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One-step immersion for fabrication of superhydrophobic/superoleophilic carbon felts with fire resistance: Fast separation and removal of oil from water

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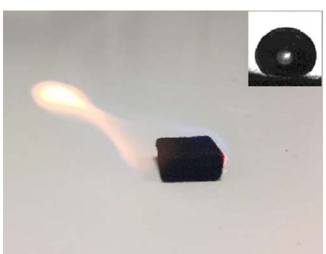
GRAPHICAL ABSTRACT

Carbon felt functionalized with 4-(heptadecafluorooctyl)aniline through an immersion process led to the formation of a superhydrophobic/superoleophilic material with good performance for oil/water separation and excellent fire retardant properties.



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ABSTRACT

The paper reports on a simple, low cost, and easy to perform methodology for the preparation of superhydrophobic/superoleophilic carbon felts. The technique relies on a one-step immersion process of carbon felt in 4-(heptadecafluorooctyl)aniline ethanolic solution for 24 h at room temperature. The functionalized carbon felt surface displays a superhydrophobic character with a static water contact angle higher than 150°. Importantly, the superhydrophobic property is retained upon immersion of the surface for 1 day in harsh environments over a pH range of 1–14, in salt solutions such as KCl, Na_2SO_4, $MgCl_2$, $ZnSO_4$, and in various organic solvents such as hexane, toluene, acetone, chloroform, diethyl ether and DMSO. Additionally, the modified carbon felt exhibits good thermal stability and flame retardant properties. In fact, igniting an ethanol-soaked carbon felt has no impact on the material shape/morphology after burning the ethanol away. Finally, the superhydrophobic/superoleophilic properties of the carbon felt are successfully applied for fast and efficient organic solvent separation from water. The high performance of the carbon felt associated with the simple and cost-effective functionalization process hold great promise for potential applications in environmental remediation.

1. Introduction

With the rapid development of industry, oil spills and chemical leakage accidents occur frequently, leading to the environmental pollution and ecological destruction [1–3]. Aside from environmental and ecological negative impact, the spilled oil and organic compounds can

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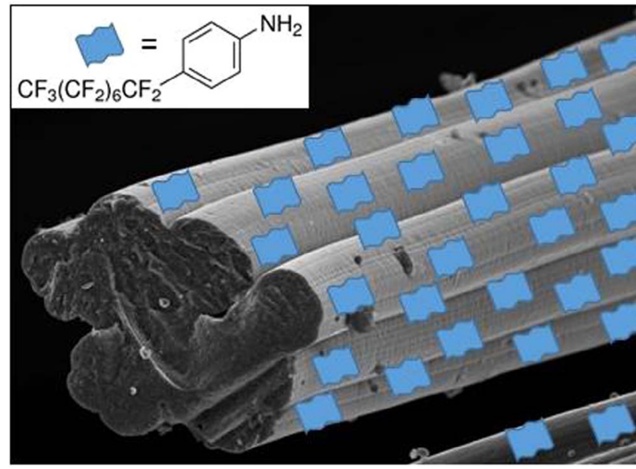
potentially cause fire and explosion, because most of crude oil and organic components are highly flammable. Therefore, it is urgent to develop effective, facile and low-cost methods to clean up the oil spills and chemical leakage for environmental and water source protection. In general, three methods, including burning, mechanical collection and the use of dispersants, have been used in removing oils and organic solvents from water. However, these traditional strategies are less efficient and tedious to implement.

Recently, novel materials with superhydrophobic and superoleophilic properties have attracted much attention due to their low cost, high performance and chemical durability. For example, modified polyurethane sponge [4–12], graphene-based sponge [13–16], metal mesh [17–22], etc. have shown high efficiency for oil/water separation and removing oil/organic solvent from water. Wu et al. prepared magnetic, durable, and superhydrophobic polyurethane (PU) sponge in a multi-step process consisting of chemical vapor deposition of tetraethoxysilane to tightly bind Fe_3O_4 nanoparticles on the sponge and then dip-coating in a fluoropolymer aqueous solution; the modified PU was successfully applied for oil adsorption and oil/water separation [10]. Jayaramulu et al. reported on superhydrophobic/superoleophilic composites obtained from highly fluorinated graphene oxide (HFGO) and nanocrystalline zeolite imidazole framework ZIF-8; the hybrid material displayed a high water contact angle of 162° and low oil contact angle of 0° and thus revealed good adsorption for nonpolar/polar organic solvents and oils from water [13]. Liu et al. fabricated a superhydrophobic/superoleophilic copper mesh with excellent oil-water separation efficiency through electrodeposition and surface modification with lauric acid [19]. However, most of the reported techniques require tedious processes, harmful and expensive precursors or complex equipment, which hamper their large scale production. Therefore, there is a continuous demand for the development of facile and low cost methods for the preparation of versatile materials with superhydrophobicity and superoleophilicity, which cannot only separate oil/water mixture, but also remove oil/organic solvent from water. This field will experience a huge and continuous growth in the coming years as issues related to environmental remediation will become central in the modern life. This is even true, because human health is highly impacted by all types of pollutions.

In this study, we investigate for the first time the use of functionalized carbon felt, consisting of random carbon fibers, as a base material to enable separation of oil from water. The initial hydrophobic carbon felt is turned into a superhydrophobic/superoleophilic material through a simple immersion process in an ethanolic solution of 4-(heptadecafluorooctyl) aniline under ambient conditions. The choice of this molecule (chemical structure depicted in Scheme 1, inset) is made based on the presence of i) a long fluorinated alkyl chain to ensure a low surface energy coating and ii) an aromatic core to achieve strong interaction with the graphitic carbon fibers composing the carbon felt.

The resulting superhydrophobic/superoleophilic carbon felt exhibits excellent thermal stability, corrosion resistance in acid, alkaline and high salt solution as well as in organic solvents. Additionally, it shows good performance for oil/water separation and oil/organic solvents removal from water. Importantly, the combination of superhydrophobicity with flame-retardant properties is desirable for many aspects, and particularly when the spilled oil or organic solvents are susceptible to cause fire and explosion [23–25]. We demonstrate further that the functionalized carbon felt soaked with ethanol is not affected upon ignition and burning of ethanol. The good performance of the superhydrophobic/superoleophilic carbon felt material for oil/water separation along with its flame-retardant properties and commercial availability hold great promise for industrial applications where these properties are desirable.



Scheme 1. Illustration of the carbon felt functionalization with 4-(heptadecafluorooctyl) aniline ($C_{14}H_6F_{17}N$).

2. Experimental section

2.1. Materials

All chemicals were reagent grade or higher and were used as received unless otherwise specified. Carbon felt (carbon content: 99.9%; density: 0.088 g/cm^3) was purchased from Mersen France Gennevilliers S.A.S. 4-(Heptadecafluorooctyl)aniline ($C_{14}H_6F_{17}N$, ≥95.0%), rhodamine B, absolute ethanol (≥99.8%), hexane, acetone, toluene, chloroform, diethyl ether, dimethylsulfoxide (DMS), potassium chloride (KCl), sodium sulfate (Na_2SO_4), magnesium chloride ($MgCl_2$), zinc sulfate ($ZnSO_4$), hydrochloric acid (HCl) and sodium hydroxide (NaOH) were purchased from Sigma-Aldrich. The water used throughout the experiments was purified with a Milli-Q system from Millipore Co.

2.2. Fabrication of superhydrophobic/superoleophiliccarbon felt

The carbon felt (1.0 cm × 1.0 cm × 0.5 cm) samples were successively washed in an ultrasonic bath with ethanol for 10 min, and dried in an oven at 50 °C for 20 min. Then, the cleaned carbon felts were immersed into 10 mM 4-(heptadecafluorooctyl)aniline ethanol solution (20 mL) for 24 h with shaking at 150 rpm. Finally, the carbon felts were rinsed with ethanol three times and dried in air at room temperature.

2.3. Characterization

The morphology and composition of the modified materials were analyzed using scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) spectroscopy using a FEI Nova NanoSEM 450 scanning electron microscope with FEG (field emission gun, Schottky type) system and an ULTRA 55 (Zeiss) equipped with an energy dispersive X-ray analysis device (EDX analysis).

Fourier transform infrared (FTIR) spectra in grazing-angle attenuated total reflectance mode (g, ATR) were recorded using a Thermo Scientific FTIR instrument (Nicolet 8700) equipped with a VariGATR™ accessory (Harrick Scientific Products Inc.). Spectra were collected at