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Designing high-caliber nonwoven filter mats for coalescence filtration of oil/water emulsions

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ABSTRACT

Oily wastewater is a major problem in industries that can cause severe environmental pollution without proper treatment. In this paper, an easily fabricated, robust and durable nonwoven fibrous filter mat was prepared for coalescence filtration of four kinds of oil-in-water emulsions, namely hexadecane/water, octane/water, soybean oil/water and engine oil/water. An aromatic thermoplastic polyurethane resin (TPU) was selected to bind the fibers together for structure integrity of the mat. Special roughening treatment to the mat was conducted to deliver the required wettability for effective coalescence separation of these emulsions. The filter mat thus prepared reached a tensile strength of 2.99 MPa, 20 times stronger than that of the pristine mat. The mat surface was made both hydrophilic and superoleophilic, allowing it to have separation efficiency as high as 99.61% in a single pass flow. Moreover, even pre-saturated with oil, the mat remained effective in separation and permitted high flow, proving its high caliber for potential industrial applications.

1. Introduction

Oily wastewaters are produced in numerous industrial activities such as food processing, petroleum production, and mechanical and metallurgical industries [1]. High quantities of oily wastewater are not allowed to discharge to land or ordinary sewage systems because they pollute surface and underground water [2]. Oil in wastewater can be found in various forms and is generally divided into three categories, free, emulsified and soluble oil. Soluble oil is in very small amount, which typically requires deep treatment with adsorbents, nanofiltration (NF) or reverse osmosis (RO), and even advanced oxidation technology. As far as oily water is concerned, people are more interested in removing free and emulsified oil. Free oil is easier to treat by physical techniques such as gravity separation and skimming [3]. In contrast, emulsified oil is more difficult to separate due to its high stability in water [4]. Many removal technologies have been applied, but they all have disadvantages such as secondary pollutant generation in chemical treatment [5], wide space requirement and temperature and pH sensitivity in biological treatment [6], as well as membrane fouling in membrane filtration [7–9]. Among the physical technologies, coalescence seems to be an attractive method due to its feasibility, low operating cost and effectiveness [10–12].

Coalescence causes small oil droplets to collide and adhere to other droplets on the coalescing material. Subsequently larger droplets form and then migrate through the depth of the material. Finally the matured droplets are released and separated downstream of the material by buoyancy force due to density difference of oil and water. This happens mainly because the affinity of oil and water to the coalescing material at work is different [13]. Based on Stokes Law equation, oil droplet size has a significant impact on droplet rising velocity that is proportional to the square of droplet diameter. Therefore, it is desired to increase the droplet size as large as possible by coalescence to effectively separate oil-in-water emulsions [2].

Traditional coalescing material is either granular or fibrous, fabricated as a filtration bed [14,15]. The length/thickness of the bed plays a great role in separation efficiency. Many researchers hold the opinion that the efficiency increases with increasing bed height for it directly extends coalescing time. But Mathavan and Viraraghavan [16] found that the coalescence efficiency in a peat bed decreased with an increase in the depth of the bed. Sokolovic [17] established the minimal bed length for steady-state coalescence. Kang [2] discovered that coalescence efficiency did not increase when the bed exceeded a certain height. In addition, a long bed requires a higher pumping cost. Therefore, it is

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always better to minimize the length of the coalescence material, and nonwoven filter media and membranes seem to be better choices. For example, Hlavacek [18] used a polypropylene hollow fiber microfiltration membrane as a coalescer to break down an industrial emulsion from aluminum industry. The physical mechanism of coalescence using a membrane was observed by Nitsch [19] for oil-in-water emulsion separation. The events occurring in series across the membrane thickness included wetting, coalescence and finally formation of a W/O-domain emulsion at the membrane outlet. Shin and Chase [20] investigated the coalescence performance of a nonwoven glass fiber filter medium augmented with polymeric nanofibers and found that higher wettability and lower drag force were better quality factors in coalescence filtration.

Recent oil/water separation studies focused more on the surface wetting behavior. Many interesting methods to gain superwettability were innovated [21–23]. For example, Jin and Jiang [24] developed superhydrophobic and superoleophilic PVDF membranes for effective separation of water-in-oil emulsions. Liu [25] researched on poly (sulfobetaine methacrylate)- grafted glass fiber filter medium that was superoleophobic underwater for oil-in-water emulsion separation. The methods mentioned above, among many others [26–28], rely on sieving instead of coalescence as the main filtration mechanism, to separate either water droplets from oil or oil droplets from water. Yang et al. [29] constructed a hierarchically roughened surface structure on stainless steel fiber felt first, and then chemically treated the surface for oil-in-water emulsion separation. They found that the sturdy metal felt thus prepared could remove fine oil droplets well through the non-sieving coalescence mechanism. However, stainless steel felt may not resist corrosion in water in long term, the layer-by-layer (LBL) self-assembly method applied in their work is still not simple enough although controllable, so robust materials with the flexibility of easy fabrication are much desired.

In this paper, a durable fibrous filter mat was fabricated by simple wet-laying process for the treatment of oil-in-water emulsions. Several properties of the mat such as components, porosity, surface morphology, tensile strength, wettability, and absorption capacity were carefully designed and examined, and the coalescence separation efficiency was subsequently determined. By using a fiber binder, the tensile strength of the as-prepared filter mat was increased by 20 times compared with the pristine mat. A rough surface was constructed by forming polymeric nanoparticles on the mat via emulsification–solvent evaporation method. Uniform pore size distribution and good wettability for both water and oil of the mat contributed to the successful coalescence separation of oil-in-water emulsions. Different types of oil were used to test the separation efficiency of the freshly prepared mats, and the mats pre-saturated with oil as well to cancel out the absorption effect on separation. It was discovered that the pre-saturated mats still maintained good separation efficiencies, indicating their consistent performance even under severe challenging conditions.

2. Experimental

2.1. Materials

The fibers used to prepare the filter mat are glass wool (diameter 0.6 μm, Shenyang Dongxiang Glass Fiber Co., Ltd.), glass fiber (diameter 7 μm, length 6 mm, Taishan Fiberglass Inc.), cellulose fiber (diameter 11 μm, length 6 mm, Lenzing Fibers (Hong Kong) Ltd.) and polypropylene(PP) fiber (diameter 10 μm, length 6 mm, Changsha Bosaite Construction Materials Co., Ltd.). Meltblown polyester(PET) nonwoven medium was obtained from Shandong Laifen Nonwoven Co., Ltd. Isopropyl alcohol (IPA) was purchased from Xilong Chemical Co., Ltd. Cyclohexanone, octane, and cetyl trimethyl ammonium bromide (CTAB) were purchased from Sinopharm Chemical Reagent Co., Ltd. Hexadecane was purchased from Haltermann GmbH. Soybean oil and engine oil were purchased from local stores. Aromatic type of thermoplastic polyurethane resin (TPU) was obtained from Taiwan Sheen Soon Co., Ltd.

2.2. Preparation of filter mat

The filter mats were made with total mass of 6 g of glass wool, glass fiber and cellulose fiber based on a mass ratio of 2:1:1 using a conventional wet-laying process, which involves fiber dispersing, dewatering and drying. All the fibers were washed with water before use; they were then dispersed in 500 mL water and mixed uniformly with an electric extractor for 5 min. The fiber dispersion was then transferred to a handsheet former containing 10 L water, stirred up and down for remixing with a homogenizer for 10 min, and finally filtered through a fine copper screen by simply draining water from the flat bottom of the former. The partially dewatered filter mat collected on a discrete porous support on the screen was removed, put on a plate dryer and dried at 80 °C for 2 h. The dried filter mat was cut into several pieces with a diameter of 25 mm, and was designated as GGC (glass–glass-cellulose) for further use.

2.3. Preparation of GGC-TPU-R

TPU was used as a binder for the filter mat. It was cleaned with DI water and dried before use. A binder solution of 10% TPU was made by completely dissolving TPU in cyclohexanone with magnetic stirring. The GGC was then fully immersed in the binder solution for 2 s, lifted up to drain the residual solvent, and then ventilated to dry to completely remove the solvent. This mat was designated as GGC-TPU. To make roughened mat, binder impregnated GGC was partially dried to avoid complete binder solidification, and then put into 30 mL IPA mixed with 0.02 g CTAB. The mixture was ultrasonicated for 30 min at room temperature. The mat was then picked up and rinsed with pure IPA three times and ventilation-dried. Mat processed this way was designated as GGC-TPU-R, i.e. roughened GGC-TPU.

2.4. Oil-in-water emulsion separation

The oil-in-water emulsion was prepared by mixing oil and water using a homogenizer (Shinetek Instruments Co., Ltd.) operating at 15,000 rpm for 10 min. The initial oil concentration was 1000 mg/L. The droplet size of the oil in the emulsion was in the range of 1–18 μm as observed with an optical microscope (6XB-PC) (Fig. S1) made by Shanghai Optical Instrument Factory. The average emulsion size was about 2–3 μm. The emulsion stayed stable for almost 2 h as can also be seen in the supplementary document (Fig. S1). To conduct filtration experiments, the circular filter mat (diameter 25 mm; effective area 283.5 mm^2; thickness and basis weight are showed in Table 1) was mounted inside a syringe filter holder with guaranteed seals. Then 50 mL oil-in-water emulsion was drawn with the syringe which was attached to the filter holder immediately. Then the emulsion was pushed through the mat by hand in 60 s at a constant speed. A container was used to

Table 1
Physical properties of GGC, GGC-TPU and GGC-TPU-R.

	GGC	GGC-TPU	GGC-TPU-R
Dry tensile strength (MPa)	0.15	6.94	2.99
Wet tensile strength (MPa)	Not measurable	1.58	0.40
Basis weight (g m^{-2})	175 ± 2	395 ± 5	275 ± 3
Thickness (μm)	1000	1100	1100