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Hydrogen sorption by single-walled carbon nanotubes prepared by a torch arc method

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Abstract

Single-walled carbon nanotubes (SWCNTs) were prepared by a novel torch arc method operated in open air and isotherms of hydrogen sorption were volumetrically measured at 295 and 77 K under pressures below 110 kPa. The averaged diameter of SWCNTs was 1.32 nm, as measured by high-resolution transmission electron microscopy, and the dominant presence of the (9,9) armchair nanotubes accompanied by the (10,10) armchair nanotubes was suggested by the breathing mode Raman scattering. The maximum hydrogen concentrations reached 0.932 wt.% at 295 K under 106.7 kPa and 2.37 wt.% at 77 K under 107.9 kPa with the hydrogen uptakes not yet saturated. It is likely that the sorbed hydrogen molecules resided on the exterior surfaces of SWCNTs. The higher surface area of SWCNTs by a factor of 10 as compared to the graphitic raw material would play a dominant role for hydrogen sorption to bring about the higher hydrogen wt.% by a factor of 10. The high graphitic nature of SWCNTs would also play an important role, but the observed surface H/C ratio, 0.680 at 77 K under 100 kPa, exceeded the expected value, 0.333, for the commensurate adsorption structure, and the uppermost value, 0.49, for the densest monolayer, which suggested the presence of unknown sorption mechanism(s) other than physisorption on carbon hexagons. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

The necessity of lightweight hydrogen storage materials has been keenly felt these years. According to Chahine and Benard [1], a hydrogen density of 6.5 wt.% is required for an electric vehicle powered by a hydrogen fuel cell to achieve a range of 500 km. Before the finding of ‘Laves phase related BCC solid solution’, conventional hydrogen storage alloys have not reached more than 2 wt.% of hydrogen at room temperature, as reviewed by Akiba and Iba [2]. At slightly higher temperatures (>423 K), metal-doped sodium aluminum hydride, NaAlH₄, reversibly store hydrogen and 4.2–3.1 wt.% of reversible hydrogen capacities have been achieved during 35 dehydrogenation–rehydrogenation cycles [3,4]. The low reaction kinetics has been improved by mechanically grinding the hydride with an addition of carbon [5], but relatively high temperatures

(>423 K), high hydrogenation pressures (6–15 MPa) and cyclic instability have not significantly improved.

Before the discovery of a plausibly high hydrogen storage density in single-walled carbon nanotubes (SWCNTs) at room temperature and 41 kPa [6], hydrogen sorption by carbons have been studied mostly at cryogenic temperatures. Substantial amounts of chemisorption was first found at temperatures above 200 K for alkali metal–graphite intercalation compounds [7,8], and then significant physisorption was observed at lower temperatures, 77–113 K [9,10]. The hydrogen capacities of these compounds are so small as exemplified by C₈KH_{0.67} [11], C₂₄KH₄ [10,12] and C₄KH_{0.67–0.8} [13,14], that they are not applicable to hydrogen storage. Hydrogen adsorption on activated charcoals was first studied in an engineering point of view [15], and hydrogen capacities began to be compared favorably with a compressed gas system at 40 MPa (4.7 wt.%) [16,17]. While Chahine and Bose claimed that lowering the storage temperature from 298 to 77 K would increase the adsorption by a factor of 15 [18] and that the application of adsorption of hydrogen on activated

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