Separation and Purification Technology 172 (2017) 59–67

Contents lists available at ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Restructuring the surface of polyurethane resin enforced filter media to separate surfactant stabilized oil-in-water emulsions via coalescence

Dan Hu [a,b], Lei Li [a,b], Yanxiang Li [a], Chuanfang Yang [a,*]

[a] Key Laboratory of Green Process & Engineering, State Key Laboratory of Biochemical Engineering, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, PR China

[b] University of Chinese Academy of Sciences, Beijing 100049, PR China

ARTICLE INFO

Article history:
Received 26 February 2016
Received in revised form 23 June 2016
Accepted 29 July 2016
Available online 2 August 2016

Keywords:
Resin emulsification
Wettability
Coalescence
Surfactant
Oil/water emulsions

ABSTRACT

Reported here is a facile resin emulsification method of treating polyurethane enforced filter media by controlling the initial level of solvent volatilization for resin solidification. The treated media are endowed with varying degrees of lipophilicity as determined by their lipophilic to hydrophilic (L/H) values. An optimized treatment condition was pinned for coalescence separation of surfactant-stabilized oil-in-water emulsions using a surfactant free emulsion as the bench mark. The starting separation efficiency was found to depend on the type of surfactant involved in the order of non-ionic surfactant Tween 80 > cationic surfactant cetyl trimethyl ammonium bromide (CTAB) > anionic surfactant sodium dodecyl benzene sulfonate (SDBS). Quartz crystal microbalance (QCM) experiments showed that Tween 80 had the largest adsorption on the polyurethane coating, while least adsorption occurred to SDBS, and mediocre adsorption to CTAB. However, Tween 80 resulted in the lowest time-weighted averaged efficiency as compared to the other surfactants that barely caused any efficiency changes with filtration time. To make the resin emulsification treatment more environmental friendly, more polar solvents were tested, and ethanol was identified as a good alternative to isopropanol.

1. Introduction

Oil exploration generates numerous oily wastewaters, direct discharge of these waters into the environment is a serious concern [1]. Other oily wastewaters produced by oil spills and organic contamination involved in different industrial activities are also environmental hazards threatening the ecological system [2,3]. Separation of oils/organics from these waters is therefore necessary and oftentimes mandated, and economically viable treatment technologies are highly demanded. The conventional oil-in-water separation technologies, such as gravity separation, centrifugation, flotation, filtration and electrochemical flocculation either involve great external energy consumption to drive the separation, or are less effective than required, and sometimes generate secondary pollutants [4]. In recent years, the rapid development of super wetting materials, generally in the form of two-dimensional membranes [5], has triggered enormous interests. Jiang and coworkers [6] revealed that a hierarchical structure combined with proper chemical composition could result in superhydrophobicity, a mimic lotus effect that promotes self-cleaning. This finding inspired them to conduct the very early work to develop a superwetting material to separate oil-water mixtures [7]. At present, materials like this are further divided into three types: superhydrophobic-superoleophilic, superhydrophilic-underwater superoleophobic, and superhydrophilic-superoleophobic. They can be in different forms such as metallic mesh and felt [2,8], polymeric materials [9], non-woven and woven fabrics [10,11], sponges and foams [4,12], carbon derived materials [3], and various particles and powders [13,14]. Moreover, smart materials with switchable wettability were also developed that attracted increasing attention for their significance [15,16]. The controllable surface wettability can be achieved by applying an external stimulus such as light illumination [17], temperature [18], electrical potential [19], and pH [2,20]. However, in many cases, these specially designed surfaces/materials were applied toward the separation of regular oil-water mixtures. They are not always suitable for emulsified oil/water separation, especially surfactant-stabilized emulsions, either because of low separation efficiency, or surface fouling that alters the surface properties and blinds the pores of the materials.

Many oily wastewaters contain some kind of surface active substances, causing increased stability of the formed emulsions, and the resultant difficulty in oil-water separation. A number of researchers have been dedicated to solving this issue. Tuteja

* Corresponding author.
E-mail address: cfyang@ipe.ac.cn (C. Yang).

http://dx.doi.org/10.1016/j.seppur.2016.07.051
1383-5866/

et al. [21] developed an oleophobic membrane based operation that allowed for on-demand separation of various oil-water mixtures using gravity as the only driving force. They were able to achieve separation efficiencies over 99.9% for both oil-in-water and water-in-oil emulsions stabilized with nonionic surfactants. Jin et al. [22] fabricated an underwater superoleophobic poly (acrylic acid)-grafted PVDF membrane using salt-induced phase-inversion method. The membrane showed high separation efficiency of sodium dodecyl sulfate (SDS) and Tween 80 stabilized oil-in-water emulsions. It is also noticed that superhydrophobic/superoleophilic iron particles [23] and under-water superoleophobic cellulose sponge [24] were designed to separate cetyl trimethyl ammonium bromide (CTAB)/Tween 80 stabilized emulsions by sorption. More work can be found in literature [25–30] that was dedicated to the separation of emulsions involving surfactants such as Tween 80, SDS and Triton X-100. However, these separations are mostly size exclusion-based, membrane fouling by surfactants is an emerging problem that necessitates special care to maintain the separation efficiency. Coalescence, as an opposite process to membrane filtration, is an effective way to achieve high flux, low-pressure and fouling less separation of liquid-liquid dispersions. Huck et al. [31] reported a non-fouling capture-release coalescence material based on polymer brushes. This material could capture oil droplets on the modified surfaces by strong adhesion, and allow the captured droplets to coalesce with other oil droplets for eventual self-release by buoyancy. Chawaloesphonsiya [32] studied the effects of media shape, size, and packing density on coalescence separation efficiency. They revealed that media wetting properties and filter bed permeability were key influencing factors. Dong and co-workers [33] investigated the effects of demulsifier dosage, CO_2 pressure, and mixing time for high pH water-in-oil emulsion separation. They found CO_2 accelerated the release of the droplets by changing the pH of the aqueous phase of the emulsion, and it also elevated the whole coalescence process by decreasing the viscosity of heavy oil thanks to the gas solubilization in the oil. Krebs et al. [34] studied the coalescence and compression of emulsions under enhanced gravity with in situ optical microscopy and provided insight into the droplet dynamics in dense systems. Yang et al. [8,35] developed self-assemblies onto ordinary stainless steel fiber felt, providing the surface with special micro/nano structure and low surface energy. The felt thus obtained could effectively separate non-surfactant emulsified oil through coalescence. They further made an effort to uncover the sensitivity of coalescence to surface wettability and pore size of the modified felt, in an attempt to gain more insights of coalescence working mechanism.

When treating oily wastewater, traditional fibrous coalescing technology has the advantages of convenient operation, long service life and high oil removal efficiency. In addition, this technology does not need additional reagents, hence high ability in value maintenance of the recovered oil is made conveniently possible. While industrial practice with this technology has never stopped, research work in this area has less visibility, overshadowed by membrane filtration. Nonetheless, published literatures can be seen from time to time [36–39]. With commercial filter media, Agarwal [38,39] looked into factors including surface energy, pore size, porosity, surface roughness and operating conditions on coalescence separation of isooctane-in-water emulsions, and found that an effective coalescing filter should (1) be hydrophobic and oleophilic; (2) orient vertically and (3) have fiber surface roughness for better oil wettability. Coalescence filter media tend to be thick to allow sufficient residence time for droplet to grow, while thinner media such as sintered stainless steel fiber felt [8,35], membranes [40], or resin bonded non-woven [41] are attractive in reducing pressure drop and the size of the finished filter products. They are expected to become more popular for large-scale coalescence separation in oil extraction field, hydrometallurgical plant and pharmaceutical industry, where the recovery of un-dissolved organics is required, and where the discharge of treated water must meet regulation guidelines. Since surfactants are often existent in these oily wastewaters, better filter media technology needs to be developed to effectively separate surfactant-stabilized emulsions.

In this paper, we went beyond our previous research [41] that centered on the separation of surfactant-free oil-in-water emulsions. We explored herein the facile solvent volatilization-resin emulsification technique and optimized it for treating wet-laid filter media made in house to separate surfactant stabilized oil-in-water emulsions. The media were endowed with surface roughness, formed by resin islands created by the emulsification process. An interesting relationship between the resin binder solidification and the initial coalescence separation efficiency was discovered. Three surfactants, namely Tween 80, CTAB and sodium dodecyl benzene sulfonate (SDBS), representatives of non-ionic, cationic and anionic ones, were applied to form the oil-in-water emulsions. The wetting behavior of the filter media was characterized by the lipophilic to hydrophilic value (L/H) measured experimentally. It was found that when the value was close to 2, best coalescence separation of these emulsions could be achieved. Explanation of the different separation efficiency corresponding to different surfactant stabilized emulsions was assisted by quartz crystal microbalance (QCM) studies that showed different surfactant adsorption behavior. Finally, more environment friendly solvents for resin emulsification were examined as alternatives to the more volatile isopropanol (IPA), and ethanol was found to be a good choice.

2. Experiments

2.1. Materials

The fibers used to prepare the filter media are glass wool (diameter 0.6 μm, Shenyang Dongxiang Glass Fiber Co., Ltd.), glass fiber (diameter 7 μm, length 6 mm, Taishan Fiberglass Inc.), and cellulose fiber [diameter 11 μm, length 6 mm, Lenzing Fibers (Hong Kong) Limited]. Aromatic type of thermoplastic polyurethane resin (TPU) was obtained from Taiwan Sheen Soon Co., Ltd. IPA was purchased from Xilong Chemical Co., Ltd. Cyclohexanone, SDBS and CTAB were purchased from Sinopharm Chemical Reagent Co., Ltd. Tween 80 was purchased from Gaungdong Guanghua Sci-Tech Co., Ltd. Ethanol was purchased from Beijing Chemical Works. Hexadecane was purchased from Haltermann GmbH. All these chemicals were used as received without further treatment unless otherwise indicated.

2.2. Preparation of filter media

The preparation of the pristine fibrous filter media was introduced in our previous work [41]. A total mass of 3 g of glass wool, glass fiber and cellulose fiber based on a mass ratio of 2:1:1 was dispersed in 500 mL water and mixed uniformly with an electric extractor for 5 min. The fiber dispersion was then transferred to a hand-sheet former containing 10 L water, stirred 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 medium (200 mm diameter) 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 material was cut into several pieces with a diameter of 25 mm for further treatment. TPU was cleaned with deionized water and dried before use. 10% resin solution was made by completely dissolving TPU in