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Research on the Development and Application of Oily Wastewater Biological Treatment Technology

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In this paper, composition and harm of oily sewage are analyzed, it was concluded that the research and application of oily wastewater biological treatment technology is of great significance. Based on the analysis of state and characteristics of oil pollutants in oily wastewater, in oily wastewater biological treatment technology, mainly microbe is used for the elimination of water surface oil and decomposition of petroleum hydrocarbons which are dissolved in water. The key points for biological treatment technology are selection of microbe for degradation oil pollutants, assessment of microbe's degrading ability, as well as determination of factors affecting efficiency of oil pollutants in degraded water. At present, commonly used oily sewage biological treatment technologies include activated sludge method, bio-film method, land treatment method and genetic engineering modification method, the author listed newest application of various new technologies, and analyzed on research trend and application prospect of the oily sewage biological treatment technology. Finally, the development of oily wastewater biological treatment technology and the trend of its applied research were summarized.

Keywords: Oily sewage; Biological treatment technology; Development; Application.

In human life and production activities, the water whose natures change have been polluted. The reason for natures change and use value is not available is that various pollutants are added. Oil pollutants are mainly from oily sewage and petroleum products[1]. Oily sewage refers to those which contain natural oil, petroleum products, tar and its fractions, edible vegetable oil and fat. Oily wastewater is one of key industrial pollution sources, when oil in water reaches 0.01 mg/L, fish will have a special odor and become inedible. When oil content is a little more, oil film is formed on the surface of the water. Air is separated from water, leading to hypoxia. Oil film is attached to fishes gill. Fishes face difficulties in breathing, and even die. Marine oil pollutants not only affect growth of marine living beings, reduce marine self-purification ability, but also affect coastal environment[2]. Oil, suspended solids, heavy metal and other substances cause great damage to growth of aquatic organisms, soil, human health and crop. Therefore, the treatment of oily wastewater is related to survival and development of enterprises.

Since 1980s, the traditional activated sludge process began to be adopted in oily wastewater treatment[3]. In recent years, with the development of oily wastewater treatment technology, biology technology is becoming more and more popular in oily wastewater treatment due to its low cost and high efficiency. So far, it can be applied in fields of printing and dyeing wastewater, brewing wastewater, paper making wastewater,

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petrochemical oil-containing sewage and oil wastewater[4]. The oily wastewater biological treatment technology does not have some shortcomings as physical and chemical methods, therefore, its research and application is becoming a hot topic in oily wastewater treatment in recent years[5]. Therefore, the development and application of oily wastewater biological treatment Technology have great prospect.

Characteristics of oily wastewater

Characteristics of oily wastewater as follows

State of oil pollutants in water

Oil pollutants in water generally exist in three states, namely, oil slick, emulsified oil and soluble oil.

Oil slick

Dispersed particles of oil is large, the size is greater than 100. The oil is the main part of oil pollutants in water and can be easily separated from water[6].

Emulsified oil

Dispersed particle of oil in water is very small, it exists in emulsive state, emulsified oil is relatively stable, and it is difficult to be separated from water[7].

Dissolved oil

A small portion of oil exists in dissolved state, it is called dissolved oil, the solubility is 5~15mg/L. Generally most of oil wastewater is treated through the process: oil separation, coagulation (DAF), filtration and treatment technology. The treatment of oily wastewater which is generated in well drilling mainly relies on coagulation process[8].

Characteristics of oily wastewater

Oil extraction wastewater

With longer oilfield exploitation period, moisture content of crude oil is more and more high, especially in later stage of oilfield development, at present, for most of Chinese oilfields, the comprehensive water content is up to 80%, and even 90% for some oilfields. Every year, 410 million tons oil sewage is produced, which has become the main source of oil sewage[9]. The oil sewage is mainly composed of oil slick, emulsified oil, colloidal dissolved substances and suspended solids.

Characteristics of refinery wastewater

In the production process of oil refining, oil gasÿcondensate separation water, washing and reaction water are main sources of oily sewage in refineries. In addition, oily wastewater which is produced in atmospheric and vacuum distillation and catalytic cracking equipments is also the source[10]. The main pollutants are composed of oil, sulfide and phenol, the oil content can be up to 1000 (mg/l) or higher, or as low as 10 mg/L. Drilling wastewater mainly comes from the loss of drilling fluid in drilling operation, drilling circulation system's leakage, drilling cutting and flushing water, at the same time, characteristics and pollution degree of drilling oily sewage are closely related to the drilling fluid system. According to relevant statistics, regardless of drilling depth and drilling fluid system, the main pollutants in refinery wastewater are suspended substance, chromium and phenol, oil drilling sewage contains a lot of COD whose treatment is difficult.

Application of oily wastewater biological treatment technology

Major technical methods of oily wastewater biological treatment have their own advantages and disadvantages, as well as applicable conditions, none of them is the best and most advanced, or can be applicable to any condition. The key to oily waster treatment is removing oil slick and emulsified oil[11]. The processing methods contain physical, chemical and biological methods. The use of physical methods mainly relies on oil adsorption ship and high performance oil adsorption material and other means, it involves large equipments and high expense[12]. Chemical method mainly involves the use of synthetic chemical dispersants, in fact, chemical pollutants are added into water, causing new pollution to some degree. Using biological method is mainly eliminating oil on water surface and dissolving petroleum hydrocarbons with the use of microorganism.

Microbial treatment technology is well favored by a large number of researchers because of its advantages of good treatment effects, stable system, easy operation, convenient management and low cost[13]. According to related experimental research, optimum operation parameters and affecting factors of anaerobic, aerobic biochemical reactor should be determined, on the basis of success in screening of oil-degraded microorganism. The purpose of oily wastewater biological treatment is to make organic pollutants in oil sewage convert into harmless substances through microbial metabolism[14]. In oily sewage, most of the organic substances and some inorganic substances can be used by microorganisms as nutrient source. Those substances are generally called substrate (or matrix). If the substrate in the oily sewage can be degraded by microorganisms, it means biological method of harmless treatment is feasible[15]. The biological treatment also applies to oilfield oily wastewater treatment. The biological treatment process can effectively remove solvable organics, and realize legal discharge of remaining oily sewage in oilfields. Oily wastes produced in different oilfields vary, but there are certain similarities[16]. Determining COD of sewage is the key to legal discharge through water quality study. The main biological treatment technologies are as follows:

Activated sludge method

Activated sludge method refers to a process in which activated sludge is evenly distributed in aeration tank, mucus outside microorganisms absorb organic pollutant with aeration or hydraulic agitation (anaerobic conditions),organic compounds begin metabolic conversion with biological enzyme, microorganisms realize sound growth and reproduction, at the same, harmless treatment of oily wastewater is achieved[17]. Since the advent of activated sludge treatment technology, certain improvements have been made in oxygen supply, feeding mode, treatment process and energy saving, efficiency and so on.

In aerobic activated sludge treatment of oily wastewater, the pH value is about 7. The reactor is a 1 L cylindrical cylinder. SBR (Sequencing Batch Reactor) is adopted, with built-in rigid filler. Valve control water decanter is used. In order to make sure activated sludge has full access to oily sewage, goldfish aerator is used for oxygenation. The temperature is controlled between 30 and 35°C, NH_4Cl, KH_2PO_4 nutrients are added according to certain proportion, the proportion of oily sewage in the system is gradually enhanced, change of COD in sewage sludge is measured after acclimation for 7 days. SBR process is an advanced biological treatment process in the world, in 1971, Erwin and others from Notre Dame University in Indiana State describe and defined the SBR reactor. With the large-scale application of oily water treatment automation technology, it has obtained great attention and federal financial aid in the United States, Canada, Australia, Japan, France, Germany and other countries, SBR reaction pool does not need double sedimentation tank and sludge return system, thus reducing occupied land and construction cost. SBR adopts automation technology, has the advantages of simple process, convenient operation and management, strong capacity of resisting impact load, as well as great treatment effect[18]. Its system operation and sludge cultivation and domestication are also relatively easy.

In order to trace change in microbes in the reactor, sampling biological phase microscopy is made in acclimation of activated sludge, microscopy results show that, in early stage of cultivation, flocculating constituent of activated sludge hasn't formed, sludge is sticky, microorganism are mostly bacteria and algae. In middle stage of cultivation, compendium, paramecium, flagellate, vertically and protozoa appear in the actor. In the late stage of acclimation, activated sludge becomes mature, biological flock structure is good, microorganisms in mixture have been concentrated in activated sludge flocks, few free microorganisms, water is clear, rotifers and other metazoans begin to appear in the actor, this indicates that activated sludge with good activity has appeared. It is also found that in water-inflow period and idle period (oily wastewater is in hypoxia), a lot of aerobic microorganisms die, facultative microorganisms reproduce quickly[19]. In the first few days of operation, due to loss of sludge, the SBR reactor discharge dirty water. The water becomes relatively clear 3 weeks later. According to comparison in input and output water, input water usually have black mud and is a little smelly, effluent COD concentration gradually decline, after observing mixed liquid, a lot of light brown aerobic sludge is found.

The main water quality indicators in the 3 weeks (average) are as follows: influent COD is 324 mg/L, effluent is COD 229 mg/L. The removal rate is only 29.3%. With the increase of activated sludge in the SBR pool, the processing ability of the system enhances. Although increased load leads to lower quality of output water within a short period of time, but the quality return to the normal quickly. Finally, when water flow reaches 2 L/d, the

SBR cycle is 6 h, water input is 1 h, aeration 3.5 h, precipitation 1 h, decanting 30 min. Average volume load is 0.65 kg COD/m3.d, stable operation for 3 weeks. Through experimental monitoring, biological observation and data analysis, we found that starting can be basically completed in twentieth days, and aerobic system can basically achieve desired treatment effect.

Microorganisms can survives and reproduce in oily sewage through cultivation and domestication, degrade and convert organics and generate stable CO_2, H_2O, NH_3, etc., finally reaching the purpose of eliminating pollution. Research results show that, after SBR treatment of oily wastewater, the effluent can meet national emission standards. The effluent COD average value is 72 mg/L, the removal rate is 77.7%. BOD declines from 90 mg/L to 14.4 mg/L, the removal rate is 84%. SS declines from 200 mg/L to 47 mg/L, the removal rate is 76.5%. Therefore, oily sewage treatment using biological technology is feasible in technology.

Conventional activated sludge process has the disadvantages of small bearing capacity against quality change and local impact, likely sludge bulking and poisoning in petrochemical wastewater treatment, in recent years, many domestic and foreign researchers have made a great deal of efforts to improvement the traditional method. The typical examples are anaerobic baffled reactor, plug-flow activated sludge system and ASBR. Anaerobic baffled reactor can adapt to shock of toxic organics, plug-flow activated sludge system has strong shock resistance, large treatment depth, few possibility of sludge expansion and low cost, and is suitable for high concentration organic wastewater. This is a practical technology of value of popularization, and has been successfully applied in beverage, printing and dyeing, chemical industry, synthetic wastewater. Good effects should also be realized in oily sewage field. ASBR (Anaerobic Sequencing Batch Reactor) is a new type of highly efficient anaerobic reactor which was proposed and developed by the professor Richard. Daunt from Lowe State University in 1990s. It is composed of one or several ASBR reactor. When running, oily water enter the reactor, through biochemical reaction with anaerobic sludge, a cycle of operation is finished and ends with discharge of supernatant. A complete cycle is divided into four stages, namely water inflow phase, reaction period, sedimentation period and drainage period. It has the advantages of good solid-liquid separation effect, clear output water, simple process, small occupation area, low construction cost, great resistance to load shock, great adaptability, large adaptation range (5~65), great activated sludge and strong capability of processing. According to water quantity and the way of discharge, ASBR method can realize achievement through single, serial or parallel connection[19]. Nowadays, the development and application of liquid sensor, electromagnetic valve control device and electronic computer allow the realization of automatic operation management of ASBR method and direct treatment of high concentration organic oil wastewater, such as brewing oily wastewater, papermaking oily wastewater, pharmaceutical oily wastewater, considering characteristics of oily sewage, its application in oily sewage field should be very promising.

Biological membrane method

When oily sewage flows through solid filler, bio-film forms on the filler, the bio-film breeds a large number of microorganisms which purify oily wastewater. The main structures are: biological filter, biological turn plate, biological contact oxidation bed and biological activated carbon fluidized bed. In recent years, in order to improve efficiency of bio-filter, a new type of plastic module with high porosity, high surface area and high performance of water distribution is adopted[20]. At the same time, microorganism in filter bed is selected and optimized. In addition, improvement and restructuring are made in process, such as the abolition of filter return system, using membrane mud A/O process. The biological membrane method has long been used in oily sewage treatment in city, now. Good effects have been realized in oily sewage treatment with the use of the method.

Only wastewater has poor biochemical performance. The adoption of anaerobic - aerobic contact oxidation technology in treatment of oily wastewater finally makes BOD, COD, SS and oil reach relevant emission standards. Practice has proved that: anaerobic - aerobic contact oxidation technology is feasible in oily wastewater treatment. In view of this, advanced process from other industries can be tried in oily wastewater treatment. Promotion can be carried out after gaining some experience. In anaerobic hydrolysis-high load trickling filter, the upper part of anaerobic hydrolysis tank is sludge bed, the low part is used for filling WY plastic packing, and aerobic biological filter is a new type of high load trickling filter without circulation. Filter bed also adopts WY plastic module packing, the packing's porosity is above 95%, the attached area is 130~150 m^2/m^3, and has excellent water distribution. The filler has corrosion resistance, pressure resistance, light weight, cheap price, easy to manufacture and install and other good technical and economic performance. The method adopts high-efficiency plastic module packing, it enhance processing efficiency, at the same time, largely reducing investment and operation cost, sludge treatment and disposal cost[21]. Earthworm ecology filter makes use of artificial ecological system established in the filter bed, various forms of pollutants are treated and converted in the most economical and reasonable way through coordinating functions of earthworms and other microorganisms, its treatment effects are better than other biological treatment process, it also has the advantages of zero sludge emission, low energy consumption and low electricity consumption. In recent years, the technology has been successfully tested in France, Chile and China.

Land treatment method

Soil and its microorganisms and plant roots are used to absorb and decompose pollutants in oily sewage. Soil and plant system has effective function of removing phosphorus and nitrogen, phenol, arsenic and other chemical substances. It includes stabilization pond, overland flow and wetland treatment.

Stabilization pond method refers to decomposition of organics in oily sewage through metabolic clearance effect of microorganism. Stabilization pond can be divided into aerobic pond, facultative pond, anaerobic pond and aerated pond. Stabilization pond's treatment effect is affected by light, temperature, season and other factors, generally it is high in summer, and low in winter.

Artificial wetland is one treatment method for oily sewage land, its purification mechanism is complex, and needs to be further studied, it is generally believed that there are combined effects of physical deposition, chemical reaction and biochemical reaction. Oily wastewater treatment system includes pretreatment and artificial wetland. The pretreatment is composed of grille, settling basin, sedimentation tank and anaerobic pond. It has the advantages of low investment, low operating cost, low repair requirement, making artificial wetland system have great prospect in oily sewage treatment.

The use of conventional processing techniques in oily wastewater treatment can not meet corresponding requirements, due to constraint of economic conditions. Application of new technology is not feasible, while wetland ecological system can solve the problem very well. After adsorption, filtration, settlement and biodegradation of wetland system, $CODC_r$, BOD_5, oil, sulfide, volatile phenol and other major indicators reduce sharply, realizing a high processing efficiency. Kelt Oil Company of Columbia applies reed root filtration system to remove contaminants in produced water, reed root absorb pollutants, the water can be used for irrigation of paddy field after the treatment[22]. The system is built in Casanare grassland area in which there are many rivers and high ground water level, the biological technology research and Development Company Transform offers certain assistance. The company announces that they system can remove 90% of phenol pollutants 1 year later, and show all effects 3 years later, and SS, COD, Phenolics can all decline to different degrees.

Genetic engineering modification method

With further study, foreign scientists have found many microorganisms with many special degrading capabilities, the required enzyme in the process of organic pollutants degradation is coded by degradation plasmid, instead of chromosome. In 1976, Friclo etc. lacerated the microbial strain which can degrade oil with the introduction of plasmid. Its objective is to use this microorganism to degrade oil in place where there is oil pollution. Although compared with natural degradation microorganisms, its degrading ability is not prominent, but the microbe has a patent to protect commercial value of the research. Friclo's work means that the genetic engineering method can solve environmental pollution issue in new research fields. Since then, A.M. Chakrabayty from the United States uses the genetic engineering method and introduces degradative plasmid of 4 pseudomonas bacteria into the same microbial strain, constructs a super microorganism with strong degradation ability, the super microbe has the functions of degrading alkanes, aromatic hydrocarbons, polycyclic aromatic hydrocarbons and others. The super microorganism has strong reproduction ability. Its efficiency of metabolic degradation of organic pollutants is much higher than natural microorganisms. In the 4 type of degradation plasmids found so far, white Pseudomonas microorganisms of the genus can degrade petroleum and petroleum products. On the basis of a lot of past studies on aerobic microbial degradation, further study on anaerobic microbial population and its characteristics are conducted. Gene engineering technology extracts, transforms constructs and transfers degradation plasmids in natural super microorganisms, cultivates super microorganisms with special functions, and studies acclimation of microorganisms in seawater environment to enhance their ability and efficiency of degrading of oil pollutants. The research and development of oily pollutants biological treatment device are also made.

Application of dominant mixed microorganisms has extensive degradation performance and better degradation efficiency, its requirements on temperature and pH value are also lower, and it is easy to maintain advantages in various environments. In view of this, we should cultivate corresponding dominant microbial population in the treatment of oilfield oily sewage to enhance the efficiency of biological treatment.

Mechanism for microbial degradation of oil pollutants: The mechanism for microbial degradation of oil pollutants (hydrocarbons) has been clarified elementarily. It starts with catalyzation by Oxygenase, in this process, molecular oxygen is necessary. Aliphatic hydrocarbons converts into fatty acid, then forms acetyl coenzyme A through beta oxidation, and enters the three carboxylic acid cycles. Aromatic hydrocarbon by hydroxylation, split ring forms various intermediate metabolites. The difficulty for microbial degradation of hydrocarbons varies, generally, linear alkanes (C_{10}~C_{26}) is the most easy to be degraded, alkanes is more easy than cycloalkane isoalkane or olefin, aromatic hydrocarbon with low molecular weight can be also easily degraded. The degradation of hydrocarbon with many branches or more aromatic ring compounds is relatively difficult. Numerous studies demonstrate that degradation rate is inversely proportional to chain length.

4 Key technical points of biological treatment method

Screening of oil pollutants degradation microorganisms

According to reports, after screening and testing, it has been found that there are over 200 microbes that can degrade petroleum hydrocarbons. Among the discovered microorganisms, types and quantity of Gram-negative microorganisms are more than those of Gram-positive microorganisms[23]. The common negative microbics are: Pseudomonas microorganisms of the genus, arc microorganism of the genus, immobile microorganism of the genus, yellow bar microorganism of the genus, aeromonas microorganism of the genus, colorless rod microorganism of the genus, alcaligenes rod microorganism of the genus. Gram-positive microorganisms are: Corynebacterium microorganism, microbial species, bacillus microorganism of the genus, Staphylococcus microorganism of the genus, Lactobacillus microorganism of the genus, milk bar microorganism of the genus.

Evaluation of microbial degradation capacity of oil pollutants in water (petroleum hydrocarbons)

Common methods of assessing microbial degradation ability at home and abroad are infrared spectrophotometry, fluorescence spectrophotometry, ultraviolet spectrophotometry and gravimetric method. Walker etc. used gas chromatography method for evaluation of microbial degradation capacity of oil pollutants. There are also some reports about 14C tracer method, liquid chromatography/mass spectrometry, fluorescence detector chromatograph[24]. Generally speaking, oil pollutants (petroleum hydrocarbons) can be totally biodegradable with microorganism, but residual parts are inert material of low toxicity which causes not damage to the environment.

Therefore, the ration between total

hydrocarbons and hydrocarbon which is difficult to degrade, or change in total hydrocarbons can be used as basis for assessing the ability and effect[25].

Environmental factors influencing microbial degradation of oil pollutants in water

Besides ability of microbial degradation of oil pollutants, environmental factors affecting degradation effect and rate are dissolved oxygen, nutrient, pH value, temperature and pollutant toxicity[26]. Dissolved oxygen: the process of microbial degradation of oil pollutants is mostly aerobic process, certain oxygen concentration is important to maintain the activity of microorganism. Lack of oxygen can affect normal metabolism of microorganism. Generally, dissolved oxygen concentration of 2 ml/L or so is appropriate. Nutrient: microbial metabolism needs certain nutrient, proper amount of nitrogen, phosphorus and other trace elements offer nutrients for the growth of microorganisms[27]. PH value: in microbial aerobic process, pH value is generally 6.5~9.0, when PH value is less than 6.5, ekaryotic microorganism compete with microorganism, when pH value is lower than 4.5, ekaryotic microorganism is completely in dominant position. When pH value is more than 9, rate of metabolism of microorganism is inhibited. Water temperature: water temperature is an important factor affecting microbial metabolic activity. Relevant studies show that in microbial aerobic biochemical process, the best water temperature is 20~30°C. When it is above 35°C or below 10°C, degradation effect will be worse[28]. Toxicity of pollutant: biological toxicity of organic pollutant is closely related to biodegradability of oil pollutants, larger biological toxicity of organic pollutant means lower biodegradability. Generally, BOD5/COD ratio is used to assess biodegradability of pollutants in water.

CONCLUSIONS

Through the above research on development and application of oily wastewater biological treatment technology, research trends on oily wastewater biological treatment technology can summarized as follows:

(1) In oil pollutants biological treatment technology, the study at early stage is about biological treatment of organic pollutants. Since the nineteen seventies, the US leads the study on elimination of oil pollutants in water with microorganism, later many countries carried out corresponding studies. At the initial stage, selection of microorganism which is capable of oxidative degradation of oil pollutants (petroleum hydrocarbons), measure of degrading ability and environmental conditions for microbial degradation of oil pollutants are explored. Then research on the use of genetically modified microorganism is conducted.

(2) Research tendency for biological treatment technology of oil pollutants in water is continued screening of highly effective natural microbial species which can decompose hydrocarbon organics and microbial strain substrate. In microbial strain immobilization technology, development and research on cheap immobilization carrier, composite immobilization, mixed immobilization of microorganism algae, highly efficient immobilization reactor will be further carried out.

(3) Super microorganism with a variety of degradation functions or stronger degradation functions can be obtained through genetic engineering modification, the development of oil pollutants biological treatment device indicates that people can construct and cultivate super microorganism which can degrade oil pollutants biologically and apply it into practice, which offer a new research field for technical methods of environmental protection. At the same time, we should pay attention to clean production, process industrial oily sewage from the source so as to reduce processing pressure at the end. With shortage of water resources and increasingly serious pollution, oily sewage treatment technique and its comprehensive utilization will be an important research topic that needs urgent exploration and practice.

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