The advanced oxidation processes of oilfield wastewater: A review

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Abstract. Oilfield wastewater is the injection water from underground that brought to the surface during oil or gas extraction. It is generated in large amounts and has a complex composition, containing various toxic organic and inorganic compounds. Oilfield wastewater is currently treated in conventional methods such as phase separators, decanters and cyclones, which do not achieve more restrictive limitations related to the reuse of the effluent (reinjection into extraction wells). Therefore, advanced oxidation processes (AOPs) have been used in oilfield wastewater treatment for its excellent oxidizability. This review tries to provide an overview of AOPs for wastewater treatment. These oxidants include ozone, Semiconductor photocatalytic, Fenton and ferric iron. Optimum operating conditions and processing efficiency were also discussed.

1. Introduction
In the late stage of the oil field, the water injection development method is basically adopted, that is, injecting high-pressure water to maintain the reservoir pressure and driving the crude oil to be extracted from the well [1]. After a period of water injection, some of the crude oil flows out with the injected water, which is called oil field production water. With the increasing water content of oil field, the water produced by the oilfield becomes the main pollutant. In order to improve the recovery rate, chemical flooding technology has been widely used in various oil fields, which has led to chemical flooding in the oil field [2]. Compared with the traditional oil field produced water, chemical flooding produced water shows different characteristics, which include residual oil residual displacer such as hydrolysed polyacrylamide (HPAM), surfactants or high content alkali, which composition is complex and biological diversity is lager. There are many kinds of organic matter in the PW of oil field, such as high oil concentration, large scale, high suspended solids and high treatment requirements [3-6]. These characteristics increase the difficulty of chemical flooding significantly. Direct external drainage can cause serious environmental problems which endanger human health consequently.

At present, the treatment of water extraction from chemical flooding field is mainly focused on from three aspects: agent, equipment and technology [7-9]. Treatment agents mainly include the development of new agents such as demulsifier, oxidant and adsorbents, development of different structures or types of reagents and new methods for evaluation of pharmaceutical screening, also include optimization of operational condition [10]. Treatment equipment mainly includes the inclined the inclined tube oil remover and the ultrasonic demulsification degreasing apparatus, fine filter equipment such as a polymer film, an inorganic ceramic membrane, and a surface modified membrane, and integrated processing equipment. The treatment technology includes advanced oxidation
technology, electrochemical method and other combination process. Using medicament treatment chemical flooding produced water still have problems such as uneconomical, low oil recovery, large generation of sludge, difficulties in sludge treatment, material blockage caused by flocs and accompanied secondary pollution. And produced water treatment equipment has good function of oil-water separation, but still can’t meet standard when dealing with emulsive, complex, high oil displacement agent + and suspended solid chemical flooding produced water. In recent years, AOPs have become pilot in environmental science research and have been widely used in water treatment for its great potential and significant unique advantages [11-13].

Among the innovative wastewater treatment technologies, advanced oxidation processes played an important role. AOPs have several advantages when compared with traditional methods and AOPs for wastewater treatment. The AOPs has a good degradation effect on wastewater which has high density, high toxicity, and poor biodegradability [14,15]. At the same time, it has significant advantages in the treatment of trace amounts of harmful chemical substances such as environmental hormones. AOPs has attracted attention in engineering applications because of its advantages such as the ability of complete oxidation, high degradation efficiency, and no secondary pollution [14,16].

2. Advanced oxidation process for the treatment of oil field wastewater

2.1. Oxidation by ozone(O₃)

The earliest use of ozone in wastewater treatment can be traced back to 1906, the world's first ozone disinfection process was used in the French Nice water plant, and subsequently, it has been widely used in domestic sewage, industrial wastewater and other treatment [17]. Ozone technology is widely used in advanced wastewater treatment due to its strong oxidation ability. Ozone is a strong oxidant and has good effects in disinfection, color removal, deodorization, removal of organic matter and COD. Ozone-based advanced oxidation processes, such as O₃/H₂O₂ process and homogeneous catalytic ozonation, have been applied in wastewater treatment due to the high oxidizability and no secondary pollution [18]. The reaction can be described as follows [19,20]:

\[
\begin{align*}
O_3 + H_2O + hv & \rightarrow H_2O_2 + O_2 \\
H_2O_2 + hv & \rightarrow 2OH^- \\
O_3 + H_2O_2 & \rightarrow 2OH^- + 3O_2
\end{align*}
\]

Ozone oxidation is used in oilfield wastewater treatment because it contains persistent organic substances, such as surfactants, which cannot be decomposed by microorganisms, but ozone is very easy to oxidize and decompose these substances. Especially in oilfield wastewater containing phenolic compounds, ozone oxidation treatment can remove the stench produced by phenol. In recent years, the research on advanced ozone oxidation process includes ozone and combined ozone oxidation (UV/O₃, O₃/H₂O₂, UV/ O₃/H₂O₂, Ultrasound/ O₃) [14,21].

The studies that used O₃ and combined O₃ processes to treat different types of oilfield wastewater r is presented in table 1. Concerning of the studies that treated oilfield wastewater by O₃, we found that the treatment effect is the best when PH is alkaline in the advanced oxidation of ozone. The effluent can basically satisfied the standard through the ozone advanced oxidation treatment.

2.2. Semiconductor photocatalytic method

TiO₂ under light conditions can decompose water into H₂ and O₂ that first discovered by Fujishimia and KHonda in 1972, which technology was quickly applied in wastewater treatment. Many studies have proved that many refractory organics can be efficiently removed or degraded under the photocatalytic oxidation [30]. TiO₂ has become the most widely used material in the field of photocatalytic environmental because of its low cost, low toxicity, stability, and high catalytic activity. The photocatalytic process in the presence of oxygen is shown as following equations [31].
The 4th International Conference on Water Resource and Environment (WRE 2018) IOP Conf. Series: Earth and Environmental Science 191 (2018) 012098 doi:10.1088/1755-1315/191/1/012098

$$\text{SMP} + h_v \rightarrow \text{SMP}(e_{CB}^- + h_v^+) \quad (4)$$

$$\text{HO}^- + h_v^+ \rightarrow \text{HO}^+ \quad (5)$$

$$\text{H}_2\text{O} + h_v^+ \rightarrow \text{HO}^- + \text{H}^+ \quad (6)$$

$$\text{O}_2 + e_{CB}^- \rightarrow \text{O}_2^- \quad (7)$$

Table 1. Comparison of \textit{O}_3 oxidation method based on oilfield wastewater.

| Process     | Conditions          | Efficiency | Comments/additional information                                           | Refs. |
|-------------|---------------------|------------|---------------------------------------------------------------------------|-------|
| \textit{O}_3 | Qm \textit{O}_3 2.88 mg/L | 95\%       | The performance of ozone is better under alkaline conditions.             | [21]  |
| \textit{O}_3 | Qm \textit{O}_3 0.4-0.6 mg/L | Sulfate-reducing bacteria can basically be eliminated. | When the contact reaction time \( t > 2.5 \text{ min} \), \( C_t > 2.0 \text{ mg· min}^{-1} \), can meet the standard of water injection. | [22]  |
| UV/\textit{O}_3 | Qm \textit{O}_3 20-31 mg/L UV 253.7 nm | COD degradation rate of 52.2\%, turbidity removal rate of 90\%, color removal rate of 95\% | When the pH reaches about 11, COD degradation rate of wastewater reaches the best effect. | [23]  |
| \textit{O}_3 | Qm \textit{O}_3 6 mg/min | 37.98\% COD removal in 20min | When the pH reaches about 9, COD removal rate of wastewater reaches the best effect. | [24]  |
| \textit{O}_3/\textit{H}_2\textit{O}_2 | Qm \textit{O}_3 5 mg/min | 32.2\% COD removal, 82.8\% PAM, 82.1 oil removal in 20min | When the pH is 8, \textit{H}_2\textit{O}_2's dose is 0.4 mL/L and the temperature is 20 degrees centigrade, the treatment reaches the best effect. | [25]  |
| \textit{O}_3/\text{Sonolysis} | Ultra sound strong 1.6 W/cm² Qm \textit{O}_3 7.5 mg/L | 86.1\% viscosity reduce in 15min | This method is not good for the removal of suspended solids. | [26]  |
| \textit{O}_3 | Qm \textit{O}_3 12 mg/L pH 9 | More than 55\% COD removal in 60min | The discharge can be achieved (GB 8978-1996). | [27]  |
| Heterogeneous catalysis \textit{O}_3 | Qm \textit{O}_3 12 mg/L Catalyst A3 1000 mg/L pH 10.8 | 79.4\% COD removal in 50min | The discharge can be achieved (GB 8978-1996). | [28]  |
| \textit{O}_3/\text{Sonolysis} | Three-frequency quadrature field (20 kHz +28 kHz +40 kHz) | 86.1\% viscosity reduce in 15min | The discharge can be achieved (GB 8978-1996). | [29]  |

Table 2 lists all studies on photocatalytic processes. The studies shows that control conditions have a great influence in semiconductor photocatalytic reactions. In general, the treatment effect is better. The reason why there is a bad treatment effect is that no suitable semiconductor mix ratio has been found. Therefore, when using this method to treat sewage, the best mix ratio should be done first, which can achieve economical and practical purposes.

Table 2. Comparison of Semiconductor photocatalytic method based on oilfield wastewater.

| Process     | Conditions          | Efficiency | Comments/additional information                                           | Refs. |
|-------------|---------------------|------------|---------------------------------------------------------------------------|-------|
| \textit{TiO}_2/\textit{UV} | (\textit{TiO}_2) 2 g/L | 61\% COD removal in 60 min | The effluent reached (GB 8978-1996 level 2). | [32]  |
| \textit{TiO}_2/\textit{UV} | (\textit{TiO}_2) 0.05 | 7.2\% COD removal in 50 min | COD removal rate is extremely low, | [33]  |
2.3. Fenton oxidation

The Fenton method is a typical advanced oxidation technique. It is a pretreatment method that uses Fenton reagent to improve the biodegradability. The oxidation system formed by \( \text{H}_2\text{O}_2 \) and catalyst \( \text{Fe}^{2+} \) is usually called Fenton reagent. Fenton reagent has a very strong oxidizing ability because hydrogen peroxide can produce highly oxidized hydroxyl radicals in the presence of iron ions [36-38]. In addition, hydroxyl radicals have high electronegativity or electrophilic, and their electron affinity is 569.3 KJ, which has a strong addition reaction characteristic. The reaction can be described as follows:

\[
\text{H}_2\text{O}_2 + \text{Fe}^{2+} \rightarrow \text{Fe}^{3+} + \text{OH}^- + \text{OH}^- \tag{8}
\]

\[
\text{OH}^- + \text{Fe}^{2+} \rightarrow \text{Fe}^{3+} + \text{OH}^- \tag{10}
\]

Fenton oxidation is the most practical and widely used method for advanced oxidation. It has extremely strong oxidizability and is suitable for the treatment of high-concentration wastewater that is difficult to degrade and has biological toxicity. Table 3 compares Fenton oxidation treatment of different types of oilfield wastewater. The results show that the treatment effect is better under alkaline conditions.

Table 3. Compares Fenton oxidation treatment of different types of oilfield wastewater.

| Process | Conditions | Efficiency | Comments/additional information | Refs. |
|---------|------------|------------|---------------------------------|-------|
| Fenton’s reagent (Fe\(^{2+}\) + \( \text{H}_2\text{O}_2 \)) | FeSO\(_4\) 3.6 mmol/L, pH 4 | the viscosity of the system | The control of temperature and the amount of hydrogen peroxide are the most suitable. | [35] |
| Fenton’s reagent (Fe\(^{2+}\)/\( \text{H}_2\text{O}_2 \) (mole ratio)=1:10, pH 3) | 34.13% COD removal in 45 min | | H\(_2\)O\(_2\)/COD\(_{Cr}\) (mass ratio) and pH value were the main factors affecting the removal rate of COD\(_{Cr}\). | [25] |
| UV/Fenton/ \( \text{C}_2\text{H}_2\text{O}_2\^- \) | Fenton reagent 10Q, pH 7 | More than 70% HPAM removal; More than 90% oil removal | The effect of oil removal is better in ternary complex flooding. | [39] |
| Fenton’s reagent (Fe\(^{2+}\)/\( \text{H}_2\text{O}_2 \) (mole ratio)=1:2.5, I 1.5A, pH 3) | 34.13% COD removal in 30 min used Fenton; 93% COD removal in 30 min used Electric Fenton | | The effect of the test treatment effect was the reaction time: >\( \text{H}_2\text{O}_2 \) and FeSO\(_4\) mol ratio > \( \text{H}_2\text{O}_2 \) thick >pH. | [40] |
| Electric Fenton Fenton | | | | |
| Fenton’s reagent (Fe\(^{2+}\)/\( \text{H}_2\text{O}_2 \) (mole ratio)=1:10, pH 3) | COD\(_{Cr}\), polymer and oil removal rates were 90%, 95% and 92% respectively in 40 min. | | The discharge can be achieved (GB 8978-1996). | [41] |
| Fenton’s reagent (Fe\(^{2+}\)/\( \text{H}_2\text{O}_2 \) (mole ratio)=1:3, Quality ratio=1:3) | The quality concentration of COD\(_{Cr}\) is controlled below 50 mg/L and the mass concentration of nh3-n is lower than 3 mg/L. | | The pH is set at 2.2 ~ 2. Fenton oxidation was the most efficient. | [42] |
| Fenton’s | Fe\(^{2+}\)/\( \text{H}_2\text{O}_2 \) | 60% COD removal in 7h | The discharge can be achieved | [43] |
2.4. High iron oxidation

Iron is an environmentally harmless particle, which accounts for about 5.1% of the earth. Iron is usually present in the form of metal or Fe (II) and Fe (III). In 1702, German chemist Stahll [44] first discovered Fe(IV), more than 100 years later, Thompsont obtained the high purity and high yield of Fe(VI) in the laboratory for the first time using hypochlorite oxidation trivalent iron. After that, there have many industrial methods, such as electrolysis, high temperature peroxides and potassium perchlorate.

Fe(VI) is a new kind of water treatment agent that not like chlorine source water treatment agent such as liquid chlorine, bleaching powder that No secondary pollution and good safety. Table 4 lists all studies on high iron oxidation treatment of different types of oilfield wastewater.

Table 4. Compares High iron oxide treatment of oilfield wastewater.

| Process                  | The synthesis of Fe(VI) | Conditions                        | Efficiency     | Comments/additional information                                                                 | Refs. |
|--------------------------|-------------------------|-----------------------------------|----------------|-----------------------------------------------------------------------------------------------|-------|
| Fe(NO3)_3·9H2O           | Hypochlorite oxidation  | Iron salt is added to 85%-95%; KOH 10.5 mol/L; | 70% COD removal in 30 min | The treatment for oily wastewater and containing polymer sewage has unique advantages. | [39]  |
| Fe(VI)                   | Potassium hypochlorite oxidation | K2FeO4 0.003 mol/L, pH 3 | 30% COD removal in 40 min | The polymer flooding PAM wastewater reaches the national level II pollutant discharge standard. | [45]  |
| Fe(VI)                   | Potassium hypochlorite method | K2FeO4 0.003 mol/L, pH 3 | The degradation rate of PAM decreased rapidly in 30 min. | The polymer flooding PAM wastewater reaches the national level II pollutant discharge standard. | [23]  |

3. Conclusions

Different types of AOPs used in oilfield wastewater treatment were reviewed in this paper. AOPs include heterogeneous and homogeneous photocatalysis, Fenton and Fenton-like processes, ozonation, the use of ultrasound, microwaves and g-irradiation, electrochemical processes and wet oxidation processes. This review mainly summarizes the application of oil fields in different advanced oxidation processes and draw following conclusions:

- Advanced oxidation of ozone is the earliest application of advanced oxidation processes. The analysis results of ozone application imply: 1) The performance of ozone is better under alkaline conditions; 2) Ozone combined with other catalysts can achieve better result than pure ozone.

- In oilfield wastewater treatment, TiO2 is the common semiconductor material used in photocatalytic method. The analysis results show that the amount of semiconductor material has great influence on eliminate of pollutants. Thus it is of great importance for researchers to find out a proper dosage of semiconductor material in application.

- Fenton oxidation has been widely used in diverse wastewater treatment recently and achieved excellent results. Similarly, in the treatment of oilfield wastewater, Results of some researches result show that factors such as the ratio of Fe2+ and H2O2, PH, temperature and reaction time have effects on the treatment of oilfield wastewater. The results also suggest that when dealing...
with oilfield wastewater, fenton oxidation technology can obtain better result under acidic conditions.

- Similarly, acidic conditions are also more suitable for high iron oxidation to achieve better results in oilfield wastewater treatment.

**Acknowledgments**

This research was supported by China NSF (Program No. 51401165) and Natural Science Basic Research Plan in Shaanxi Province of China (Program No. 2014JM2-2018).

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