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Research Progress on Treatment of Alcohol-Free Produced Water in Changqing Gas Field

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Abstract

Changqing gas field is the largest oil and gas field in China. The discharge of alcohol-free produced water continues to grow with the expansion of mining scale. This kind of wastewater has the remarkable characteristics of ' high turbidity, high salinity, strong corrosion and low pH value '. The composition is complex and the treatment is difficult. The national environmental protection policy has tightened the promotion of wastewater treatment from underground reinjection to ground standard treatment and comprehensive utilization. Efficient treatment and resource utilization have become the key to green mining of gas fields. In this paper, the water quality characteristics and three treatment methods of produced water without alcohol in gas field are systematically sorted out. The five main treatment technologies of physics, chemistry, physics and chemistry, biology and compound, as well as the special treatment technologies such as Fenton method and double alkali system method and the reuse technology of fracturing flowback fluid are expounded in detail. The optimization measures of corrosion control are analyzed, and the future development direction of technology is prospected. This review can provide a reference for the efficient treatment and resource utilization of alcohol-free produced water in Changqing gas field and similar gas fields.

Keywords

Gas Field; Produced Water Without Alcohol; Processing Technology; Water Quality; Resource Utilization.

1. Introduction

In natural gas production, methanol is often used as a hydrate inhibitor to prevent pipeline blockage, thereby forming alcohol-containing produced water. The produced water without alcohol is the groundwater directly produced in the process of gas field exploitation. With the expansion of natural gas exploitation, its emissions continue to grow. As the largest oil and gas field in China, Changqing Oilfield is located in the Loess Plateau where water resources are scarce. After treatment, the annual surplus of alcohol-free produced water reaches $4.5\times10^6 m^3$, and its discharge will pose a potential threat to the local fragile ecological environment. With the continuous tightening of national environmental protection policies, the treatment of gas field wastewater has gradually shifted from underground reinjection to comprehensive utilization after ground standard treatment. The efficient treatment of produced water without alcohol is imminent. The gas field produced water without alcohol has unique water quality characteristics and is difficult to treat. How to achieve its standard treatment and resource

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utilization has become a key technical problem for green mining of gas fields. This paper focuses on the treatment technology of alcohol-free produced water, which provides support for related research and engineering application.

2. Water Quality Characteristics and Treatment Methods of Alcohol-Free Produced Water in Gas Field

Gas field produced water refers to the groundwater taken away from the surface during the exploitation of natural gas[1]. The natural gas of all gas wells in Changqing gas field is ' gas-liquid mixed transportation ' through the two main lines of the south and north, and then the gas and liquid phases are separated by different treatment methods. According to the difference of mining methods, the produced water of the gas field can be divided into two types, namely, the produced water without alcohol and the produced water with alcohol. For these two kinds of water quality, each has a completely different treatment process. How to correctly deal with the produced water in gas field has always been a difficult point in gas field exploitation.

2.1. Analysis of Water Quality Characteristics

The produced water without alcohol in the gas field is derived from the groundwater brought out in the process of natural gas exploitation, and its water quality is significantly affected by the geological conditions of the gas field, mining methods and other factors. The main domestic gas field represented by Changqing gas field, the produced water without alcohol shows distinct characteristics of ' high turbidity, high salinity, strong corrosion and low pH value ', which is usually a kind of acidic colloidal state. At the same time, there are $NaHCO_3$, Na_2SO_4 and other water properties, mainly $CaCl_2$[2].The specific water quality analysis data are shown in Table 1.

Table 1. List of produced water quality in Changqing gas field

Index	Jingbian gas field	Yulin gas field	Sulige gas field
pH	5.0-6.0	6.0-6.5	6.0-7.5
HCO_3^-(mg/L)	426	421	294
CL^-(mg/L)	41000	43000	24000
SO_4^{2-}(mg/L)	490	38.3	1850
K^++Na^+(mg/L)	8500	1063	4340
Ca^{2+}(mg/L)	15010	1534	5720
Mg^{2+}(mg/L)	1752	129	249
Total iron content (mg/L)	115	38.9	17.5
Mineralization (mg/L)	75000	7086	38976

It can be seen from Table 1 that the pH value of water in each block of Changqing gas field ranges from 5.0 to 7.5, and the salinity can reach up to 75000 mg/L. At the same time, it contains high concentrations of Cl^-, Ca^{2+}, Mg^{2+} and total iron. In addition, some of the produced water is also accompanied by suspended solids, oil substances and trace organic matter. These characteristics not only increase the difficulty of treatment, but also put forward strict requirements for the corrosion resistance of the treatment equipment.

2.2. Treatment Way of Produced Water Without Alcohol in Gas Field

According to the research, in the process of natural gas exploitation, in the face of the problem of produced water pollution, Western developed countries mainly exploit it with strong economic strength and scale. At the same time, some expensive treatment technologies are used to control it during mining. And so far, these treatment technologies are still continuously

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enriched and improved. At present, there are three main ways to control the produced water in various gas fields in China[3], namely, comprehensive treatment, reinjection of strata and discharge after treatment.

2.2.1. Comprehensive Management

This model takes the resource utilization of produced water as the core, uses the treated wastewater to produce salt, and extracts trace elements such as bromine (Br), iodine (I) and lithium (Li) from the enriched brine, which realizes the resource value-added of wastewater. It is suitable for gas fields with suitable water quality and high resource recovery value.

2.2.2. Reinjection Formation

For some gas production, oil production wastewater and some oilfield wastewater with high content of heavy metals, arsenic and sulfide, which can not be directly discharged into the river due to high salt content, the method of reinjection into the formation has been adopted one after another, and the reinjection index is shown in table 2. However, the reinjection reservoir not only has higher requirements for water quality, but also has higher requirements for the condition of the reinjection well itself and the selection of the reinjection layer. With the adjustment of environmental protection policy, its application is gradually limited.

Table 2. Reinjection technical indexes of produced water in Changqing oil and gas field

Years	Implementation standards	Indicators
Before 2004	'Sewage reinjection index control standard ' Changqing Petroleum Exploration Bureau No. 10, 1999	Suspended Solid ≤ 10mg/L Oil Content ≤ 35mg/L
2004-May 2017	'Gas field water reinjection method' SY / T 6596-2004	Suspended Solid ≤ 15mg/L Oil Content ≤ 30mg/L
After May 2017	'Gas field water injection technical requirements' SYT 6596-2016	According to the geological data of the reinjection well, it is determined by injection test.
	'Gas field gathering and transportation design specification 'GB50349-2015	

2.2.3. Post-treatment Efflux

Various physical, chemical and biological methods are used to comprehensively treat the pollutants in the water body, so that the content of pollutants in the water body reaches the national standard. This is the most radical way in the process of natural gas production, which is the same as the current international trend of ' protecting the environment'.

3. Treatment Technology of Produced Water without Alcohol in Gas Field

Sewage treatment technology at home and abroad generally includes three processes of separation, conversion and utilization. The treatment technology of produced water without alcohol in gas field can be divided into five categories: physical treatment, chemical treatment, physical and chemical treatment, biological treatment and composite treatment. The principles and application characteristics of various technologies are different.

3.1. Physical Processing Method

The physical treatment method is used to treat the sewage. The main purpose is to remove the minerals in the water and most of the suspended solids and oil substances. In the process of treatment, the substances will not undergo phase change, and the separation coefficient is very large. At present, the commonly used physical treatment methods are as follows[4].

(1) Gravity separation method

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This method is to use the density difference between the oil phase and the water phase and the incompatibility of the two-phase substances to achieve the separation of suspended solids, oil and water three-phase substances in the case of static or flowing sewage, and the separation effect has a great relationship with the settling time. This method is suitable for the treatment of floating oil and dispersed oil with particle size > 60 μm. The advantages of this method are that it does not require external power, large processing capacity, no secondary pollution, low operation and maintenance costs, and has a strong tolerance for the suspension and oil concentration of the treated sewage. It can remove such impurities in a large amount. The disadvantage is that it covers a large area.

(2) Centrifugal separation method

The principle is to use the physical law that the centrifugal force generated by the mass difference of different phases in the wastewater is not the same, and the impurities in the wastewater are separated from the wastewater by the strong centrifugal force field. Because the quality is different, so it is subject to different centrifugal force. Larger particle impurities will be subjected to greater centrifugal force, while smaller particle impurities will be smaller. Therefore, in the process of centrifugal separation, if the center of the force field is taken as the origin, then as the particle mass increases, the distance between the position where it is thrown away and the center point will also increase accordingly, so as to achieve the purpose of separating particle impurities of different mass into different regions.

(3) Mechanical filtration method

The mechanical filtration method refers to the use of a filter with a filter material to treat the sewage. The filter material has a certain interception and pollution capacity for the oil droplets and suspended solids in the water, so as to achieve the purpose of purifying the sewage. This method is usually used in the secondary or advanced treatment of oily wastewater, and is mainly used to remove dispersed oil and emulsified oil. Among these filter materials, the fiber ball filter material is the most noticeable. The fiber ball filter material not only has a strong ability to remove solid particles and oil, but also does not lose the filter material.

(4) Coarse-grained method

The coarse-grained method takes advantage of the affinity difference between the oil-water two-phase polymer materials to achieve the purpose of separation. With the flow of sewage, some hydrophobic and lipophilic substances coalesce and adsorb with them, so that the size of oil droplets continues to increase. The coarse granulation method is more suitable for the treatment of dispersed oil and emulsified oil with oil particle diameter greater than 10 μm. This method is simple in operation, small in equipment and small in investment. However, it also has a flaw, that is, the coarse-grained material is easy to block, so the filter material has a certain life. In industrial production, the most common are quartz sand and anthracite.

(5) Membrane separation method[5]

The membrane method is based on screening. By selecting different membrane materials, the separation of pollutants in water is realized. At present, there are many kinds of membrane materials used in industry, and they have attracted much attention due to their advantages of simple equipment and low energy consumption. At present, domestic and foreign scholars have carried out a lot of research on non-polluting high-efficiency membrane materials. In this study, multi-stage membrane separation was also used to treat the produced water without alcohol into a water body that can be returned to the fracturing fluid.

3.2. Chemical Treatment Method

In the conventional water treatment process, because the wastewater contains more suspended solids, and the wastewater is acidic or alkaline, therefore, in order to combine the removal of impurities in the wastewater with the removal of acid and alkali, chemical

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substances are added to the water treatment process, and the wastewater is purified by the action of chemical substances. This process is called chemical treatment. According to the different action principles of the agent, such as oxidation principle, acid-base neutralization, flocculation and sedimentation, it can be divided into the following[6].

(1) Advanced oxidation technology

The technology takes ' oxidation ' as the core and gives full play to the strong oxidation of ' hydroxyl '. In general, most of the organic matter in the wastewater to be treated has a high molecular weight and is difficult to degrade. If it is directly discharged into the environment without effective treatment, it will cause irreparable damage. Although there are differences in the production mode of (·OH) and the reaction conditions of oxidation technology, the ultimate goal of advanced oxidation processes is to oxidize and degrade pollutants[7] into small molecules with weak toxicity or even non-toxicity.

(2) Chemical precipitation method

This method is mainly to remove impurity ions and dissolved pollutants in water. The principle of chemical sedimentation method is composed of three steps : adding chemical reagent, chemical reaction and precipitation. After using this technology to treat wastewater, there will be two significant changes: first, the pollutants in wastewater change from dissolved state to precipitated state; second, the pollutants change from soluble state to insoluble state ; this method has the characteristics of low cost and mature technology, so it has been widely used.

(3)Chemical flocculation method

Chemical flocculation is an important means of sewage treatment[8], which is suitable for the treatment of dispersed oil and emulsified oil with oil particle size greater than 10 μm in water. The basic principle of chemical flocculation is to use small colloidal particles such as impurities and colloids in wastewater to destabilize and coalesce under the conditions of physical adsorption and neutralization of flocculants, and then remove the generated large particle aggregates by precipitation or air flotation. This method is low in cost and stable in operation, but for sewage with violent water quality fluctuations, it is necessary to strictly control the number of injected agents to achieve the purpose of effectively dealing with water quality fluctuations.

3.3. Physicochemical Treatment

Through this method, pollutants in water can be removed. The essential principle is to combine physical and chemical effects to achieve mass transfer of pollutants between the two phases. Commonly used methods include stripping, electrolysis, etc.[9].

(1) Stripping method

Compared with the traditional back-extraction method, the difference between the traditional back-extraction technology and the traditional back-extraction method is that it uses water vapor as a medium to remove volatile toxic substances in water, and the back-extraction technology needs to ensure that the wastewater is in full contact with water vapor to achieve the back-extraction goal.

(2) Electrolysis method

Electrolysis technology, including electrolytic suspension technology and electrolytic flocculation technology. The electrolytic air flotation method refers to the microbubbles of hydrogen and oxygen generated by electrolysis of water in the cathode and anode under the action of a DC power supply. These microbubbles can oxidize and reduce pollutants in the water. At the same time, because the microbubbles of the microbubbles, small oil droplets, suspended particles, etc. will adhere to the surface of the microbubbles and be removed by air flotation. The electrolytic flocculation method produces flocculant-like metal ions and their hydrolysates by depleting the oxidation of the electrode to achieve the separation and removal of pollutants.

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3.4. Biological Treatment

In the field of water treatment, by combining the aerobic and anaerobic properties of microorganisms, or the combination of two properties[10], microorganisms are used as intermediate media to realize the decomposition and conversion of pollutants, so as to purify sewage. Microorganism is the core material of biological treatment process. A/O and UASB are the two most commonly used methods[11]. The former is a combination of anaerobic and aerobic methods, which has a very high removal rate in the process of removing suspended solids and organic pollutants in water. Under the action of biodegradation, the wastewater is biodegraded, three-phase mixed and then three-phase separated[12]. The biological treatment method is not only simple to operate and does not produce other pollutants, but also has a low investment cost in industrial applications. Therefore, it is widely used at home and abroad[13].

3.5. Composite Treatment Method

Based on the increasingly complex types and composition of industrial wastewater[14], composite water treatment technology is a development trend of water treatment technology at home and abroad. The process is a combination of a variety of water treatment processes, which can carry out advanced treatment of wastewater, so it can effectively remove suspended solids in water and organic pollution.

3 Special treatment technology of alcohol-free produced water in gas field[15]

(1) Fenton method :

Hydrogen peroxide (H_2O_2) is used as an oxidant to form a Fenton reaction with ferrous (Fe^{2+}) ions[16]. The Fenton reaction has a strong oxidizing activity and can achieve effective removal of COD in raw water without causing secondary pollution. At the same time, due to the enrichment of Fe^{2+}, the consumption of the agent can be reduced to a certain extent. However, the pH value of Fenton reaction has certain limitations. Too high or too low pH value will lead to the generation of hydroxyl radicals (·OH), which will affect the oxidation of the system. Therefore, it is generally believed that the pH value of Fenton reaction is set between 2 and 4, and the treatment effect of organic pollutants will be better.

(2) NaOH-$NaCO_3$ double alkali system method :

There are many ways to reduce the hardness. The most common way is to add chemical agents to remove the cations that contribute to the hardness by precipitation. The current way to reduce the hardness of the process is to adjust the water to alkaline by adding sodium hydroxide. The cations in the water combine with hydroxide to form hydroxide compounds to settle, but the hardness removal effect of sodium hydroxide on calcium ion compounds is poor. In order to further enhance the hardness reduction effect, sodium hydroxide-sodium carbonate double alkali method can be used to reduce the hardness. Firstly, sodium hydroxide is added to remove cations such as magnesium ions in water, and then sodium carbonate is added. Carbonate ions are combined with calcium ions in water to reduce the content of calcium ions in water and further reduce the hardness of standard water. Sodium carbonate is a strong alkali and weak acid salt, which is cheaper than sodium hydroxide and can play a role in regulating pH with sodium hydroxide.

There are many ways to reduce the hardness. Among them, the most used method is to add sodium hydroxide (NaOH) to make the water weaker, make the water alkaline, and make the cations in the water react with OH-to form hydroxide compounds. However, the NaOH-$NaCO_3$ double alkali system has little effect on Ca^{2+}. Therefore, NaOH should be added first to remove cations such as Mg^{2+} in the water. After that, $NaCO_3$ is added, and CO_3^{2-} will combine with Ca^{2+} in water, so that the content of Ca^{2+} in water will decrease, and then the hardness of water will be reduced to standard.

(3) Solar evaporation technology

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Evaporation is a method of using thermal energy to convert liquid water into gas. It has broad prospects in the fields of pure water, seawater desalination, sewage treatment, evaporative crystallization and so on. Because Changqing is located in the Ordos Basin and the Shaanxi-Gansu-Ningxia Basin, it is sunny and suitable for the conversion and application of solar energy. Combining solar energy instead of fossil energy with evaporation technology, the standard water of the gas field water plant is treated in depth. On the basis of green and low carbon, the salt, organic matter and other substances in the standard water can be further removed to improve the water quality of the produced water[17]. However, due to the complex composition of the gas field water sample and the easy pollution of the evaporation device, it is necessary to apply it to the actual wastewater treatment. A breakthrough in the anti-fouling properties of the material is still needed[18].

4. Present Situation of Fracturing Flowback Fluid Reuse Treatment Technology

Fracturing flowback fluid is an important part of alcohol-free produced water. Its reuse treatment can reduce the cost of liquid preparation and realize the recycling of water resources. In 2022, Li Bin carried out research on the flowback fluid in the fracturing process in the Sulige gas field[19].In view of the characteristics of the flowback fluid in the Sulige gas field and the water quality requirements of the prepared fracturing wastewater, the treatment technology of ' pretreatment-coagulation-precipitation-two-stage filtration-disinfection-bacteria ' was proposed. The comparison of gel breaking performance is shown in Table 3. The treated flowback fluid can be made into guanidine gum type and polymer type fracturing fluid, and its high temperature resistance, shear resistance, viscoelasticity, gel breaking and other properties can meet the requirements of national regulations.

Table 3. Gel breaking performance of fracturing fluid

Item	Test result	
	Guanidine gum fracturing fluid	Polymer fracturing fluid
Gel breaking time/h	0.5-2 Controllable	0.5-2 Controllable
Viscosity of gel breaking liquid /($mm^2 \cdot s^{-1}$)	1-2	1-3
Surface tension of gel breaking liquid /($mN \cdot m^{-1}$)	23-25	23-27
Kerosene interfacial tension/($mN \cdot m^{-1}$)	0.8-1.4	0.8-1.8
Residue rate/($mg \cdot L^{-1}$)	300-450	220-350

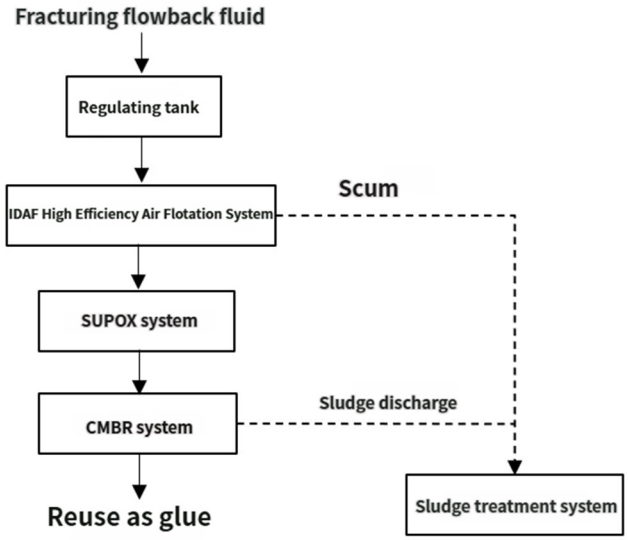

Fig 1. Fracturing flowback fluid reuse treatment process

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Lun et al.used IDAF air flotation-SUPOX-CMBR technology to efficiently treat the fracturing flowback fluid[20].After treatment, the suspended solids content in the water sample was less than 2mg/L, and the oil content was less than 3mg/L, which met the influent standard. IDAF flotation-SUPOX-CMBR combined process technology was used. The oil content of the treated water was less than 3mg/L, the suspended solids content was less than 2mg/L, and the turbidity was less than 1NTU. The treated water sample was used as water for compounding, and then the compounding experiment was carried out, and the performance of the viscosity change after compounding was evaluated. Finally, it was determined that it met the requirements for use. The specific process is shown in Figure 1.

5. Present Situation of Alcohol-free Produced Water Treatment Process

At present, the widely used gas field non-alcohol produced water treatment process is mainly divided into two categories: three-stage and transitional[21]. The main process of the three-stage sewage treatment technology is shown in Fig.2.

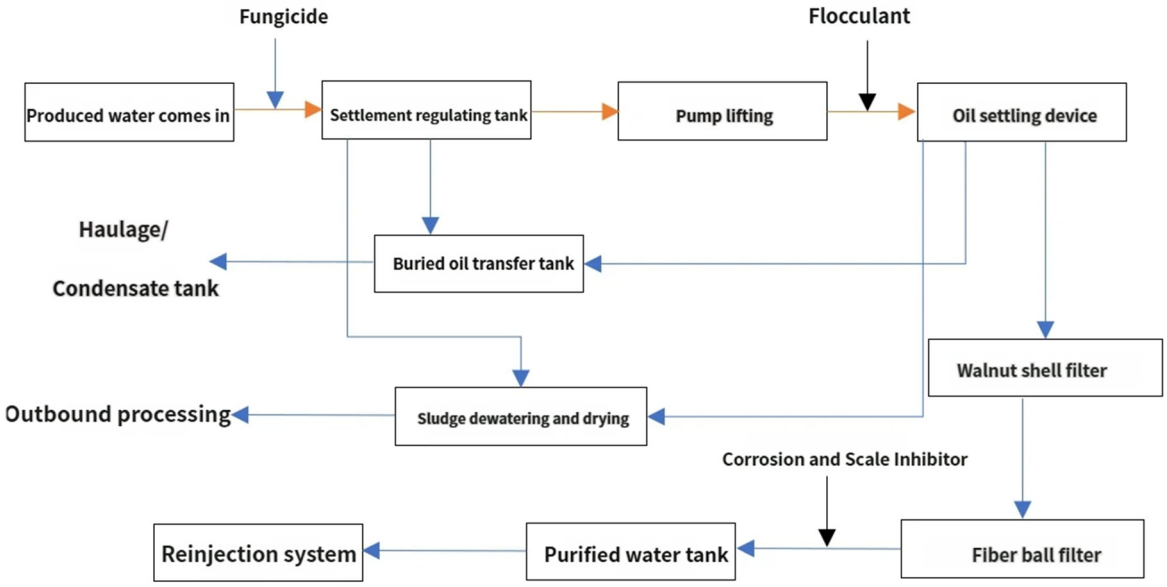

Fig 2. ' Secondary sedimentation oil removal + secondary filtration ' process flow chart

The typical non-alcohol produced water treatment process is as follows[22]:

(1) introducing the produced water without methanol into a closed discharge device, such as an unloading tank ; some impurities were removed by precipitation separation.

(2) the secondary precipitation entering the regulating water tank through the rotary pump;

(3) The static separation was carried out by adjusting the water tank, and the produced water without methanol was pumped into the integrated oil removal sedimentation tank by using the booster pump to realize the oil removal sedimentation.

(4) feeding the solid impurity and the solid impurity into the automatic filtering equipment, and removing the solid impurity and the oil pollution in the produced water without methanol;

(5) adding the bactericide to the pure water tank and adding it to the inlet part;

(6)The water in the pure water tank is tested, and the water is returned through the return water system.

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6. Corrosion Control and Treatment Optimization

The high corrosivity of alcohol-free produced water is a key issue affecting the stable operation of the treatment system and the safety of reuse. Targeted control measures need to be taken:

(1) Considering the corrosiveness of reclaimed water, it is necessary to optimize the proportion of reclaimed water in reclaimed water to ensure that the hardness of reclaimed water is within an acceptable index range.

(2) Because the sewage contains a large amount of iron salt, the iron content in the sewage is too high. Therefore, the recycled water in the sewage must be replaced by corrosion-resistant materials.

(3) If the K^+ method is used for enrichment, the calculation of K^+ concentration in water replenishment needs to be corrected, and the corrected enrichment factor is more accurate and effective.

After deep treatment, the produced water of the gas field is successfully applied to the circulating cooling water system, so as to achieve zero discharge of the produced water of the gas field, which plays a major role in environmental protection[23].

7. Summary

The treatment technology of alcohol-free produced water in gas fields has formed five major technical systems: physical, chemical, biological, physicochemical and composite treatment. Each technology has its own emphasis on principle, applicable scenarios and treatment effects. Among them, composite treatment, membrane separation technology and special technology for fracturing flowback fluid reuse have become research hotspots. In view of the water quality characteristics of ' four low ' gas fields such as Changqing gas field, the process of ' secondary sedimentation oil removal + secondary filtration ' has been applied in engineering, but it still needs to be improved in terms of efficient de-hardening, deep COD reduction and material anti-pollution.

In the future, the development direction of alcohol-free produced water treatment technology in gas fields includes : first, to develop high-efficiency, low-cost, anti-pollution membrane materials and composite membrane separation technology to improve the efficiency of deep treatment ; the second is to optimize the combination of composite treatment processes to achieve accurate water quality adaptation ; third, strengthen the combination of clean energy such as solar energy and evaporation technology to promote green and low-carbon treatment; the fourth is to deepen the research on the recycling technology of special wastewater such as fracturing flowback fluid and improve the recycling rate of water resources. Through the combination of technological innovation and engineering practice, it will further promote the development of alcohol-free produced water treatment in gas fields to the direction of high efficiency, resource utilization and greening, and provide technical support for green mining and ecological environment protection of gas fields.

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