Abstract

In this study, a Carbon Nanotube (CNT) dispersion method which can be adopted during the synthesis of a CNT/polysulfone Mixed Matrix Membrane (MMM) resulting in optimal performance in the application of oil and water separation is described. Furthermore, the influence of the CNT particle size on MMMs performance is reported. The two aspects of CNT/polysulfone MMMs considered in this study were investigated in two parts. In the first part of this work, CNT/polysulfone MMMs with 5% multi-walled CNT loading and pure polysulfone membranes were synthesized using the phase inversion method, three CNT dispersion methods (Method 1, Method 2 and Method 3 were adopted during synthesis of the MMMs). Characterization of the membranes and CNT was carried out to examine the morphology, the surface chemistry as well as the hydrophilic properties. Scanning Electron Microscopy (SEM) micrographs depicted both porous cross sections and surface morphologies in MMMs and polysulfone membranes. However, the degree of CNTs dispersion within the polysulfone matrix was less pronounced for MMMs prepared using CNT dispersion Method 2. The functional groups observed for the MMMs and CNTs using Fourier Transform Infrared Spectroscopy (FTIR) spectra included carboxylic acid groups (O−H) and carbonyl group (C=O) which are responsible for hydrophilic properties. The contact angles measured for the MMMs using CNT dispersion Method 1, Method 2 and Method 3 were $76.6\pm5.0^{\circ}$, $77.9\pm1.3^{\circ}$ and $77.3\pm4.5^{\circ}$ respectively, while $88.1\pm2.1^{\circ}$ was measured for the pure polysulfone membranes. The oil rejection obtained for MMMs synthesized by adopting CNT dispersion Method 1, Method 2 and Method 3 were 48.71%, 65.86% and 99.88% respectively. The oil rejection obtained for the pure polysulfone membrane was 84.92%. The pure water and oil water flux increased with an increase in trans-membrane pressure for all the membranes. The oily water permeability of the MMMs

was 26.4 L m^{-2} h^{-1} bar^{-1}, 113 L m^{-2} h^{-1} bar^{-1} and 2.3 L m^{-2} h^{-1} bar^{-1} for MMMs prepared using CNT dispersion Method 1, Method 2 and Method 3 respectively, while the pure polysulfone membrane oily water permeability was 6.9 L m^{-2} h^{-1} bar^{-1} at 6.9 bar.

The competition between water and oil to occupy the porous sites of the membranes was confirmed by the low oil rejection result for M_4, indicating that the surface chemistry of the membranes favours the absorption of oil over water. The low rejection observed for M_1 and M_2 were attributed to poor CNT dispersion and formation of interfacial defects between the CNTs and polymer matrix, resulting in an increase in the free fraction volume that enhanced the permeation flux as well as the permeability. The adoption of CNT dispersion Method 3 has produced MMMs with exceptional oil rejection as well as good permeability.

In the second part of this work, CNT dispersion Method 3 was adopted during the synthesis of MMMs with different CNTs particle size while the CNT loading was kept at 5%. CNT I, OD 6-9 nm **x** L 5 nm, and CNT II, D 110-170 nm **x** L 5-9 μm , were used in the study. Characterisation techniques and performance evaluation methods used in the first part of this work were also employed in part two. FTIR spectra confirmed that the functional groups present in both the CNTs were similar and most importantly included carboxylic acid groups (O−H). The contact angles measured for MMMs from CNT I and CNT II were 77.3 ± 4.5° and 78.8 ± 5.6° respectively. The oil rejection obtained was 99.88% and 99.76% for CNT I MMMs and CNT II MMMs respectively. The oily water permeability was 2.11 L m^{-2} h^{-1} bar^{-1} and 2.20 L m^{-2} h^{-1} bar^{-1} for CNT I and CNT II respectively, at 8.28 bar. Fouling of CNT II MMMs was observed from a trans-membrane pressure at 9.66 bar which resulted in a decrease in permeability, while the oil permeation flux for CNT I MMMs increased with increasing TMP above 9.66 bar.

This study demonstrates that the CNT dispersion method employed during synthesis influences the performance of MMMs in oil-water mixture separation, as well as the hydrophilic properties, which was indicated by the differences in the MMM contact angles. An interesting observation was that the incorporation of CNT in CNT/polysulfone composites can also be detrimental to the performance of the MMM, which was illustrated by the poor oil rejection obtained for MMMs prepared using CNT dispersion Method 1 compared to the pure polysulfone results. Furthermore, the CNT particle size has been shown to influence the performance of CNT/polysulfone MMMs. Increasing the particle size of the CNTs resulted in improved oil permeation flux as well as permeability at low pressure, however at a TMP above the 8.28 bar, MMMs with bigger CNTs foul more rapidly. The best CNT dispersion can be obtained in CNT/polysulfone MMMs by employing CNT dispersion Method 3 during synthesis and using small CNT diameters.