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Ultra-wetting graphene based ultrafiltration membranes for efficient waste-water treatment

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Abstract

Graphene, an sp2-hybridized, two-dimensional carbon material is gaining much attention in the field of membrane science and engineering. Theoretical analysis have also predicted that graphene based membranes may exhibit orders of magnitude with greater permeability than the current state of the art membranes. However, most of these studies are based on a single layer of graphene sheet. Experimental studies also show that it is difficult to fabricate leak-free porous graphene membranes with large surface area. In this work, we report a facile method to fabricate graphene-based composite ultrafiltration membrane in real downstream application. In order to achieve this, the wettability of graphene was increased by amine and carboxyl functionalization. Graphene was first carboxylated, using highly concentrated acid mixture (hydrochloric acid and sulphuric acids). The carboxylic group was further modified to acid chloride. Carbon nanomaterials such as graphene and its derivatives based membranes in real applications are still far from reality due to their hydrophobic nature and limitations in their fabrication process. Here, we have devised a simple and feasible fabrication method to bring the high end nanocarbon based material to real downstream applications. In order to achieve this objective, the wettability of graphene was initially increased to an ultra-wetting level by incorporating amine and carboxyl functionality onto the graphene. The amine and carboxylated graphene is then covalently attached to a polymer matrix to fabricate a water filtration membrane. The characterization of the modified supported graphene-based membrane indicates that significantly higher hydrophilicity than previously expected is achieved, with the water contact angle reduced to zero. The ultra-wetting graphene increases the water permeability of the membrane by 126% without any changes in the selectivity. Based on our findings, we conclude that the ultra-wetting graphene will be an ideal material for new generation water filtration membranes. Oil pollution in water and separation of oil from water are receiving much attention in recent years due to the growing environmental concerns. Membrane technology is one of the emerging solutions for oil-water separation. However, there is a limitation in using polymeric membrane for oil water separation due to its surface properties (wetting behaviour), thermal and mechanical properties. Here, we have shown a simple method to increase the hydrophilicity of the polyethersulfone (PES) hollow fibre ultrafiltration (UF) membrane by using carboxyl, hydroxyl and amine modified graphene attached poly acrylonitrile-co-maleimide (G-PANCMI). The prepared membranes were characterized for its morphology, water and oil contact angle, liquid entry pressure of oil (LEPoil), water permeability and finally subjected to a continuous 8 h filtration test of oil emulsion in water. The experimental data indicates that the G-PANCMI play an important role in enhancing the hydrophilicity, permeability and selectivity of the PES membrane. The water contact angle (CAw) of the PES membrane is reduced from 63.7 ± 3.8° to 22.6 ± 2.5° which is 64.5% reduction while, the oil contact angle was increased from 43.6 ± 3.5° to 112.5 ± 3.2° which is 158% higher compared to that of the PES membrane. Similarly, the LEPoil increased 350% from 50 ± 10 kPa of the control PES membrane to 175 ± 25 kPa of PES-G-PANCMI membrane. More importantly, the water permeability increased by 43% with >99% selectivity. Based on our findings we believe that the development of PES-G-PANCMI membrane will open up a solution for successful oil-water separation. Graphene, an sp2-hybridized, two-dimensional carbon material is gaining much attention in the field of membrane science and engineering. Theoretical analysis have also predicted that graphene based membranes may exhibit orders of magnitude with greater permeability than the current state of the art membranes. However, most of these studies are based on a single layer of graphene sheet. Experimental studies also show that it is difficult to fabricate leak-free porous graphene membranes with large surface area. In this work, we report a facile method to fabricate graphene-based composite ultrafiltration membrane in real downstream application. In order to achieve this, the wettability of graphene was increased by amine and carboxyl functionalization. Graphene was first carboxylated, using highly concentrated acid mixture (hydrochloric acid and sulphuric acids). The carboxylic group was further modified to acid chloride. Finally, the acid chloride modified graphene oxide was amine functionalized by using ethylene diamine. The functionalized graphene oxide was then attached to a highly hydrophilic water insoluble polymer (poly-acrylonitrile-co maleic-anhydride). The graphene oxide grafted poly acrylonitrile co maleimide (G-PANCMI) was used to prepare the dope solution. The hollow fibre ultrafiltration membranes were prepared by dry wet spinning.

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