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### **ACCEPTED MANUSCRIPT**

## **Block Copolymer Membranes with Catecholic Bolaamphiphile Assemblies**

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#### **Abstract**

We report the first use of the assemblies of catecholic bolaamphiphilic compound, i.e., *bis-* (*N*-α-amido-3,4-dihydroxyphenylalanine)-1,7-heptane dicarboxylate (DOPA-C7), as an organic filler for gas separation membranes. Specifically, the membranes were prepared by adding the assembly fillers to a block copolymer matrix, i.e., nonpolar polystyrene-*b*-polybutadiene-*b*-polystyrene (SBS) or polar poly (ether-*b*-amide) (PEBAX). The interaction between the filler and matrix was investigated regarding the structure, morphology, and CO<sub>2</sub>/N<sub>2</sub> separation performance. The strongly interacting PEBAX/DOPA-C7 membranes showed a typical trade-off behavior, i.e., a decrease in CO<sub>2</sub> permeability and increase in CO<sub>2</sub>/N<sub>2</sub> selectivity with the filler contents. In contrast, the weakly interacting SBS/DOPA-C7 interestingly showed an improved CO<sub>2</sub>/N<sub>2</sub> selectivity, from 14.1 to 21.1, with a slight increase in CO<sub>2</sub> permeability (from 347.5 to 349.7 Barrer) owing to the catechol group of DOPA-C7 that can function as a Lewis base. This indicates that very strong interactions between the polymeric matrix and filler could have a negative impact on the gas separation performance. This work not only explores the importance of a polymer matrix, but also opens

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