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## Enhancement of CO<sub>2</sub>/CH<sub>4</sub> separation performances of 6FDA-based co-polyimides mixed matrix membranes embedded with UiO-66 nanoparticles

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### Abstract

Metal-organic frameworks (MOFs) incorporation into mixed matrix membranes (MMMs) is gaining more attention due to the combined advantages of high separation performance and easy processability. Nanoparticles (NPs) of CO<sub>2</sub>-philic MOF UiO-66 (Zr-BDC) were synthesized with high surface area and ca. 50 nm particle size (and also for comparison with 100 and 200 nm sizes). They were incorporated into three 6FDA-based co-polyimides (namely 6FDA-BisP, 6FDA-ODA, and 6FDA-DAM), forming MMMs with loadings in the 4 – 23 wt.% range. The NPs and MMMs were characterized accordingly by XRD, BET, SEM, TEM, FTIR, and TGA. CO<sub>2</sub> and CH<sub>4</sub> isotherms on the NPs were measured by a static volumetric method at the pressure up to 10 bar. Fractional free volume (FFV) was calculated using solid density, measured by pycnometer. Gas separation performance was evaluated using a feed composition of 50%:50% CO<sub>2</sub>:CH<sub>4</sub> binary mixture at 35 °C and a pressure difference of 2 bar. The presence of UiO-66 NPs in the continuous 6FDA-BisP and 6FDA-ODA co-polyimides improved both CO<sub>2</sub> permeability and CO<sub>2</sub>/CH<sub>4</sub> selectivity by 50 – 180% and 70 - 220%, respectively. In the case of 6FDA-DAM MMMs, the CO<sub>2</sub> permeability was significantly improved by 92%, while maintaining the CO<sub>2</sub>/CH<sub>4</sub> selectivity. The best results in terms of CO<sub>2</sub>/CH<sub>4</sub> selectivity were 41.9 for 6FDA-BisP (17 wt.% filler loading, 108 Barrer of CO<sub>2</sub>), 57.0 for 6FDA-ODA (7 wt.% filler loading, 43.3 Barrer of CO<sub>2</sub>) and 32.0 for 6FDA-DAM (8 wt.% filler loading, 1728 Barrer of CO<sub>2</sub>). The study confirmed the UiO-66 NPs incorporation into these co-polyimides has brought the improvement of the dense membranes, without jeopardizing their positive attributes.

**Keywords:** Gas separation, 6FDA-based co-polyimide, Metal-organic framework, UiO-66, Mixed matrix membrane.

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