Water Research 123 (2017) 332–344

Contents lists available at ScienceDirect

Water Research

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

Green technological approach to synthesis hydrophobic stable crystalline calcite particles with one-pot synthesis for oil–water separation during oil spill cleanup

Min-Nan Wu [a], Jyoti Prakash Maity [b, c, **], Jochen Bundschuh [c, d], Che-Feng Li [b], Chin-Rong Lee [a], Chun-Mei Hsu [e], Wen-Chien Lee [e], Chung-Ho Huang [f], Chien-Yen Chen [b, *]

[a] Department of Physics, National Chung Cheng University, 168 University Road, Ming-Shung, Chiayi County 62102, Taiwan
[b] Department of Earth and Environmental Sciences, National Chung Cheng University, 168 University Road, Ming-Shung, Chiayi County 62102, Taiwan
[c] School of Civil Engineering and Surveying, University of Southern Queensland, West Street, Toowoomba 4350, Australia
[d] Deputy Vice-Chancellor's Office (Research and Innovation), University of Southern Queensland, West Street, Toowoomba 4350, QLD, Australia
[e] Department of Chemical Engineering, National Chung Cheng University, 168 University Road, Ming-Shung, Chiayi County 62102, Taiwan
[f] Department of Civil Engineering at the National Taipei University of Technology, No. 1, Sec. 3, Zhongxiao E. Rd., Da'an Dist., Taipei City 106, Taiwan

ARTICLE INFO

Article history:
Received 1 March 2017
Received in revised form
14 May 2017
Accepted 6 June 2017

Keywords:
Water–oil separation
Calcite nanoparticles synthesis
Characterization
Hydrophobic-oleophilic $CaCO_3$
Oil removal
Isothermal and kinetics

ABSTRACT

The process of separating oil and water from oil/water mixtures is an attractive strategy to answer the menace caused by industrial oil spills and oily wastewater. In addition, water coproduced during hydrocarbon exploitation, which can be an economic burden and risk for freshwater resources, can become an important freshwater source after suitable water-oil separation. For oil-water separation purposes, considerable attention has been paid to the preparation of hydrophobic-oleophilic materials with modified surface roughness. However, due to issues of thermodynamic instability, costly and complex methods as well as lack of ecofriendly compounds, most of hydrophobic surface modified particles are of limited practical application. The study presents a facile procedure, to synthesize crystalline particles of calcite, which is the most stable polymorph of $CaCO_3$ from industrial $CaCO_3$ using oleic acid as an additive in a one-pot synthesis method. The XRD results show that the synthesized particles were a well-crystallized form of calcite. The FTIR results reflect the appearance of the alkyl groups from the oleic acid in synthesized particles which promotes the production of calcite with 'rice shape' (1.64 μm) (aggregated by spherical nanoparticle of 19.56 nm) morphology with concomitant changes in its surface wettability from hydrophilic to hydrophobic. The synthesized particles exhibited near to super hydrophobicity with ~99% active ratio and a contact angle of 143.8°. The synthesized hydrophobic calcite particles had an oleophilic nature where waste diesel oil adsorption capacity of synthesized calcium carbonate (HCF) showed a very high (>99%) and fast (7 s) oil removal from oil-water mixture. The functional group of long alkyl chain including of C=O bounds may play critical roles for adsorption of diesel oils. Moreover, the thermodynamically stable crystalline polymorph calcite (compared to vaterite) exhibited excellent recyclability. The isothermal study reflects the comparatively high value of correlation coefficient ($R^2 = 0.94$) for the Langmuir isotherm compared to those of the Freundlich isotherm ($R^2 = 0.82$) showed that the adsorption of diesel oil onto the hydrophobic $CaCO_3$ adsorbent was much better described by the Langmuir isotherm. The kinetics study of second-order rate expression ($R^2 = 0.99$) more fitted with the experimental data compare to first-order model ($R^2 = 0.92$). The synthesized calcite exhibited a significant dual oleophilic and hydrophobic nature that can be applicable for oil adsorption/or removal purpose in oil contaminated areas in environment and/or industrial oily wastewater for green, simple, and inexpensive environmental cleanup.

* Corresponding author.
** Corresponding author. Department of Earth and Environmental Sciences, National Chung Cheng University, 168 University Road, Ming-Shung, Chiayi County 62102. Taiwan.
E-mail addresses: jyoti_maity@yahoo.com (J.P. Maity), chien-yen.chen@oriel.oxon.org (C.-Y. Chen).

http://dx.doi.org/10.1016/j.watres.2017.06.040
0043-1354/

1. Introduction

Consequences from energy production through hydrocarbon exploitation can influence the environment and economies in adverse way. Catastrophic oil spills have caused great damage to life and environment (Sandrini-Neto et al., 2016; Duke, 2016; Jenny et al., 2016; Olson et al., 2016; Zhu et al., 2016a; DeLorenzo et al., 2016; Joye et al., 2016) and water coproduced during hydrocarbon exploitation can be an economic burden and, if not properly managed, can have severe negative environmental impacts; however, after suitable treatment, it can become an important freshwater source. Generally, the spilled oil or oil-containing coproduced water affects the biotic life in multiple ways which are either directly from the oil or due to cleanup process (Bautista and Rahman, 2016). There was a major oil spillage which was well documented in numerous reports and research publications in the Gulf of Mexico during April 20, 2010–July 15, 2010 when millions of liters (60,000 barrels of oil per day were leaking into the water) of oil were released due to the explosion of Deepwater Horizon (Harding et al., 2016; Smith et al., 2014; Bergin, 2012). In April 2013, almost 113 m^3 of crude oil were spilled from a Shell pipeline that entered a waterway that connects to the Gulf of Mexico (https://www.rt.com/usa/shell-pipeline-oil-texas-409/). On March 2016, oil was spilled from a ship that polluted the northern coastline of Taiwan (http://news.thewildeast.net/taiwans-north-east-coast-polluted-by-oil-spill/) and on 14th September 2016, a catastrophic oil spill occurred from vessels in Kaohsiung port when Typhoon Meranti swept through the port city (http://news.thewildeast.net/oil-spills-pose-environmental-threat-after-typhoon/). Unfortunately in every case, the spilled oil affects the bio-geosphere and the ecological balance in the environment (Barry, 2007; Sandrini-Neto et al., 2016; Duke, 2016; Jenny et al., 2016; Olson et al., 2016; Zhu et al., 2016a; DeLorenzo et al., 2016; Joye et al., 2016); results include unemployment and negatively impacts the fisheries industry and associated industries. As a consequence, in the case of the Deepwater Horizon spill ("deadly" 2010 oil spill) in Texas, later on in July 2015, BP agreed to pay (US) $18.7 billion in fines, which is the largest corporate settlement in U.S. history (http://www.reuters.com/article/us-bp-gulfmexico-settlement-idUSKCN0PC1BW20150702). In response to the severity of such environmental disasters environmental management to selectively remediate oil contaminated water is required, which must also be both a cost-effective and large-scale technology.

Freshwater production from water coproduced during oil exploitation – the world average is 3 barrels of water produced per barrel of oil (Veil et al., 2004) – which could contribute to alleviate the ever increasing freshwater demand. This is particularly true for irrigation, which consumes over 70% of the total freshwater production. Globally, there are millions of oil and gas wells. An older (1999) study showed that more than 210 million barrels (33.4 million m^3) water is coproduced each day (Veil et al., 2004). Using these production data and a nominal value of (US) $0.50 per barrel for disposal cost (the cost of its disposal varies widely from 0.10 to 2.00 US$ per barrel (Arnold et al., 2004)), then 38.3 billion US$ would be the annual cost for water disposal/management (Arnold et al., 2004). Therefore, finding low-cost solutions for removing oil from co-produced water would be a favorable option for freshwater production and an economical and environmental highly beneficial solution for oil produced water management.

Several traditional techniques for cleaning up oil spillages and to separate oil and water phases in water coproduced during hydrocarbon exploitation have previously been utilized including physical processes (flotation, vacuum and centrifuge, hydrocyclone) (Li et al., 2007; Al-Shamrani et al., 2002; Harvey and Stokes, 1973; Bai et al., 2011), filtration membrane (Prince et al., 2016; Zhu et al., 2016b), bioremediation (use of microorganisms or biological agents) (Yang et al., 2009; Pasumarthi et al., 2013; Bovio et al., 2017; Kasai et al., 2002; Crisafi et al., 2016; Jean et al., 2008; Liu et al., 2010; Majumder et al., 2014) and *in situ* burning (Lin et al., 2002, 2005; Guénette, 1997; Potter and Buist, 2008. Fritt-Rasmussen et al., 2015). Chemical dispersants can be used to dissipate oil slicks (Chapman et al., 2007). Other process such as dredging, beach raking, booms and solidifying, and other sustainable approaches of management plans can be used for oil cleanup and recovery, (Rahsepar et al., 2016; Al-Majed et al., 2012). The main disadvantages of the physical remediation technologies to combat marine oil contamination is that they are (i) Highly expensive, (ii) Labor intensive, (iii) Low efficient in high waves, (iv) Difficult to store and clean, and (v) Low flexible for towing (Dave and Ghaly, 2011). There are several drawbacks in conventional processes such as bioremediation since they are slow processes and *in situ* burning is only applicable under low wind conditions and results in air-borne pollutants. The dispersants process causes the spilled oil to rapidly break up into smaller tiny droplets which are rapidly mixed into the aquatic system and spread throughout the surface water. Dispersed oil droplets infiltrate into surface water and deeper water and can lethally contaminate aquatic life (plant, animals, corals, etc.). The adsorbent (nanoparticles of zeolite, activated carbon, silica, synthetic polymers, etc.) is quite effective and frequently used to separate oil-water mixtures, because of their simplicity in use and low cost. However, the adsorbent material is required to have several key properties such as high oil-adsorption capacity, high stability, low density, good recyclability, and it must also be environmental-friendly. Numerous earlier studies have reported several synthetic and natural adsorbent materials (zeolite, activated carbon, silica, organophilic clays and synthetic polymers, etc.) for oil pollution control (Muir and Bajda, 2016; Inagaki et al., 2014; Siddiqa et al., 2015). Nevertheless, an important challenge for current research is to increase hydrophobicity and oleophilicity and to improve the stability of absorbing materials. For these reasons cost-effective production of materials is targeted, which must also be ecofriendly and effective in real-world practical applications, are key strategies of materials synthesis.

Numerous researchers have synthesized different materials for oil adsorption using various processes (Wang et al., 2009; Li et al., 2013; Tian et al., 2011; Kong et al., 2015; Song, 2016; Xu et al., 2016; Hu et al., 2009; Sarkar and Mahapatra, 2014). The metal copper meshes used hydrophobic and oleophilic surfaces for water and oil mixture separation which are prepared by etching with nitric acid and modified by 1-hexadecanethiol (HDT) (Wang et al., 2009). ZnO and micro/nanoscale array-coated mesh (stainless steel mesh) films have been used for the separation of water and oil (Li et al., 2013; Tian et al., 2011). Hydrophobic cuprous oxide nanostructures on phosphor-copper meshes have been applied for oil-water separation (Kong et al., 2015). There are several micro/nanoscale adsorbents available for oil-water separation (Song, 2016; Xu et al., 2016; Hu et al., 2009; Sarkar and Mahapatra, 2014; Wang et al., 2013). Recently, a hydrophobic metal wire mesh (SMWM) was prepared by the electrospray method for separating oil from water (Song, 2016). Silica nanoparticles and decanoic acid-modified TiO_2-coated cotton fabric has been used for oil-water separation (Xu et al., 2016). However, only very limited research has investigated simple, low-cost synthesis processes of adsorbent materials as well as targeting their (adsorbent material) higher stability, low density, better recyclability, and environmental-friendly properties. Surface modification of $CaCO_3$ has received recent attention in oil removal applications because of its versatility (many applications as textiles, rubber, paints, polymer and dentifrices, etc.), as well as being highly abundant naturally as a biomineral and therefore cost-effective. The micro-sized $CaCO_3$