



ORIGINAL RESEARCH

Recent advances in hydrogen storage technologies based on nanoporous carbon materials

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Abstract Hydrogen is a promising energy carrier that can potentially facilitate a transition from fossil fuels to sustainable energy sources without producing harmful by-products. Prior to realizing a hydrogen economy, however, viable hydrogen storage materials must be developed. Physical adsorption in porous solids provides an opportunity for hydrogen storage under low-stringency conditions. Physically adsorbed hydrogen molecules are weakly bound to a surface and, hence, are easily released. Among the various surface candidates, porous carbons appear to provide efficient hydrogen storage, with the advantages that porous carbon is relatively low-cost to produce and is easily prepared. In this review, we summarize the preparation methods, pore characteristics, and hydrogen storage capacities of representative nanoporous carbons, including activated carbons, zeolite-templated carbon, and carbide-derived carbon. We focus particularly on a series of nanoporous carbons developed recently: metal–organic framework-derived carbons, which exhibit promising properties for use in hydrogen storage applications.

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1. Introduction

The pace of fossil fuel consumption, particularly oil and gas, has increased in recent decades, which poses several challenges to our energy systems, including dwindling finite resources and the production of environmentally harmful by-products, especially carbon dioxide emissions [1–4]. These challenges give rise to strong demands for more efficient energy sources. To address the problems of global warming, pollution, and fossil fuel resource depletion, a next-generation energy source must overcome the stringent criteria of renewability, environment-friendly

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by-products, and abundance. The use of hydrogen as an energy carrier has been widely discussed because it stores the highest energy density by weight, it can be produced from a variety of sources (e.g., water, biomass, and organic materials) [1,2], and its by-product is pure water. The realization of a “hydrogen economy” in the near future is considered to be a possible solution to the looming energy crisis.

Hydrogen, a tasteless, colorless, and nontoxic molecule, provides a clean combustion resource that generates neither air pollutants nor greenhouse gases. Hydrogen has a very high gravimetric energy density of 120 kJ g^{-1} , about three times the energy density of gasoline and almost seven times the energy density of conventional fossil fuels [2]. The main concern associated with realizing a “hydrogen economy” is the very low volumetric energy density of hydrogen: moving a vehicle requires a far larger fuel tank than typical gasoline-fueled vehicles. Hydrogen storage and transport is a major bottleneck in the development of hydrogen-based technologies, e.g., fuel cells [1–6]. The development of viable technologies and materials for the effective, safe, and stable storage of hydrogen constitutes a crucial step toward realizing a “hydrogen economy”.

Several approaches to hydrogen storage are available: (i) high-pressure tanks, (ii) cryogenic liquefaction of molecular hydrogen, (iii) chemical solid storage materials, and (iv) physically adsorbing porous materials [1,2,7]. High-pressure tanks require pressures of 350–700 bar for hydrogen compression, however, even at such high pressures, the energy density is below that of conventional energy sources. The storage tanks must be prepared from strong lightweight materials that can withstand pressurized conditions, thereby increasing the application costs. The cryogenic liquefaction of hydrogen has been considered as a viable on-board storage option for the automotive industry; however, hydrogen liquefaction occurs only at very low temperatures (20 K). Ambient heat flow into the storage tank readily evaporates hydrogen, thereby increasing the internal pressure of the tank and causing fuel loss (boil-off) and safety challenges. The chemical storage of solid materials is safer than compression or liquefaction and is based on dissociative hydrogen adsorption to the solid metals (usually lithium, magnesium, or aluminum) [1,2]. This method provides a high gravimetric hydrogen storage capacity at ambient temperatures and pressures. Unfortunately, desorption of hydrogen from these materials suffers from a large endothermic energy barrier (MgH_2 : $-75.3 \text{ kJ mol}^{-1}$), thus requiring unacceptably high temperatures to release the hydrogen, which reduces the energy efficiency (poor release kinetics and storage material recycling) [1,2]. Physically adsorbing porous materials provide another option for safe hydrogen storage without thermodynamic energy inefficiencies. One challenge is that the adsorption processes rely on van der Waals interactions, which are intrinsically low-energy. Nonetheless, most current efforts toward the development of hydrogen storage materials have focused on the design of nanomaterials that reversibly store hydrogen molecules at temperatures approaching ambient conditions [8–10].

Until recently, hydrogen storage methods that rely on physically adsorbing materials have maximized the surface textural characteristics of the adsorbent. In general, the hydrogen storage capacities that utilize physical adsorption alone appear to be governed by the specific surface area (SSA)

and pore characteristics (pore shape and size distribution). Possible target materials that satisfy these requirements include microporous materials, such as activated porous carbons [11], zeolites [12], porous polymers [9], or metal–organic frameworks (MOFs) [10]. Among these candidate materials, highly porous adsorbents based on carbonaceous materials have attracted considerable interest and have been extensively investigated due to their attractive features, including a high SSA, good thermal and chemical stabilities, and mass production (they are easily synthesized from a variety of organic precursors) [5,11,13]. Importantly, carbonaceous adsorbents are lightweight and provide excellent kinetics and a very high SSA. They have, thus, attracted extensive attention for use in hydrogen storage applications.

The scope of the discussion in the present review is limited to recent advances in hydrogen storage using porous carbonaceous adsorbents that have received significant attention from the hydrogen storage research community. The prominent carbonaceous adsorbents are categorized as follows: (1) carbon nanomaterials, carbon nanotubes and fullerene, (2) zeolite-templated carbons (ZTCs), and (3) carbide-derived carbons (CDCs), and the preparation methods, pore characteristics, and hydrogen storage behaviors are described. In the following section we introduce the recently developed MOF-derived carbons (MDCs) that exhibit promising H_2 storage capacities better than those of previously reported porous adsorbents. We concentrate on the preparation and pore characteristics of MDCs to provide guidelines for the design of suitable porous carbon materials for hydrogen storage.

2. Carbon materials for hydrogen storage

2.1. Carbon nanomaterials

Numerous reports have described the uses of various carbon nanomaterials, including single- and multi-walled CNTs, nanohorns, graphite nanofibers, graphene, and fullerenes, as efficient hydrogen storage media [14–18]. Among these, CNTs may satisfy the criteria for on-board hydrogen storage materials set by the US Department of Energy (DOE). Dillon et al. [14] and Chambers et al. [17] reported that SWCNTs and graphite nanofibers store, respectively, 10 wt% and 67 wt% hydrogen at ambient temperatures. Hydrogen storage in carbon nanomaterials, particularly CNTs, has since been further optimized using theoretical and experimental approaches by various research groups. Since the beginning of 2000, however, reports critical of the hydrogen storage capacities of carbon nanomaterials began to emerge [18]. Tibbetts et al. conducted independent investigations of the hydrogen storage properties of several carbon nanomaterials, stating in their article: “we believe that in all cases so far claims of more than 1 wt% at room temperature are erroneous and due simply to experimental error” [15]. It is now widely accepted that the previous spectacular results arose from water impurities and erroneous instrumentation (see Fig. 1) [15,18]. Nonetheless, theoretical approaches have suggested that transition metal atom-doped graphene and fullerene may have the potential to store hydrogen at room temperature [19–21]; thus, these materials are competitive candidates for hydrogen storage applications.

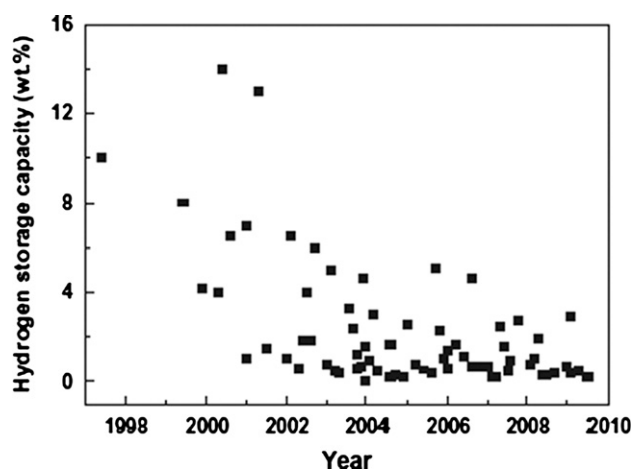


Fig. 1 A plot of the reported hydrogen storage capacities of CNTs from the literature versus their year of publication. Reprinted with permission from [18].

2.2. Activated carbons (ACs)

Activated carbons (ACs) are highly porous, amenable to large-scale preparation, and very stable to chemicals and heat. For these reasons, ACs are considered to be the most commercializable targets for hydrogen storage applications. ACs can be synthesized from a variety of organic precursors, including agricultural wastes, such as coconut shells and fibers, jute fibers, nut shells, soybeans, and oil seeds [13,22–24]. Typical synthetic methods include the generation of highly porous structures via physical or chemical activation [11]. Physical activation involves the gasification of carbonaceous materials in the presence of suitable oxidizing agents, CO_2 and steam. In chemical activation, porosity is generated via chemical reactions between reagents (KOH is most widely used) and carbonaceous materials: $6\text{KOH} + 2\text{C} \leftrightarrow 2\text{K} + 3\text{H}_2 + 2\text{K}_2\text{CO}_3$, followed by the production of CO_2 from the decomposition of K_2CO_3 , thereby generating further porosity [11]. The chemical activation method has several advantages over the physical activation method, including a lower processing temperature, shorter processing time, and higher porosity. Chemical activation was, thus, adopted to improve the porosities of various carbonaceous materials [11,22–24].

Pores are classified according to size, and are usually divided into three groups: (i) macropores, with an average pore diameter exceeding 50 nm, (ii) mesopores, with an average pore diameter in the range of 2–50 nm, and (iii) micropores, with an average pore diameter less than 2 nm. Micropores are further divided into supermicropores (0.7–2.0 nm) and ultramicropores (<0.7 nm). Typical ACs contain mostly micropores with a very high SSA (>3000 m^2/g based on BET measurements) and a very high total pore volume (>1.5 cm^3/g) [5,11,25]. The adsorption potentials on the pore walls overlap, yielding a relatively high adsorption energy. The preponderance of micropores in ACs is, therefore, believed to positively influence hydrogen storage applications.

Cazorla-Amorós et al. prepared a series of ACs from various coals (anthracite and bituminous coal) activated by KOH treatment [25]. The resulting materials exhibited very high BET SSAs exceeding 3000 m^2/g with high micropore volumes greater than 1 cm^3/g . In this work, KUA5 showed a

very high hydrogen storage capacity at room temperature (298 K) and 200 bar (1.2 wt%) whereas KUA6 displayed an excellent cryogenic (77 K) hydrogen storage capacity at 40 bar (5.6 wt%). This work pointed out that the micropore evolution and narrow pore size distributions via KOH activation played an important role in the hydrogen storage capacity (at both 77 and 298 K). Gao et al. applied both physical and chemical activation methods to commercial ACs, showing that unlike CO_2 activation (physical), KOH activation (chemical) remarkably altered the pore structures of the ACs by promoting the development of very narrow micropores (<1 nm) [11]. The resultant AC-K5 displayed hydrogen uptakes of 2.49 wt% at 77 K and 1 bar or 7.08 wt% at 77 K and 20 bar that were among the largest values reported for porous carbons. Considering that the BET SSA of AC-K5 was lower than the SSA of other ACs, such as KUA6 and Maxsorb-3000, a hydrogen storage capacity of 7.08 wt% demonstrated that the number of narrow micropores (<1 nm) was critical for determining the hydrogen storage capacity [8,11,26].

2.3. Zeolite-template carbons (ZTCs)

As an alternative to the disordered pore structures of ACs, microporous carbons with highly ordered regular pore arrays have been produced using hard templating methods and inorganic frameworks [27]. The porosities of carbons may be controlled using hard templating materials, such as zeolites and aluminosilicate materials, which are attractive due to their ordered and uniform interconnecting sub-nanometer pore channels that are readily accessible to a variety of carbon precursors. Kiyotani et al. described the typical fabrication of zeolite-templated carbon (ZTC) as follows [28]: (i) a carbon precursor was incorporated into the pores and channels of selected zeolites via either chemical vapor deposition (CVD) or liquid impregnation. (ii) Polymerization, followed by carbonization of the incorporated carbon precursors was conducted. (iii) After carbonization, the used zeolite template was completely removed by successive washing with strong acids, such as hydrochloric acid, as shown in Fig. 2a.

The obtained microporous ZTCs exhibited high BET SSAs (>2000 m^2/g). Unlike the amorphous morphologies of ACs, ZTCs displayed morphologies similar to those of the original zeolite template particles, as observed using scanning electron microscopy (SEM, Fig. 2b) [28,29]. The XRD patterns of the ZTCs generally demonstrated that the long-range ordered structures of the zeolite template were replicated in the ZTC [29,30]. The excellent ordered structures permitted characterization of the thin carbon walls by transmission electron microscopy (TEM). The textural characteristics of the ZTCs included a very high micropore volume (>1 cm^3/g) [28–30], suggesting that a large fraction of the pores in the carbon were in the micropore range. Another textural feature of the ZTC is that most of the micropores consisted of pores with diameters between 1 and 2 nm. The high structural regularity of the pores in the ZTCs was assured by the large pores (>0.6–0.7 nm) of the selected zeolite. Consequently, most reported ZTCs with higher structural ordering yielded a pore size distribution 1–2 nm in width, despite the fact that pore size could be reduced by adopting zeolite with smaller pores [30,31].

Mokaya et al. prepared microporous ZTCs via impregnation with furfuryl alcohol, followed by carbonization and further

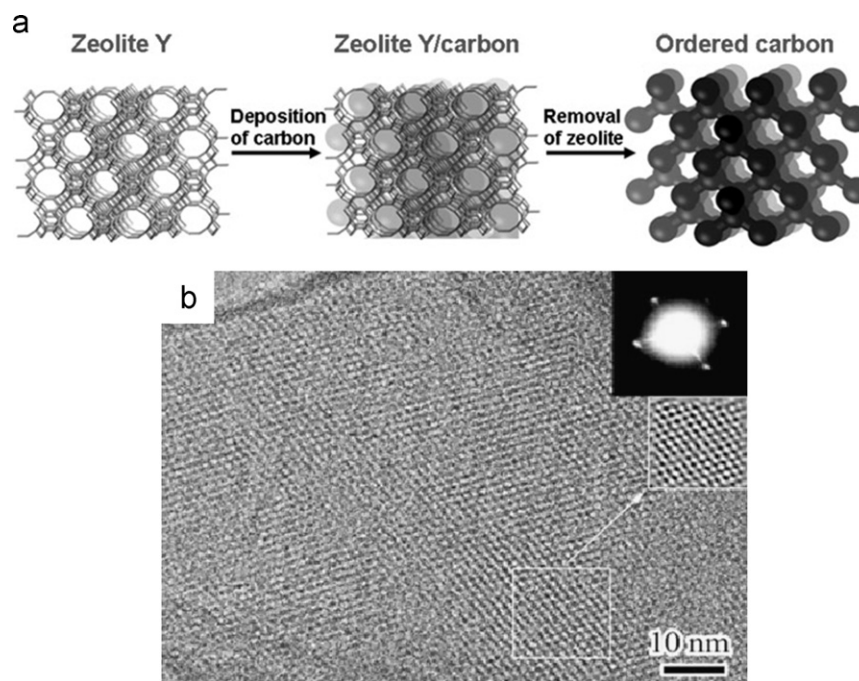


Fig. 2 (a) Schematic explaining the overall template synthetic procedure for microporous carbons using a zeolite Y template. (b) High-resolution transmission electron microscopy (HRTEM) image of the ordered microporous carbon prepared following the procedure reported. The inset corresponds to a diffraction pattern taken from this image. Reprinted with permission from [27].

carbonization through CVD methods under an acetonitrile flow in argon [30]. The obtained ZTCs displayed high BET SSAs ($>2000 \text{ m}^2/\text{g}$) and micropore volumes ($>0.8 \text{ cm}^3/\text{g}$) concurrently with nitrogen atom doping. The ZTCs exhibited a hydrogen uptake of 5.4 wt% at 77 K and 20 bar. Kyotani et al. introduced ultramicropores into the ZTCs using chemical activation methods to obtain highly porous activated ZTCs (CA850, Ac-ZTC1, and P7(2)-H displayed BET SSAs of 3189, 3064, and $3800 \text{ m}^2/\text{g}$, respectively) [29,32]. The resultant activated ZTCs displayed enhanced H_2 storage uptakes due to the evolution of ultramicropores, which was beneficial for hydrogen storage.

2.4. Carbide-derived carbons (CDCs)

The selective etching of metal atoms from metal carbides leads to the formation of porous carbon with a density lower than that of graphite, called carbide-derived carbon (CDC) [33]. CDCs were first thought to be an undesirable by-product; however, Gogotsi et al. identified a useful application of CDCs as molecular sieves, gas storage adsorbents, catalysts, and supercapacitors [33–35]. This group developed mass production methods for CDCs. CDCs were typically produced by the chlorination of metal carbide according to the reaction: $\text{MeC}_x + x/2\text{Cl}_2 (\text{g}) = \text{MeCl}_x + \text{C}$, where Me indicates an extracted metal atom (see Fig. 3). The carbide used during processing (such as SiC, TiC, ZrC, B_4C , TaC, and Mo_2C) and the heating temperature during the chlorination process significantly affected the microstructures of the obtained CDCs [33–35].

The characteristics of CDCs are unique in their pore structure, which is tunable at the atomic level, and their narrow pore size distribution [33,34]. Unlike ACs and ZTCs, well-controlled CDCs exhibit a high ultramicropore volume

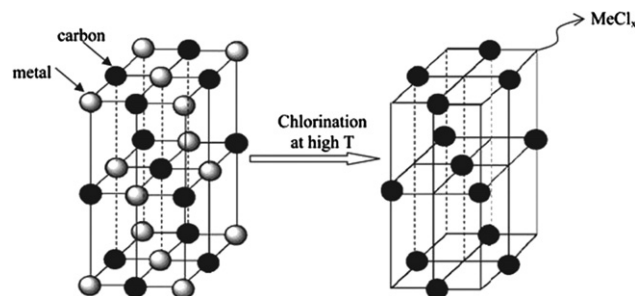


Fig. 3 Schematic view of preparation of carbide-derived carbons by chlorination of metal carbides. Reproduced from [35] with permission of The Royal Society of Chemistry.

with a very narrow pore size distribution. The sizes of ultramicropores are readily controlled by the periodic ordering of the metal atoms, which depends on the type of metal carbide used and the chlorination temperature. Despite the attractive tunability of the ultramicropores, however, CDCs do not display high BET SSAs ($<2000 \text{ m}^2/\text{g}$) [34,36]; rather, the SSAs are lower than the values for ACs and ZTCs.

Hydrogen preferentially adsorbs in ultramicropores, as supported by many experimental and theoretical approaches; therefore, the pore characteristics of CDCs were expected to be particularly suitable for hydrogen storage applications. The experimentally measured H_2 uptake at 77 K of an annealed TiC-CDC was $336 \text{ cm}^3 \text{ g}^{-1}$ (3.0 wt%), even at low pressures (~ 1 bar) [34]. This material had outperformed all other porous adsorbents, including ACs, ZTCs, and MOFs, before PCN-12 reported [37]. The average H_2 storage capacity of the CDCs at 77 K and 1 bar ($1000 \text{ m}^2/\text{g} < \text{BET SSA} < 2000 \text{ m}^2/\text{g}$) was estimated to be about 2.0 wt%, which is significant from the

viewpoint of the unit capacity (the capacity normalized by the SSA). Notwithstanding these promising results, the H_2 uptakes of the CDCs quickly saturated below 10 bar because the CDCs had an intrinsically relatively low pore volume [34,36]. To increase the pore volumes of CDCs, Kockrick et al. produced ordered mesoporous CDCs by chlorinating ordered mesoporous SiC, prepared by nanocasting using hexagonally ordered silica SBA-15 as a template [38]. The resultant CDCs (CDC-3–1273 H_2) exhibited an enhanced H_2 storage capacity of 50.9 mg/g at 77 K and 40 bar.

2.5. MOF-derived carbons (MDCs)

A new type of carbon, MDCs, was first proposed by Xu et al., who used MOF-5 as a template and furfuryl alcohol as the carbon precursor [39]. The impregnated furfuryl alcohol was polymerized and carbonized inside the micropores of an MOF. After carbonization, the template was eliminated using HCl treatment to produce a unique nanoporous carbon material. The procedure was similar to the preparation of ZTCs, with the exception that a MOF was used as the template. Unlike the zeolite hard template, the MOFs itself provided an excellent source of carbon via carbonization of the organic ligand. Park et al. [40] and Yamauchi et al. [41] developed enhanced microporous carbon materials via the direct carbonization of MOFs without the use of carbon precursors. A simple heat treatment at a specific temperature yielded a highly porous carbon. According to the procedures of Park et al. heat treatment of a selected MOF (MOF-5) under an inert atmosphere and at 600 °C resulted in the formation of zinc oxide at amorphous carbons [40,42,43]. Further increases in the

temperature produced a highly porous carbon, as shown in Fig. 4. The reduction of zinc oxide occurred in the presence of the carbonaceous materials, and the remaining zinc metals were easily vaporized because their boiling points were lower than that of zinc oxide [40]. In other cases, the metal oxides remaining after whole carbonization could be etched away using a strong acid [39,41].

Unlike the narrow micropore size distributions of ACs, ZTCs, and CDCs, MDCs displayed an unusual hierarchical porosity with an exceptionally high pore volume. The hierarchical porosity included almost all pore size ranges, including ultramicropores, supermicropores, mesopores, and macropores [39–41]. PCP-800 (produced from an Al-based MOF) exhibited a superbly high surface area (5500 m^2/g) and pore volume (4.3 cm^3/g), the highest values among porous carbons [41]. MDC-1 (produced from a Zn-based MOF) displayed an extremely high ultramicropore volume (0.63 cm^3/g), comparable to the ultramicropore volumes of CDCs, and high meso- and macroporosities [40].

Encouraged by the high SSA, pore volume, and favorable pore size distributions, the H_2 storage capacities of the MDCs were tested. The H_2 storage capacities of MDCs were found to exceed those of other benchmark materials, surpassing the capacities of all other porous materials characterized to date. MDCs (C1000) based on a zeolite imidazolate framework (ZIF) yielded a 2.77 wt% H_2 storage capacity at 77 K and 1 bar, the highest value reported at the time among ACs and ZTCs, excluding CDCs [44]. One year later, MDC-1 yielded an all-time high H_2 storage capacity of 3.25 wt% at 77 K and 1 bar due to the highly developed ultramicroporosity [40]. Interestingly, the H_2 storage behavior of MDC-1 at 1 bar increased steadily, as shown in Fig. 5, whereas other benchmark materials,

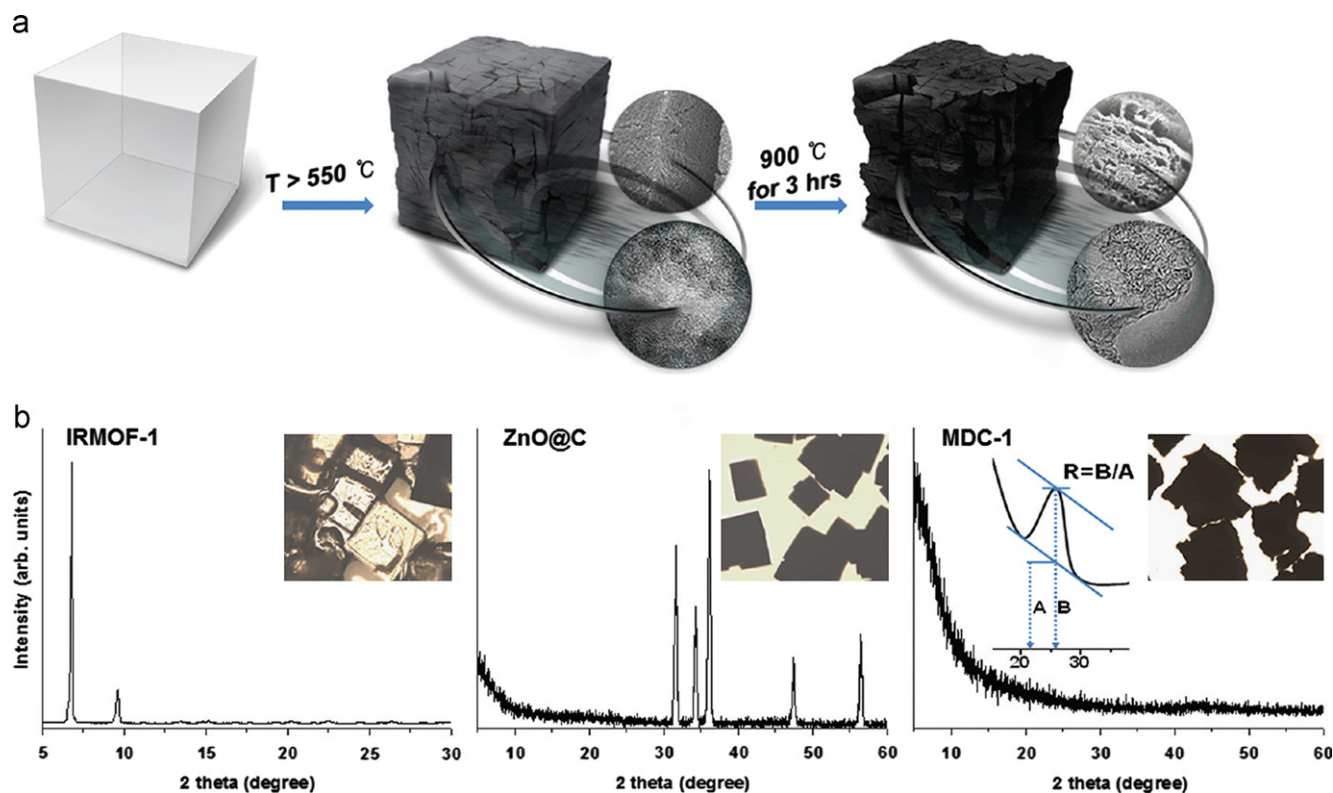


Fig. 4 Structural changes of MOF-5 with heat-treatment to yield MDC-1: (a) schematic view and (b) powder X-ray diffraction patterns with corresponding optical micrographs. Reprinted with permission from [40]. Copyright 2012 American Chemical Society.

especially CDC, displayed saturated behavior at this pressure [40]. This behavior resulted from the hierarchical porosity of the MDCs, suggesting that higher H₂ uptakes may be expected at higher pressures. In general, the benchmark materials that exhibit high H₂ storage capacities at 1 bar have shown relatively low H₂

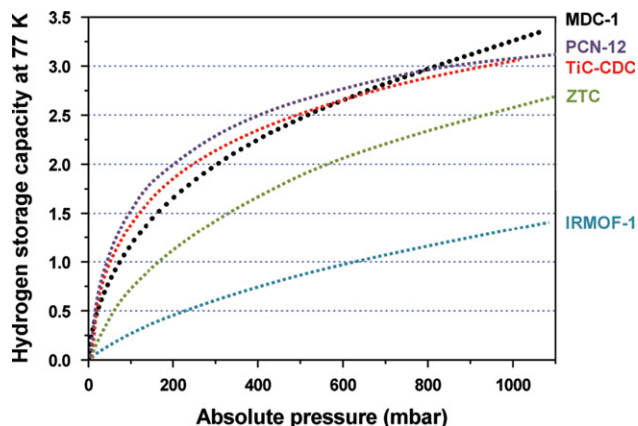


Fig. 5 Hydrogen storage behavior at 77 K and 1 bar of MDC-1 and benchmark materials. Reprinted with permission from [40]. Copyright 2012 American Chemical Society.

storage capacities at high pressures due to early pore saturation in the limited pore volume. Consistent with this expectation, the high-pressure H₂ storage capacities of MDC-1 at room temperature also surpassed the performance of H₂ storage materials at room temperature and high pressures, as shown in Table 1. These results indicate the suitability of hierarchical MDC pore systems for H₂ storage applications at both low and high pressures.

3. Conclusions and outlook

Recent progress in the synthetic methods, pore characteristics, and hydrogen storage capacities of various porous carbon materials was briefly reviewed. None of the porous carbon materials considered above (or any other porous adsorbent for that matter) has yet satisfied the criteria set by the DOE for hydrogen storage applications; however, the slow pace of technological advancement in the design and manufacture of highly pressurized cryogenic tanks may render porous carbon material adsorbents highly practical.

A rough comparison of the pore characteristics of different porous carbons is given in Table 2. By selecting an appropriate pore system for storing hydrogen molecules under given conditions, a variety of carbon materials may potentially be

Table 1 Preparation methods, pore characteristics, and H₂ storage capacities of the selected nanoporous carbons.

Material	Preparation	BET SSA (m ² /g)	Micropore vol. (cm ³ /g)	H ₂ uptake (wt%)		Ref.
				77 K	298 K	
<i>Activated carbons</i>						
AC-C2	CO ₂ activation	1488	0.6	1.7 (1 bar), 4.5 (20 bar)		[11]
AC-K5	KOH activation	3190	1.09	2.49 (1 bar), 7.08 (20 bar)		[11]
Maxsorb		3178	1.2		1.05 (200 bar)	[25]
AX21		2513	0.93	4.8 (40 bar)	1.1 (200 bar)	[25]
KUA5		3183	1.1	5.2 (40 bar)	1.2 (200 bar)	[25]
KUA6		3808	1.27	5.6 (40 bar)	1.05 (200 bar)	[25]
A5-CMK-8	Activation of mesoporous carbon	2700	1		0.75 (100 bar)	[45]
<i>Zeolite-templated carbons</i>						
AN8.5(2)-H	Zeolite Y	1.16	2900		0.71 (100 bar)	[29]
P7(2)-H	Zeolite Y	1.58	3800		0.87 (100 bar)	[29]
CA850	Zeolite alpha	0.71	3189	2.3 (1 bar) 6.0 (20 bar)		[46]
CB850h	Zeolite beta	1.13	3150	2.4 (1 bar) 6.9 (20 bar)		[46]
750	Zeolite Y N doping	0.79	1926	5.4 (20 bar)		[30]
800	Zeolite Y N doping	0.83	2022	5 (20 bar)		[30]
<i>Carbide-derived carbons</i>						
SiC1200	SiC	0.49	1279	2.1 (1 bar)		[47]
B4C800	B4C	0.99	2012	1.91 (1 bar)		[47]
ZrC600	ZrC	0.48	859	1.62 (1 bar)		[34]
ZrC600*	ZrC, H ₂ annealed	0.65	1388	2.58 (1 bar)		[34]
TiC800	TiC	0.82	1566	2.55 (1 bar)		[34]
TiC800*	TiC, H ₂ annealed	0.94	1943	3.02 (1 bar)		[34]
TiC-CDC	TiC, CO ₂ activation	1.34	3038	4.7 (60 bar)		[36]
AC-900	ZrC, KOH activation	0.98	2770	2.7 (1 bar) 6.2 (20 bar)		[48]
<i>MOF-derived carbons</i>						
NPC	FA/MOF-5		2872	2.6 (1 bar)		[39]
C800	FA/ZIF-8		2169	2.33 (1 bar)		[44]
C1000	FA/ZIF-8		3405	2.77 (1 bar)		[44]
MDC-1	IRMOF-1	3174	1.01	3.25 (1 bar)	0.94 (100 bar)	[40]
MDC-3	IRMOF-3	1678	0.66	2.1 (1 bar)	0.6 (100 bar)	[40]

Table 2 Comparison of pore characteristics of various nanoporous carbons^a

Adsorbent	Ultramicroporosity	Supermicroporosity	Mesoporosity	Macroporosity
ACs	++	++	+	–
ZTCs	+	+++	+	–
CDCs	+++	+	–	–
MDCs	+++	++	++	+

^aQualitative evaluation of the relative porosity evolution of the nanoporous carbons, with +++, ++, +, and – (+++ best, – worst).

useful for meeting the DOE requirements. The new DOE targets released in 2009 recommended ultimate operating pressures of 0.4–1.2 MPa and a H₂ delivery temperature of between –40 °C and 100 °C. Very high ultramicropore and supermicropore volumes will be required to accomplish these target storage capacities. From this perspective, the pore characteristics of MDCs are highly desirable. MDCs are still in the early stages of development. By analogy to the development of MOFs, wherein several thousands of MOFs have been reported to date thus, MDC performance optimization is expected to yield fruitful results.

A great deal of progress has been achieved over the past decade in the field of solid porous adsorbents for hydrogen storage applications. Although it is possible that adsorptive storage on high-surface area adsorbents has already reached the limits determined by physical constraints, and, therefore, additional progress in this field is unlikely, recent experimental and theoretical studies suggest that the pore size distributions may be optimized. Small pore volumes, rather than high surface areas, decisively affect the hydrogen storage capacity near room temperature. Current research directed toward the development of ultrahigh-surface area materials must, therefore, be reconsidered. Concurrent considerations associated with chemical approaches, such as spillover and alkali metal doping, provide exciting opportunities for realizing viable H₂ storage carbon materials.

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