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Removal of oil from oil-water emulsion by hybrid coagulation/sand filter as pre-treatment

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ABSTRACT

The removal of oil from water is of increasing importance in many industries, such as oil and gas, petrochemical and food industries, because of the large volumes of oily wastewater they produce. The aim of this work is to evaluate and compare the performance of ferrous and aluminium sulfate coagulations and to compare between sand filter, coagulation and a hybrid coagulation/sand filter process when used as pre-treatment options. When the concentration of oil was low in the oil-water emulsion, the treatment by sand filter alone was adequate (oil ≤ 50 mg/L). On the other hand, when the oil concentration was greater than 50 mg/L, advanced treatments such as coagulation are required as post-processes to reach good water quality. Aluminium sulfate was observed to be more efficient (about 5% to 7% higher than the use of ferrous sulfate) and less costly than ferrous sulfate to remove oil from oil-water emulsion using coagulation. The optimal technology for pre-treatment to remove oil from oil-water emulsion was found to be a coagulant dosage of aluminium sulfate combined with sand filter at pH 8 during various concentrations as hybrid. Optimal conditions were found to occur at mixing duration and speed of 120 s and 250 rpm respectively.

1. Introduction

The use of pre-treatment separation for the removal of oil from water is becoming very important in many various industries such as in the oil and gas industry, petrochemical industries and some food industries. These industries produce large amounts of oil-water emulsion, such as oil well produced water and the hydrocarbon concentration in oily wastewater from various industries usually ranges between 50-1,000 mg/L [1–3]. Hence, this oil-water emulsion should be treated before disposal or reuse, especially in water-stressed areas. There are several methods available for treatment of oil-water emulsions including coagulation/flocculation by air floatation, ultrasonic separation, and chemical de-emulsification. These methods are considered primary methods of water treatment [4,5]. Another advanced method for separating oil from oil water emulsions are pressure driven membrane separation processes. However, a major problem for such processes is fouling [6]. For all but low concentration oil-water mixtures pre-treatment is needed to remove the bulk of the oil, with membranes used to remove residual oil to produce a high-quality product.

To date, there have been many studies about processes for treatment of oil-water emulsions. For instance, the roles of aluminium and ferric sulfates as coagulation agents for oil–water emulsions were investigated in terms of oil removal by Al Shamrani et al. [7]. Suzuki & Maruyama [8] tested coagulation by poly-aluminium chloride whilst adding casein before the foam separation stage, noting a dramatic improvement in oil removal.

However, these methods have some drawbacks, such as high cost, especially coagulation/flocculation by air floatation, because the flotation process requires energy, and generation of secondary pollutants, especially by chemical de-emulsification processes [9,10]. This work examines the treatment of oil-water emulsions for the reuse of water by coagulation with a sand filter as a pre-treatment step, with vegetable oil used to form the oil-water emulsions.

The objective of this study was to reduce fouling when membrane techniques are used after the pre-treatment process because membrane fouling is the major problem during membrane separation processes [6,11]. When the concentration of oil is low in the oil-water emulsion, a pre-treatment step alone is enough for treatment [5,12]. In this work the concentration of oil that needed the pre-treatment filtration process was oil < 500 mg/L. When the oil concentration is greater than 500 mg/L, advanced treatments, such as membrane separation, are performed as post-processes to reach good water quality, based on the United States Environmental Protection Agency (USEPA) standards.

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2. Materials and methods

2.1. Materials

Aluminium sulfate ($Al_2(SO_4)_3.16H_2O$) was purchased from Fisher Scientific UK Ltd. The purity and molecular weight of this product was > 97% and 630.39 g/mol, respectively. Ferrous sulfate (FeSO4 · 7H2O) was purchased from Sigma-Aldrich Company Ltd. The purity and molecular weight of this product were ≥99% % and 278.01 g/mol, respectively. The chemicals used for pH control were sodium hydroxide (0.1M) (Fisher Scientific, UK) and hydrochloric acid (0.1M) (Fisher Scientific, UK). The pH meter was used to control solution pH at different solution chemistry and it was purchased from Fisher Scientific UK Ltd. Metal salts used were copper (II) nitrate, cadmium nitrate, iron (III) nitrate, nickel (II) nitrate, and zinc nitrate (Sigma-Aldrich Ltd, Dorset, UK). Sodium Chloride (NaCl) was purchased from Fisher Scientific-UK, with purity higher than 99.5%. Eight concentrations of commercial vegetable oil were used to make oil/water emulsions at 0.546, 50, 200, 500, 650, 800 and 1000 ppm concentrations.

2.2. Jar test procedure

Oil-water emulsions were prepared by mixing commercial vegetable oil and deionised water at 1500 rpm in a magnetic stirrer (Fisher Scientific, UK) for 10 min. After adjusting the pH to the appropriate value, the oil-water emulsion was mixed with the coagulant in a standard jar-test apparatus (Bibby-Stuart Flocculator SW6) for 120 s at 250 rpm as rapid mixing, followed by slow mixing for 18–20 min at 30 rpm. After 20 min of settling, the floc formed and the sample was taken from approximately 3 cm below the liquid surface. Oil concentration was determined by a Shimadzu Total Organic Carbon (TOC) analyser (Model TOC-L, Shimadzu). This procedure was applied when the coagulation process was to be used without sand filter and when using the sand filter after coagulation. Samples were also taken after purification using the sand filter. In addition, the zeta potentials of oil droplets were measured in prepared emulsion using a Zeta-Sizer 3000 HS (Malvern Instruments, UK).

2.3. Sand filtration study

The sand column used in this study had dimensions (70 mm Inside Diameter (ID) and 400 mm length) (Fig. 1). The sand filter consisted of three layers: sand layer (depth approx. 8 cm); the second layer consisted of gravel (diameter ≈ 4 mm and layer depth ≈ 20 mm); and the third layer was composed of glass particles (diameter ≈ 18 mm, total number of 25, and depth ≈ 40 mm). Stainless steel mesh (Aperture 0.039 mm, The Mesh Company (Warrington) Ltd, UK) was placed at the bottom of the sand column and between the layers (Fig. 2). The ratio between the depth of the sand and glass gravels was (2:1) that ratio based on the depth.

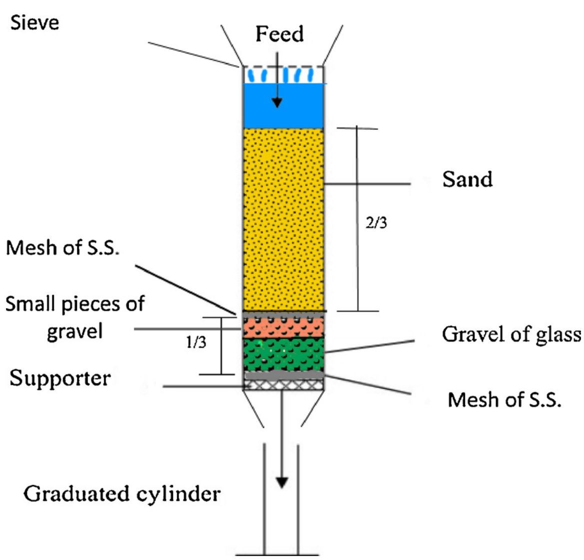

Fig. 1. Schematic diagram for sand filter.

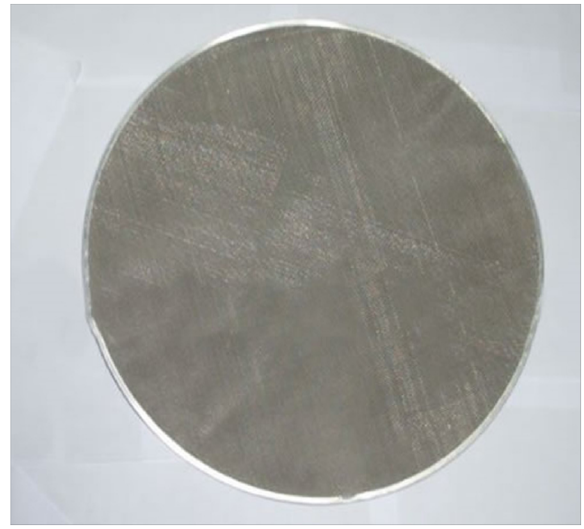

Fig. 2. Stainless steel mesh.

At this condition, the filtration rate for clean water was estimated to be around 0.94 m^3/m^2.hr with the driving force for sand filtration supplied by gravity. The solution after coagulation was fed slowly (about 50 ml in every 5 min). Normally after each experiment, the sand required cleaning after each oil concentration experiment. To clean the sand 200 ml distilled water, followed by 100 ml 0.1 M NaOH, then 200 ml distilled water were rinsed through the sand filter column.

Sand filtration will remove both oil droplets and oil flocs (after coagulation process) by capturing suspended particles on to the surface of sand grains as they pass through the sand filter. Therefore, when removing oil droplets from emulsion by using sand filter there are three mechanism steps [13,14]. The first step, is the transport of the particles onto the sand grains and is a physical process. The second step is the attachment of particles to the grain surface (collector) and is mediated by a combination of the electrostatic attraction force, mass attachment force and adhesion force. These forces collectively hold oil droplets on the surface of the sand grain. The final step is the purification process where undesirable materials are collected around the sand grains.

3. Result and discussions

3.1. Pre-treatment by sand filter

The effect of the sand filter pre-treatment on oil removal between initial concentration of oil and final oil concentration was investigated. The initial concentration of oil was 0.546 (concentration of oil from Ras Tanura in Arabian Gulf) [15,16], 50, 200, 350 (similar to the concentration of oil in the oil fields in Saudi Arabia) [17], 500, 650, 800 and 1000 ppm. These concentrations were used to evaluate the sand filter oil absorption.

Table 1 and Fig. 3 present the results obtained from the pre-treatment process by using the sand filter. As the initial oil concentration (C_0 mg/L) was increased the concentration of the oil after the sand filter treatment increased and the oil removal decreased.

The final oil concentrations after using the sand filter at initial oil concentrations of 0.546 and 50 mg/L were about 0.003 and 1.86 mg/L respectively. According to the United States Environmental Protection