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Full Length Article

One-step assembly of Fe(III)-CMC chelate hydrogel onto nanoneedle-like CuO@Cu membrane with superhydrophilicity for oil-water separation

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ABSTRACT

The research of superhydrophilic interface is developing rapidly, but the preparations of superhydrophilic surfaces through simple methods are still challenging. Herein, we reported a facile, rapid and environmentally-friendly approach for preparing a novel superhydrophilic and underwater superoleophobic membrane *via* the thermal oxidation of Cu mesh and one-step coordinated assembly of Fe(III)-CMC chelate hydrogel. Superhydrophilicity was attributed to the hydrophilicity of Fe(III)-CMC chelate hydrogel and nanoneedle-like rough structure of CuO@Cu membrane. The membrane was used to separate a variety of oil/water mixtures and exhibited excellent separation performance. Moreover, the membrane exhibited the excellent durability and superior stability against corrosion conditions. We envision that the Fe(III)-CMC@CuO@Cu membrane with good underwater superoleophobicity could provide a candidate not only for oil/water separation but also many other potential applications such as underwater oil manipulation, self-clean, and bio-adhesion control.

1. Introduction

With the development of industry and economy, the petroleum and its products were widely applied in national economy and human daily life [1]. At the same time, oily waste-water has become one of the most serious pollution problems of worldwide environment, which was mainly from industrial waste, domestic sewage and oil spill. Among them, oil spill was the main source of oil pollution, which could directly poison marine life and cause serious damage to air, soil and groundwater [2]. Thus, it was urgent to solve the problem of oil pollution. Traditional techniques for oil-water separation, such as adsorption [3], gravity separation [4], oxidation [5], electrification method [6], and biological treatment [7], were limited by low separation efficiency and complex instruments. Membrane separation was widely used in waste-water separation, due to the low-cost, high-efficiency and excellent repeatability [8–10]. However, the application of traditional membrane materials was limited by low flux and easy fouling. Recent research found that the use of superwetting materials was the most efficient way to solve these problems [11–13]. This technology separated the oil phase and the aqueous phase from oil-water mixture utilizing the oleophilicity or hydrophilicity of superwetting membrane, with the advantages of normal temperature operation, no phase change, no additional impurity and energy saving [14]. The various superwetting membranes were fabricated for oil-water separation, mainly including superhydrophobic and superoleophobic materials [15–17]. The superhydrophobic materials were easily polluted by oil droplets and quick decrease of separation efficiency during the separation process [18]. Therefore, superhydrophobic materials were unsuitable for the separation of water-rich oil/water mixtures or oil-in-water emulsions. Superoleophobic and superhydrophilic material may be an excellent alternative with the good self-cleaning ability, instead of the superhydrophobic material [19]. Thus, it was significant to prepare superhydrophilic and superoleophobic materials for oil-water separation.

The synthesis methods of superhydrophilic/superoleophobic materials were divided into two categories. The most effective way to achieve superoleophobicity was by introducing fluorinated low surface-energy chemicals on micro/nano-hierarchical structures [20]. But this method brought some environment hazards and superoleophobic property of these materials would be lost under water. Inspired by the superoleophobicity behavior of oil

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droplets on fish scales in water, the researchers synthesized underwater superoleophobic surfaces through the combination of suitable surface roughness and high-surface-energy chemistry [21]. Many underwater-superoleophobic membrane materials were fabricated, such as hydrogel coated mesh [22,23], porous nitrocellulose membranes [24], inorganic nanowire hair copper mesh [25], and PMAPS-g-PVDF membrane [26]. However, these methods have some drawbacks, e.g. the cumbersome preparation process, the more expensive experimental reagents and the use of toxic reagents, which may cause environmental pollution and limit their

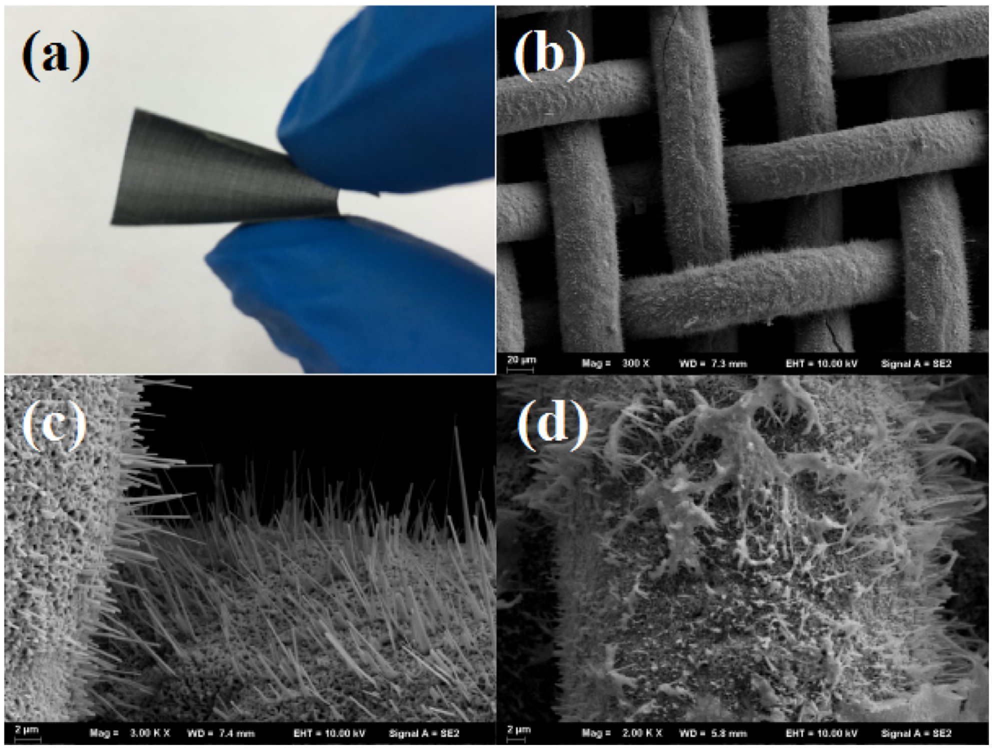

Fig. 1. Physical image of Fe(III)-CMC@CuO@Cu membrane (a), and SEM image of CuO@Cu membrane (b) and Fe(III)-CMC@CuO@Cu membrane (c, d).

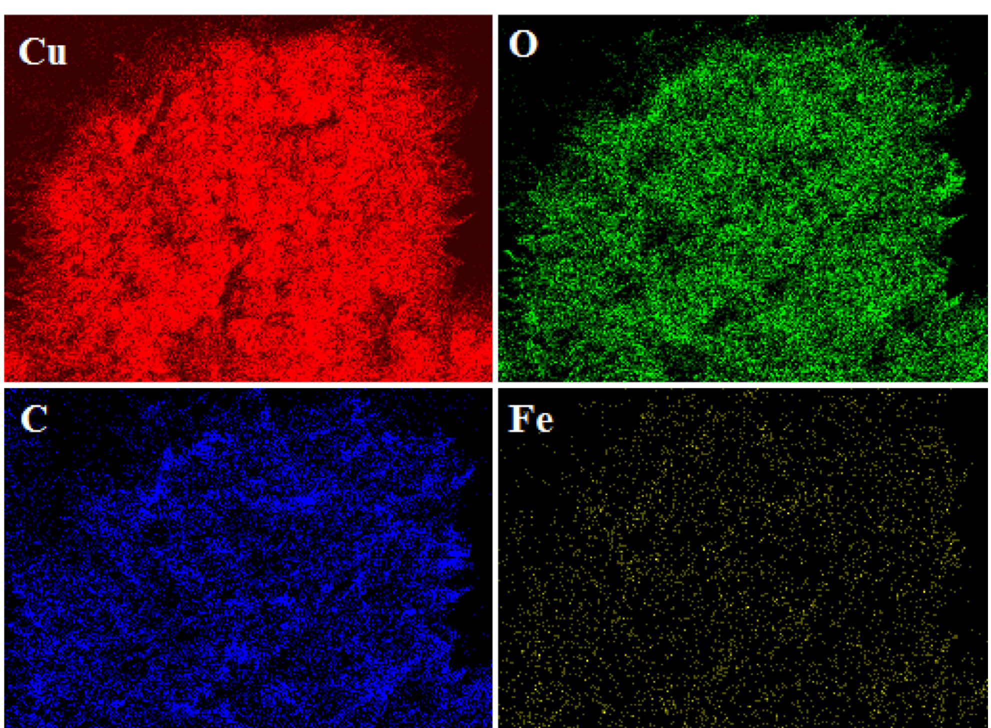

Fig. 2. EDS elemental Mapping images of Fe(III)-CMC@CuO@Cu membrane (Fig. 1d).