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A modular variable-process treatment system for operation liquid waste: A case study



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ABSTRACT

The components of petroleum wastewater are complex and variable, which makes it hard to meet the reinjection requirements based on a fixed treatment process. Therefore, an efficient wastewater treatment system adapting to variable water quality is necessary. In this study, the types and composition of wastewater from the Changqing oilfield were systematically analyzed. The characteristics of different types of wastewater were clustered by a water quality matrix. Based on the results, viscosity is an important limiting factor in wastewater treatment. Six types of wastewater could be divided into high contaminative wastewater (HCW), medium contaminative wastewater (MCW) and low contaminative wastewater (LCW). Thus, a modular and variable-process wastewater treatment system was developed. The system consists of five modular units including oil removal, viscosity reduction, DAF/DOF, two-stage filtration, and ultraviolet disinfection. Each module can be flexibly used for the wastewater treatment according to the variable water quality and characteristics of raw water. The system was stable and reliable during 160 days of field operation. The suspended solids (SS), petroleum concentration, and median particle size of treated wastewater were less than 2.0 mg/L, 6.0 mg/L and 1.5 µm, respectively, which meet the requirements of the reinjection standard. The treatment system can improve the wastewater treatment efficiency with variable water quality in the oilfields and protect regional environmental safety.

1. Introduction

At present, acidification, fracturing and well washing are the main methods that are employed to increase oil production [1]. They become more important for the development of unconventional oilfields characterised by low pressure, low permeability, and low abundance [2]. During processing, a large amount of operation liquid waste (OLW) is produced [3]. In general, OLW has a high pollutant concentration, complex composition, and strong stability. It will cause significant environmental problems if discharged without treating [4,5]. It was widely reported that contamination of surface water and shallow groundwater following spills of hydraulic fracturing fluids in certain regions [6,7]. It was realized that OLW should be disposed to meet the standards, otherwise it will cause harm to the environment.

Several operations such as well washing, acidification, and fracturing are used alone or combined to increase oil production depending on geological conditions and the characteristics of the well. Both of well washing and acidification process are common operations for production increasing, and they also can be employed to remove blockages of the pipes. For fracturing, the pipeline should be cleaned by well washing or acidification to ensure smooth. The kinds of fracturing fluids mainly depend on the selected operation, which is also closely related to geological conditions. Therefore, due to the different operation stages of the wellsite, the quality of the produced fracturing wastewater is greatly diverse. Consequently, the OLW is discrete because the operation process is not continuous.

Physical, chemical, physicochemical, and biological methods can be used to deal with the OLW [8,9]. Yang et al. [10] used a combination of flocculation, Fenton oxidation, and sequencing batch reactor (SBR) processes to treat OLW. Rosenblum et al. [11] reported that coagulation–adsorption have a good removal efficiency for organic compounds and turbidity of OLW. Lei et al. [12] treated potential toxic pollutants in OLW with multifunctional iron–biocarbon composite materials. Li et al. [13] used Chlorella to treat OLW. Andrew et al. [14] reported that it can obtain high economic benefits by treating hydraulic fracturing fluids together with acid mine drainage [14]. Biofilm technique is an attractive approach to facilitate the degradation of diverse compounds in OLW, with strong tolerance of high salt and metal concentrations [15]. In summary, the conventional methods are always combined with several new technologies in OLW treatment.

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Fig. 1. Generic framework of the fixed stream treatment system of OLW.

Note: Waste liquid of washing well(WLWW), Acidizing waste fluid (AWF), Guar waste liquid(GWL), EM waste liquid(EMWL), Thickened waste liquid(TWL), Biopolymer Fracturing Fluid(BFF).

Most studies treat OLW with the fixed processing, however, the types of OLW are various, such as acidising waste fluid (AWF), waste liquid from the washing well (WLWW), fracturing wastewater (guar waste liquid (GWL), EM waste liquid (EMWL), thickened waste liquid (TWL) and biopolymer fracturing fluid (BFF)). Fixed treatment system has strong pertinence and low cost. In general, a certain type of specific OLW is typically treated by fixation system and the effluent parameters as SS, petroleum, corrosion rate, median particle size, can meet the reinjection underground limitation (SY/T 5329-2012) [16]. However, due to the complex characteristics of OLW wastewater, it always needs much coagulant, or longer hydraulic retention time in treatment process, as shown in Fig. 1. In order to treat different types of OLW and meet the reinjection standard, HRT has to be prolonged and the dosage of reagents needs to be increased. These measures increase the cost of treatment. Moreover, it is difficult to distinguish the types of OLW in the actual treatment, due to the great differences of water quality and treatability. Therefore, it is necessary to treat the OLW separately in each process.

Changqing oilfield covers an area of $370,000 \text{ km}^2$ which is distributed in Ordos Basin, China. It is a typical unconventional oilfield with low pressure, low permeability, and low abundance. According to statistics, more than $350,000 \text{ m}^3$ of OLW are produced in Changqing oilfield every year. The OLW from the Changqing oilfield was classified in this manuscript based on its quality. A water quality matrix was used to study the characteristics of different OLW. Based on the concept of source separation, a modular and variable-process in OLW treatment system was constructed. The treatment system can improve the efficiency of wastewater with variable water quality in the oilfields.

2. Materials and methods

2.1. Sample collection and water quality analysis

Based on the division of Changqing Oilfield, several typical wellsites were selected for sampling, and 283 wastewater samples were collected for Water matrix analysis, including 44 AWF, 70 GW L, 44 EMWL, 47 TW L, 37 BFF, and 41 WLWW samples.

The suspended solids (SS), petroleum contents, corrosion rate, median particle size, contents of total growth bacteria (TGB), sulphatereducing bacteria (SRB), and iron bacteria (IB) in wastewater samples were determined with reference to the "Recommended water quality standard and methods for the analysis of oilfield injecting waters in clastic reservoirs" (SY/T 5329-2012) [16]. The pH, chemical oxygen demand (COD) and viscosity were determined according to the recommended standard methods [17], the colour of the OLW was examined by the UV spectrophotometer.

In general, water quality indicators of OLW from different processes can be divided into four categories, including physic-chemical indicators, organic pollutants indicators, microbial indicators and other indicators. For example, the physic-chemical indicators are pH, SS and colour; the organic pollutants indicators are COD and petroleum pollutants; the microbial indicators are TGB, SRB and IB; and the other indicators are TDS, median particle size, viscosity and corrosion.

2.2. Water matrix analysis

We have adopted this suggestion. Specific revision can be seen in 2.2. Water quality matrices have been recognized for the evaluation of treatment processes for various wastewater types. Based on the theory, the water quality matrix is established. "Water quality standard and practice for analysis of oilfield injecting waters in clastic reservoirs" (SY/T 5329-2012) is taken as the constraint condition. In this study, SS, Oil content, Viscosity, SRB, Corrosion rate, Median particle size were used to evaluate the water quality of various wastewater [18,19].

According to the water quality index, the excess times of pollutants were calculated. Comparing the excess multiple with the standard value, the judgment matrix was constructed, and the calculations as well as distribution of pollutant weight were examined.





Fig. 3. a) COD and b) petroleum pollutant concentration distribution range of OLW for different processing steps in oilfield.

3. Results and discussion

3.1. Water quality of OLW

3.1.1. Analysis of the physic-chemical indicators

As shown in Fig. 2a, the pH of GWL, EMWL, TWL, BFF and WLWW are neutral (7.1 ~ 7.4) and have small fluctuations. In contrast, the pH of AWF is in a relatively low range (4.3 ~ 5.2), which is mainly due to the utilization of large amount of acid during the acidification process. Fig. 2b shows the analysis results of SS content in different wastewater samples. It can be seen that the SS distribution range of GWL is relatively wide (180 ~ 2700 mg/L), with an average value of 1340 mg/L. While, the SS values of the other five wastewaters have relatively narrow fluctuations (80 ~ 1230 mg/L) and small mean values (260 ~ 580 mg/L). As shown in Fig. 2c, the colour values of AWF, GWL, EMWL, TWL and BFF are very close, and they are distributed in the range of 190 ~ 240 times, which are much higher than that of WLWW (<40 times).

3.1.2. Analysis of the organic pollutants

Figs. 3a and b shows the concentration distribution of COD and petroleum pollutants of OLW for different processing steps in oilfield. It can be seen that the COD concentrations of AWF, EMWL, TWL and BFF are similar, and their average values are all around 4000 mg/L. In contrast, the COD value distribution of GWL is relatively wide, and its average value is about 6500 mg/L, which is relatively higher than other wastewaters. This is mainly because the fracturing fluid used in the fracturing process contains a large amount of organic additives, such as guar gum and cellulose, etc. While, the COD value of WLWW is at a lower level than other wastewaters, and its average value is about 235 mg/L. As shown in Fig. 3b, the distribution of petroleum pollutants in these wastewaters varies widely. Among them, GWL and BFF have the highest content of petroleum pollutants, with average values of 58 and 48 mg/L, respectively. The content of petroleum pollutants in EMWL and TWL is at an intermediate level, and their average values are 30 and 25 mg/L, respectively. In contrast, the content of petroleum pollutants in AWF and WLWW is relatively low, and their average values are both around 8 mg/L.

3.1.3. Analysis of the microbial indicators

TGB, SRB and IB are the three most common microorganisms in oilfield wastewater that pose a potential threat to the operating system. As shown in Figs. 4a-c, the presence of these three microorganisms was detected in all wastewater samples. Moreover, there are significant differences in the content of TGB in different wastewater samples. It can be seen that the TGB content in AWF and WLWW is relatively low, and their average value are 850 m L⁻¹. The TGB content in EMWL, TWL and BFF is relatively high, and their average values are around 2500 m L⁻¹. In contrast, the TGB content in GWL is the highest, with an average value of 10,000 m L⁻¹. As shown in Fig. 4b, except for the relatively

low SRB content of the WLWW, the SRB content of the other wastewaters is relatively high, and their average values are about 1700 m L^{-1} . As shown in Fig. 4c, the content of IB in AWF, GWL and TWL is relatively close, and their average values are about 1600 m L^{-1} . In contrast, IB content in EMWL, BFF and WLWW is relatively low, with an average value of about 800 m L^{-1} .

3.1.4. Analysis of the other indicators

In addition to the above indicators, we also further analysed other water quality indicators that affect the effectiveness of oilfield wastewater treatment. Fig. 5a shows that the viscosity of GWL, TWL, and EMWL is higher than other kinds of wastewater, ranging from 2.2–21.6 mPa·s. The viscosity of AWF, BFF, and WLWW is lower, ranging from 1.4–8.8 mPa·s. As shown in Figs. 5b-d, TDS, median particle size and corrosion rate, etc. the distribution of these water quality indicators in all wastewater samples is also significantly different.

Based on the above analyses, we can demonstrate that the water quality indicators in Changqing Oilfield's operating wastewater have a wide range of distribution, and there are significant differences in pollutant indicators between different types of wastewaters. For the efficient treatment of oilfield wastewater, it is often necessary to consider tasks such as pH adjustment, turbidity removal, viscosity reduction, sterilization and corrosion rate control. However, in the case of large differences in water quality, the existing fixing process is not targeted, and problems such as inadequate or excessive treatment of wastewater often occur, resulting in substandard water quality or high operating costs. Therefore, it is urgent to develop a more flexible treatment process to achieve efficient and low-cost treatment of oilfield wastewater.

3.2. Treatability of OLW

The treatment characteristics of different wastewater were evaluated in this study to lay a foundation for constructing a flexible treatment process. Based on the water quality test results of OLW, and combined with "SY/T 5329-2012" water quality standard, the excess of multiple pollutants was calculated, as shown in Table 1.

Then, the relative importance of any two of the indicators is determined according to the size of the multiple of the super standard, and a judgment matrix is constructed, and the judgment matrix is assigned in combination with the scale assignment rule to obtain the evaluation matrix assignment diagram of oilfield operation wastewater treatment characteristics as shown in Fig. 6.

As shown in Fig. 6, the characteristic vectors of SS, petroleum, viscosity, SRB, corrosion rate and median particle size are 2.9349, 0.3244, 0.5147, 1.7894, 0.1613, and 0.8389, respectively. The eigenvectors W = (0.4486, 0.0507, 0.0779, 0.2717, 0.0233, 0.1275) can be used as weight vectors(P = 0.0969 < 0.1). The concentration of the pollutants in all types of wastewater is used as the allocation object and the pollutant discharge of all types of wastewater is applied as the



Fig. 4. a) TGB, b) SRB, and c) IB content distribution range of OLW for different processing steps in oilfield.

allocation object. The weights of the wastewater are GWL (0.287), BFF (0.176), TWL (0.17), AWF (0.154), EMWL (0.129), and WLWW (0.084), indicating that the treatability of OLW is quite different (Table 2).

3.3. Construction of variable processes for operational wastewater treatment

3.3.1. Feasibility of variable processes for operational wastewater treatment Although the characteristic vector of viscosity is not the highest, viscosity significantly affects the fluidity of wastewater and has a negative impact on the subsequent process. Coagulant is difficult to diffuse in wastewater rapidly, which is not conducive to the removal pollution from the wastewater. Therefore, viscosity is an important limiting factor in OLW treatment.

The evaluation of GWL is 0.287 which is much higher than other types of wastewater. It indicated that the treatability of the GWL is significantly different from others. The evaluation of wastewater from TWL, AWL and BFF is between 0.15 and 0.2. For EMWL and WLWW, the evaluation value is below 0.15, indicating that all of these wastewaters can be treated easily. Overall, based on the evaluation of water quality matrix, six types of OLW can be divided into three categories:

HCW, MCW and LCW.

In general, organic compounds with carboxyl and hydroxyl groups are easy to coagulate [20]. However, the water quality of OLW is intricate and contains many kinds of complex organics, which is difficult to remove by coagulation alone [21–23]. Therefore, it is necessary to modify the wastewater to increase the proportion of oxygen-containing functional groups of aromatic rings of organic compounds and improve coagulation efficiency.

3.3.2. Proposed treatment configuration

Based on the investigation of OLW, a framework of multilevel system with source separated process to treat the OLW was proposed. A modular and variable-process wastewater treatment system was developed, as shown in Fig. 7.

The system comprises five units: oil removal, viscosity reduction, separation, filtration, and disinfection. Oil removal and viscosity reduction can be used as pre-treatment to remove oil and viscosity to ensure the stable operation of subsequent processes. Separation and filtration can be used as solid-liquid separation to remove particles. Disinfection can remove microorganism effectively.

The coalescer bed has been successfully applied for oil removing



Fig. 5. a) viscosity, b) corrosion rate, c) TDS and d) median particle size of OLW for different processing steps in oilfield.

Table 1

The excess standard rates of the OLW quality evaluation.

Parameter	AWF	GWL	EMWL	TWL	BFF	WLWW	Average
SS	208.955	673.383	238.848	290.247	152.593	122.057	281.014
Oil content	0.409	9.162	3.577	2.071	7.021	-0.002	3.706
Viscosity	1.992	7.774	7.228	8.933	2.618	1.546	5.015
SRB	163.034	209.169	114.483	160.5	214.65	84.842	157.78
Corrosion rate	-0.566	-0.689	-0.548	-0.685	-0.497	-0.800	-0.631
Median	13.354	12.947	9.574	12.528	15.139	11.733	12.546
particle size							

		SS	Oil content	Viscosity	SRB	Corrosion rate	Median particle size
SS	ſ	1	7	6	3	9	5
Oil content		1/7	1	1/3	1/6	4	1/4
Viscosity		1/5	3	1	1/3	7	4
SRB	-	1/3	6	3	1	7	4
Corrosion rate		1/9	1/4	1/7	1/7	1	1/5
Median particle size		1/5	4	1/4	1/4	5	1

Fig. 6. Judgement matrix assignment graph for the normalisation of the treatability of OLW.

Table 2

Evaluation of wastewater from the oilfield industry and modular treatment system for different OLW.

Category	HCW	MCW		LCW		
Items Result Evaluation Characteristic	GWL 0.287 E > 0.2 High viscosity, difficult to treat	TWL 0.17 0.15 < Mediun easy to	BFF 0.176 E < 0.2 m viscosity o treat	AWF 0.154 y, not	EMWL 0.129 E < 0.15 Low visc to treat	WLWW 0.084 osity and easy

coagulation are carried out simultaneously in a single unit. During this process, Al-based coagulants such as PAC can act as catalyst to promote the decomposition of ozone to produce more hydroxyl radicals and achieve the efficient removal of organic pollutants. On the other hand, ozone will change the hydrolysis form of coagulant, as a result, it improves the coagulation efficiency [30]. A dual-membrane system is widely used in wastewater treatment [31,32]. The filter unit consists of a two-stage filtration system: sand filtration and ultrafiltration [33]. Ultraviolet disinfection can be used for disinfection [34].

4. Case study

[24,25]; anthracite has been used as filter material. The NH_4ClO_4 is used as viscosity reducer. The dissolved air flotation (DAF) is widely used in the water treatment field combination with filtration and other processes to improve the water quality [26]. Ozone can adjust the functional groups of organic matters in wastewater, which makes the pollutants easier to be coagulated. [27–29]. In the DAF, ozonation and

Accordingly, the highest removal efficiency and the optimal performance can be achieved by selecting and combining appropriate treatment units in this system. To verify the treatment performance of the variable-process system, a modular and variable-process wastewater treatment system was constructed. During 160 days of field operation, the combined process was flexibly adjusted based on different



Fig. 7. Modular, variable-process treatment system for OLW.



Fig. 9. Profiles of the SS, petroleum oil, viscosity, medium particle size removal for MCW.

wastewater types. During the 160 days operation, AWF accounted for 12.5 % of total wastewater, GWL accounted for 18.9 %, EMWL accounted for 16.9 %, TWL accounted for 14.5 %, BFF accounted for 22.7 %, and WLWW accounted for 14.5 %.

The viscosity of GWL is very high, which indicated that GWL is difficult to be removed by coagulation. The GWL is treated through oil removal, viscosity reduction, dissolved ozone flotation (DOF), double membrane filtration and UV disinfection process. Fig. 8 shows that the



Fig. 10. Profiles of the SS, petroleum oil, medium particle size and corrosion rate removal for LCW.

viscosity of final effluent is lower than $1 \text{ mPa}\cdot\text{s}$, and SS, petroleum contents and median particle size are less than 2.0 mg/L, 6.0 mg/L, and $1.5 \mu\text{m}$, respectively.

The TWL, BFF and AWF are treated by oil removal, viscosity reduction, DAF, double membrane filtration and UV disinfection process. Fig. 9 shows that the viscosity of final effluent is lower than 1 mPa·s, and SS, petroleum contents and median particle size are less than 2.0 mg/L, 6.0 mg/L, and $1.5 \mu \text{m}$, respectively.

The EMWL and WLWW are treated by oil removal, DAF, double membrane filtration and UV disinfection process. Fig. 10 shows that SS, petroleum contents and median particle size are less than 2.0 mg/L, 6.0 mg/L, and $1.5 \mu \text{m}$, respectively.

The influent pH of the operation wastewater varies between 2 and 9, and the effluent pH ranges from 6 to 9. The SRB, IB, and TGB contents in the wastewater are $90 \sim 5300/\text{mL}$, $100 \sim 6000/\text{mL}$ and $120 \sim 22000/\text{mL}$, respectively. After UV disinfection, the SRB, IB, and TGB in the effluent water are $0 \sim 10/\text{mL}$, $2 \sim 65/\text{mL}$ and $30 \sim 70/\text{mL}$, respectively. The contents of the three microorganisms are much lower than those listed in SY/T 5329-2012.

The system ran steadily under water quality variation during the 160 days of field operation. The SS is less than 2.0 mg/L, the petroleum content is less than 6.0 mg/L, the median particle size is less than $1.5 \,\mu\text{m}$, and the water quality meets the reinjection requirements. The modular and variable-process wastewater treatment system is suitable for the treatment of wastewater from unconventional oilfields.

5. Conclusion

In this study, the types and composition of OLW from the Changqing oilfield were analysed, and the treatability were clustered using a water quality matrix. Based on the analysis results, six types of wastewater were divided into HCW, MCW and LCW. A modular and variable-process wastewater treatment system was developed. The system consists of five units: oil removal, viscosity reduction, DAF/DOF, two-stage filtration, and UV disinfection. Each module of the system can be flexibly used for the wastewater treatment according to the variable water quality and characteristics of the raw water. During 160 days of field operation for the treatment of OLW with variable water quality, the quality of the effluent meets the reinjection requirements of the standard with SS < 2.0 mg/L, petroleum particle contents < 6.0 mg/L and median particle size < 1.5 μ m. The treatment system can improve the wastewater treatment efficiency with variable water quality in the oilfields and protect regional environmental safety.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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