ISOC2013
Qingdao, China

油田化学
与提高原油采收率新进展

Advances in Oilfield Chemistry and Enhanced Oil Recovery

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本书按四个专题编排：提高采收率、调剖堵水、油水井处理和钻井液完井液化学，内容基本反映了近年来提高采收率和油田化学领域所取得的成果。所选论文涉及化学驱油剂研发及作用机理研究、驱油体系性能评价、堵水调剖新进展和新方法、油水井处理用化学剂和作用机理、钻完井新型处理剂和作用机理等领域。论文资料翔实珍贵，内容丰富，适用性强，应用范围广。

本书适合油田化学领域的工程技术研究人员参考，也可供石油与天然气工程学科相关领域的师生参考。

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前言

2013国际油田化学新进展（ISOC 2013）学术研讨会于2013年6月26～28日在山东青岛中国石油大学（华东）隆重召开。会议主题为：油田化学与提高采收率面临的技术创新与挑战。

会议由中国石油大学（华东）主办，中国石油大学（华东）石油工程学院油田化学系承办，中石油钻井工程技术研究院、中海油服油田事业部、中石化胜利油田采油院协办。共有来自美国、英国、加拿大、挪威、荷兰、哥伦比亚、科威特、墨西哥以及国内的40多家石油企业、科研院所及大专院校的200余位专家学者参会，中国石油大学（华东）副校长李兆敏致开幕词。会议收到国内外中英文论文共计80余篇，本书精选其中27篇英文论文和19篇中文论文收入论文集。

围绕本次会议主题，与会专家学者就国内外油田三次采油理论与技术的新进展、钻井液完井液新技术、油水井处理剂以及非常规油气（低渗、高温高盐、稠油油藏、页岩气、煤层气等）开采中油田化学工程面临的技术挑战等重大议题进行了交流研讨。会议共安排34场大会学术报告，其中美国得克萨斯大学奥斯汀分校、密苏里科技大学、堪萨斯大学、新墨西哥大学、加拿大卡尔加里大学、荷兰代尔夫特大学、哥伦比亚国家石油公司、美国麦克巴石油公司、科威特石油公司，以及国内北京大学、四川大学等高校和石油公司的专家应邀做了英文主题报告。通过会议期间的交流与讨论，加深了与会者对油田面临的化学挑战、应对策略和未来重点发展方向的理解。

通过举办国际油田化学新进展学术研讨会，为中国石油大学（华东）60周年校庆营造浓厚的学术氛围，增强了中国石油大学（华东）与海内外科研院所、高校和技术公司的合作，推动油田化学理论与技术研究的发展，为促进国内外油田化学工作者合作交流做出了积极贡献。

本论文集得到了组委会和学术委员会的大力支持。参与人员有赵明伟、宫厚健、范海明、江琳、耿杰、陈五花、孙明波、黄维安、徐加放、樊泽霞、王增宝、邱正松、曹杰。研究生赵昊阳、季岩峰、胡雷雷、孟令伟、魏绍龙、张斌、赵健、杜山山、马艳哲、蔡志彪等人也参与了相关工作。特别感谢 Barbie Kuntemeier 对英文的仔细修改。

由于时间仓促，文中不足之处在所难免，恳请读者批评指正。

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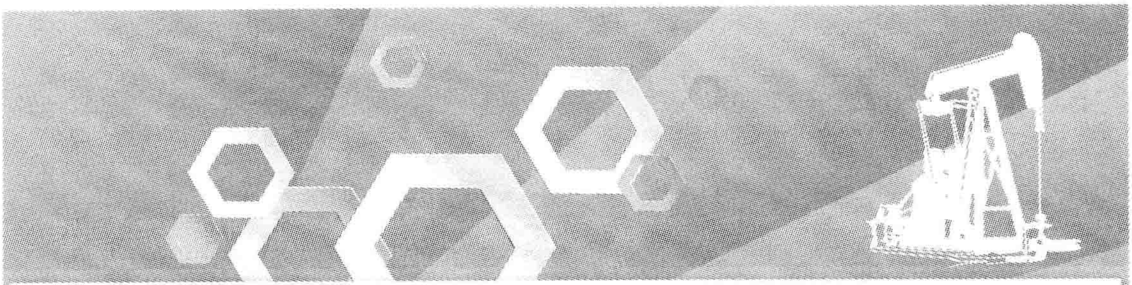

目录

Section 1
Chemical Enhanced Oil Recovery

International Symposium on Oilfield Chemistry
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Chemical Flooding Applied in the Shengli Oilfield

Xulong Cao, Xinwang Song, Jichao Zhang, Jing Shi, Fuqing Yuan

(Geological Scientific Research Institute of Shengli Oilfield Company, SINOPEC, Dongying 257015, P. R. China)

Abstract: The original oil in place suitable for chemical flooding in the Shengli oilfield is 16.05×10^8 ton. Though the resource is abundant, the oil recovery is poor due to rigorous reservoir conditions, including high temperature, high salinity, high divalent-cation content, high oil viscosity and severe heterogeneity. In an effort to counteract the rigorous reservoir conditions, polymer flooding technologies for class Ⅰ and Ⅱ reservoirs has been improved, and surfactant/polymer flooding has been implemented industrially. Heterogeneous combination flooding and foam flooding technologies have made great breakthroughs. A series of chemical flooding technologies have been developed, and these methods play a significant role in maintaining oil production in mature fields.

Key words: chemical flooding; polymer flooding; surfactant/polymer flooding; heterogeneous combination flooding, foam flooding

The original oil in place (OOIP) suitable for chemical flooding in the Shengli oilfield is 16.05×10^8 ton [1]. Though the available chemical flooding resources are tremendous, the reservoir conditions are difficult in terms of high temperature, high salinity, high divalent-cation content, high oil viscosity and severe heterogeneity. Stricter requirements have been established for reservoir recognition and residual oil distribution studies, heat-resistant and salt-tolerant capabilities and the viscofication of displacement systems. To counteract the rigorous reservoir conditions, a unique series of chemical flooding technologies have been developed.

1. Chemical flooding in the shengli oilfield

By the end of March 2013, 50 chemical flooding projects with an overall OOIP of 4.46×10^8 ton had been implemented in the Shengli oilfield. Chemical floods have contributed to an annual oil increment of 150×10^4 ton for nine continuous years. The cumulative oil increment due to chemical floods has reached 2273×10^4 ton, equivalent to 5.1% incremental recovery. It is estimated that the total incremental recovery will be 8.1%, and the recoverable reserve will increase by 3615×10^4 ton.

2. Chemical flooding technologies

With the continuing large-scale industrial application of chemical flooding, the remaining reservoir conditions are becoming increasingly harsh. This situation has spurred the development of new displacement systems with higher sweep efficiency and displacement efficiency. To help counteract the challenges of class Ⅲ high-temperature and high-salinity reservoirs, class Ⅳ high-permeability reservoirs, and reservoirs after polymer flooding, novel chemical flooding systems, such as surfactant/polymer flooding systems, heterogeneous combination flooding systems, and polymer enhanced foam flooding systems, have been developed.

2.1 Polymer flooding

It is well known that the mobility ratio plays an important role in oil recovery [2]. However, the viscosity of the polymer solution changes constantly during percolation, making the mobility ratio of polymer flooding difficult to determine [3,4]. To solve this issue, the correlation between the viscosity ratio of the polymer solution to oil and the enhanced recovery was studied. A reasonable viscosity ratio range was determined to be 0.15~0.5, with a comparatively large recovery increment magnitude. Numerical and physical simulations yielded consistent results.

Partially hydrolyzed polyacrylamide (HPAM) with a molecular weight of 15×10^6 was chosen for class I reservoirs. Its viscosity satisfied the requirements of 20mPa · s at 1500mg/L. The formulation of polymer flooding for class I reservoirs was 0.05PV × 2000mg/L + 0.20PV × 1500mg/L. HPAM was prepared using fresh water and injected using produced water. A pilot polymer flooding pilot was conducted at Gudao's ZYQ Ng3 (a class I reservoir) in 1992. The reservoir parameters were as follows: oil viscosity at the surface of 46.3mPa · s, formation temperature of 70℃, salinity of 5293mg/L, divalent cation content of 102mg/L, and OOIP of 165×10^4 ton. An injection of 1389.5 ton of polymer yielded a maximum water cut reduction of 21.2% (from 92.1% to 70.9%) and an increase in oil production from 122 ton/day to 351 ton/day. The cumulative oil increment was 20×10^4 ton, while the enhanced recovery was 12.0% over waterflooding, and the incremental oil per ton of polymer was 143 ton.

HPAM with a molecular weight of 20×10^6 was suitable for class Ⅱ reservoirs. Its viscosity reached 20mPa · s at 1500mg/L. In contrast, the viscosity of 1500mg/L HPAM with a molecular weight of 15×10^6 was only 9.3mPa · s. A pilot polymer flooding application was conducted at Shengtuo's SYQ (a class Ⅱ reservoir) in 1998. The reservoir parameters were as follows: oil viscosity at the surface of 25mPa · s, formation temperature of 80℃, salinity of 21000mg/L, and divalent cation content of 311mg/L. From April 1998 to September 2001, 7005.7 ton of polymer was injected. After applying polymer flooding, the maximum water cut reduction reached 14.2% (from 95.9% to 81.7%), and oil production increased from 261 to 608 ton/day. The cumulative oil increment was 71.5×10^4 ton. The incremental recovery was 6.6% OOIP over waterflooding, and the incremental oil per ton of polymer was 102 ton. It is estimated that the final enhanced recovery and oil increment per ton of polymer will be 7.0% and 109 ton, respectively.

So far, 29 polymer flooding projects have been implemented in class Ⅰ and Ⅱ reservoirs, with an OOIP of 2.91×10^8 ton. The cumulative oil increment was 1743×10^4 ton, equivalent to 6.0% OOIP of recovery. The final incremental recovery will be 7.1%.

2.2 Surfactant/polymer flooding

Research on combination flooding, mainly alkaline/polymer/surfactant (ASP) and surfactant/polymer (SP) flooding, began in the late 1980s. It has been suggested that ASP and SP techniques could improve oil recovery by 12%～20% OOIP.

Considering the high acid value of Shengli crude oil, ASP flooding was considered first. The first pilot of ASP flooding in China was conducted at Gudong in 1992. The water cut of the central wells had already exceeded 98% for three years before the pilot. After applying the ASP system, the water cut of the central wells declined from 98.5% to 85%, and the cumulative incremental oil production increased to 1740 ton. The enhanced oil recovery was 13.4%, and the oil increment per ton of polymer was 110 ton. By the end of the ASP pilot, 76.2% OOIP was obtained in the central wells [4,5]. After the great success of the pilot test, ASP flooding was extended to western Gudao in 1997. From May 1997 to April 2002, the cumulative incremental oil production and recovery were 29.6×10^4 ton and 15.0%, respectively.

Despite the high recovery factor of ASP flooding, the scale deposit in the injection system was a serious issue. Additionally, the produced fluids contained a large number of emulsions, which were difficult to break. Therefore, the industrial application of ASP flooding was restricted. To avoid the problems caused by ASP flooding, an alkali-free surfactant/polymer system was developed. The first SP pilot was conducted at Gudong's QQX in 2003. The parameters were as follows: oil-bearing area of 0.94km^2, OOIP of 277×10^4 ton, 10 injectors and 16 producers with well spacing of 300m, oil viscosity at the surface of 40.1mPa·s, formation temperature of 68℃, total salinity of the formation water of 7883mg/L, and divalent cation content of 244mg/L. The total water cut was 98.2% before the SP pilot. After the application of SP flooding, the water cut in the central wells dropped to 60.4%, and oil production increased from 10.7 to 127 ton/day. The cumulative oil increment in the central wells was 12.2×10^4 ton, and the enhanced recovery was 18.0% OOIP. The cumulative oil increment over the entire pilot area was 24.7 × 10^4 ton, and the enhanced recovery was 8.9%.

The large-scale industrial application of SP flooding began in 2007. By the end of March 2013, 17 SP projects had been conducted in the Gudao, Gudong, Shengtuo and Chengdong fields. The producing reserves were 1.39×10^8 ton, with a cumulative incremental oil production of 538×10^4 ton. The oil increment of SP flooding in 2012 accounted for 62% of the chemical flooding oil increment in the Shengli oilfield. The recoverable reserves and oil recovery had increased by 1052×10^4 ton and 10.2%, respectively. SP flooding has become an important contribution to oil production in the Shengli oilfield.

2.3 Heterogeneous combination flooding

The OOIP of reservoirs after polymer flooding in the Shengli oilfield was 2.9×10^8 ton.

The average oil recovery was only 46.0%, implying that 54.0% of reserves remained underground. Core tests and dynamic monitoring data have revealed that the heterogeneity becomes more severe after polymer flooding. A novel heterogeneous combination flooding (HCF) system was developed that consists of branched preformed particle gel (B-PPG), polymer and surfactant. Physical simulation experiments showed that the HCF system an improve oil recovery by 13.6% over polymer flooding.

The HCF pilot was conducted at Gudao's ZYQ Ng3 in October 2010 with 15 injectors and 10 producers. It had a line drive well network with well spacing of 135m×150m. The water cut and oil recovery before the pilot were 98.3% and 52.3%, respectively. After applying the HCF system, the water cut in the central wells dropped to 83.5%, and oil production increased from 4.5 to 75.6 ton/day. The enhanced oil recovery was 2.8% and is estimated to be 8.5%. The final oil recovery could reach 63.6% OOIP.

2.4 Foam flooding

Foam has several favorable features, such as good heat/salt resistance and high blocking efficiency. Additionally, foaming agents can reduce the oil-water interfacial tension in order to yield good displacement efficiency. Research on foam flooding (FF) began in 2000. The mechanisms of foam generation, destruction and migration in the porous medium were explored. The interaction between nitrogen and foaming agents was examined, and a nitrogen FF system was developed. Physical simulation experiments showed that polymer-enhanced nitrogen FF can improve oil recovery by 26.0% over water flooding and 16.7% over polymer flooding [6-9]. A FF pilot was implemented at Chengdong's XQ in October 2004 with 4 injectors, 3 central producers and 11 benefit producers. The parameters were as follows: oil viscosity at the surface of 74mPa • s, formation temperature of 60℃, and a total water cut and oil recovery before the FF pilot of 97.7% and 40%, respectively. The main slug had been injected since July 2005. The cumulative injection volume was 0.32 PV. The injection pressure increased from 7.6 to 13.6 MPa. The resistance coefficient of the FF system was 4.2, considerably higher than that of polymer flooding (only 1.6) under similar circumstances. Thus, the percolating resistance and blocking ability improved. According to the injection profile test, the water injection profile improved, and the vertical heterogeneity was adjusted. The water cut in the central wells decreased from 97% to 82.4%. The cumulative incremental oil production was 2.1×10^4 ton, equivalent to 5.4% of oil recovery.

3. Conclusions and perspectives

In reservoirs in which oil recovery is difficult, such as those with high temperature and high salinity, class Ⅲ, Ⅳ and Ⅴ reservoirs, and reservoirs after polymer flooding, it is recommended to integrate chemical flooding material development based on theoretical and experimental research. Polymer flooding and SP flooding have achieved great success but require further improvements in order to be more applicable under severe reservoir conditions.

HCF and foam flooding have potential as new EOR opportunities for high-temperature, high-salinity and heterogeneous reservoir applications.

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Study of the Novel Oil-displacing System of the β-cyclodextrin Inclusion Complex

Yanfeng Ji, Wanli Kang[❶]**, Leilei Hu, Shaolong Wei, Jian Zhao**

[China School of Petroleum Engineering, China University of Petroleum (East China), Qingdao, 266580, P. R. China]

Abstract: A supramolecular recognition system was constructed based on the water-soluble β-cyclodextrinpolymer P(AM/β-CD/NaA) as host and hydrophobically modified polyacrylamide P(AM/C18/NaA) as guest. The influences of salt concentration and temperature on the viscosity of the mixture also were studied. O/W crude oil emulsions stabilized by the inclusion complex solutions were prepared, and a laser particle size analyzer and Turbiscan lab stability analyzer were employed to study their stability. The results indicated that these emulsions became more stable than pure amphiphilic polymer. This likely occurred because the elastic polymer gel structure in the continuous phase has the ability to hold oil droplets.

Key words: cyclodextrin polymer; amphiphilic polymer; inclusion association; O/W crude oil emulsion

1. Introduction

Cyclodextrins (CDs) are cyclic oligosaccharides consisting of six to eight glucose units linked by 1,4-α-glucosidic bonds forming a torus-shaped ring structure[1]. The β-cyclodextrin (β-CD) containing seven glucose units has been used widely in organic chemistry and polymer chemistry[2]. The primary and secondary hydroxyl groups in β-CD form a complex network of intramolecular hydrogen bonds that yield a polar hydrophilic outer shell and a relatively hydrophobic cavity. Thus, β-CD can generate host/guest inclusion complexes through its inclusion with suitable hydrophobic molecules[3].

This paper reports on acrylamide/allyl-β-cyclodextrin/sodium acrylate terpolymer-P(AM/A-β-CD/NaA) as a host polymer and acrylamide/N-*n*-octadecylacrylamide/sodium acrylates terpolymer-P(AM/C_{18}/NaA) as a guest polymer. The inclusion complex solution, when amphiphilic polymer at a low concentration (below the critical aggregation concentration), exhibits good viscosity. The novelty of the present work is that the stability of the network structure under high-mineralization oilfield conditions has been improved to achieve enhanced oil recovery.

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❶ Address correspondence to: Wanli Kang, School of Petroleum Engineering, China University of Petroleum (East China), Qingdao 266580, P. R. China. Fax: +86-532-86981196; E-mail: kangwanli@126.com.

2. Experimental section

2.1 Materials

This study utilized reagent-grade sodium chloride, calcium chloride and sodium hydroxide purchased from the Aladdin Chemical Reagent Factory (Shanghai, China). Acrylamide/allyl-β-cyclodextrin/sodium acrylates terpolymer-P(AM/A-β-CD/NaA) as a host polymer were prepared using a method described in the literature[4]. Acrylamide/N-*n*-octadecylacrylamide/sodium acrylates terpolymer-P(AM/C_{18}/NaA) as a guest polymer were prepared using a method that our group previously reported[5]. The experimental oil was Daqing Oilfield dehydration crude, and the experimental water was Daqing Oilfield simulated formation water.

2.2 Stability experiments

The properties of P(AM/C_{18}/NaA) and the inclusion complex solutions were evaluated through stability experiments of salt resistance. Viscosity measurements were performed with a DV-Ⅱ viscometer (Brookfield, 6 rpm, 45℃).

2.3 Stability of the O/W crude oil emulsion

The O/W crude oil emulsion was prepared by mixing the Fuyu crude oil and mation water containing the inclusion complex (1 : 4, v/v) at 45℃ using a FM-200 homogenizer (Fluko Corp.). The particle size distribution was analyzed by a Rise-2006 laser particle size analyzer (Jinan Rise Science & Technology Co., Ltd.). The stability of the O/W crude oil emulsion stabilized by the inclusion complex solution was monitored by TSI (Turbiscan Lab Expert).

3. Results and discussion

3.1 Effect of salt concentration

The two investigated polymer systems reached their maximum viscosity when the concentration of inorganic salt (NaCl and $CaCl_2$) increased. The viscosity change of the P(AM/C_{18}/NaA) solution was more significant than that of its mixture with P(AM/A-β-CD/NaA). The solution viscosities of P(AM/C_{18}/NaA) peaked when the concentration of NaCl was 5000mg/L, and the maximum viscosity of the inclusion system was approximately 8000mg/L (Fig. 1). However, when $CaCl_2$ was added into the P(AM/C_{18}/NaA) solution and the inclusion system, the viscosity decreased significantly. As Fig. 1 illustrates, the viscosity of the inclusion system had less effect than that of the P(AM/C_{18}/NaA) solution on the concentration of NaCl and $CaCl_2$. Therefore, the inclusion system had a higher salinity tolerance than the P(AM/C_{18}/NaA) solution.

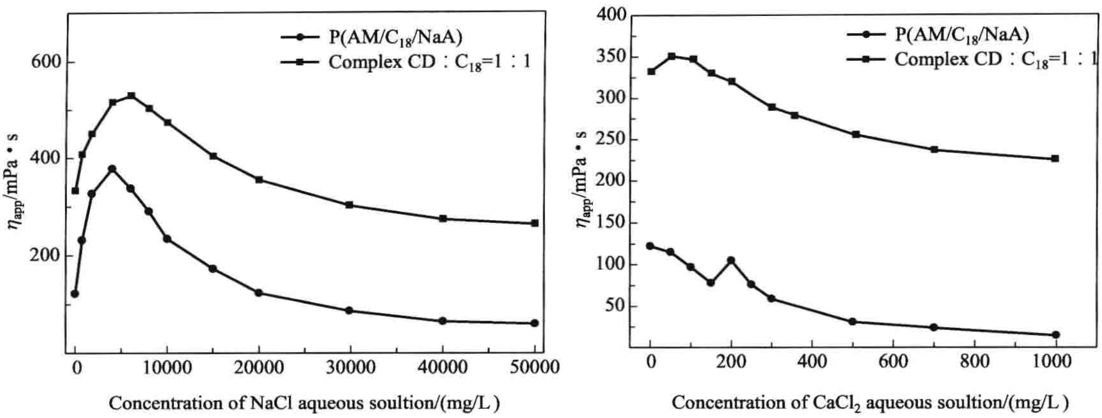

Fig. 1 Effect of NaCl and $CaCl_2$ concentration on the viscosity of polymer and inclusion complex: the concentration of P(AM/BHAM/NaA) is 600mg/L

3.2 Effect of temperature

An inclusion association is a type of non-permanent, reversible bonding effect. Certain temperatures can cause the inclusion to separate. Fig. 2 shows that as the solution temperature increased, the complex began to undergo the process of inclusion separation, which caused the solution viscosity to decline. According to the Arrehenius equations, the flow activation energy of the complex, which was a mixture of P(AM/C_{18}/NaA) and P(AM/A-β-CD/NaA), at a molar ratio of CD and alkyl of 1 : 1, was 75.7kJ/mol. However, the flow activation energy of amphiphilic polymer P(AM/C_{18}/NaA) was 242.7kJ/mol, which was obtained by the viscosity and temperature curves of the amphiphilic polymers. The flow activation energy reflects the amphiphilic polymer solution's temperature sensitivity. The greater the flow activation energy, the stronger the temperature dependence of the polymer. The results of activation energy of the P(AM/C_{18}/NaA) solution is significantly higher than the host/guest inclusion complex. The activation energy differed mainly because the host and guest polymers had different effects on the inclusion association. The inclusion system exhib-

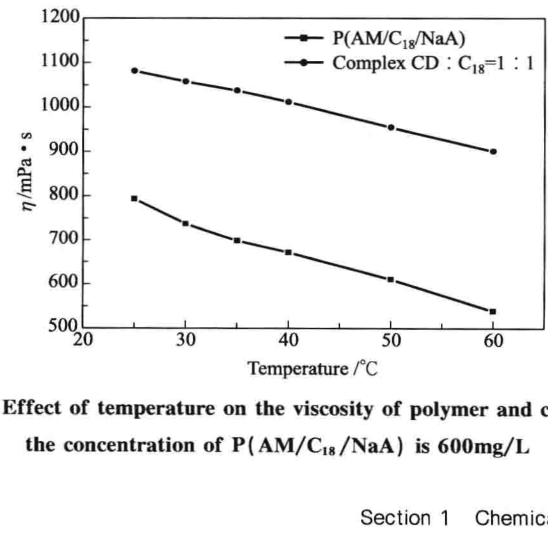

Fig. 2 Effect of temperature on the viscosity of polymer and complex: the concentration of P(AM/C_{18}/NaA) is 600mg/L

ited good temperature resistance.

3.3 Stability of O/W crude oil emulsions

Fig. 3 shows the influence of P(AM-C_{18}-NaA) and its inclusion system on the particle size distribution of the O/W crude oil emulsions. The oil droplets became more uniformly distributed and their average size decreased when P(AM/C_{18}/NaA) was included wtih P(AM/A-β-CD/NaA). The results show that the O/W crude oil emulsions stabilized by the inclusion system became more stable than the pure amphiphilic polymer at a low amphiphilic polymer concentration.

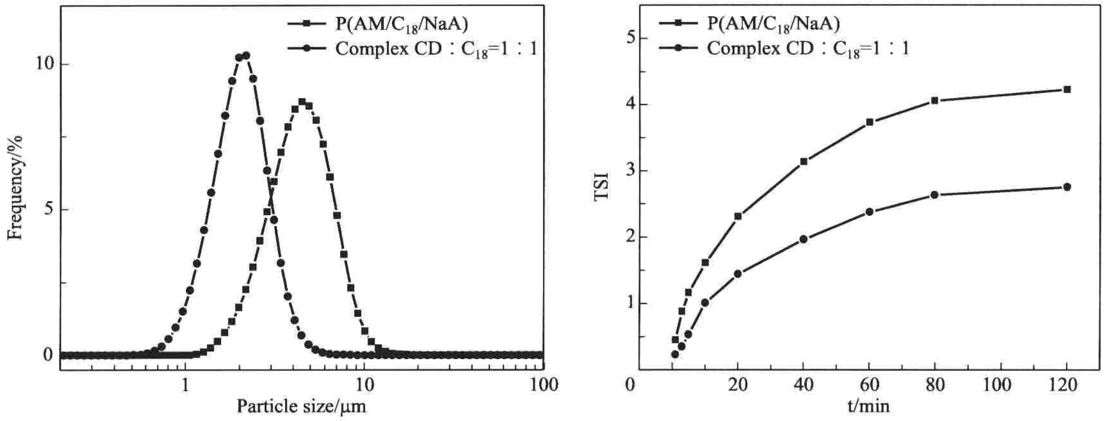

Fig. 3 Particle size distribution and TSI values of O/W crude oil emulsions: the concentration of amphiphilic polymer is 600mg/L

The global stability of all of the emulsion samples stabilized by P(AM/C_{18}/NaA) and of the inclusion system is compared in Fig. 3. The TSI values of O/W crude oil emulsions stabilized by P(AM/C_{18}/NaA) and its inclusion system indicate that the stability improved when mixed with P(AM/A-β-CD/NaA) to form an inclusion system.

4. Conclusions

The stability experiments of P(AM/C_{18}/NaA) and its inclusion complex show that the P(AM/C_{18}/NaA) mixed with P(AM/A-β-CD/NaA) could yield higher salt and temperature tolerance. The O/W crude oil emulsion of the P(AM/C_{18}/NaA) inclusion complexes (β-CD : C_{18} = 1 : 1) had the best stability. The O/W crude oil emulsion of the inclusion complex was more stable than the pure amphiphilic polymer emulsion, probably because its elastic polymer gel structure in the continuous phase can hold oil droplets.

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