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# Bio-based superhydrophilic foam membranes for sustainable oil–water separation†

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The development of a low-cost, high-performance, biobased membrane technology has been attempted to treat environmentally sensitive wastewater streams. Novel foam membranes (FMs) were derived from agarose (Agr) and gelatin (Gel) in combination with a non-toxic fruit extract and natural crosslinker, genipin (G). FMs were successfully tested for their oil–water separation efficiencies. FMs attained unique capillary microstructures (10–45 μm) as a result of the controlled lyophilization process, which allows selective permeation of water. Stable microporous membranes with nominal pore sizes in between the microfiltration and ultrafiltration range generated as high as >500 L $m^{-2}$ $h^{-1}$ continuous flux with ~98% pure product water. One of the advantages of these FMs is that after oil–water separation, they can undergo an easy membrane cleaning process, thereby retaining surface activity for long term performance.

## 1. Introduction

The last two decades have seen more oil spill incidences than ever before, except those in war-like (Gulf War, 1991) situations.[1] Increasing oil spill accidents pollute oceans on a daily basis, causing severe complications to ecosystems, which proves catastrophic to native marine wildlife in particular. Constant dissipation or scattering of oil into the marine environment over time has proven deadly to marine life such as fish, birds, invertebrates, mammals, reptiles, plants and algae. One accident could leave a huge quantity of oil being spilt in one place, inducing a long-lasting impact on the environment.[2–4] Moreover, numerous terrestrial activities, including industrial oily wastewater discharge, oil refineries, automotive industrial release, shipping travel, domestic drains and dumping, cause lasting impact on the day-to-day life of sweetwater reservoir.[5–7] Unending deep water horizon oil spill accidents and their aftermath prompts robust and cleaner approaches.[8] Furthermore, there are issues concerning stable emulsions, which pose serious challenges to meet the desired specifications for oil content in the product water or *vice versa*. The use of demulsifier additives to attain high quality water increases operational cost, in addition to the demand for the post recovery step.

On the other hand, researchers are constantly looking for new materials and techniques to solve problems that are threatening our environment. In a unique attempt, nanoporous PTFE was used for the separation of two immiscible liquids based on the segmented flow microchemistry principle. Selective wettability resulted in successful separation of aqueous-organic/fluorous liquid mixtures.[9] Recently, a detailed review has been reported highlighting advanced membranes used for separation of complex emulsified oil–water mixtures and effluents.[10] Ceramic membranes in the ultrafiltration (UF) and microfiltration (MF) range have been developed to allow successful separation of both immiscible effluents and complex emulsions. Among these, $Cu(OH)_2$ nanowire-haired membranes yielded flux as high as ~100 000 L $m^{-2}$ $h^{-1}$.[11] On the other hand, ultrafast separation of stable emulsions was achieved using carbon nanotube (CNT)-based membranes. With different sets of membranes >96% purity to oil was achieved.[12] A hydro-responsive membrane prepared with a simple, dip-coating technique. Polyester fabric dip-coated using an organosilicon (f-POSS) in combination with cross-linked poly(ethylene glycol) diacrylate (x-PEGDA) resulted in variable surface characteristics. As-prepared flexible fabrics were successfully tested for both oil-in-water and water-in-oil emulsion separation applications.[13]

In addition to this, conventional electrospun polyacrylonitrile (PAN)-based nanofibrous membranes[14] and solution-cast

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†Electronic supplementary information (ESI) available: Membrane swelling study results, long term performance in crossflow module and membrane surface generated images. See DOI: 10.1039/c4gc01070a