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WHAT'S HOT IN... CHEMISTRY, march/april 2010

Mining the Lower Levels Reveals a Real Gem

by John Emsley

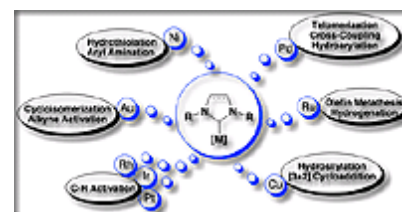


Inspection of the current list shows that papers on superconductivity occupy four slots, and **graphene** accounts for the remainder. The former group of papers is about the remarkable iron/lanthanum superconductor which was first discussed in the **Nov/Dec 2008** issue of *Science Watch*. The latter group were dealt with in the **Jan/Feb 2010** issue, but among them now is one that is new to the list: paper #7. This reports a chemical method, using hydrazine, for reducing graphite oxide to graphene sheets which exhibit semiconducting behavior.

Although the glamour of superconductors and graphene electrodes holds center stage, some excellent conventional chemistry is waiting in the wings at position #14: R.A. Kelly, *et al.*, *Organometallics*, 27(2): 202-10, 28 January 2008, with 18 citations this period and 66 overall. This paper concerns iridium complexes with N-heterocyclic carbene (NHC) ligands and represents the collective work of researchers in chemistry departments at the University of New Orleans (Louisiana), the Institute of Chemical Research of Catalonia (Spain), the University of Salerno (Italy), and the University of Miami (Florida). The lead author of the paper is **Steven Nolan**, who is now at the University of St. Andrews in Scotland.

Carbenes are rather remarkable entities, with a central carbon atom with two covalent bonds plus an electron pair, in theory making them ideal ligands. (All the NHCs of paper #14 are heterocyclic molecules with two nitrogens attached to the carbene carbon.) These carbene complexes of transition metals have attracted considerable interest as catalysts for various reactions, including olefin metathesis, hydrogenation, hydrosilation, cross-coupling, and isomerization. Although carbene complexes are overshadowed by the more common phosphine complexes, there is now evidence that they are actually better in many respects. Not only are they more stable towards air and moisture, but some of them have unique catalytic abilities.

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Organometallic Chemistry and Catalysis.

Figure from the **Nolan Group**, University of St. Andrews, Scotland, U.K.

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The NHCs were first isolated in 1991 but have gained popularity greatly since then, and Nolan has shown just how versatile their ruthenium, palladium, and copper complexes are as catalysts. When he turned his attention to iridium-NHC complexes things became clearer as to why they are superior, and as a result this work is now being highly cited.

Paper #14 concerns the iridium complexes of nine different NHCs of the form [(NHC)Ir(CO)₂Cl] which were produced in yields as high as 90%. They were made via the corresponding cyclooctadiene complexes. The focus of #14 is the carbonyl stretching frequencies of the products, and these are used to examine and rank the electron-donating strength of the NHC entities. The result reveals that all are more strongly donating as ligands than even the strongest donating tertiary phosphines. What is also shown is that the other constituents of the heterocyclic ring of the NHC have only minor influences on the molecule's electron donating power. The carbene with bulky adamantyl groups attached to the two nitrogens gave the lowest stretching frequencies of the CO bonds, and the reason seems to be their steric requirements.

So why is this paper being strongly cited now?

Says Nolan, "It deals with fundamental properties of a ligand family that is becoming very popular in organometallic chemistry and homogeneous catalysis. Some of these NHC complexes (Ru, Pd, Cu, and Au) can now be synthesized on a large scale and are commercially available. I believe we are addressing some fundamental concepts that are widely applicable to these important areas. It is our hope that a basic understanding of this ligand family will help move towards a sustainable green chemical industry."

Currently Nolan is directing his research toward understanding and maybe even predicting the reactivity of transition metal complexes. If he succeeds, he believes it will then be possible to design catalysts with improved performance in the many areas where they are being used and possibly even designing novel chemical transformations.

Since paper #14 appeared, Nolan has moved on to studying NHC complexes of gold. In his paper in the *Journal of the American Chemical Society* (N. Marion, *et al.*, 131[2]: 448-9, 2009), he shows that these will act as catalysts for the hydration of alkynes, and in acid-free conditions, and at levels of only parts per million. In *Chemistry – A European Journal* (R.S. Ramon, *et al.*, 15(35): 8695-7, 2009), he looks at the way such gold catalysts permit the hydration of nitriles to form amides.

Dr. John Emsley is at the Department of Chemistry, Cambridge University, U.K. 🇬🇧

Chemistry Top 10 Papers

Rank	Paper	Citations This Period (Sep-Oct 09)	Rank Last Period (Jul-Aug 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	109	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	42	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	38	4
4	C. Lee, <i>et al.</i> , " Measurement of the elastic properties and intrinsic strength of monolayer graphene, " <i>Science</i> , 321(5887): 385-8, 18 July 2008. [Columbia U., New York, NY] *327FB	31	†
5	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	29	3
6	D.C. Elias, <i>et al.</i> , " Control of graphene's properties by reversible hydrogenation: Evidence for graphane, " <i>Science</i> , 323(5914): 610-3, 30 January 2009. [U. Manchester, U.K.; Inst. Microelectronics Tech., Chernogolovka, Russia; Cambridge U., U.K.; U. Nijmegen, Netherlands] *400JB	23	†
7	S. Gilje, <i>et al.</i> , " A chemical route to graphene for device applications, " <i>Nano Letters</i> , 7(11): 3394-8, November 2007. [U. Calif., Los Angeles] *232ZI	22	†
8	R.R. Nair, <i>et al.</i> , " Fine structure constant defines visual transparency of graphene, " <i>Science</i> , 320(5881): 1308, 6 June 2008. [U. Manchester, U.K.; U. Minho, Braga, Portugal] *309DO	22	†
9	A.A. Balandin, <i>et al.</i> , " Superior thermal conductivity of single-layer graphene, " <i>Nano Letters</i> , 8(3): 902-7, March 2008. [U. Calif., Riverside] *273QS	21	†
10	C. Gomez-Navarro, <i>et al.</i> , " Electronic transport properties of individual chemically reduced graphene oxide sheets, " <i>Nano Letters</i> , 7(11): 3499-503, November 2007. [Max Planck Inst. Solid State Res., Stuttgart, Germany; U. Siegen, Germany; EPFL, Lausanne, Switzerland] *232ZI	18	†

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