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CO₂ adsorption on N-doped graphene oxide

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Introduction. Among the anthropogenic greenhouse gases (CO₂, CH₄, NO_x), CO₂ is the main because of its highest concentration in the atmosphere. Aqueous amine solutions is a technology widely used for CO₂ uptake, however, the high cost to regenerate the solutions is a disadvantage.¹ This problem can be overcome using adsorption process.^{2,3} In this work, we synthesized a nitrogen-doped porous graphene oxide (N-graphene, Fig. 1 (a)) and investigated its adsorption capacity towards CO₂ and H₂. The CO₂ adsorption capacity of N-graphene was compared with the results obtained for MCM-41 and ZIF-8 commercially obtained. N-graphene was prepared from a hydrothermal synthesis containing a mixture of GO and polyalilamine, followed by chemical activation at 800 °C with KOH.

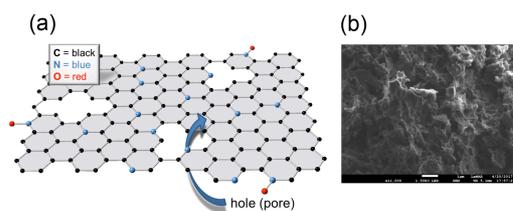


Figure 1. (a) N-graphene and its (b) SEM images.

Characterizations. High-resolution XPS C1 showed the presence sp² carbon, epoxy, carbonyl and carboxylic acids at 284.66, 286.38, 287.52 and 288.80 eV, respectively. The high-resolution XPS N1 spectrum exhibited the pyridinic (399.72 eV), and grafitilic (401.58 eV), showing successful an introduction of nitrogen atoms. N₂ adsorption/desorption experiments revealed type IV isotherms for the N-graphene and MCM-41, indicating the presence of mesoporous. ZIF-8 showed type I isotherm, indicating the presence of microporous. Surface areas of ZIF-8, N-graphene and MCM-41 were 1534, 1155, and 856 m² g⁻¹. The large surface area of the N-graphene is due to the

activation process that removed carbon atom from within graphene structure, generating pore in the material. The pore structure of N-graphene was confirmed by SEM image (Fig. 1 (b)).

Adsorption Experiments. CO₂ isotherms, at 25 °C, Fig. 2 (a), showed higher CO₂ adsorption for the N-graphene in all range of the pressure compared to MCM-41 and ZIF-8, suggesting CO₂ loading does not depend only on the surface areas of the materials. The values of isosteric heats (Q_{st}) obtained for N-graphene were between -72 and -27 kJ mol⁻¹, Fig. 2 (b). These values suggest the existence of strong interactions between CO₂ molecules and nitrogen atoms into N-graphene. The low Q_{st} values reported for MCM-41 (-20 to -16 kJ mol⁻¹)¹ and ZIF-8 (-30 to -18 kJ mol⁻¹)³ are characteristics of weak interactions. Reusability experiments showed N-graphene is quite stable under various adsorption/desorption cycles.

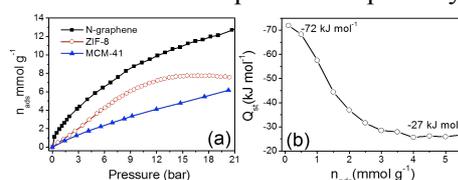


Figure 2. (a) CO₂ isotherms of N-graphene, ZIF-8 and MCM-41 at 25 °C. (b) Q_{st} for N-graphene in different CO₂ loaded.

Conclusion. The insertion of nitrogen atom into the structure of graphene was confirmed by XPS technique. CO₂ adsorption experiments showed higher CO₂ adsorption for N-graphene than that for MCM-41 and ZIF-8, which is due to high surface area and Q_{st} of N-graphene.

Acknowledgments

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