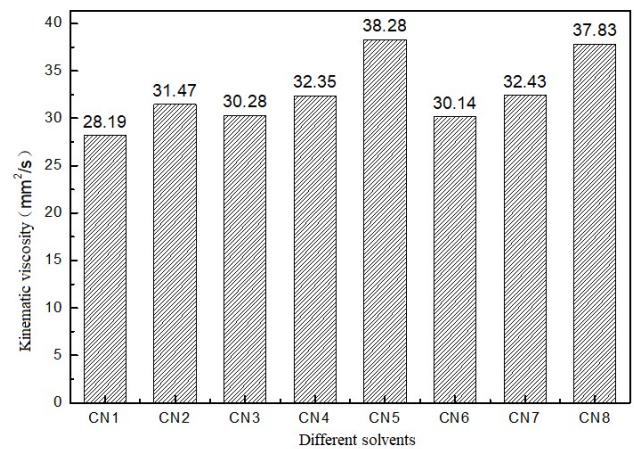
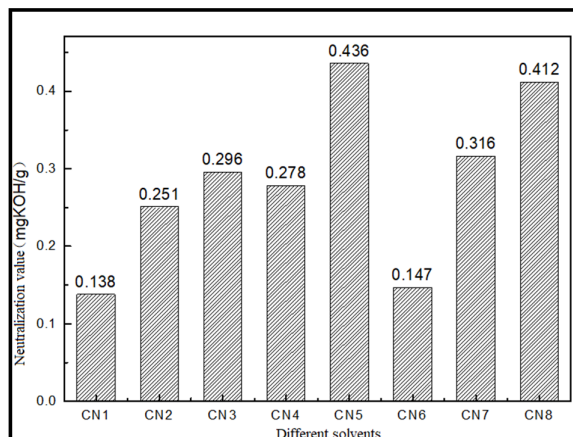


Figure 2 Comparison of different solvents and kinematic viscosity



As can be seen from Figure 1, there are differences in the removal of acetone insoluble matter in heat transfer oil by eight solvents under their respective optimum process conditions. In addition to the solvent CN5, CN8, the other six solvents have a good effect on the removal of acetone insoluble heat transfer oil. Among them, the solvents CN1 and CN6 have the best effect. The solvent CN1 can reduce the content of acetone insoluble matter from 3,942 mg/100 ml to 560 mg / 100 ml, and the solvent CN6 can reduce the content of acetone insoluble matter from 3,942 mg / 100 ml to 662 mg / 100 ml, which has a good removal effect.

Figure 3 Comparison of different solvents and acid values



As for viscosity and acid value, it can be seen from Figure 2 and Figure 3 that except CN5 and CN8, other solvents can reduce the kinetic viscosity and neutralisation value of heat transfer oil to the normal level. Neutralisation of CN1 and CN6 heat transfer oil can be dropped to below 0.15 mg KOH/g, and on the improvement of the viscosity degree is also evident in the relative to other solvents, suggests that heat transfer oil acid mainly exist in the colloid asphalt composition, and the main reason for the excess heat transfer oil viscosity is also due to the heat transfer oil has more colloid asphalt.

CN1 is a mixture of C3 and C4 ketone organic extraction solvent commonly used in industry with small molecules. It has a large difference in molecular weight from the gelatine asphaltene. It has a poor solubility to the gelatine asphaltene and a good solubility to the ideal components in heat transfer oil, so it has a good solvent removal effect on the acetone insoluble substances in heat transfer oil. However, CN3 is a mixture of C4 and C5 ketones, but its molecular weight is larger, and its polarity is stronger than CN1, and it has a certain solubility to gelatinous asphaltene. Therefore, its acetone insoluble matter removal effect is poor.

CN2, CN4 and CN7 are non-polar alkanes, CN4 is the mixture of C4 and C5 alkanes, CN2 is C5 alkane and CN7 is C7 alkane. Because the solvent CN4 has the smallest molecular weight and the smallest solubility of acetone insoluble matter among the three solvents, its acetone insoluble matter removal effect is better than CN2 and CN7. However, the molecular weight of CN4 is still larger than

that of solvent CN1, so the removal effect of insoluble matter in acetone is worse than that of CN1.

Polarity solvent CN6 to the strongest ketone compounds, it belongs to ketone, also belong to amide, and molecular weight is bigger, the colloid asphalt has a very good solubility in oil, the heat transfer oil polarity difference of non-ideal component in poor solubility, and CN6 density is greater than 1 g/cm³, easy to heat transfer oil in the colloid asphaltene precipitation down, therefore, CN6 has a good solvent refining effect.

The solvent CN5 has medium polarity and high molecular weight. It has a good solubility to the rational components of heat transfer oil and a certain solubility to the colloid asphaltene. Therefore, the refining effect of the solvent is not good. The solvent CN8 is a mono-cyclic aromatic hydrocarbon compound, which has a good solubility to both colloidal asphaltenes and ideal components of heat transfer oil. Therefore, the extraction effect is poor.

Through the experimental and theoretical analysis, the initial screening of the best solvent is CN1 and CN6.

4 Analysis of influencing factors of extraction process conditions

The extraction effects of the two solvents were further studied, and the removal effects of acetone insoluble matter were investigated under different solvent ratio, settling time and settling temperature, and the improvement of neutralisation value and kinetic viscosity as well as the yield of heat transfer oil were compared.

4.1 Effect of solvent ratio on extraction efficiency

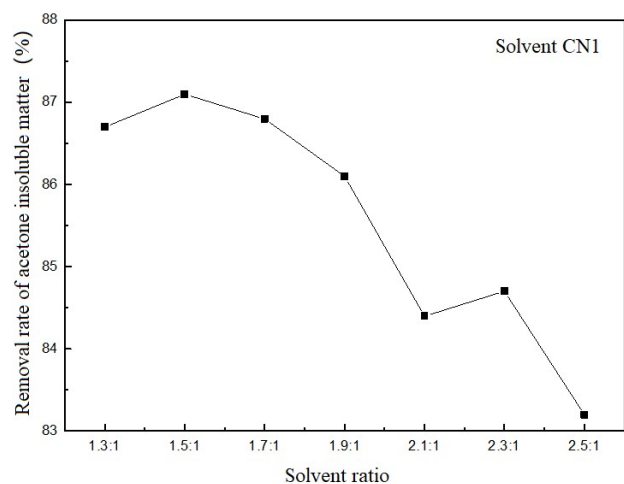
According to the orthogonal experiment, the optimal conditions for CN1 were as follows: solvent ratio 2:1, settling temperature 100°C, settling time 0.5h; The optimal combination of conditions for CN6 was as follows: solvent ratio 2:1, settling temperature 140°C, settling time 1h. Therefore, when the solvent ratio was investigated, the fixed temperature of CN1 was 100°C, the time was 0.5h, and the solvent ratio was 1.3:1, 1.5:1, 1.7:1, 1.9:1, 2.1:1 and 2.3:1. CN6 was fixed at 140°C for 1h, and the solvent ratios were 1.6:1, 1.8:1, 2.0:1, 2.2:1, 2.4:1, and 2.6:1. In order to choose the best solvent ratio, the most important evaluation index is the removal rate of acetone insoluble matter. Meanwhile, the neutralisation value and the improvement of kinematic viscosity of heat transfer oil are also investigated under the same conditions.

It can be seen from Figure 4 that as the solvent ratio increases, the removal rate of acetone insoluble first increases and then decreases. The absolute value of the removal rate of acetone insoluble is not much different. The solvent ratio of CN1 is 1.5:1. The extraction effect is the best. The removal rate of acetone insoluble matter is 87.1% through one extraction and sedimentation. It can also be observed that the optimal solvent ratio of CN6 is 2.4:1. At this solvent ratio, the acetone insoluble matter will be

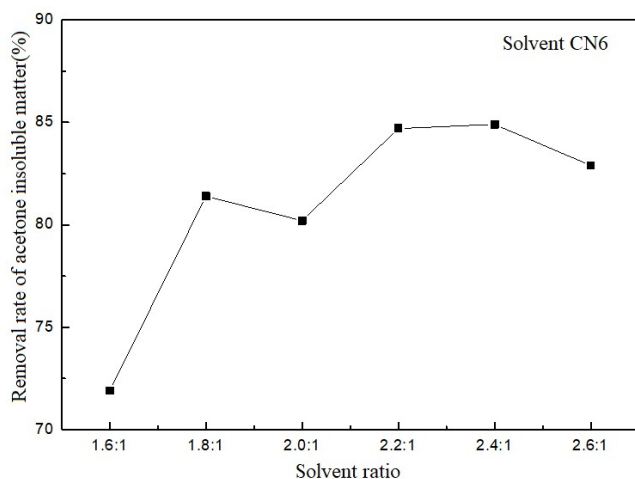
removed through one extraction and sedimentation. The removal rate reached 84.9%.

It can be seen from Figure 5 that the change rule of the neutralisation value is similar to the removal rate of acetone insoluble matter, indicating that the acidic substances in the heat transfer oil mainly exist in the gum asphalt. After removing the gum asphalt, the neutralisation value of the heat transfer oil can be effectively reduced.

Figure 4 The removal rate of acetone insoluble solids comparison in different solvents ratio about CN1 and CN6 solvents (a) removal rate of solvent CN1 insoluble in acetone, (b) removal rate of solvent CN6 insoluble in acetone



(a)



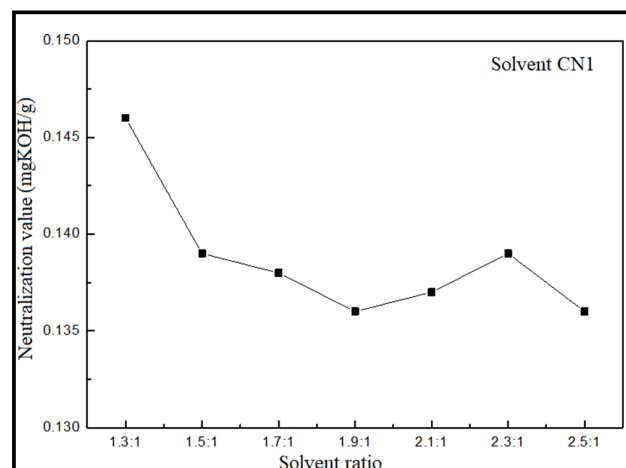
(b)

It can be seen from Figure 6 that after solvent extraction, the kinematic viscosity of the heat transfer oil can be reduced to the normal index of 13.0–39.5 mm²/s. The excessive kinematic viscosity of the heat transfer oil is mainly caused by the gum asphalt in the heat transfer oil. Caused by the high content, after removing the acetone insoluble matter, the kinematic viscosity of the heat transfer oil has been significantly improved.

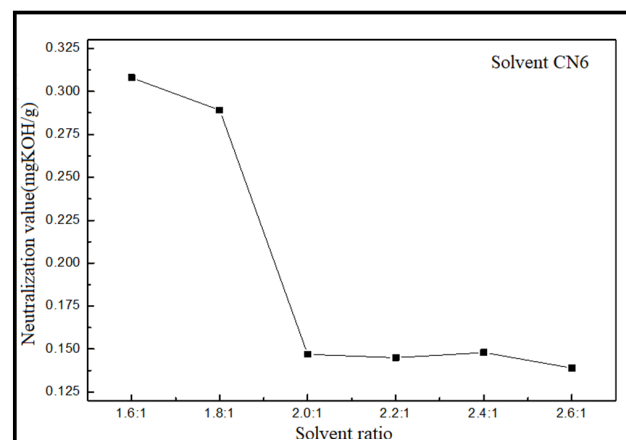
From Figure 7, based on the heat transfer oil extraction sedimentation experiment under the different solvent ratio,

it can be concluded that the CN1 solvent in solvent ratio of 1.5:1, CN6 solvent in solvent ratio of 2.4:1 under the highest acetone insoluble content removal rate. From Figures 5, 6 and 7, under this solvent ratio, settlement after an extraction, two kinds of solvents in the acid value of heat transfer oil and kinematic viscosity drop to normal levels and can maintain high yield.

Figure 5 The acid value comparison of HTF in different solvent ratio about cn1 and CN6 solvents (a) neutralisation value of solvent CN1, (b) neutralisation value of solvent CN6



(a)



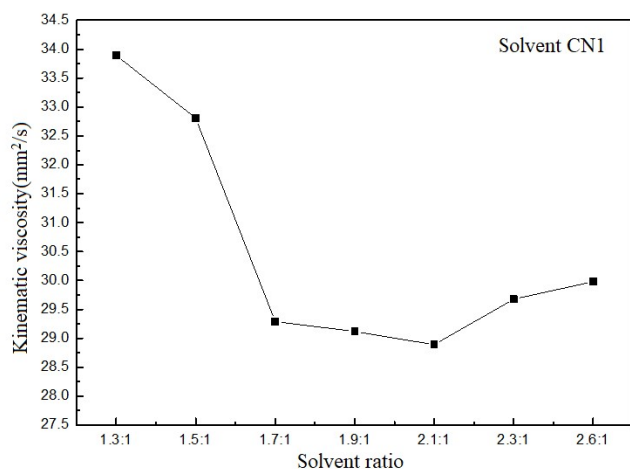
(b)

4.2 Effect of settling temperature on extraction efficiency

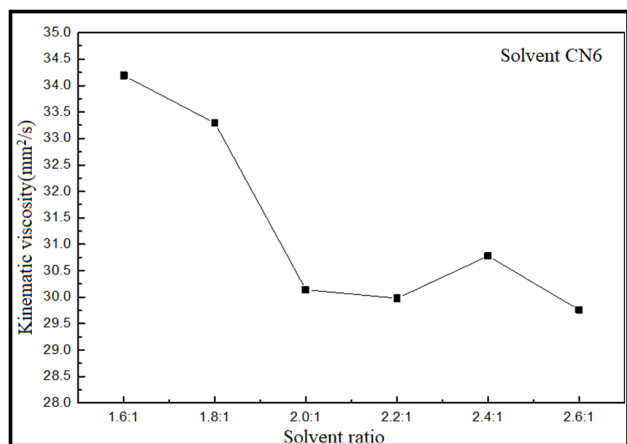
In the experiment, the optimal solvent ratio of CN1 is 1.5:1, and the optimal solvent ratio of CN6 is 2.4:1. Due to the large difference between the boiling points of the two solvents and the different optimum settling temperatures obtained by the orthogonal test, CN1 is 100°C and CN6 is 140°C, the conditions selected for the investigation of settling temperature are as follows: The solvent ratio of CN1 was 1.5:1, the settling time was 0.5h, the settling temperature was 80°C, 90°C, 100°C, 110°C, 120°C, 130°C; The solvent ratio of CN6 was 2.4:1, the settling time was

1h, and the settling temperature was 130°C, 140°C, 150°C, 160°C, 170°C and 180°C.

Figure 6 The kinematic viscosity comparison of HTF in different, (a) kinematic viscosity of solvent CN1 (b) kinematic viscosity of solvent CN6

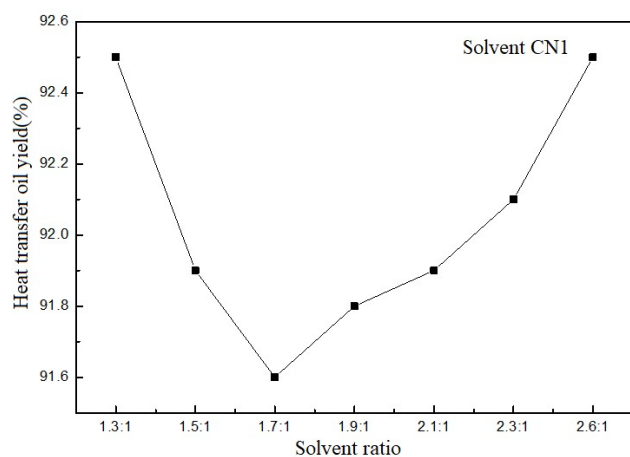


(a)



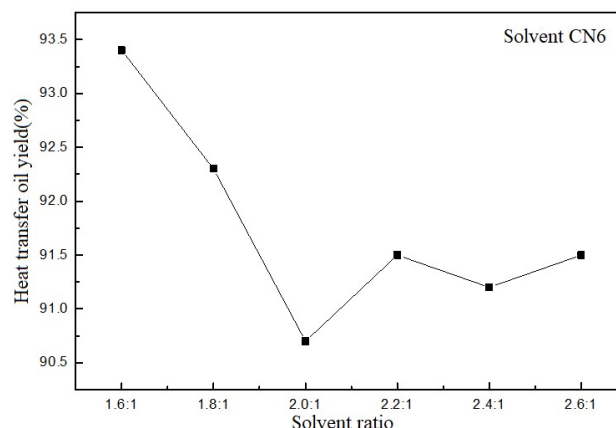
(b)

Figure 7 HTF yield comparison in different solvent ratio about CN1 and CN6 solvents, (a) solvent CN1 heat transfer oil yield (b) solvent CN6 heat transfer oil yield



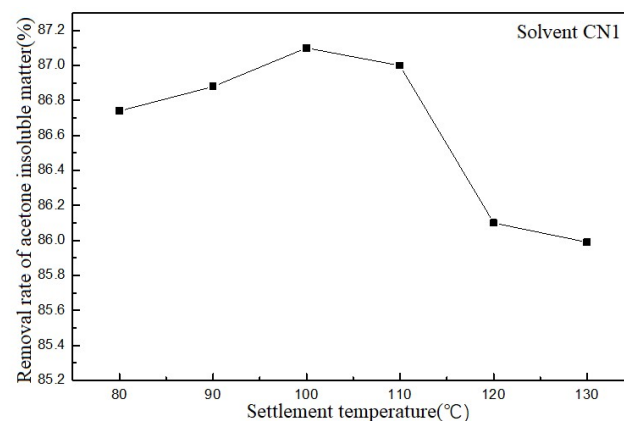
(a)

Figure 7 HTF yield comparison in different solvent ratio about CN1 and CN6 solvents, (a) solvent CN1 heat transfer oil yield (b) solvent CN6 heat transfer oil yield (continued)

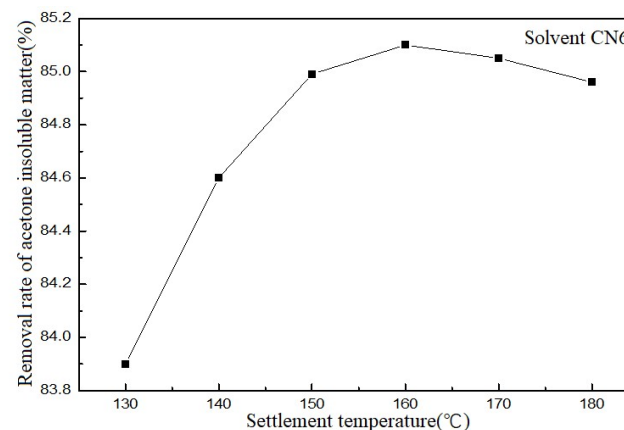


(b)

Figure 8 The removal rate of acetone insoluble solids comparison in different settling temperature about CN1 and CN6 solvents, (a) Removal rate of solvent CN1 insoluble in acetone (b) Removal rate of solvent CN6 insoluble in acetone



(a)

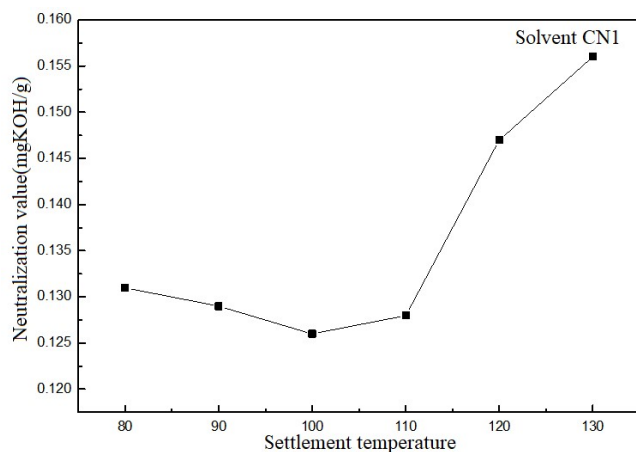


(b)

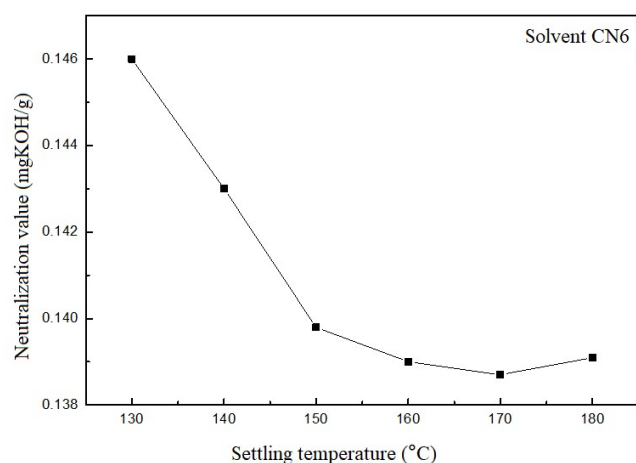
It can be seen from Figure 8 that when the sedimentation temperature of CN1 solvent is 80°C to 110°C, the sedimentation temperature has little effect on the removal

rate of acetone insoluble matter. When the temperature is higher than 110°C, the removal rate of acetone insoluble matter is obvious. When the temperature is 100°C, the removal effect of acetone insoluble matter is the best, which is 87.1%. If the temperature is too low, the solubility of the solvent is too low to extract and separate the heat transfer oil from the acetone insoluble; if the temperature is too high, the solubility of the solvent to the oil will increase, which is not conducive to the removal of the acetone insoluble. The best sedimentation temperature of solvent CN6 is 160°C, and the removal rate of acetone insoluble matter at this temperature is 85.1%. Unlike the solvent CN1, the solvent CN6 has a high boiling point. Therefore, the optimal sedimentation temperature also appears in a higher temperature region, indicating that the solvent's ability to dissolve the heat transfer oil at different temperatures is closely related to the boiling point of the solvent itself.

Figure 9 The acid value comparison of extracted HTF in different settling temperature about CN1 and CN6 solvents, (a) neutralisation value of solvent CN1 (b) neutralisation value of solvent CN6

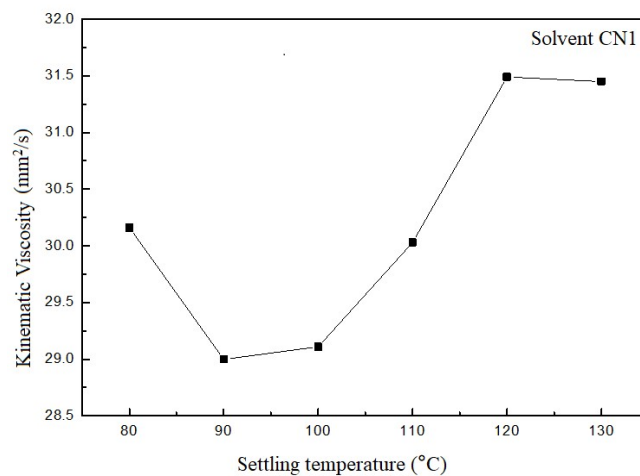


(a)

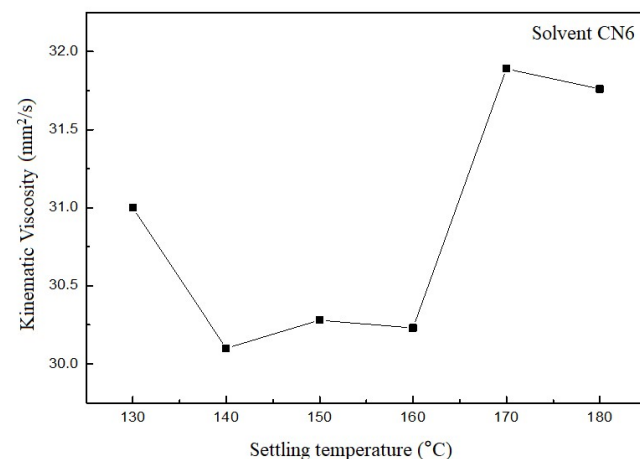


(b)

Figure 10 The kinematic viscosity comparison of extracted HTF in different settling temperature about CN1 and CN6 solvents, (a) kinematic viscosity of solvent CN1 (b) Kinematic viscosity of solvent CN6



(a)

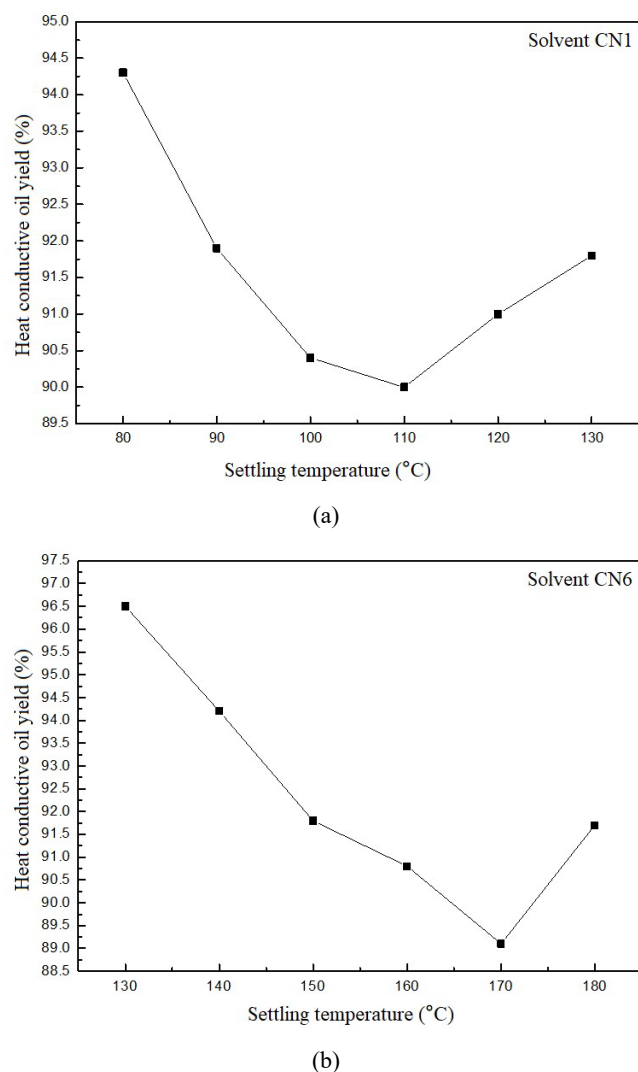


(b)

It can be seen from Figure 9 that the solvent CN1 has a low boiling point, so temperature changes in the low temperature range will have a greater impact on the solvent's solubility. The solvent CN6 has a high boiling point, so the best removal temperature for the acidic substances in the heat transfer oil also appears in a higher temperature range. When the sedimentation temperature is 170°C, the removal of the acidic substances in the heat transfer oil has the best effect.

The gum asphalt in the heat transfer oil are removed through the solvent extraction process to improve the viscosity of the heat transfer oil. It can be seen from Figs. 10 and 11 that although the viscosity of the heat transfer oil changes steadily as the temperature changes. Although the yield of heat transfer oil changes significantly with the change of the sedimentation temperature, the minimum yield can still be maintained at 90% and above.

Figure 11 HTF yield comparison in different settling temperature about CN1 and CN6 solvents, (a) solvent CN1 heat transfer oil yield (b) solvent CN6 heat transfer oil yield



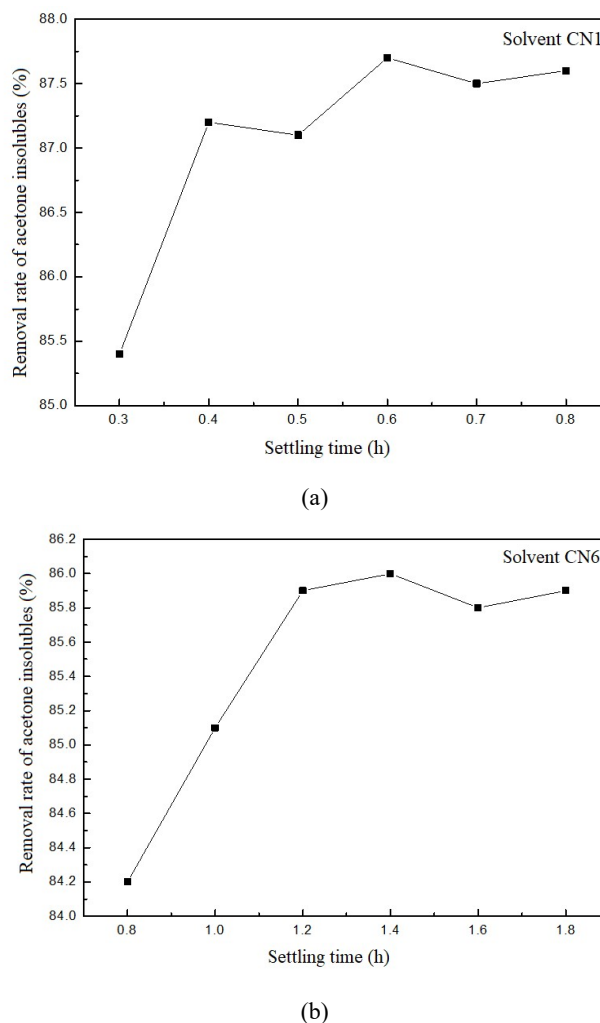
4.3 Effect of settling time on extraction effect

In general, an appropriate increase in settling time is beneficial to the precipitation of acetone insoluble substances, but too long settling time will make a part of the light gum asphalt back to the solvent. The heat transfer oil used in this experiment has been in online circulation, so a long settling time will also affect the normal application of the heat transfer oil. In order to ensure the effect of solvent extraction without affecting the normal use of heat transfer oil, it is necessary to investigate the solvent extraction settlement time of heat transfer oil.

Through the investigation of solvent ratio and settling temperature of CN1 and CN6, the best solvent ratio of extraction and settling temperature of CN1 is 1.5:1 and the best settling temperature is 100°C. The optimum solvent ratio of CN6 was 2.4:1 and the optimum precipitation temperature was 160°C. In the previous orthogonal experiment, the optimal settling time of solvent CN1 is 0.5h, and that of solvent CN6 is 1h. Therefore, in the investigation of time, the experimental conditions of solvent

CN1 were: solvent ratio of 1.5:1, settling temperature of 100°C, settling time of 0.3h, 0.4h, 0.5h, 0.6h, 0.7h, 0.8h; The experimental conditions of solvent CN6 were as follows: solvent ratio 2.4:1, settling temperature 160°C, settling time 0.8h, 1h, 1.2h, 1.4h, 1.6h, 1.8h.

Figure 12 The removal rate comparison of acetone insoluble solids in different settling time about CN1 and CN6 solvents, (a) removal rate of solvent CN1 insoluble in acetone (b) removal rate of solvent CN6 insoluble in acetone

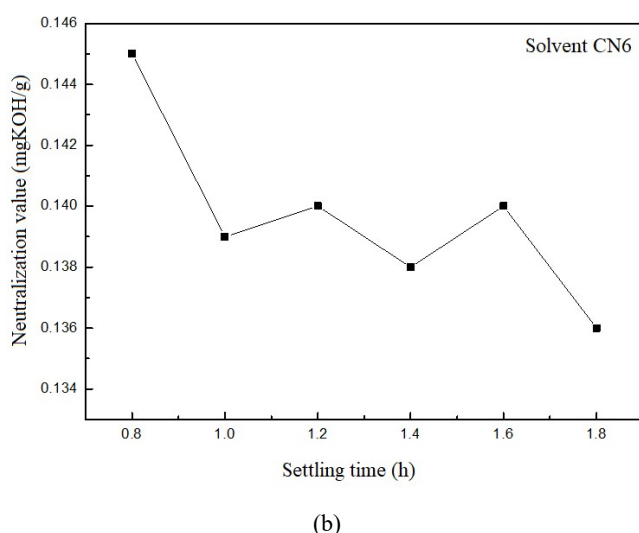
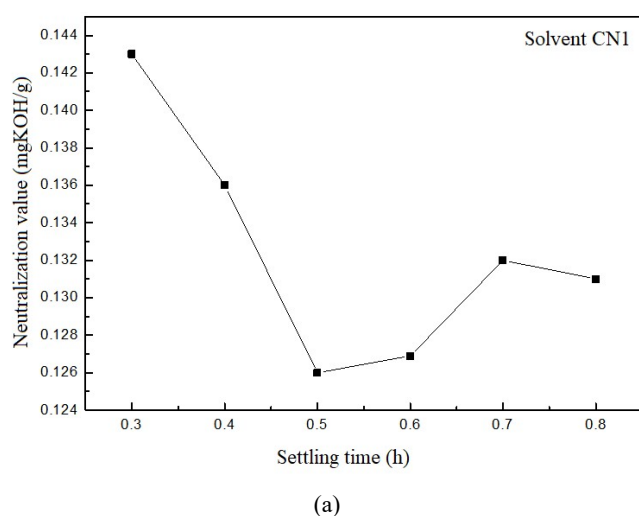


It can be seen from Figure 12 the best sedimentation time of solvent CN1 is about 0.6h. Under this sedimentation time, after one extraction sedimentation, the removal rate of acetone insoluble matter can reach 87.7%; the best sedimentation time of solvent CN6 is about 1.4h, this sedimentation time is lower, after one extraction and sedimentation, the removal rate of acetone insoluble matter can reach 86%. In summary, when the solvent ratio is 1.5:1, the sedimentation temperature is 100°C, and the sedimentation time is 0.6h, the solvent CN1 has the best removal effect on the acetone insoluble matter in the heat transfer oil; the solvent CN6 has the solvent ratio of 2.4:1 and the sedimentation temperature is 160°C, the sedimentation time is 1.4h, the effect of removing the acetone insoluble matter in the heat transfer oil is the best.

From Figures 13–15, under these settling time, settlement after an extraction, two kinds of solvents in the acid value of heat transfer oil and kinematic viscosity drop to normal levels and can maintain high yield.

Through the investigation of solvent ratio, settling temperature and settling time, it can be found that solvent CN1 can achieve the best removal effect of acetone insoluble matter under the conditions of solvent ratio of 1.5:1, settling temperature of 100°C and settling time of 0.6h. The solvent CN6 can achieve the best removal effect of acetone insoluble matter under the conditions of solvent ratio of 2.4:1, precipitation temperature of 160°C and deposition time of 1.4 h.

Figure 13 The acid value comparison of extracted HTF in different settling time about CN1 and CN6 solvents, (a) neutralisation value of solvent CN1 (b) neutralisation value of solvent CN6



The properties of the heat transfer oil obtained by the two solvents under the best extraction conditions are shown in Table 4.

Figure 14 The kinematic viscosity comparison of extracted HTF in different settling time about CN1 and CN6 solvents, (a) Kinematic viscosity of solvent CN1 (b) Kinematic viscosity of solvent CN6

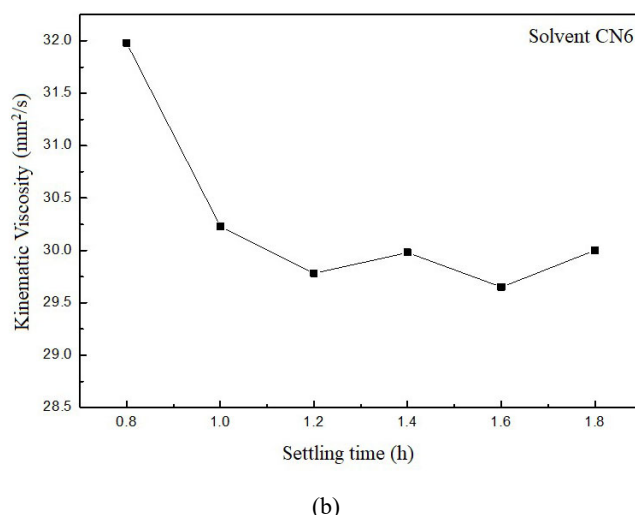
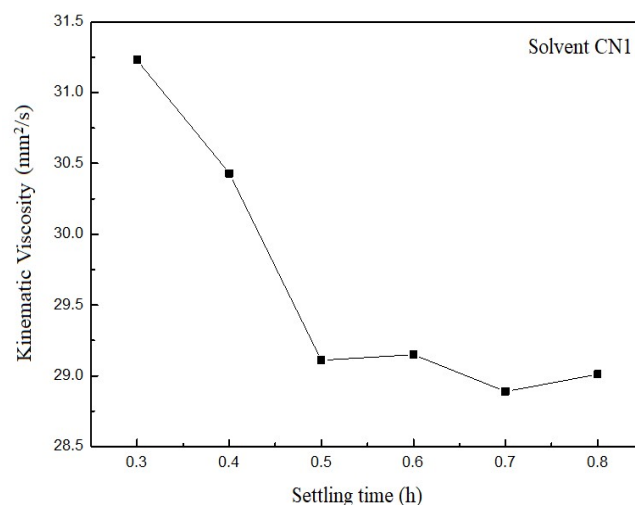
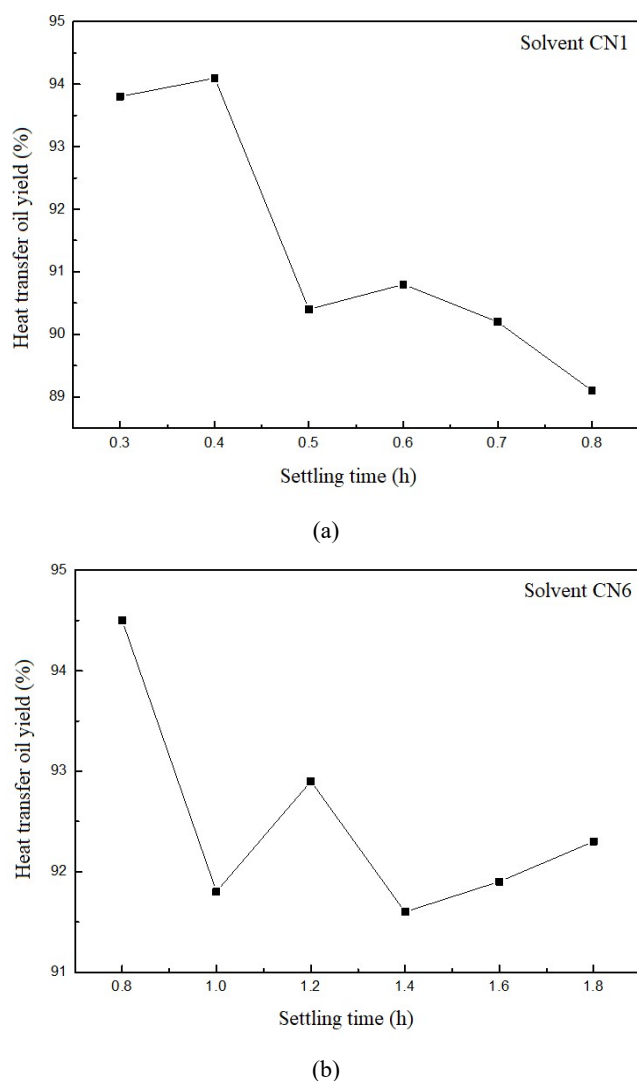


Table 4 Relevant properties data of extracted HTF under optimal conditions about CN1 solvent and CN6 solvent

Index	Removal rate of acetone insoluble matter (%)	Kinematic viscosity	Neutralisation value	Yield (%)
CN1	87.7	29.15	0.126	90.8
CN6	86	29.98	0.138	91.6

As can be seen from the data in Table 4, under the best conditions, CN1 has a higher removal rate of acetone insoluble matter through a single extraction settlement. At the same time, both solvents can adjust the kinetic viscosity and neutralisation value of heat transfer oil to normal levels, and both have a high yield of heat transfer oil. Comparing the optimal extraction conditions of the two solvents, it can be seen that solvent CN1 has better performance.

Figure 15 HTF yield comparison in different settling time about CN1 and CN6 solvents, (a) solvent CN1 heat transfer oil yield (b) solvent CN6 heat transfer oil yield



5 Numerical simulation of asphalt settlement separation tank

5.1 Establishment of geometric model of settling tank

The separation of oil in the settlement separation tank and the gelatinous asphaltene occupies a very important position in the heat transfer oil online regeneration system (Yu and Mao, 2014). Due to the limitation of experimental conditions, the experimental device of the extraction settlement experiment is simple, and the experiment is carried out in the static state. In practical industrial applications, settling tank is the key equipment for online regeneration of heat transfer oil. There is an inlet velocity for mixed fluid to enter the settling tank. If the settling effect of settling separation tank is poor, the purpose of removing gum asphaltene cannot be achieved. The heat transfer oil is separated according to the actual operating conditions of the heat transfer oil. The processing capacity of the heat transfer oil is 0.5 m³/h, the solvent used is 0.8 m³/h according to the experiment, and the actual oil

temperature of the heat transfer oil is 190°C. According to the treatment process, the solvent and heat transfer oil can be separated at reduced pressure after the extraction. At the same time, the temperature of the mixture in the settling tank can provide part of the heat of the solvent vaporisation, so that the solvent and heat transfer oil are fully separated, to meet the requirements of the process, so the settling tank temperature is selected as 120°C. In actual working conditions, the pressure of heat transfer oil pipeline is 0.8 MPa. In order to ensure the full mixing of solvent and heat transfer oil and the state of solvent liquid in the settling tank, the inlet pressure is selected as 0.8 MPa. Considering that the solvent ratio is the main influencing factor, the solvent ratio is investigated. Sedimentation time has little effect on acetone removal rate and the actual situation of the server, the calculation time is controlled to 20 minutes. At present, the research on settlement separator has been mature, and it is often used in oil and water separation. Therefore, Fluent software is used to simulate and analyse the separation tank.

5.1.1 Simplification of settlement tank model

The structure of the settlement separation tank is simple and belongs to the centre symmetry structure. The internal flow field is distributed in three dimensions. In actual working conditions, there will be a gas phase space in the upper part of the separation tank. The purpose is to achieve pressurisation and avoid sudden pressure change, which has no influence on the separation effect of the internal flow field. Therefore, the gas phase space is not considered, but only the liquid phase space is considered (Shabani and Mazahery, 2012; Polderman et al., 1997). The separation tank is initially filled with liquid, and the fluid is treated as an incompressible continuous fluid. The separation process is carried out under the condition of constant temperature, and all phases are in the state of heat transfer equilibrium. Therefore, the separation process is regarded as an adiabatic process. Different internals are generally placed in settlement separation tanks to improve fluidity by optimising internals. This tank body does not consider the influence of internal components, the entrance is selected elbow, internal set overflow plate, the model is simplified.

5.1.2 Sedimentation tank model and physical property parameters

The model selected in this section is obtained through calculation and selection. The geometric dimensions of the main body of the asphalt separation tank are as follows: the length of the cylinder is 1,910 mm, the diameter of the cylinder is 1,000 mm, the height of the head is 275 mm, the height of the overflow plate is 750 mm, and the diameter of the inlet and outlet pipes is 25 mm. The original model is shown in Figure 16, and the simplified three-dimensional model is shown in Figure 17.

After the heat transfer oil and solvent are fully mixed, the flow rate of 1.3 m³/h enters the asphalt separation tank. The inlet pressure of the asphalt separation tank is 0.8 MPa,

the outlet pressure is 0.1 MPa, the gravity acceleration is the negative direction of Y, the mixing phase concentration is 96%, the density of the gum asphaltene is 1,100 kg/m³, and the concentration is 4%.

Figure 16 Original model

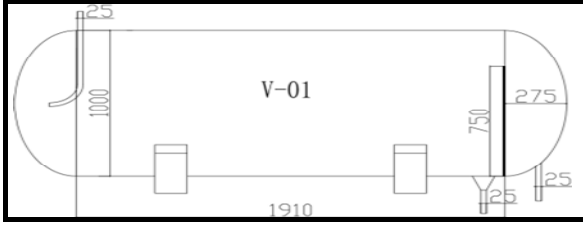
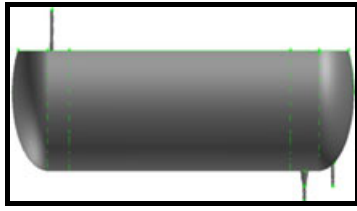


Figure 17 Simplified three-dimensional model



- a The velocity of the inlet fluid calculated from equation (1):

$$v = \frac{Q}{s} = \frac{Q}{\pi r^2} \quad (1)$$

where, Q represents flow rate, m³/h; s represents the entrance sectional area, m²; R represents the inlet pipe radius, m.

- b Density of the mixed phase

The heat transfer oil and solvent are mixed according to the solvent ratio, and the density of their mixing needs to be calculated, which can be obtained by equation (2):

$$\rho_{mix} = \rho_o V_o + \rho_s V_s \quad (2)$$

where ρ_o and ρ_s respectively represents the density of heat transfer oil and solvent, kg/m³; V_o and V_s represent the volume fractions of the heat transfer oil and the solvent (the sum of the two is 1).

- c Dynamic viscosity of mixed phase

The dynamic viscosity of the mixed phase is determined according to the solvent ratio, and the corresponding volume fraction of the components is different with the different solvent ratio. Therefore, the viscosity calculation is based on the Arrhenius viscosity model, which can be calculated by equation (3):

$$\log \mu = \sum_i x_j \log \mu_i \quad (3)$$

- d Reynolds coefficient

The magnitude of Reynolds coefficient determines laminar flow or turbulent flow, so it can be calculated by equation (4):

$$R = \frac{\rho D v}{\mu} \quad (4)$$

where, ρ represents the density of the mixed phase, kg/m³; D stands for inlet pipe diameter, m; μ

Represents the dynamic viscosity of the mixed phase, kg/m s.

- e Turbulence intensity coefficient

Turbulence intensity coefficient is calculated based on Reynolds coefficient, and the calculation Equation is shown in equation (5):

$$I = 0.16(R)^{-0.125} \quad (5)$$

where, I is the turbulence intensity coefficient; Re is the Reynolds coefficient.

Four kinds of solvent CN1, CN2, CN4 and CN6 with better test results were selected through orthogonal test. When the mixing temperature of sedimentation tank was 120°C (determined according to the process), According to the above calculation equation, the boundary conditions of four solvents in the same solvent ratio (volume ratio) of 2:1 and CN1 solvent (optimal in the experiment) in different solvent ratios of 0.5:1, 1:1 and 2:1 were calculated respectively. Table 5 lists the summary table.

Table 5 Boundary conditions value

Solvent	$V(m/s)$	$\rho_{mix}(kg/m^3)$	$\mu(10^{-4}kg/m \cdot s)$	$Re(10^4)$	$I(\%)$
CN1(0.5)	0.736	758.61	8.659	1.61	4.77
CN1(1)	0.736	735.19	5.658	2.39	4.54
CN1(2)	0.736	711.76	3.697	3.54	4.32
CN2(2)	0.736	663.58	4.232	2.054	4.43
CN4(2)	0.736	624.46	3.322	3.46	4.33
CN6(2)	0.736	899.52	8.423	1.96	4.65

Through the data in the table, we can quickly set the inlet and outlet boundary conditions in Fluent.

5.1.3 Setting of boundary conditions

1 Inlet boundary conditions

In the asphalt separation tank, the inlet diameters of oil and asphalt phases are equal, both of which are 25 mm. The inlet boundary conditions are consistent. The inlet velocity is 0.736 m/s, the direction is the negative direction of the Y- axis, and the pressure is set as 0.8 MPa.

2 Exit boundary conditions

The fluid in the asphalt separation tank is incompressible, and the outlet diameter of oil and asphalt is equal, both of which are 25 mm. The mixture stays in the separation tank for a period of time, so the oil phase adopts the velocity outlet boundary, and the

outlet pressure is set at 0.1 MPa, while the asphalt phase is discharged regularly, so the outlet is set as the wall surface.

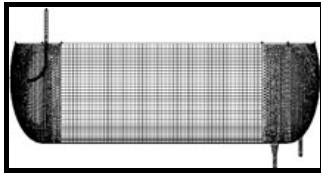
3 Wall conditions of asphalt separation tank

The place where the oil phase and gas phase contact in the asphalt separation tank is not considered, so it is set as a free flow outlet, the overflow plate is set as the wall surface, and the others are set as the wall surface by default.

5.1.4 Selection of meshing internals of the model

Asphalt separating tank 3D model of meshing is done in gambit software, the settlement of separating tank structure is simpler, symmetry and centre. At the time of mesh, we need to model, cut into parts, ground up whole consists of structured and unstructured meshes, this can not only achieve the aim to reduce the number of grids, but also guarantee the quality of the grid (Reiterer and Kalab, 2021). Grid division of asphalt separation tank is shown in Figure 18.

Figure 18 Separation tank grid



The quality of the 3D grid of the asphalt separation tank is composed of 415,825 grids, and there is no negative volume grid. The maximum deflection angle of the grid is 0.763761, which meets the calculation requirements.

The inlet of asphalt separation tank is selected as elbow pipe, and the inner component is only overflow plate, as shown in Figure 19.

Figure 19 Elbow and overflow plate

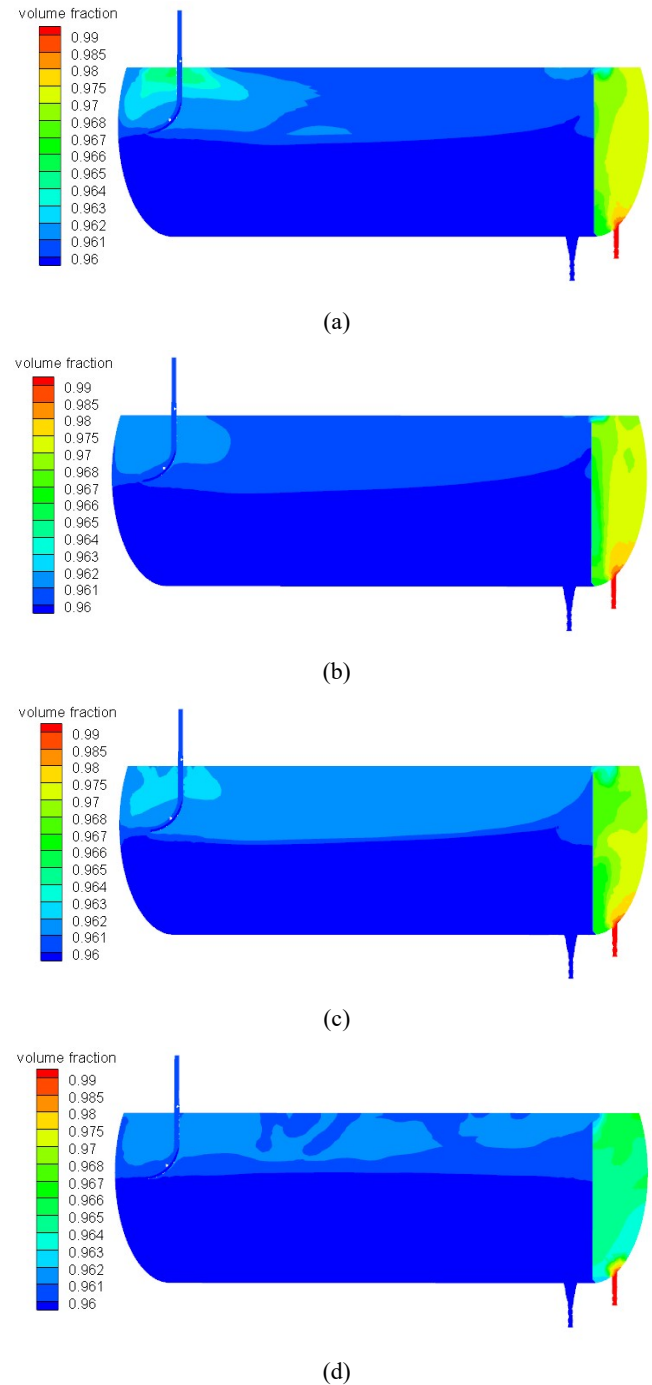


5.2 Simulation calculation results and analysis of settlement tank

5.2.1 Concentration field analysis of several different solvents under the same solvent ratio

When the solvent ratio of CN1, CN2, CN4 and CN6 is 2:1, considering the long-time of simulation calculation, the simulation calculation time is selected as 20 minutes. The volume fraction cloud diagrams of the oil phase after settling and separating with four solvents for 20 minutes are shown in Figure 20.

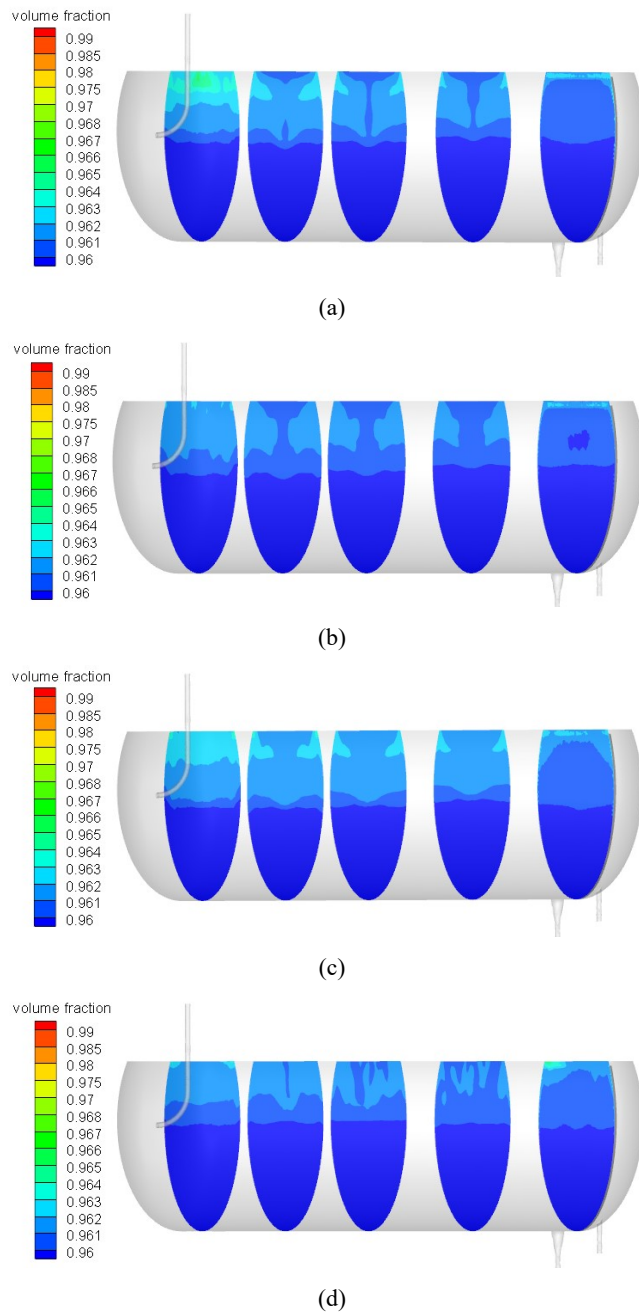
Figure 20 The oil phases volume fraction cloud chart of different solvent in $Z = 0$ cross section, (a) CN1 solvent $Z = 0$ cross-section (b) CN2 solvent $Z = 0$ cross-section, (c) CN4 solvent $Z = 0$ cross-section (d) CN6 solvent $Z = 0$ cross-section (see online version for colours)



As shown in Figure 20, the oil phase volume fraction cloud diagram with the solvent ratio of 2:1 of the four solvents show that the separation effect of these four solvents is not very ideal after 20 minutes of separation, but the effect of CN1 solvent is better than the other three solvents. In the cloud figure, the volume fraction of oil phase in CN1 solvent reaches 96.5%, while the volume fraction of oil phase in other three solvents does not reach 96.3%. The separation effect near the overflow port except CN6 solvent is basically like that of other three solvents. The separation

effect of the four solvent oil phase exports is ideal, the volume fraction of oil phase reaches more than 99%, and the separation rate reaches more than 80%. According to the cloud map, the settling effect of CN1 of the four solvents is the best and consistent.

Figure 21 The volume fraction of oil phases cloud chart in different cross sections of different solvents along x axis cross section, (a) cross-section cloud image of CN1 solvent (b) cross-section cloud image of CN2 solvent (c) cross-section cloud image of CN4 solvent (d) cross-section cloud image of CN6 solvent (see online version for colours)



However, the effect of CN6 solvent was better than that of CN4 solvent in the experiment, and the effect of CN4 solvent was slightly better than that of CN6 solvent in the simulation, which may be related to the properties of the

solvent itself. The boiling point of CN6 solvent is higher than that of CN4 solvent. When the temperature is 120°C, the density of CN4 solvent changes more than that of CN6 solvent. Moreover, the extraction mechanism of CN4 solvent and CN6 solvent is different, so that the mixed phase of CN4 solvent is easier to separate from the asphalt phase.

The separation effect of the four solvents is related to the separation time and inlet speed, so the separation effect of 20 minutes when the inlet speed of the four solvents is not good, but it is comparable.

Figure 21 shows the cross section of the four solvents along the X-axis.

As can be seen from Figure 21, the volume fraction of oil phase of these four solvents is constantly changing along the X-axis at $X = 0.1$ m, $X = 0.5$ m, $X = 0.9$ m, $X = 1.4$ m and $X = 1.9$ m, but the concentration field distribution is relatively similar. Except for CN6, the distribution of the concentration field in the cross section of other solvents is symmetric. It can also be seen from the figure that the separation effect of the middle part of the tank is not necessarily optimal, but the separation effect of the two sides is better. The separation effect at the overflow port is basically the same. The location of the overflow port represents the actual discharge position of asphalt phase, and the volume fraction of the oil phase reaches 96.3%, indicating that the separation effect is not ideal, but it can be seen that the separation effect of CN1 solvent is good, so it can provide guidance.

As can be seen from Figures 20 and 21, the separation effect of CN1 solvent is the best, which is consistent with the best sedimentation effect of CN1 solvent obtained in the orthogonal test.

In order to obtain the separation effect of CN1 solvent in different solvent ratio, it is necessary to ensure the same separation time and sedimentation temperature. Therefore, the separation results of CN1 solvent under the solvent ratio of 0.5:1, 1:1 and 2:1 are compared and analysed below, and the optimal solvent ratio of 2:1 is verified.

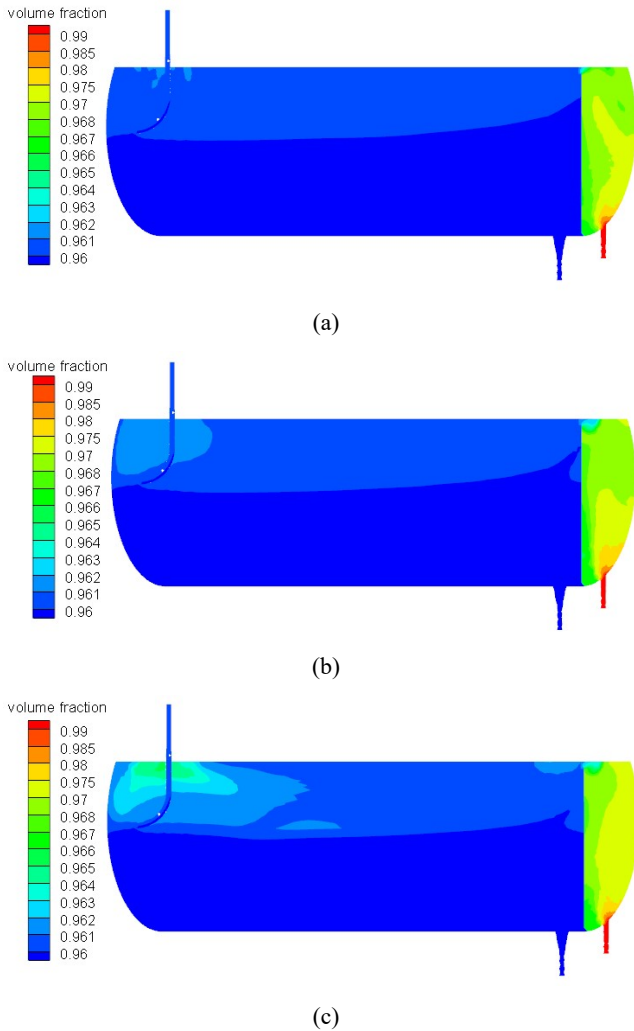
5.2.2 Oil phase volume fraction cloud graph analysis of CN1 solvent at different solvent ratios

Figure 22 shows the oil phase volume fraction cloud diagrams of CN1 solvent separated for 20 minutes at the ratio of three solvents.

According to Figure 22 cloud diagram of oil phase volume fraction of CN1 solvent with solvent ratio of 0.5:1, 1:1 and 2:1, it can be seen that the separation effect of CN1 solvent with solvent ratio of 2:1 is significantly better than that of the other two solvent ratios after settling and separation for 20 minutes. However, the settlement separation time is short, so the overall separation effect of tank is not ideal. The separation effect of the inlet part and the part near the overflow port is better than that of the middle part of the tank. The oil phase concentration at the export is above 99%, indicating that the oil phase at the export is well separated from the asphalt phase, and the

asphalt separation rate is above 80%. From the nephograph, it can be seen that the separation effect of solvent ratio 2:1 is better than 1:1 and better than 0.5:1. Since the density of solvent CN1 is less than that of heat transfer oil, the density of the mixed phase decreases with the increase of the solvent ratio after the mixing of the two, and the settlement separation considers the density difference. Therefore, with the increase of the solvent ratio, the density difference between the mixed phase and the asphalt phase will increase, making the separation effect of the two phases become better. The separation effect of solvent CN1 is better when the solvent ratio is 2:1, which is consistent with the solvent ratio of 2:1 obtained in the orthogonal test under the optimum process combination conditions, indicating that the simulation results are basically accurate.

Figure 22 The volume fraction of oil phases in different ratio about cn1 solvent in $z = 0$ cross section, (a) CN1 solvent ratio 0.5:1 (b) CN1 solvent ratio 1:1 (c) CN1 solvent ratio 2:1 (see online version for colours)

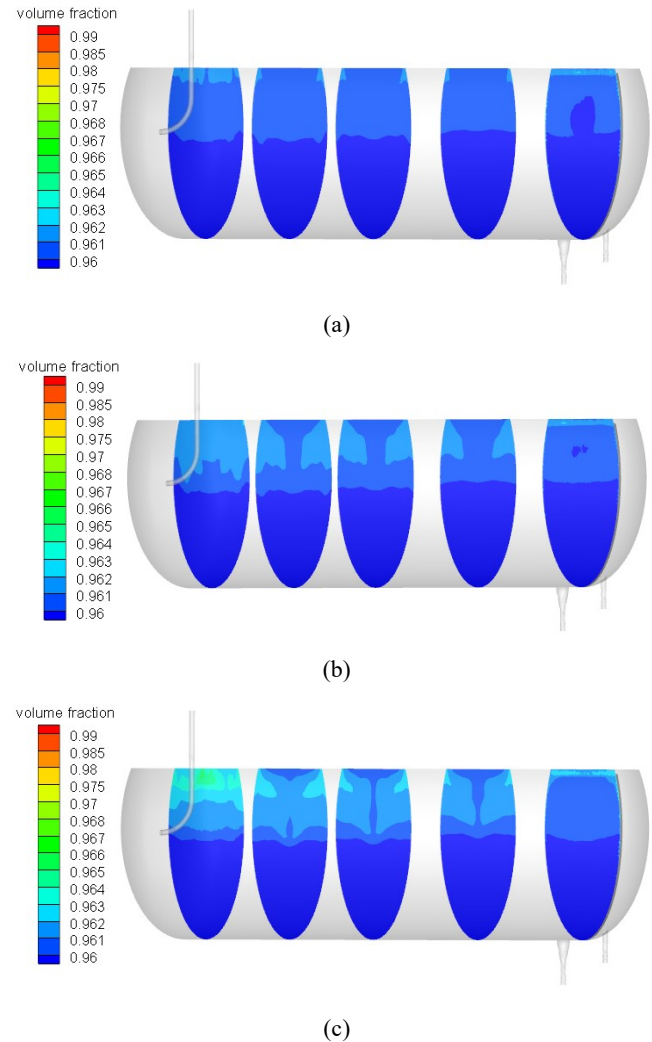


When the solvent ratio of CN1 is 0.5:1, 1:1 and 2:1, the oil phase volume fraction cloud diagram of the settling tank along the X-axis at different cross sections are shown in Figure 23. As can be seen from Figure 23, the oil phase volume fraction at $X = 0.1$ m, $X = 0.5$ m, $X = 0.9$ m, $X = 1.4$ m and $X = 1.9$ m along the X-axis direction under

the three solvent ratios is significantly different. The distribution of concentration field of CN1 solvent in different solvent ratios is axisymmetric, and the separation effect at the inlet is obviously better than that at the middle part of the tank, and the effect near the overflow plate is similar, and the separation effect is better than that at the tank part. However, the effect of separation for 20 minutes is mediocre, and the settlement time should be increased for separation.

According to the processing requirements of the actual working conditions of FPSO ship, the fluid has a speed. In order to ensure the full separation of asphalt phase, the settlement separation time is selected as 1h to provide guidance for field application.

Figure 23 The volume fraction of oil in different section about CN1 solvent, (a) CN1 solvent ratio 0.5:1 cross-section cloud image (b) CN1 solvent ratio 1:1 cross section cloud (c) CN1 solvent ratio 2:1 cross-section cloud image (see online version for colours)



5.2.3 Analysis of the concentration field of CN1 when the solvent ratio was 2:1

The oil phase volume fraction cloud diagrams of CN1 after 1h solvent deposition are shown in Figure 24.

As can be seen from Figure 24, the solvent ratio of CN1 is 2:1. After settling and separating for 1 hour, the overall separation effect is good, the stratification is obvious, the asphalt phase is basically at the bottom of the tank, and the oil phase is at the upper part of the tank. The position near the overflow plate represents the actual discharge position of asphalt, so the oil phase concentration here can best reflect the actual situation. It can be seen that the volume fraction of oil phase here reaches 98.7%, and the separation rate reaches 67.5%. The oil phase volume fraction reached 99.5% and the separation rate reached 90%, indicating that the separation effect of two phases was very good.

Figure 24 Cn1 solvent settling one-hour concentration field,
(a) cross-section cloud image of CN1 along axis $Z = 0$
(b) cross-section cloud image of CN1 along the X-axis
(see online version for colours)

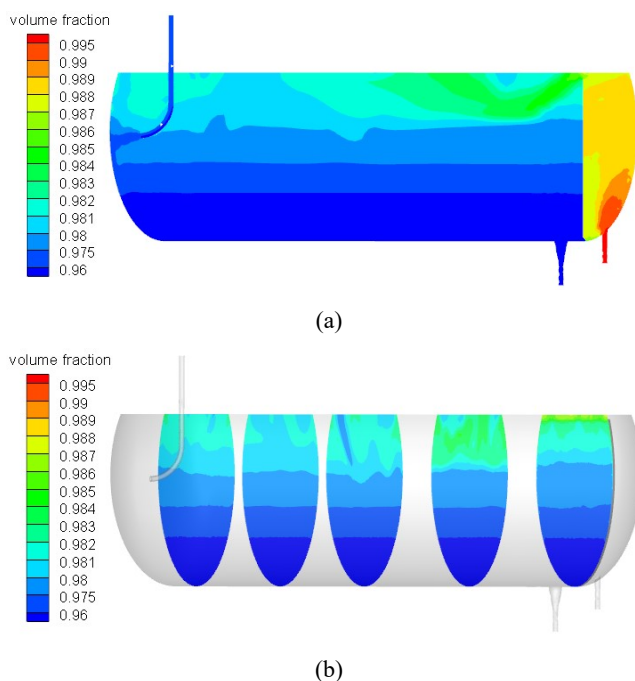
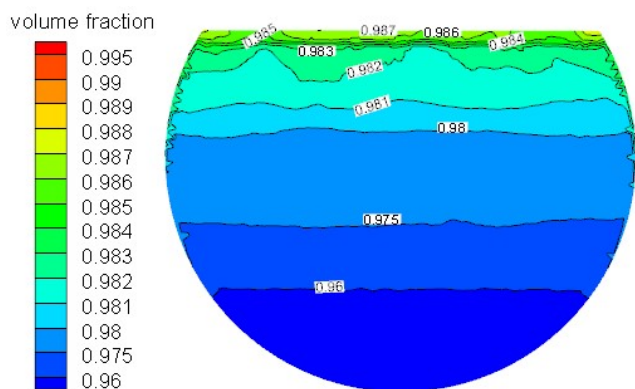


Figure 25 Oil concentration field about cn1 solvent in $x = 1.9$ m
(see online version for colours)



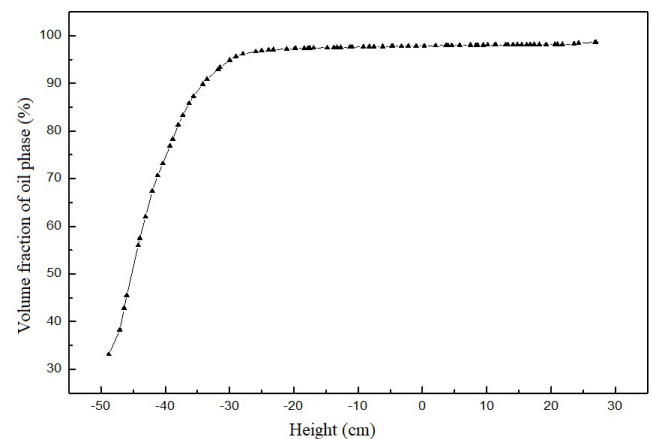
The distribution of the concentration field along different sections of the X-axis is non-axisymmetric. The middle and lower layers of the tank are more uniform, but the upper layers of the tank are more chaotic, which may be related to the existence of eddy current or back mixing. The overall

separation effect of the tank is quite different, the closer to the overflow plate, the more obvious the stratification. It can be seen from the cloud graph that increasing the settlement time is beneficial to the separation of two phases, so the appropriate settlement separation time is of great significance to industrial production.

Figures 25 and 26 show the simulation of oil phase concentration field and volume fraction longitudinal distribution of CN1 solvent along $X = 1.9$ m section

It can be seen intuitively from the oil phase concentration field cloud diagram and the longitudinal distribution simulation diagram of volume fraction in Figures 25 and 26 that the separation effect of CN1 solvent is obvious when the solvent ratio is 2:1. The volume fraction of oil phase reaches 98.7%, the separation rate reaches 67.5%, and the acetone insoluble matter removal rate is 85.8%. There is a certain deviation between simulation and test, and the deviation is 21.3%. Because the effects of extraction process and internal component parameters are not considered in the simulation process and the simulated solvent ratio is volume ratio while the experimental solvent ratio is mass ratio, the deviation is acceptable. Although the simulation with Fluent has limitations and cannot accurately reflect the real situation, the simulation results can provide reference for industrial production.

Figure 26 Simulation value of oil phase volume fraction distribution about cn1 solvent in $x = 1.9$ m (see online version for colours)



6 Conclusions

Aiming at the problem of performance deterioration of heat transfer oil during long-term use, this paper achieves the goal of online recycling and reuse of heat transfer oil through the solvent extraction regeneration method. To realise the regeneration of heat transfer oil, a set of extraction and sedimentation devices was designed to conduct sedimentation experimental research on different solvents. Then, the effectiveness of the method was verified by the extraction component analysis experiment, the related index detection experiment of the extraction heat transfer oil performance and the numerical simulation

analysis. Through orthogonal experiments, two extraction solvents were preliminarily determined. Through further experiments, CN1 solvent can reduce the insoluble heat transfer oil and acetone from 3,942 mg / 100 ml to 485 mg / 100 ml, which meets the requirements of use.

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