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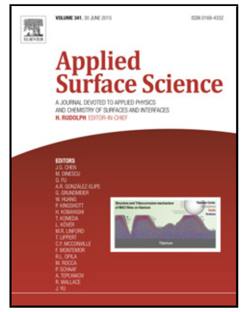

Title: Theoretical investigation of gas separation in functionalized nanoporous graphene membranes

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<AT>Theoretical investigation of gas separation in functionalized nanoporous graphene membranes

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<ABS-Head><ABS-HEAD>Graphical abstract
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<ABS-HEAD>Highlights► The capability of functionalized graphene for gas separation was computationally investigated; ► The novel mass-transport mechanism of the graphene membranes were revealed; ► The graphene membranes have great potential applications in the flue gases and natural gas processing.
<ABS-HEAD>ABSTRACT

<ABS-P>Graphene has enormous potential as a membrane-separation material with ultrahigh permeability and selectivity. The understanding of mass-transport mechanism in graphene membranes is crucial for applications in gas separation field. We computationally investigated the capability and mechanisms of functionalized nanoporous graphene membranes for gas separation. The functionalized graphene membranes with appropriate pore size and geometry possess excellent high selectivity for separating CO_2/N_2, CO_2/CH_4 and N_2/CH_4 gas mixtures with a gas permeance of ~10^3–10^5 GPU, compared with ∼100 GPU for typical polymeric membranes. More important, we found that, for ultrathin graphene membranes, the gas separation performance has a great dependence not only with the energy barrier for gas getting into the pore of the graphene membranes, but also with the energy barrier for gas escaping from the pore to the other side of the membranes. The gas separation performance can be tuned by changing the two energy barriers, which can be realized by varying the chemical functional groups on the pore rim of the graphene. The novel mass-transport mechanism obtained in current study may provide a theoretical foundation for guiding the future design of graphene membranes with outstanding separation performance.
<KWD>Keywords: graphene; density functional theory; molecular dynamics; gas separation; diffusion barrier

<H1>1. Introduction
Graphene, as a two-dimensional sheet of sp^2-bonded carbon atoms, is the thinnest membrane (3.4 Å), which makes it promising for potential application as a gas membrane-separation material, because its infinitesimal thickness promises transport resistance minimization and flux maximization.[1-4] Because perfect graphene is impermeable, nanoporous graphene (graphene-related materials with nanopores in the plane) is used to achieve gas permeability.[5, 6] Recently, theoretical and experimental work has suggested that such nanoporous graphene