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Inorganic impurity removal from waste oil and wash-down water by *Acinetobacter johnsonii*

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HIGHLIGHTS

- Process was developed to cleanse waste oil using *Acinetobacter johnsonii*.
- Inorganic impurity in waste oil was reduced below 0.5% after biotreatment.
- Turbidity and sulfides in wash-down water were controlled within 100 NTU and 1 mg/L.
- Inorganic impurity in oil phase was removed by wettability reversal.
- Inorganic impurity in water phase was removed by bioflocculation.

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ABSTRACT

The removal of the abundant inorganic impurities in waste oil has been one of the most significant issues in waste oil reclamation. *Acinetobacter johnsonii* isolated from waste oil in aerobic process was employed to remove the inorganic impurities in waste oil and wash-down water. The biological process was developed through the primary mechanism research on the impurity removal and the optimization of the various parameters, such as inoculum type, inoculum volume and disposal temperature and time. The results showed that waste oil and wash-down water were effectively cleansed under the optimized conditions, with inorganic impurity and turbidity below 0.5% and 100 NTU from the initial values of 2% and 300 NTU, respectively. Sulfide, the main hazardous matter during waste oil reclamation, was also reduced within 1 mg/L. After the biotreatment, the oil–water interface was clear in favor of its separation to benefit the smooth reclamation of waste oil and wash-down water.

1. Introduction

Large amount of waste oil, of high recovery value, in the form of dropped crude oil, oil loam and wash-down water, is generated in petroleum production [1–3], and usually reclaimed by gravitationally settling inorganic impurities at 353 K (in winter) or 363 K (in summer) [4]. However, the complex inorganic impurities of high concentration could not be effectively removed, so the residual impurities, especially pyrite, widely distribute over the oil–water interface, resulting in serious emulsification [5]. In order to avoid the industrial accidents, such as the damage of the plate electrodes and the havoc on the combined-stations, many researches focus on surfactants [6]. To some degree, such method is effective to solve these troubles through demulsification. However, the effect is limited because almost no surfactant can effectively demulsify waste oil due to the different quality between waste oil and produced oil. Also, the difficulties of waste oil reclamation do not only lie in oil–water emulsification. It is almost not efficient for the impurity removal, which would bring much more troubles to the follow-up disposal of oil and water. Thus this method is far from satisfaction in practice. Besides, the centrifugal process, the membrane filtration and the skid-mounted unit have also been tried to reclaim waste oil [7,8]. These methods have not been put into practice for their poor efficiency, limited disposal capacity or high cost.

Biotreatment of inorganic impurities has attracted much attention and provided promising methods of high efficiency and low cost [9–11]. It has been reported that many microorganisms could produce bioflocculants to flocculate toxic organic and inorganic compounds such as diatomite, chalcopyrite, sphalerite, galena and quartz from aqueous phase [9–13]. Suspended solids could also be deposited effectively and selectively by bioflocculation [9,10,13].

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Moreover, the wettability reversal of inorganic impurities in waste oil by microorganisms and some metabolites was in favor of their transport into water phase, during which waste oil could be utilized as carbon source for cell growth [14–16]. Thus, the impurity removal in oil phase through wettability reversal and in water phase through flocculation by microorganisms may be a novel, effective and practical method, concerning which little information was available [17].

A. johnsonii, one of the most popular microorganisms capable of utilizing various alkenes and aromatic pollutants to produce bioactive molecules as well as coaggregating with other free-floating bacteria or microflocs, showed high potential application in the waste oil recovery [18–25]. And this bacterium has been isolated from waste oil in flotation machine with aerobic environment and identified by Institute of Microbiology Chinese Academy of Sciences previously.

This work was to study the inorganic impurity removal in waste oil and wash-down water by *A. johnsonii*. Operating parameters were optimized and the primary cleansing mechanism in oil phase was also investigated.

2. Materials and methods

2.1. Cultivation conditions

The synthetic medium was composed of (g/L): 15 glucose, 1 yeast extract, 0.4 K_2HPO_4, 0.2 KH_2PO_4, 0.1 NaCl, 0.1 $MgSO_4$, 0.01 $MnSO_4{\cdot}H_2O$, 0.04 $FeCl_3{\cdot}6H_2O$, 0.01 $Na_2MoO_4{\cdot}2H_2O$, 0.4 $(NH_4)_2SO_4$. The initial pH of all the media was adjusted to 7.2 before sterilization which was performed at 121 °C for 20 min. Subculture of *A. johnsonii* was harvested as inoculum at OD_{600} = 1.2 after two continuous transfer cultivations in the shaking flasks at 200 rpm and 310 K [26].

2.2. The waste oil cleansing tests

Waste oil and wash-down water were collected from waste oil recovery pool in the 1st Oil Extraction Factory of Daqing Oilfield. Experiments were conducted in a thermostat water bath with a gas flowmeter. About 70 g waste oil and 140 mL wash-down water were combined into 250 mL wide-necked bottles with rubber stopper, and then the biotreatment was performed with the subculture of *A. johnsonii* as inoculum and the aerating velocity of 2 L/min at 310 K. Additionally, aeration outlet was conclusively placed at the oil–water interface to ensure the intensive mixing of the oil and water phases. After the overnight biotreatment, it was heated up to 323 K and kept constant as soon as the aeration stopped. Controls were also conducted at the same conditions without inoculation.

Inorganic impurity, sulfide and turbidity were determined for samples and controls. Various factors on the removal of inorganic impurities, including inoculum type, inoculum volume, biotreatment time and temperature and sedimentation time were investigated based on the basic operating procedures above.

2.3. Analytical methods

Cell density was monitored spectrophotometrically by measuring the optical density at 600 nm [27]. Inorganic impurity in oil was determined by the standard Petroleum Products and Additives-Determination of Mechanical Impurities-Gravimetric Method (GB/T 511-2010). In tests, waste oil and wash-down water were sampled from Recovery Pool in different period. The removal rate of impurity in oil phase was calculated as follows:

$$\text{removal rate}(\%) = \frac{\text{control} - \text{disposed}}{\text{undisposed}} \times 100$$

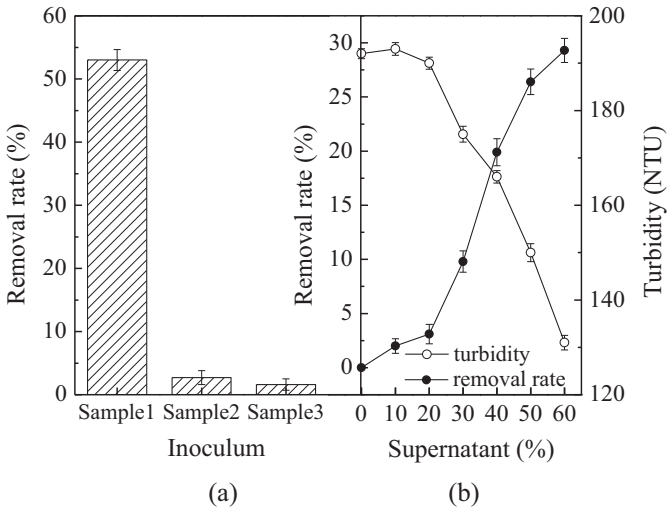

Fig. 1. Removal potentials of inorganic impurity using different inoculum and inoculum volume of cell free supernatants. Sample 1 is subculture, Sample 2 is supernatant, and Sample 3 is resting cells.

where "disposed" was the inorganic impurity concentration in waste oil after biotreatment, and "undisposed" was the initial impurity concentration of samples.

Sulfide was monitored by the enterprise standard Non-Water Determination of Sulfide-Methylene Blue Spectrophotometric Method (Q/SY DQ1127-2006). Turbidity in water was measured using High Accuracy Attributes Data Nephelometer (model HI 98703, Hanna, Italy).

2.4. Statistics

All experiments including the controls were repeated three to five times. The data shown in the corresponding figures and tables were mean values and the error bars presented the standard deviation.

3. Results and discussion

3.1. Effects of different inocula

The effects of three different inocula on the impurity removal were investigated. The three inocula were the subculture containing cells and intermediate metabolites (10 mL), the cell free supernatants only containing intermediate metabolites (10 mL) after the centrifugation of the subculture at 5000 rpm for 10 min, and the cells washed twice with 0.1 mol/L sodium phosphate buffer (pH = 7.2) and resuspended with 5 mL sterile saline water.

As shown in Fig. 1(a), compared with the other two samples, Sample 1 demonstrated much higher removal rate of about 53%, in which the subculture of 5% was inoculated and then some natural substrates as start carbon and energy source promoted cell growth to overcome the toxicity of organic compounds. Furthermore, the transport of impurities from oil phase into water phase occurred by the reversal of metabolite excreted by cells consuming a little oil.

In Sample 2, low removal rate was observed, but it has been reported that some intermediate metabolites benefited the impurity removal [9,10,12,13]. As shown in Fig. 1(b), the more cell free supernatants were inoculated, the higher removal rate was observed, but the maximum removal rate was still below 30% when the inoculum volume reached to 60%. And the turbidity of water phase decreased from 190 to 130 NTU as more supernatants were inoculated. It illustrated that intermediate metabolite in the