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Research article

Multiwall carbon nanotube reinforced teflon fibrils for oil spill clean up and its effective recycling as textile dye sorbent

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ABSTRACT

Surface functionalized multiwall carbon nanotube (MWCNT) reinforced teflon fibrils (MWCNT@Teflon) were successfully tested as an - oil - absorbent that can be used as a potential oil recovery material at the time of oil spill accidents in water. We found that oleic acid functionalization of MWCNTs was important for their adhesion onto teflon fibrils and at the same time prevented the MWCNT leaching into oil/water interface. The fibrils had displayed superior mechanical and thermal stability and provided a new insight to oil spill clean-up applications with easy recovery of absorbed oil by simple squeezing. Recycling of exhausted MWCNT@Teflon fibrils after oil recovery applications was conducted by pyrolysis under inert atmosphere in presence of magnetic clay. The magnetic clay absorbed the pyrolysis products, resulting in a heterostructured magnetic clay carbon composite (MCC) which was found super paramagnetic and chemically stable in all pH. The MCC was found capable of adsorbing textile dye from water ultra-fast with in a maximum contact time of 2 min and magnetically separable after adsorption experiments.

1. Introduction

Accidental release of liquid petroleum hydrocarbon into water, often reported as oil-spill, has severe detrimental impact on the marine environment than any other waste or spill material. When oil-spill ensue on the sea environment; floating barriers are initially used to control its spreading. When the oil spreading is under control, oil is removed via convenient means such as *in situ* burning, bio-remediation, filtration and absorption (Nevesa et al., 2015). Adsorbents offering physico-chemical interaction with spilled oil and facilitate quick physical change from fluid to semi-solid phase can make the oil removal much easier compared to bio-remediation/filtration. Adsorbents such as saw dust, vermiculite, organic gel, fly ash, poly dimethoxy siloxane sponges, fluorine functionalized graphene aerogels, polymers resins, polyurethane foams, clay composites, carbon nanotube (CNT) sponges are currently reported for oil adsorption (Cambiella et al., 2006; Mysore et al., 2005; Kizil and Sonmez, 2017; Moura and Lago, 2009; Karakasi and Moutsatsou, 2010; Choi et al., 2011; Hong et al., 2015; Nikkhah et al., 2015; Gui et al., 2010). However, oil sorbents (adsorbent/absorbent) that can be used for recovery of oil at the time of oil spill incidents at an economically and operationally practical level are limited. In most of the reported cases, poor durability, non-regenerative nature, requirement of post-processing for the extraction of absorbed oil and generation of huge amount of oil saturated wastes pose difficulties in real life applications. Among the reported oil sorbents, CNT sponges are reported to be promising in terms of repeated oil sorption efficacy (Gui et al., 2010). But with all the advantages of CNTs, limitations such as its delicate and expensive synthesis procedure, high tendency for aggregation and handling difficulty pose problems in any direct usage (Abraham et al., 2017). More importantly, because of high oleophilicity, CNT tends to leach out into the recovered oil and demands further purification procedure for both the water and recovered oil. Usage of minimal amount of CNT by immobilizing them in a safe and cheap matrix is thus much desirable, because the demand may be in huge quantities if any oil spill accidents occurred

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in sea regions. There were some attempts on chemical modification of CNTs with polymers such as poly(N-diethylaminopropyl) methacrylate and polystyrene to improve the oil sorption capacity of CNTs (Abraham et al., 2017; Gu et al., 2014). Recently Keshavarz et al., has reported that immobilizing CNT on to polyurethane foam (PU) considerably increased the oil sorption capacity (Keshavarz et al., 2015). Nevertheless, one problem with the PU foam is its high porous nature and abundance of ether and amide-groups, which make PU susceptible to absorb both oil and water (Li et al., 2012).

Here we report a teflon and multiwall carbon nanotube (MWCNT) based material which absorb huge amount of oil from water very rapidly and repeatedly. We used teflon fibrils which were the waste generated in Teflon ware manufacturing industry. Using them as a matrix to immobilize MWCNT helped to cut down the material and operational cost to a great extent. Notably, our value addition recycling approach also contributes to a solution to teflon waste disposal problem which is much more difficult due to the non-biodegradable nature of teflon. Teflon acts not only as a binding matrix to MWCNT but also as an oil absorbing component to the hybrid system due to hydrophobic nature. The fibrilar form of teflon is also preferentially useful for this application since the several fibrils entangle together to form a network structure, which can trap large amount of oil from the water like a sponge and at the same time give higher mechanical durability. Nevertheless, immobilizing MWCNT onto the teflon fibrils is very challenging because of the surface energy mismatch between teflon and MWCNT (Wang and Musameh, 2003). In order to inhibit the leaching of MWCNT, the interfacial adhesion between MWCNT and teflon fibrils should be improved. Thus it is very necessary to overcome the van der Waals potential between MWCNT particles to prevent agglomeration and to form a stable and homogenous arrangement of MWCNT on teflon fibrils. Here in we present a new approach to selectively improve the adhesion between MWCNT and teflon matrix by functionalizing MWCNT with oleic acid (OA). To the best of our knowledge, this is the first report on utilizing teflon ware manufacturing waste to prepare a super oleophilic composite by combining with MWCNT.

Generation of huge amount of oil saturated waste is another practical problem which is faced when sorbent materials are used for oil recovery applications. Generally landfilling or incineration with no material recovery is followed, which induce concern to the local community about the environmental impact. As a solution to this challenging environmental issue, we successfully tested the feasible conversion of our exhausted oil sorbent into magnetic clay-carbon bi-functional adsorbent. Magnetic component was introduced in the recycling stage to ease the operational difficulty with powder sorbent materials. The recycled product is found to be an ultra-fast dye adsorbent which can be easily recovered from water using magnetic separation.

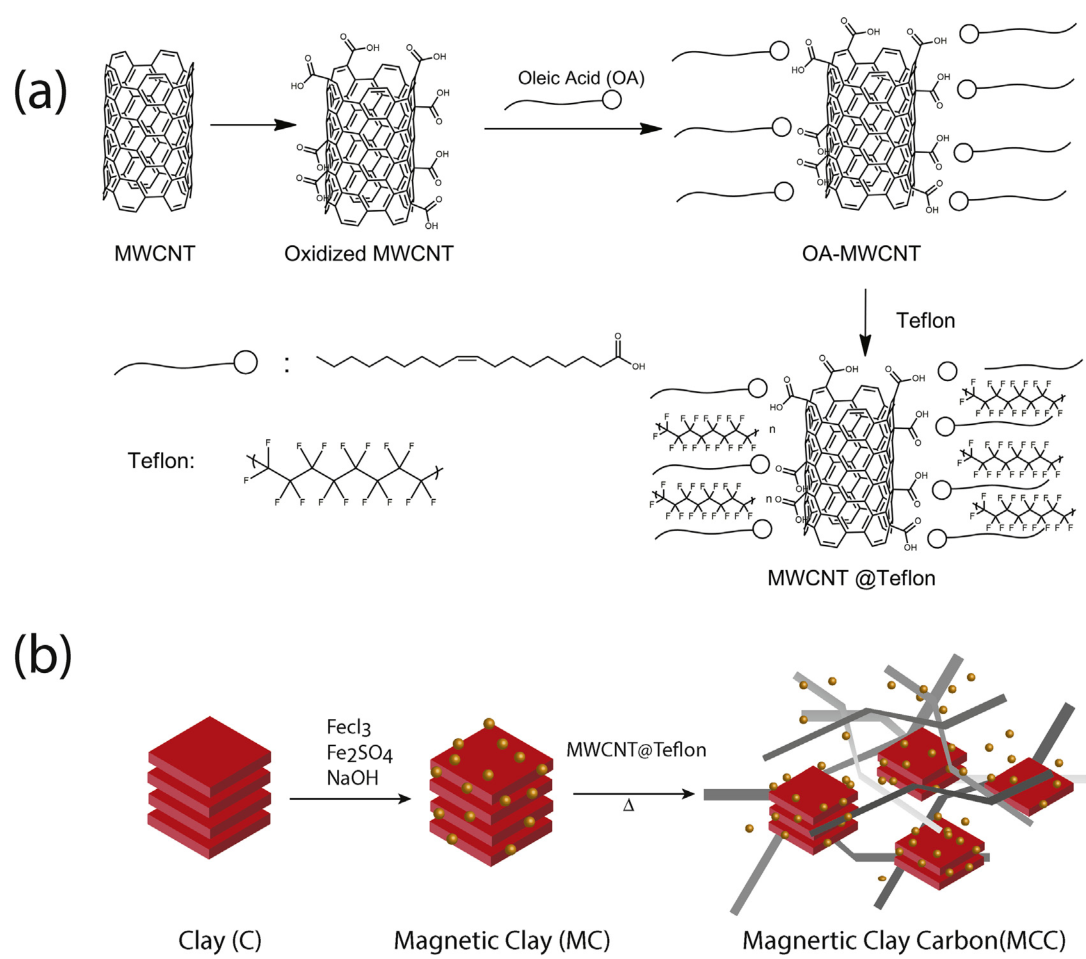

Fig. 1. Schematic representation of the synthesis route of (a) oil absorbing MWCNT@Teflon and (b) dye adsorbing magnetic clay carbon composite (MCC).