# Study and application of oily sludge harmless gel particles system for enhanced oil recovery

Liu Huaizhu<sup>1,2,a,\*</sup>, Ni Yin<sup>1,2,b</sup>, Zhao Kangning<sup>1,2,c</sup>, Hu Binbin<sup>1,2,d</sup>, Ma Ying<sup>1,2,e</sup>

<sup>1</sup>Tangshan Jiyou Ruifeng Chemical Limited Company, Jidong Oilfield, CNPC, Tangshan, China <sup>2</sup>Hebie Oilfield Chemical Agent Technology Innovation Center, Tangshan, China <sup>a</sup>liuhuaizhu007@126.com, <sup>b</sup>rfny@petrochina.com.cn, <sup>c</sup>zhkn@petrochina.com.cn, <sup>d</sup>rfhubb@ petrochina.com.cn, <sup>e</sup>rfmy@petrochina.com.cn \*Corresponding author

**Abstract:** In order to solve the problems of difficult injection and easy secondary recovery of oil-bearing sludge for profile control, the micro-morphology and composition characteristics of oily sludge in Jidong Oil Field were determined and analyzed by SEM. The influence of different dispersants on the dispersion characteristics of oil sludge was analyzed. The optimum dosage of dispersing agent  $Na_2CO_3$  is 0.8%-1.0%. The effects of 4 kinds of suspension agents on the suspension performance of oily sludge were analyzed by rheological experiments. The suitable suspending agent and its dosage were determined, and on this basis, the oil sludge gel particles profile control system was made by adding gel-forming and initiator. The salinity resistance and viscoelastic properties of oil sludge gel profile control agent was measured, and the effect of profile control was evaluated by physical simulation experiment. The results showed that the oily sludge gel particle profile control system has good resistance to different salinity, better viscoelastic and good plugging properties. The field test shows that the total oil increment of sludge profile control is 2568t.

**Keywords:** oily sludge; gel particles; dispersant; suspension agent; rheological properties; profile control results

#### 1. Introduction

Oily sludge mainly comes from sewage discharge of oil field combined station system, underground operation, pipeline leakage and tank cleaning sludge, etc. Each year, about 6800m<sup>3</sup> oily sludge is produced in Gaoshangpu oil production plant of Jidong oil field, and the historical residual amount is about  $4 \times 10^4$  m<sup>3</sup>. These oily sludge are usually transported by car to environmental protection units for treatment. It is a long way and there is secondary pollution. In recent years, many oilfields have carried out research on the technology of sludge profile control, through chemical treatment of sludge to prepare profile control agent used in water injection wells, can achieve the treatment of oil-bearing sludge and improve the effect of water injection development, it is an economical and effective sludge treatment method <sup>[1-12]</sup>. However, due to the uneven particle size of oily sludge, poor suspension, low plugging strength and other factors lead to poor field application effect <sup>[13-15]</sup>. A suitable polymer flooding re-injection system was developed, but the utilization ratio of oily sludge is not high. A new hydrophobic-ally associating polymer as thickener and organic phenol resin as cross-linking agent was used to form a sludge gel system with good suspension and strong plugging ability<sup>[16-18]</sup>. However, for high capacity channels developing regions, a single gel system can not meet the requirements, requiring a high-strength curing system to achieve more effective plugging<sup>[19-20]</sup>. Because of the complex composition of sludge and the different reservoir conditions in different oil fields, the performance requirements of sludge profile control agent are different, so there is a problem of adaptability between sludge profile control agent and formation.

In view of the development of high capacity channels developing reservoirs in Jidong oilfield, the development degree and distribution range of high capacity channels in each layer are quite different, and different strength profile control agents are needed to realize effective plugging, at the same time, in order to treat sludge in situ as much as possible, the composition and particle size of oily sludge were analyzed firstly, and the adaptability of reservoir temperature, pressure, fluid property and oily sludge plugging agent was determined.

The system is based on oily sludge as the basic raw material, adding appropriate amount of

suspending agent, dispersing agent and other agent, the oily sludge is mixed into the gel system with certain suspension and stability, which is injected into the formation at a certain depth to produce "Agglomerate structure", and to seal the deep part of the large pore, thus enlarging the swept volume of water drive, enhanced oil recovery, with temperature and salt resistance, low cost, simple construction technology, plugging effect is good; The suspension, salt resistance, shear resistance, long-term stability, plugging performance of the system were evaluated, the field scale application and evaluation are carried out in order to meet the production needs of profile control and water shutoff in high capacity channels developing reservoirs and realize the harmless treatment of sludge.

#### 2. Experiment

#### 2.1. Material and apparatus

Material: carboxymethyl cellulose CMC, non-ionic Polyacrylamide NPAM, SNF, modified Polyacrylamide potassium salts KYPAM, Na<sub>2</sub>CO<sub>3</sub>, NaOH, cetyltrimethyammonium bromide CTAB, petroleum sulfonate, oily sludge from Gaoshangpu oil field sludge tank of Jidong oil production plant.

Apparatus: Quanta 200F field emission scanning electron microscope,DV-III rotary viscometer, high-speed stirrer, Haaker RS600 rheometer, oven, electronic balance (0.001g), physical simulation of oil displacement experimental device.

#### 2.2. Experimental method

(1) Preparation of the test samples, the surface morphology of the oily sludge samples was observed by the Quania 200F field emission scanning electron microscope and the component content was analyzed.

(2)The suitable dosage of dispersant was determined by the sedimentation time, the thickness of sedimentation layer and the variation of pH value.

(3) Through the law of apparent viscosity change and sedimentation phenomenon, the suitable suspensions were selected from the carboxymethyl cellulose CMC, non-ionic Polyacrylamide NPAM, SNF and KYPAM suspensions, and their dosages were determined.

(4) Adding proper amount of gelling agent and initiator, taking oily sludge as main agent to make profile control agent with good injectivity. The physical simulation experiment of profile control was carried out to evaluate the effect of profile control.

# 3. Results and discussion

#### 3.1. The composition and particle size of oily sludge

Oily sludge samples were black paste. The solid content, water content and oil content of oily sludge were 69.09%, 24.61% and 6.3%, respectively. The analysis of the mineral composition of the solid particles in the oily sludge shows that the solid particles mainly consist of 75% cuttings, 10.3% quartz, 14.9% feldspar, and a small amount of impurities and cements. The particle size distribution of solid particles is shown in Table 1. It can be seen that the proportion of solid particles between 94  $\sim$ 106µm is about 34%, and that of particles smaller than 150µm is about 80%. The particle size distribution of solid particles is relatively concentrated, which is suitable for preparing profile control agent.

Particle size(µm)	Percent(%)	Particle size(µm)	Percent(%)
<75	15.36	(150-180]	3.49
[75-83]	6.66	(180-250]	5.55
(83-94]	11.01	(250-425]	2.66
(94-106]	33.96	(425-850]	7.73
(106-125]	6.81	>850	1.64
(125-150]	5.27		

Table 1: Distribution of solid particle size of oily sludge

#### 3.2. Selection of dispersants

There are three common types of sludge dispersants: surfactant, alkali and inorganic salts. The surfactant with both hydrophilic and lipophilic groups can reduce the oil-water interface tension and disperse the oily sludge suspension better, but the presence of metal ions and impurities in the oily sludge reduces the activity of the surfactant, NaOH and KOH are strong alkalinity, which make profile control agents in strong alkalinity environment, poor reservoir compatibility, high requirements for construction equipment and pipelines, and poor surfactant safety. Inorganic salt Na<sub>2</sub>CO<sub>3</sub> was selected as dispersant of oily sludge. The pH value, sedimentation time and sedimentation layer thickness of the suspension system with 4.0% sludge effective content were determined by adding different amount of Na<sub>2</sub>CO<sub>3</sub>. The results are shown in Figure 1.As can be seen from Fig. 1, when the Na<sub>2</sub>CO<sub>3</sub> dosage is less than 0.8%, the pH value of suspension system increases with the increase of Na<sub>2</sub>CO<sub>3</sub> dosage, and it belongs to weakly alkaline system when the concentration of Na<sub>2</sub>CO<sub>3</sub> is 0.8%-1.0%, the pH value of the system is no longer increased, and the time of delamination and particle sedimentation is prolonged, when the content of  $Na_2CO_3$  exceeds 1.0%, the time of delamination and the sedimentation time of particles decrease, and the dispersion becomes worse. Therefore, the appropriate content of Na<sub>2</sub>CO<sub>3</sub> should be controlled in the range of 0.8%-1.0%. The addition of Na<sub>2</sub>CO<sub>3</sub> changed the surface electric property of the sludge particles, enhanced the ion exchange, made the surface of the sludge particles positively charged, formed a polar adsorption layer, increased the steric hindrance between the particles, and made the sludge particles more dispersed and stable.

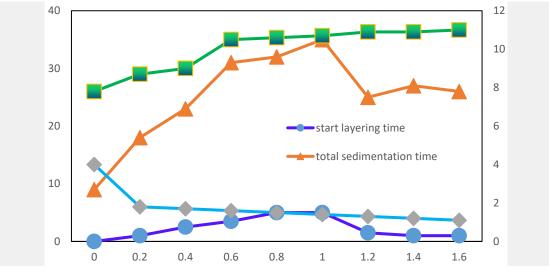


Figure 1: Dispersion performance of oily sludge

#### 3.3. Optimization of suspension agent

The preferred macro-molecule with viscosity increasing properties is the commonly used sludge suspending agent. Due to the poor biological adaptability of natural macro-molecule such as xanthan gum, sludge particles with high metal ions and impurities cannot be suspended stably, therefore, the appropriate suspension agent was selected from the four synthetic macro-molecule and the appropriate dosage was determined. Adding macro-molecule into the suspension could increase the apparent viscosity of the system. The steric resistance of the polymer chain reduced the settling velocity of the sludge particles, which was beneficial to the suspension of the sludge particles. The rheological properties and apparent viscosity with different mass fraction of suspending agent are shown in Table 1 when the effective content of oily sludge is 4%.

From Table 2, it can be seen that NPAM has the least influence on the rheological properties of the sludge system, and the apparent viscosity of the sludge system does not change with the increase of HPAM. SNF and KYPAM have a greater influence on the rheological properties of the sludge system, and with the increase of the addition amount, the apparent viscosity and dynamic shear force of the system increased obviously, but the flow index and consistency coefficient did not change obviously, which increased the injection difficulty of field construction, the rheological parameters are in a lower range and easy to control. The flocculation of KYPAM and SNF is obvious, although the system viscosity is big, but because of the adsorption bridging effect of KYPAM and SNF on clay particles, the

sludge particles quickly settle down with the form of aggregated flocculation, therefore, CMC was selected as the suspending agent of sludge system. Considering the injection capacity and application cost, the appropriate amount of CMC should be controlled at about 0.3%.

suspending agents	Concentration (%)	AV(mPa·s)	PV(mPa⋅s)	YP(Pa)	Rheological index(n)	K (mPa·sn)
СМС	0.2	3.3	3	0.307	0.87	0.01
CMC	0.3	5	4.5	0.511	0.86	0.01
CMC	0.4	7	6	1.022	0.81	0.03
CMC	0.5	9.5	9	0.511	0.93	0.02
CMC	0.6	14	12	2.044	0.81	0.05
NPAM	0.2	1.75	1.5	0.256	0.81	0.01
NPAM	0.3	1.75	1.5	0.256	0.81	0.01
NPAM	0.4	2	1.8	0.204	0.86	0.01
NPAM	0.5	2	1.8	0.204	0.86	0.01
NPAM	0.6	2	1.4	0.613	0.62	0.03
SNF	0.2	6.6	3.6	3.066	0.46	0.28
SNF	0.3	9	6	3.066	0.58	0.16
SNF	0.4	14	9	5.11	0.56	0.3
SNF	0.5	18.5	13.4	5.212	0.65	0.21
SNF	0.6	22	13	9.198	0.5	0.68
KYPAM	0.2	7.3	4.6	2.759	0.55	0.17
KYPAM	0.3	11	7	4.088	0.55	0.24
KYPAM	0.4	15.35	10.3	5.624	0.56	0.32
KYPAM	0.5	21	13.2	8.174	0.53	0.53
KYPAM	0.6	28.5	16.1	12.776	0.49	1.08

Table 2: Rheological properties of oily sludge system with different suspending agents

#### 3.4. Preparation of profile control system

According to the formula of 4.0% oily sludge (effective content) + 1.0% Na<sub>2</sub>CO<sub>3</sub>+0.3% CMC +3.0% gelling agent +0.1% initiator, a new oily sludge profile control system was made.

#### 4. Performance evaluation of oily sludge gel particle

#### 4.1. Expansion performance of oily sludge gel particle

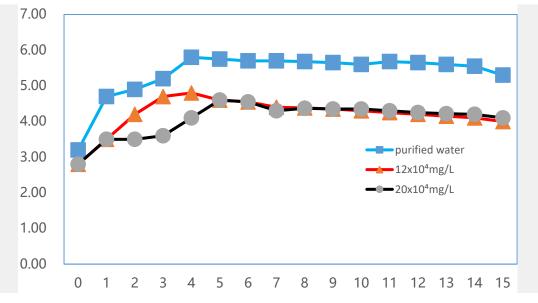
At 65°C, the expansion ratio of gel particles in purified water, injected water  $(12 \times 10^4 \text{ mg/L})$  and produced water  $(20 \times 10^4 \text{mg/L})$  is shown in Figure 2. With the increase of expansion time, the expansion times of gel particles in purified water and salt water first increased and then decreased. When the expansion time is the same, the expansion performance of gel particles in salt water becomes worse, and the expansion ratio in salt water is smaller than that in pure water. The expansion ratio of gel particles in salt water with different salinity has little difference.

#### 4.2. Viscoelasticity performance of oily sludge gel particle

At 65°C, the elastic modulus and yield stress of gel particles filled with oily sludge and normal gel

particles after 72 h immersion in salt water are 3800 Pa and 2800 Pa respectively. The elasticity and strength of oily sludge gel particles were higher than those of normal gel particles, and showed better viscoelastic properties.

# 4.3. Plugging performance of oily sludge gel particle



4.3.1. Single sand-filled pipe model

Figure 2: Effect of salinity on expansion properties of oily sludge gel particles

0.2 PV oily sludge gel particle profile control system was injected into the sand-filled pipe with different permeability which was set at 65°C for 24 hours, the breakthrough pressure, residual resistance coefficient and plugging rate were measured by water flooding, the results are shown in table 3.

No	porosity (%)	Permeabi Before injection	lity(µm2) After injection	breakthrough pressure gradient(MPa /m)	residual resistance coefficient	plugging rate(%)
1	41	0.95	0.007	8.61	136	99.2
2	39	0.21	0.006	8.46	35	97.1
3	35	0.01	0.0009	8.32	11	91

Table 3: Plugging performance under the condition of single sand-filled pipe model

From Table 3, it can be concluded that the profile control system has good plugging ability for high permeability, medium permeability and low permeability sand-filled pipes, the plugging rate is more than 90%, and the breakthrough pressure gradient after plugging is about 8.5 MPa/m.

# 4.3.2. Parallel sand-filled pipe model

0.2 PV plugging agent was injected into the parallel sand-filled pipe with different permeability gradients, and the water flooding was carried out at 65°C for 48 hours, the results are shown in table 4.

Table 4: Plugging performance under the condition of parallel sand-filled pipe model

No	Permeability (µm <sup>2</sup> )	permeability gradient	-	Water injection percentbefore injection (%)	Water injection percent after injection (%)	breakthrough pressure (MPa)	Water injection percent after breakthrough (%)
G1	3.82		41	97.6	98.7	5.8	10.7
D1	0.051	74.9	37	2.4	1.3	2.9	89.3

Academic Journal of Environment & Earth Science

# ISSN 2616-5872 Vol.6, Issue 2: 1-7, DOI: 10.25236/AJEE.2024.060201

The experimental results show that the injection of profile control system has a certain selectivity, with the increase of permeability gradient, the injection selectivity is stronger, and the plugging dose into the high permeability zone is larger, the plugging effect is also better and the damage to low permeability reservoirs can be reduced. After plugging agent breakthrough, the injection percent of low permeability reservoirs with parallel sand-filled pipes with permeability gradient of 74.9 increased from 1.3% to 89.3%, and the injection percent of low permeability reservoirs increased greatly, the injection percent of the high permeability reservoir with parallel sand-filled pipe with permeability gradient of 74.9 is reduced from 97.6% to 10.7%, and the plugging effect is very good.

#### 5. Field application

The technology has been applied in Jidong oilfield for 6 wells. The field test shows that this plugging agent system can effectively carry oily sludge, the maximum amount can reach 50% during the injection process, the injection pressure is stable, and shows good field applicability, which meets the design requirements, it can be seen from table 5 that the injection pressure of the test wells all increased over 2MPa during the test process, and the connected oil wells showed obvious effect of oil increment and water reduction, the average daily oil increment of oil wells is 1.6 t, and the average water cut is decreased by 8.8%. Compared with the conventional profile control, the average oil increment was 321t, and the effective rate was 92.3%. The total oil increment of sludge profile control is 2568t, which plays an obvious role in oil increment and water reduction.

Well number	Injection of oily sludge(m3/d)	Pressure before injection(MPa)	Pressure after injection(MPa)
G1	100	11.3	13.4
G2	120	12.5	14.2
G3	100	10.2	11.9
G4	120	11.6	13.8
G5	100	10.7	12.8
G6	150	9.6	11.8

Table 5: Effect of the pressure of injection wells on the plugging control test in oily sludge

#### 6. Conclusions

(1)The formula oily sludge gel particle profile control system was 4.0% oily sludge (effective content) + 1.0% Na2CO3+0.3% CMC +3.0% gelling agent +0.1% initiator.

(2)The new oily sludge gel particle profile control system has good resistance to different salinity, better viscoelastic and good plugging properties.

(3)The field test shows that the total oil increment of sludge profile control is 2568t, which plays an obvious role in oil increment and water reduction. At the same time, the technology can clean up all the existing oily sludge, solve the problem of safety and environmental protection, increase the output of crude oil, and obtain significant economic and social benefits.

#### References

[1] Lei G, Liu Z. Research progress on resource utilization and harmless treatment of oily sludge treatment technology [J]. Academic Journal of Environment & Earth Science, 2023, 5 (7).

[2] Shuai Z, Mei H, Aiyuan J. Feasibility study of porous media for treating oily sludge with self-sustaining treatment for active remediation technology.[J].Environmental science and pollution research international, 2023, 30(27):70131-70142.

[3] Huaizhu L, Dong C, Kangning Z, et al. The mechanisms of inhibition and lubrication of clean fracturing flowback fluids in water-based drilling fluids[J]. Green Processing and Synthesis 2023; 12: 20230062

[4] E.EB, S.EO, M.YG, et al. Distribution and Composition of High-Molecular-Mass Components in

Oily Sludge [J]. Petroleum Chemistry, 2022, 62(2):151-160.

[5] Han X, Linhui Q, Xiaonan W, et al.Oil Sludge Treatment by a Microemulsion System Containing Sodium Dodecyl Benzene Sulfonate[J].Chemistry and Technology of Fuels and Oils, 2022, 57(prepublish): 1000-1004.

[6] Yael S H, Berrin T, Shonali L. Materials and energy recovery from oily sludges removed from crude oil storage tanks (tank bottoms): A review of technologies[J]. Journal of Environmental Management, 2022, 305:114428-114428.

[7] Huibo Q, Xiaofei W, Yi-Xuan Z, et al. Insight into water-enhanced CO2 extraction in the treatment of oily sludge [J]. Journal of CO2 Utilization, 2022, 57.

[8] Cheng W, Liguo Z, Wei Z, et al.Study on Characteristics of Dispersed Oily Sludge and Its Plugging Performance[J].Petroleum Chemistry, 2022, 62 (10): 1171-1182.

[9] Biming L, Yue T, Wenbin S, et al. Novel conditioner for efficient dewaterability and modification of oily sludge with high water content[J].Environmental Science and Pollution Research, 2021, 29(17): 25417-25427.

[10] Wei L, Zhiyang L, Xiao H, et al. Synergetic effect of asphaltenes extracted from polymer containing oil sludge and HPAM at water/toluene interface[J]. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 2021, 630.

[11] Yun M, Mingzhu Y, Lu L, et al. Mechanism and Characteristics of Oil Recovery from Oily Sludge by Sodium Lignosulfonate Treatment [J]. ACS omega, 2021, 6 (39): 25819-25827.

[12] Qinghua B, Lixin H, Jianlong X, et al. Study on the treatment of oily sludge in oil fields with lipopeptide/sophorolipid complex bio-surfactant[J]. Ecotoxicology and Environmental Safety, 2021, 212 :111964-111964.

[13] Huaizhu L, Mingbang T, Baocai X, et al. Study on air foam flooding technology to enhance oil recovery of complex fault block reservoir by water injection[J] .IOP Conference Series: Earth and Environmental Science, 781 (2021) 022032.

[14] Qi Z, Qing J, Yu B, et al. Optimization and mechanism of oily sludge treatment by a novel combined surfactants with activated-persulfate method [J]. Science of the Total Environment, 2021, 800:149525-149525.

[15] Song Y, Yang S. Application Status and Prospect of Combined Treatment Technology for Oily Sludge in Domestic Oil Fields [J].IOP Conference Series: Earth and Environmental Science, 2020, 558 (2):022078.

[16] Qing T, Dongmei Z, Chunping Y. A review of the application of different treatment processes for oily sludge [J]. Environmental Science and Pollution Research, 2020, 28(1): 121-132.

[17] Ramirez D, Shaw J L, Collins D C.Oil sludge washing with surfactants and co-solvents: oil recovery from different types of oil sludges[J]. Environmental Science and Pollution Research, 2020, 28(5):1-13.

[18] Ren H, Zhou S, Wang B, et al. Treatment mechanism of sludge containing highly viscous heavy oil using biosurfactant[J]. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 2020, 585:124117-124117.

[19] Ali M A, Hassan A A M, Ibrahim R R, et al. Analysis of Solid residue and Flue Gas from Thermal Plasma Treatment of Petroleum Sludge[J].Journal of Environmental Chemical Engineering, 2019, 7(4): 103207-103207.

[20] Liu X, Yao T, Lai R, et al. Recovery of crude oil from oily sludge in an oilfield by sophorolipid [J].Petroleum Science and Technology, 2019, 37(13): 1582-1588.