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WHAT'S HOT IN CHEMISTRY, November/December 2009

MOFs Meet the Target for Mopping Up Methane

by John Emsley

Beneath the Appalachian mountains in the United States is one of the world's great gas fields, as yet undeveloped. The methane gas it contains could act as a transport fuel, and indeed in some countries compressed natural gas, which is mainly methane, is used as fuel for various forms of transport, including cars. The U.S. Department of Energy (DOE) would also like to promote it as a vehicle fuel, ideally by absorbing it into an inert material so that it can be stored at normal temperatures and pressures. The DOE has set a target ratio for such a material of 180, in other words the absorbing system must store a volume of methane 180 times larger than its own volume. This would be almost like storing liquid methane.

Carbon nanotubes, activated carbon, and zeolites have the capacity to absorb gases but they do not meet the DOE target. Metal-organic frameworks (MOFs) are much more promising, and researchers in the past decade have been focusing on these. Crystal structures based on 9,10-anthracene-dicarboxylate have enormous cavities, and theoretical calculations suggested these could meet the DOE requirement. However, when the group headed by Hong-Cai (Joe) Zhou at the Department of Chemistry at Miami University made this ultra-porous material, it fell far short of the 180 target.

Zhou, who is now at Texas A&M University, was not deterred by this setback and looked at an alternative ligand, 5,5'-(9,10-anthracenediyl)di-isophthalate, which he suspected would produce crystals with much larger pore size—and he was right. The crystals had nano-sized cavities that provided 0.87 cm³ of empty space per gram of material. His remarkable paper occupies position #11, just below the current Hot Ten (S. Ma, *et al.*, *J. Am. Chem. Soc.*, 130[3]: 1012-6, 23 January 2008; 18 citations this period, 47 overall). The new crystals were grown overnight from a solution of dimethylformamide. They absorbed methane in excess of the DOE's target and in fact achieved a storage capacity ratio of 230 after activation.

However, this storage success does not in itself solve the problem of using methane as a transport fuel. As Zhou tells *Science Watch*: "For one, the DOE storage goal refers to a systems goal. The system contains not only the absorbent but also the tank and necessary pipes. In addition, we have to lower the production cost of the new material as much as possible."

Zhou's research group is now working on MOFs with even higher adsorption capacities, and not only for methane, but for hydrogen also. The latter is the other gaseous fuel that might one day drive vehicles and is potentially the greenest fuel of all since the product of combustion is entirely H₂O. However, while the hydrogen economy has been hyped for more than 20 years, there still exist significant challenges to its implementation. Hydrogen storage is also of vital importance and a challenge for which MOFs might also provide the answer.

Recently Zhou has widened his groundbreaking work to include studies involving this gas. He and his co-workers have investigated the potential of lanthanide-based MOFs to absorb hydrogen (J. Luo, *et al.*, *J. Am. Chem. Soc.*, 130[30]: 9626, 2008), and found a way of enhancing hydrogen uptake in copper-containing MOFs (X. Wang, *et al.*, *Angew. Chem. Int. Ed.*, 47[38]: 7263-6, 2008), as well as investigating the effect of framework catenation (S. Ma, *et al.*, *J. Am. Chem. Soc.*, 130[47]: 15896-902, 2008).

His group have also been concentrating on the MOFs themselves, looking at the way these can be modified to increase their capacity to hold gaseous molecules, as described in three recent *Journal of the American Chemical Society* reports: J. Li, *et al.*, 131(18): 6368, 2009; S. Ma, *et al.*, 131(18): 6445, 2009; and D. Zhao, *et al.*, 131(26): 9186, 2009.

MOFs are a material of the future, not only useful in gas storage but for separating mixtures of gases. Says Zhou: "MOF-based mesh-adjustable molecular sieves allow selective adsorption to be tuned by temperature adjustment. This new direction toward molecular sieves can be used in highly efficient and energy-saving separation procedures." His group is now concentrating on this aspect as well as on other things, such as the potential of MOFs to act as catalysts.

One day MOFs might well play a part in CO₂ capture. We might even envisage a car of the future, which is fuelled by methane stored in MOFs, capturing the CO₂ of the exhaust gases in MOF cavities that have become vacated as the methane is consumed. ■

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from the Special Topic of Graphene →

Chemistry Top 10 Papers

Rank	Paper	Citations This Period (May-Jun 09)	Rank Last Period (Mar-Apr 09)
1	Y. Kamihara, <i>et al.</i> , "Iron-based layered superconductor La[O _{1-x} F _x]FeAs (x = 0.05-0.12) with T _c = 26 K," <i>J. Am. Chem. Soc.</i> , 130(11): 3296-7, 19 March 2008. [Tokyo Inst. Technol., Yokohama, Japan] *273SL	124	1
2	C. de la Cruz, <i>et al.</i> , "Magnetic order close to superconductivity in the iron-based layered LaO _{1-x} F _x FeAs systems," <i>Nature</i> , 453(7197): 899-902, 12 June 2008. [6 U.S. and China institutions] *311WV	47	2
3	X.L. Li, <i>et al.</i> , "Chemically derived, ultrasmooth graphene nanoribbon semiconductors," <i>Science</i> , 319(5867): 1229-32, 29 February 2008. [Stanford U., CA] *267SX	32	4
4	H. Takahashi, <i>et al.</i> , "Superconductivity at 43 K in an iron-based layered compound LaO _{1-x} F _x FeAs," <i>Nature</i> , 453(7193): 376-8, 15 May 2008. [Nihon U., Tokyo, Japan; Tokyo Inst. Technol., Japan] *301AI	28	3
5	J. Peet, <i>et al.</i> , "Efficiency enhancement in low-bandgap polymer solar cells by processing with alkane dithiols," <i>Nature Materials</i> , 6(7): 497-500, July 2007. [U. Calif., Santa Barbara] *184NH	28	7
6	S. Stankovich, <i>et al.</i> , "Synthesis of graphene-based nanosheets via chemical reduction of exfoliated graphite oxide," <i>Carbon</i> , 45(7): 1558-65, June 2007. [Northwestern U., Evanston, IL; U. North Carolina, Chapel Hill] *185XJ	27	†
7	A.I. Hochbaum, <i>et al.</i> , "Enhanced thermoelectric performance of rough silicon nanowires," <i>Nature</i> , 451(7175): 163-7, 10 January 2008. [U. Calif., Berkeley; Lawrence Berkeley Natl. Lab., CA] *249GA	26	10
8	B. Tian, <i>et al.</i> , "Coaxial silicon nanowires as solar cells and nanoelectric power sources," <i>Nature</i> , 7164(449): 885-9, 18 October 2007. [Harvard U., Cambridge, MA] *221LY	22	6
9	S.Y. Park, <i>et al.</i> , "DNA-programmable nanoparticle crystallization," <i>Nature</i> , 451(7178): 553-6, 31 January 2008. [Northwestern U., Evanston, IL; Argonne Natl. Lab., IL] *256OQ	21	†
10	A.I. Boukai, <i>et al.</i> , "Silicon nanowires as efficient thermoelectric materials," <i>Nature</i> , 451(7175): 168-71, 10 January 2008. [Caltech, Pasadena] *249GA	21	†

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