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Facile and sustainable shear mixing/carbonization approach for upcycling of carton into superhydrophobic coating for efficient oil-water separation

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ABSTRACT

The development of facile and sustainable methods to produce efficient oil-water separation materials is an important and challenging problem. This paper presents a simple and green shear mixing/carbonization approach for the upcycling of old corrugated containers into a superhydrophobic coating (OCF-600), which can be applied to a suitable sponge material and used to clean up oil spills and organic solution leaks. There is a synergetic effect between the shear mixing and carbonization processes. The water contact angle of OCF-600 ($152.57 \pm 1.50^\circ$), which is obtained by the shear mixing/carbonization process using cartons as the only precursor, is significantly larger than that of materials obtained by individual shear mixing or carbonization methods, and even larger than that of carbon material obtained by carbonization/shear mixing process ($138.85 \pm 0.85^\circ$). The shear mixing drastically enhanced the surface roughness, whereas the carbonization led to the formation of the hydrophobic groups, further strengthened its micro-roughness, and amplified the surface property to superhydrophobicity. The OCF-600 can easily be coated onto several substrates to form superhydrophobic materials for efficient oil-water separation with excellent durability and selectivity, even under corrosive and turbulent conditions. The OCF-600 coated sponges exhibited favorable adsorption capacities range from 18 to 44 times their own weight for various kinds of oils and organic solvents. The OCF-600 coated microporous membrane can separate surfactant-stabilized oil-water emulsions with a separation efficiency of greater than 99.88%. Furthermore, this approach can be extended to a series of cellulosic waste, thus could open up new prospects for the fabrication of superhydrophobic coatings through the sustainable upcycling of waste products.

1. Introduction

The global carton/corrugated box market was valued at $67 billion in 2017. The demand for cartons/corrugated boxes is expected to increase by an average of more than 4% annually over the next 5 years and will amount to almost 115 million tons of finished product by 2019 (Freedonia, 2016; Smithers Pira, 2016). In China, due to the rapid development of the express delivery industry, the number of cartons/corrugated boxes used for express packages increased from 9.92 billion in 2015 to 14.4 billion in 2016, and the amount is predicted to increase by an average of more than 50% annually (State Post Bureau of the People's Republic of China, 2016). Old corrugated container (OCCs) can be recycled to make new paper and fiber products. However, because the strength of the fiber deteriorates during the repulping process, the recycled pulp slurry cannot be used to make high-quality paper (Shimada et al., 1999). In addition, OCCs may contain a variety of extraneous materials and chemical constituents that can potentially contaminate recycled paper products. Meanwhile, the cost effectiveness of re-using OCCs is limited by the cost of box collection, transport, maintenance, and disinfection. Less than 20% of OCCs are collected and reused in China, and they are now one of the main constituents of municipal waste, causing many environmental problems (State Post Bureau of

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the People's Republic of China, 2016). Therefore, there is a strong motivation to develop facile, sustainable, and industrially scalable applications for OCCs.

Due to the development of industrial activities and changes in people's daily lives, the rapidly increasing exploitation, use, and transport of crude oils and their toxic by-products have resulted in more oily water and frequent oil spillage accidents worldwide (Atlas and Hazen, 2011; Schnoor, 2010). Oil and refined oil spills cause serious long-term effects to the marine ecosystem, some of which can last for decades. Oil spills have had disastrous economic, environmental, and social consequences around the world (Prince et al., 2016; Schaum et al., 2010). For example, 6814 dead animals were collected between April 20 and November 2, 2010, following the Gulf of Mexico spill (Gulf coast ecosystem restoration council, 2010). A recent reported revealed that in 2016, 88% of about 360 baby or stillborn dolphins within the spill area had abnormal or underdeveloped lungs, compared with 15% in other areas, indicating the long-term environmental consequences of the spill (Colegrove et al., 2016). Furthermore, spilled oil can enter the food chain through zooplankton, with long-term effects on both humans and marine life (Allan et al., 2012; Peterson et al., 2003). Hence, the effective cleaning up of oil spills, the purification of oily wastewater, and the separation of oil-water mixtures are challenging tasks worldwide and are extremely important for environmental protection, economic, and social issues.

Among the state-of-the-art oil recovery methods, the sorption method is considered to be one of the most promising approaches for cleanup of final traces of oil spill or in very small spills. Hydrophobic sorbents that can recover oil through either adsorption or absorption play an important role in oil spill cleanup, especially for small oil leaks and subsequent cleanup of smaller amounts (Fingas, 2011, 2012; Hubbe et al., 2013). Superhydrophobic materials, with a water contact angle (CA) greater than 150°, have aroused considerable interest because of the high selectivity and effective oil-water separation capacity (Lu et al., 2014, 2015; Chen et al., 2017). The wettability of the superhydrophobic materials is determined by its surface morphology together with surface energy. Such materials can be broadly categorized into two types: superhydrophobic materials with rough surface and low-surface energy that can be sprayed onto various substrates to create superhydrophobic surfaces, and superhydrophobic aerogels or foams with good selective sorption (Lu et al., 2014; Wang et al., 2015; Cao et al., 2016; Guo et al., 2016; Chen et al., 2017). However, the fabrication of such superhydrophobic surfaces usually involves post-treatment with tedious fabrication processes that use specific instruments and expensive or toxic low-surface energy polymeric coating, poly(vinylidene fluoride) for example (Table S1) (Chen et al., 2017; Huang et al., 2014; Shateri-Khalilabad and Yazdanshenas, 2013). The preparation of superhydrophobic aerogels, especially carbon aerogels, generally involves time-consuming freeze-drying and environmentally harmful acidic washing processes (Han et al., 2016; Wang et al., 2015). These disadvantages have largely restricted the practical industrial-scale application of superhydrophobic surfaces and aerogels.

In this work, we present a simple, sustainable, and environmentally friendly method of constructing robust superhydrophobic coating using OCC as the single precursor. Shear mixing the OCC in a kitchen blender for 5 min before the commonly used carbonization process resulted in a robust micro/nanostructure and enhanced superhydrophobicity due to the “lotus effect.” Unlike most of the previously reported techniques that involve long treatment times with expensive or toxic chemicals and use specific instruments (Table S1) (Huang et al., 2014; Shateri-Khalilabad and Yazdanshenas, 2013), our facile shear mixing/carbonization approach is environmentally friendly and scalable. The ethanolic suspension of the obtained OCC-derived superhydrophobic coating can be dip-coated and sprayed onto commercial polyurethane (PU) sponges and microporous membranes to create superhydrophobic materials with efficient oil absorption and oil-water separation. The obtained OCC-coated sponge quickly adsorbs a mass of oils and organic solvents from many corrosive water surfaces (1 M HCl, 1 M NaCl, and 1 M NaOH), reflecting its efficiency and stability. In addition, the OCC-coated microporous membrane can separate oil-water emulsions stabilized by Tween 80 with a separation efficiency of greater than 99.88%. A proof-of-concept study demonstrated that this facile and sustainable shear method can be easily scaled up to an industrial level for upcycling of a series of lignocellulosic waste. These obtained waste-derived superhydrophobic coatings are promising candidates for oil leakage cleanups and oily wastewater purification in complex environments.

2. Experimental

2.1. Materials

Liquid paraffin, n-hexane, petroleum ether, dichloromethane, Tween 80 and absolute ethanol were purchased from Sinopharm Chemical Reagent Co. Ltd (China) and were used as received. Peanut oil (Luhua Co. Ltd. Shandong, China), vacuum pump oil (Shanxing chemical group, Jinan, China), motor oil (Mobil 5W-30, Exxon Mobil Co. USA), PU sponge, kitchen blenders (HR2104/2017, Philips Co. Netherlands), and stainless-steel mesh (400 mesh) were purchased from commercial stores. PDMS (Sylgard 184, Dow Corning Co. Midland) was used as polymer adhesive. The cartons/corrugated boxes packaging the above reagents and materials were collected and used as the OCC.

2.2. Synthesis of OCC-derived superhydrophobic coating

The OCC-derived superhydrophobic coating was synthesized by a facile shear mixing process. In a typical experiment, the OCC was first cut into pieces and shear mixed in the kitchen blender for 5 min. The sheared OCC was then annealed in an N_2 atmosphere at 600 °C for 2 h at a heating rate of 5 °C min^{-1} in a quartz tube to obtain the OCC-derived robust superhydrophobic carbon-based coating (OCF-600).

2.3. Preparation of OCF-600 coated meshes and sponges

The PU sponge and stainless steel meshes were ultrasonically cleaned in deionized water and ethanol for 30 min and then dried at 60 °C. 25 g of OCF-600 was weighed into the kitchen blender, and 500 ml absolute ethanol and 5 g polymer adhesive were added. A kitchen blender was used for shear mixing for 10 min to obtain the ethanolic OCF-600 based superhydrophobic coating. The cleaned PU sponges were immersed in the suspension for 20 s. This ethanolic OCF-600 suspension was sprayed onto the cleaned stainless steel meshes and microporous membrane using a spray gun at the specific spray condition of 150 L/h N_2 flow rate of with a distance of 4 cm for the sake of consistency. The OCF-600 coated PU sponges, meshes, and microporous membranes were then dried at ambient temperature, with the finial coating amount of 0.64 ± 0.042 g cm^{-3}, 0.017 ± 0.002 g cm^{-2}, and 0.014 ± 0.003 mg cm^{-2}, respectively.

2.4. Material characterization

The surface morphology of the OCC-derived carbon fiber was observed by scanning electron microscopy (SEM; FEI Nova-Nano, Netherlands). The crystal structures of the OCC-derived carbon materials were determined by X-ray diffraction (XRD) (X'Pert PRO,