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ACCEPTED MANUSCRIPT

Reducing ageing of thin PTMSP films by incorporating graphene and graphene oxide: effect of thickness, gas type and temperature

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ABSTRACT

It was previously proven that small loadings of few-layer graphene (G) and monolayer graphene oxide (GO) adjust the permselectivity and reduce the ageing of thick poly(trimethyl silyl propyne) (PTMSP) membranes, which currently inhibits their application in real separations. In this work, we extend the analysis to thin film composite membranes with top layers of PTMSP/G and PTMSP/GO between 1 and 7 micrometers. The good quality of the films, obtained by spin coating the polymer solution on porous flat supports, indicates that graphenic sheets are aligned parallel to the film surface, without defects, and that the large-scale production of such composite films is feasible. The addition of G and GO to PTMSP can improve the gas permeability and selectivity, and even change the behaviour from CO_2-selective to He-selective. GO produces more repeatable effects than graphene, and it generally enhances the PTMSP permeability.

The ageing was studied by tracking permeability of 4 gases with time: thin films age faster than thick ones, because ageing is due to free volume diffusion across the film. Incorporation of 1 wt% of G and GO visibly reduces the ageing of thin PTMSP films, and the effect is repeated on many different samples. The ageing can be correlated, with a single power law, to the ratio between time and squared film thickness. The exponent of such function, that quantifies the ageing rate, decreases when G and GO are added to PTMSP thin films, by factors as high as 40%.

The temperature effect was studied, up to 60°C, on annealed samples: the membranes, CO_2-selective at room temperature, can become easily He-selective by raising the temperature. The He/CO_2 selectivity increases with temperature: such effect could be exploited syngas purification and pre-combustion capture processes.

Keywords: PTMSP; graphene; ageing; gas separation; thin films.

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