

Understanding the limits of hydrogen storage capacity achieved by physisorption in nanostructured graphene-based materials

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Carbon-based materials proposed so far as hydrogen storage media (graphite intercalated with fullerenes, carbon foams, nanotube bundles, *etc.*) perform only modestly. Their hydrogen uptake is usually within ~3.0–6.0 wt.% at 77 K [1]. For comparison, metal–organic frameworks (MOFs) are able to physisorb ~10 wt % of molecular hydrogen at similar conditions [2]. Keeping in mind that physisorption in MOFs takes place mainly on aromatic rings of organic ligands (the amount of metal centers per unit volume is very small and therefore has negligible influence on the overall gas uptake), we would like to overview different ways of how to improve the performance of graphene-based nanostructures for hydrogen storage applications.

In the last years much emphasis was put on graphene-oxide (GO) frameworks (GOFs, alternatively called pillared graphene-oxide) [3, 4]. Simulations on model systems gave promising results, namely ~5.5 wt % (at 25 bar and 77 K). However, the mechanical stability of GOFs crucially depends on the regular arrangement of “pillars” (usually, anions of dicarbonic acids) interconnecting GO sheets. Unfortunately, our results have shown that – in order to make GOFs robust with respect to “collapsing” by increasing the number of pillars in between the layers – GOFs have to become essentially non-porous (with surface areas nearly ~200 m²/g) and therefore useless as adsorbents. The results of our simulations correlate with recent experimental findings, reporting twice smaller uptake (~2 wt % at 77 K and 20 bar) [4] than that calculated for an idealized structure model.

Recently, we proposed stable and porous carbon nanotube (CNT) matrices which are able to adsorb up to 7 wt % (20 bar, 77 K) of hydrogen while the total uptake could reach 17-19 wt % (at 77 K and 100 bar) [5]. These CNT matrices could be put in between the graphene sheets, giving rise to the specific graphene-CNT composites with promising hydrogen storage properties. However, hydrogen uptake in these composites is mainly determined by the CNT matrix; graphene-(oxide) is only used to “confine” CNTs in the desired packing.

Finally, we will discuss the influence of the defects in graphene sheets on the hydrogen storage capacity and will provide the final answer to the question: “Is it really possible to go beyond 6-7 wt% (excess) of hydrogen uptake at 77-100 K in graphene-based nanostructures”?

References

- [1] K. Spyrou, D. Gournis, P. Roudolf, *ECS Journal of Solid State Science and Technology*, **2**, M3160 (2013).
- [2] O. K. Farha, A. Ö. Yazaydin, I. Eryazici, C. D. Malliakas, B. G. Hauser, M. G. Kanatzidis, S. T. Nguyen, R. Q. Snurr, J. T. Hupp, *Nat. Chem.* **2**, 944 (2010).
- [3] J. W. Burress, S. Gadipelli, J. Ford, J. M. Simmons, W. Zhou, T. Yildirim, *Chem. Commun.*, 49, 8902 (2010).
- [4] R. Kumar, V. M. Suresh, T. K. Maji, C. N. R. Rao, *Chem. Commun.*, **50**, 2015 (2014).
- [5] B. Assfour, S. Leoni, G. Seifert, I. A. Baburin, *Advanced Materials* **23**, 1237 (2011).