

Computer simulation of the role of palladium doping on the hydrogen storage capacity of nanoporous carbons

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Hydrogen adsorption on nanoporous carbon materials is one of the most promising technologies for hydrogen storage. However pure carbon materials do not meet the technological requirements due to the small adsorption energy of hydrogen to the graphitic walls of the pores. Experimental work has shown that doping with Pd atoms and clusters enhances the storage capacity of porous carbons. Therefore, it is of high interest to unravel the role played by the Pd dopant on the enhancement mechanisms. We have shown [1] that hydrogen adsorbs on Pd clusters deposited on pristine graphene following two channels i) molecular adsorption and ii) dissociative chemisorption, with moderately high adsorption energies, a feature that would justify the observed enhancement in the storage capacity of Pd-doped porous carbons. However, looking at the desorption step of the storage cycle we noticed [2] that desorption of Pd-H complexes competes with desorption of hydrogen. Desorption of Pd-H complexes would spoil the beneficial effect of the dopant. As a way to overcome this difficulty, we proposed to anchor the Pd atoms and clusters to defects of the graphene layer, for instance to graphene vacancies. Our results on the molecular adsorption and dissociative chemisorption of hydrogen on Pd atoms and small Pd clusters anchored on graphene vacancies are very promising because the Pd clusters anchored to vacancies retain their capacity of adsorbing hydrogen and completely prevent the desorption of Pd-H complexes, that was a problem in the case of Pd clusters supported on pristine graphene.

Keywords: Hydrogen; Storage; Porous Carbons; Doping

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