PÔSTER - RESUMO - SENSORES E BIOSSENSORES

STACKING-ENGINEERED HYDROGEN-SUBSTITUTED GRAPHDIYNE AS A HIGH-CAPACITY ADSORBENT AND SENSOR FOR PERSISTENT PFAS AND TOXIC GASES

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Abstract

The discovery of graphene revolutionized nanoscience due to its exceptional mechanical strength, thermal conductivity, and unique electronic properties.[1] This breakthrough spurred investigation into other two-dimensional materials, such as graphdiynes,[2] featuring acetylenic linkages that enhance electronic and mechanical performance. Graphdiyne, composed of sp- and sp^2-hybridized carbon atoms, creates larger pores facilitating molecular diffusion, advantageous for catalysis, energy storage, and environmental sensing. Recently, hydrogen-substituted graphdiyne (HsGDY) was experimentally synthesized by Zhang et al. via alcohol-thermal method, exhibiting AA-stacked configuration with ordered porous channels.[3] Fabris et al. studied its structural, electronic, and dynamical stability properties across stacking conformations (AA, AB, ABC), confirming AA as the most stable with moderate bandgap (~0.89 eV).[4]

To explore HsGDY's potential as sensor and adsorbent for toxic contaminants, we performed Density Functional Tight-Binding (DFTB+) simulations focusing

on adsorption of persistent PFAS (PFOA, PFNA) and toxic gases (NO2, SO2, H2S, HCN, CO). Calculations reveal moderate-to-strong adsorption energies and significant work function shifts (?WF), alongside DOS modifications near the Fermi level—ideal for resistive or capacitive detection. Retention capacity is evidenced by surface saturation: in a (2×2) supercell, high PFAS loadings achieve densities ~1.38 molecules/nm² for PFOA, for example, comparable to other 2D/3D carbon materials like functionalized graphene (~0.5-1.5 molecules/nm²) and GO (~3070 mg/g), and superior to porous carbons (~100-900 mg/g). Saturated gas coverages highlight NO2/SO2 selectivity with viable recovery at elevated temperatures. HsGDY's ordered porosity enables rapid diffusion, while hydrogenation opens bandgap (~1-2 eV), enhancing response. These findings position AA-stacked HsGDY—thermodynamically stable (AA > ABC > AB)—as promising for PFAS/toxic gas monitoring and remediation, aligning scalable synthesis with computational predictions.

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Palavras-chave: graphdiyne; pfas; adsorption; dftb; environmental sensors.