MALCOLM L. H. GREEN
16 April 1936 — 24 July 2020
Malcolm Green was a remarkable man—even a short meeting was enough to make this clear. He had that rare charismatic ability to connect personally with those around him, whatever their rank or status. Generous of his time with his current coworkers, he also made sure to provide help, encouragement or advice to his alumni as the need arose during their subsequent careers. Inspired by his lead, an unusually large number of them have become widely known figures in chemistry in the UK and beyond. Malcolm laid out his view on his students on one occasion when a visitor to the lab commented on how quickly a large sum had been raised by his alumni for a lectureship in his name. ‘They must really love you’, he said, to which Malcolm replied with his simple philosophy: ‘If you love them, they will love you.’

Malcolm’s infectious passion for science and his imaginative approach led him in many diverse directions during his career. Although his signature field was organometallic chemistry, he made later important contributions in nanomaterials and heterogeneous catalysis. Always interested in the reasons why Nature behaves as it does, he pioneered several new and imaginative methods for interpreting and understanding chemical bonding and reactivity, such as the covalent bond classification scheme or the rules for predicting the regiochemistry of nucleophilic attack on organometallics. He cared deeply about education and was the author of the first textbook devoted to the organometallic chemistry of the transition metals, *Organometallic compounds. Volume 2: the transition elements*, which originally appeared as early as 1968.
BACKGROUND AND EDUCATION

Malcolm Green was born in Eastleigh, Hampshire, the son of Leslie Ernest Green and Sheila Ethel Green (née Hodder). Malcolm’s father was a GP, and the family included his elder sister, Sylvia. He was educated at Smallwood Manor preparatory school and then Denstone College, an independent boarding school in Uttoxeter, Staffordshire. His mother wanted him to leave school a year early and begin earning, so he applied for a job at ICI. At the job interview, however, he was told to go for a degree instead. As a result, he entered the Acton Technical College in Middlesex, where he completed his BSc in 1956.

Moving to the Imperial College of Science and Technology, London, he obtained his PhD in 1959, focusing on organometallic chemistry. At Imperial he was inspired by his advisor, Geoffrey Wilkinson (FRS 1965), who had just returned from Harvard and was later to be awarded the Nobel Prize in 1973 for his pioneering work on organometallic compounds (Wilkinson 1974). In this way, Malcolm first came into contact with transition metal organometallic chemistry at a time when it was just beginning to gain attention as a part of the ‘renaissance’ of inorganic chemistry that is usually dated from the discovery of ferrocene (bis-cyclopentadienyl iron(II)) in the mid 1950s. Indeed, Malcolm’s doctoral thesis with Wilkinson, A study of some transition metal hydrides and olefin complexes, largely dealt with further developments of the metallocene story. He extended the range of bis-cyclopentadienyl metal complexes to include Cp₂ReH (1)* and Cp₂WH₂ (2), a complex that would prove to be an important starting point for much of Malcolm’s early independent work. Apart from the cyclopentadienyl aspect, another notable feature of this work was that many of the new species were hydride complexes, then also a novelty.

He married Jennifer Bilham in January 1965. A fellow chemist, she was appointed to a Fellowship at St Hugh’s College, Oxford, in 1969.

EARLY CAREER

After a postdoctoral year with Wilkinson, Malcolm moved to Cambridge as an assistant lecturer in 1960 and took up a Fellowship at Corpus Christi College, Cambridge, in 1961. In spite of his messy laboratory, christened ‘the jungle’ by the students, in just two years there he published a remarkable series of papers on alkyl complexes of the common transition metals, including their conversion to olefin complexes via hydride abstraction (3). In 1963 he became Septcentenary Fellow of Inorganic Chemistry at Balliol College, Oxford, and a departmental demonstrator at the university. It was at this point that the bis-cyclopentadienyl molybdenum and tungsten compounds became a central part of his work. These were soon to be made on a very large scale in apparatus specifically set aside for the work in a task that rotated among the relevant lab members and involved considerable effort. These compounds were central starting materials that allowed him to explore a very wide range of structures and reactions in a well behaved environment that enforced a cis arrangement of the non-cyclopentadienyl ligands (19).

In 1965 he advanced to a lectureship at Balliol College, where one of his early successes (1967) was demonstrating the carbene character (i.e. the Fe=–C(OH)R) form of a protonated

* Numbers in this form refer to the bibliography at the end of the text.
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iron acyl (4). He also collaborated with his wife, Dr Jennifer Green, a brilliant specialist in theoretical and physical inorganic chemistry, to probe the bonding in organometallic compounds by photoelectron spectroscopy (7). This probably helped to give Malcolm a feeling for interpreting his chemistry in terms of orbitals at a much earlier stage than most others in the field. In collaboration with Jennifer and crystallographer Ken Prout, they proposed a new bonding model (9) for the bis-cyclopentadienyl metal complexes that superseded the Ballhausen–Alcock model. He also used the known chemistry of \( \pi \)-benzene metal complexes as a starting point to explore the organometallic chemistry of the C\(_7\) aromatic, the cycloheptatrienyl cation (15). His influential textbook on transition metal organometallic chemistry (5) also dates from these years.

During this period, he also formed a connection with Portugal as a result of having Alberto Dias (8) as a DPhil student. I suspect the fact that Malcolm’s colleague, physical chemist Lionel Staveley, already had a very productive relationship with Portuguese chemistry (Calado 1983) may well have encouraged the connection. Malcolm’s rising reputation led to his attracting an increasing number of postdoctoral coworkers from abroad. For example, he and Taro Saito from Japan explored hydride chemistry, resulting in the first case of a nickel hydride and also of the use of sterically bulky ligands to stabilize otherwise sensitive compounds (6).

His feeling for chemistry was instinctive, although he was always able to provide solid arguments in favour of his position; one felt that these came after the position had been taken. Sometimes a student’s question would baffle him, in which case he might say, ‘You had better ask Jenny [his wife] about that.’ His gung-ho attitude and seat-of-the-pants approach would not be possible today (figure 1). For example, on being asked about a reaction that would not go forward as it should, he would sometimes take out his cigarette lighter to apply the flame to the flask, at which point the student in question might perhaps have been reluctant that he had asked for help. At one point, he entered the lab smoking a large cigar. A colleague and I at once pointed out that there was an open beaker of ether on the lab desk and that he shouldn’t be smoking. ‘No,’ he said, ‘there isn’t a problem. Watch this.’ He picked up the beaker and was obviously about to plunge the cigar into the ether to show how this would not lead to ignition—we were dubious and successfully intervened to hold back his arm. He was, however, firm on people wearing safety goggles to protect the eyes.

He was always ready to help, but in his own way. On another occasion an undergraduate appeared from the inorganic laboratory course with a solution that he said would not crystallize as it was supposed to do according to the instructions. Malcolm immediately seized the flask, borrowed the necessary spatula from someone’s desk and rapidly produced the crystals, working not on the bench but in mid air. Not content with that, he insisted on filtering the crystals, transferring them to a piece of paper where he summarily dried them by waving the paper about. On proudly handing the result to the astonished undergraduate, the latter expressed his grateful wonder, at which point Malcolm closed his eyes and made a sweeping gesture with his arm ‘Think nothing of it’, he said. Unfortunately, his gesture had the effect of sweeping the crystals from the student’s grasp on to the floor and the affair ended by Malcolm having to write a note to the lab course grader that the student had indeed obtained the proper result but that he, Malcolm, had upset the product.

An area with a very big future ahead of it, and which was developed at this time, was organometallic photochemistry. Irradiation of a bis-cyclopentadienyl tungsten dihydride led to the highly reactive transient, bis-cyclopentadienyl tungsten, the metal atom of which promptly
inserted into the C–H bond of \textit{para} xylene to give an alkyl hydride (12). This was a very early example (1974), and the first photochemical one, of a process that later gained great fame under the name of ‘CH activation’, a topic that remains very active right up to the time of writing (2020). This involves functionalizing normally relatively unreactive C–H bonds, ultimately those of an alkane, with a suitable metal complex.

Malcolm also discovered a somewhat related process involving CH bond cleavage: alpha elimination in a metal methyl complex to give a methylene metal hydride. Again, this was a very early example of what was later recognized as one of the standard reactions in the field. The alpha process is typically uphill thermodynamically, so its occurrence can usually only be inferred from circumstantial evidence. This is indeed the case in this example, but the evidence was persuasive on the basis of the unexpected products from a substitution reaction (13).

Another result from 1974 highlights Malcolm’s productive connection with France. Bianca Tchoubar, even though at that time well advanced in years, was a highly energetic physical chemist who identified transition metal organometallic chemistry as a topic of rising interest that was then lacking in France (Tchoubar 1964). She identified Malcolm as the right person to spread the word in France and successfully convinced him to do so. In this way, at the Centre
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Nationale de la Recherche Scientifique laboratory at Gif-sur-Yvette, near Paris, he met Hugh Felkin, noted for the Felkin–Ahn selectivity rules (Felkin & Swierczewski 1972) and they soon became good friends. Felkin had recently isolated the first compound with an iron–magnesium covalent bond that they called an ‘inorgano-Grignard’ and Malcolm soon joined in with a biscyclopentadienyl molybdenum–magnesium example (14). This led to exchanges of coworkers between the Felkin and Green groups; for example, Michel Ephritikhine joined Malcolm’s group as a postdoc and went on to a career in the French nuclear chemistry programme at Saclay, and I joined Felkin’s group as a postdoc, later to become an Attaché de Recherche.

Malcolm’s interest in materials chemistry started as early as 1977 with studies on the intercalation of organometallics into solid tantalum disulfide in collaboration with Allan Jacobson (16). Although not an important result in itself, it sowed the seed for Malcolm’s later move from organometallic chemistry into materials applications, where he was to make valuable contributions. For example, he greatly expanded the range of compounds showing large second order nonlinear optical behaviour (27).

At the same time, his organometallic work was moving in unexpected directions. Working with Steve Davies and Mike Mingos, Malcolm systematized nucleophilic attack on organometallic π ligands by introducing a set of rules (17) that successfully predict the experimental regiochemical outcome in most cases. Another adventure from this time was the development of metal vapour synthesis (10,11) for organometallic chemistry, building on the work of Peter Timms of Bristol (Timms 1970). This needed a large and impressive home-built apparatus in which an electron gun—a novelty in this application—vaporized a metal block at temperatures of up to 3500°C, after which the vapour co-condensed with a range of potential ligands such as benzene. A number of unusual compounds were first made in this way, and some can still only be made in this way, for example bis-π-benzene titanium and molybdenum (11). Malcolm was joined at Oxford by a distinguished chemist then just beginning his career, Geoffrey Cloke (FRS 2007), who made a whole series of unusual compounds in this way, such as bis-π-arene complexes of tungsten, niobium and tantalum (20) and some very unusual bis-π-arene tungsten hydride derivatives (18). Meanwhile Malcolm’s career advanced rapidly (figure 2), with a Royal Society Senior Research Fellowship in Oxford from 1979 to 1986.

His work on electronically unsaturated and minimally hindered metal alkyls from 1982 (21) resulted in the highly influential identification, definition and naming of the agostic CH bond; that is, one which is bridged with a transition metal by a 2-electron, 3-centre C–H–M bond (22). The name was suggested by his Balliol colleague, the Homer Scholar Jasper Griffin, from a word from a line in the Iliad (11.425) referring to a grasping action of the hand, and the word is now even included in the Oxford English Dictionary. These agostic interactions were soon found in a wide variety of contexts and proved to be key in the mechanism of such commercially important processes as metallocene olefin polymerization catalysis as well as being transient intermediates in a wide variety of standard organometallic reactions. This is arguably the single most influential legacy of his work and thought.

Later career

Materials science began to take a greater fraction of Malcolm’s time from the late 1980s. An early result was his 1987 finding of a large second order optical nonlinearity in a ferrocenyl-substituted styrene derivative (23), where the high donor power and polarizability of the
organometallic fragment were central to this success. In a major promotion at about this time, in 1989 he became professor and head of the Inorganic Chemistry Laboratory at Oxford and a fellow of St Catherine’s College.

In the 1990s, he became interested in heterogeneous catalysis and helped to produce a series of results in the area, of which the most influential was probably the selective oxidation of methane to synthesis gas, $\text{H}_2 + \text{CO}$, with a lanthanum ruthenium oxide catalyst (24). A related development was the conversion of methane/carbon dioxide mixtures into synthesis gas (25). A notable co-author on these papers was A. K. Cheetham (FRS 1994), another successful alumnus from his lab. Metal carbides were actively pursued as heterogeneous catalysts in this period, for example for the methane to synthesis gas reaction (33). As an example of a related $\text{C}_1$ to $\text{C}_3$ transformation, methane was converted by partial oxidation into ethylene and synthesis gas, followed by a hydroformylation step that produced propanal as the final product (26). His discovery of an extremely active Fischer–Tropsch catalyst led to the founding of the Oxford Catalyst Group, later renamed Velocys. Currently, the company is developing a project for conversion of rubbish into jet fuel.

The rising interest in carbon nanotubes from 1993 led Malcolm to enter the area; in particular he developed a general method for opening the sealed ends of these structures to permit introduction of various species into the interior of the tubes (28). This was followed by a study that showed how ultrasound introduces buckling, bending and dislocations in carbon nanotubes, ultimately leading to the formation of amorphous carbon (30). With Jeremy Sloan, he looked into introducing salts into carbon nanotubes, with a study on potassium iodide where
one-dimensional crystals could be encapsulated, imaged and the structure solved (32). As an extension of this area, he introduced silver salts followed by photolysis to give a silver nanowire encapsulated within the nanotube (31).

During this time Malcolm also returned to his covalent bond classification theme (CBC) with a detailed exposition in 1995 (29). This contribution entirely reimagined our picture of metal complexes by classifying these compounds in a new way according to their ligand set. For example, 2-, 1- and zero-electron donors such as PPh₃, H and BF₃ were designated L, X and Z ligands. Compounds could then be unambiguously assigned to certain categories, avoiding the ambiguities that often arise in using the traditional oxidation state formulation. For example, the classic Vaska compound, IrCl(CO)(PPh₃)₂, and its H₂ adduct, IrH₂Cl(CO)(PPh₃)₂, fall into the MXL₃ and MX₃L₃ classes, respectively, both very common for iridium. For each element, a chessboard diagram could then be drawn in which every compound could be put into its proper place according to its valence state and electron count, each distinct class being kept separate. Each organometallic reaction, such as oxidative addition or reductive elimination, could be identified with a specific move such as might occur in a game of chess. Favoured reactions would then be ones that involved moves between well-populated classes, while forbidden ones, such as oxidative addition for a compound already in its highest oxidation state, would involve leaving the chessboard altogether. For example, oxidative addition of H₂ to IrCl(CO)(PPh₃)₂ to give IrH₂Cl(CO)(PPh₃)₂ implies a move from one common state to another, indicating that the reaction is plausible. Malcolm taught the subject in his courses and viewed the scheme as being most useful for teaching purposes, accounting for his choosing the Journal of Chemical Education for his last word on the topic (38), this time in collaboration with his alumnus, Ged Parkin.

Perhaps the most important outcome of Malcolm’s thought on bonding was his recognition of the widespread occurrence of 2-electron, 3-centre (2e,3c) bonding in compounds in which a variety of groups bridge two metals. This only became known in the community by word of mouth because it was not published until long after the initial insight had been achieved. In 2012, with Jennifer Green and Ged Parkin, their analysis of 3-centre, 2-electron bonding appeared (37), along with a suggested method for its graphical representation. Numerous anomalies in bonding, as considered under older ideas, were cleared up by the new approach. Thus, in order to achieve 18 valence electrons on conventional counting procedures, numerous bridging hydride and carbonyl complexes require, in addition to the bridge bonding, an independent metal–metal bond. The new 2e,3c approach obviates the M–M bond. An M–H–M’ bridge, for example in [(OC)₅Cr—H—Cr’(CO)₅]⁻, is considered as being made up of an anionic 18e [(OC)₅Cr—H]⁻ unit binding to a 16e Cr(CO)₅ fragment via Cr’ in such a way that the 2e of the Cr–H bond become delocalized over Cr, H and Cr’, thus raising the electron count on Cr’ from 16e to 18e. This picture is in agreement with structural data and with sophisticated theoretical calculations that show no such independent M–M bond exists. A similar 2e,3c picture applies for bridging ligands across L, X and Z classes, where the required 2e are both supplied by a bridging L ligand, or supplied by a one-electron X ligand and one M (1e), or by both metals (1e + 1e) for a bridging zero-electron Z ligand. This is a characteristic Green insight: at once simple, original and yet profound in its implications.

Malcolm’s interest in the biological applications of carbon nanotubes is evidenced by his 2003 report (34), in collaboration with Jason Davis and Karl Coleman, both Oxford chemists, on chemical and biochemical sensing with modified single-walled carbon nanotubes (figure 3). They showed how carbon nanotubes can be nondestructively oxidized and covalently functionalized in various ways. Proteins can then adsorb noncovalently along the nanotube
to give conjugates characterizable by atomic force microscopy. Several metalloproteins and enzymes have been bound on both the sidewalls and termini of single-walled carbon nanotubes. Possible device applications were suggested because nanotube electrodes show useful electronic properties, including direct electrical communication between a redox-active biomolecule and the delocalized T system of its carbon nanotube support.

Taking official retirement in 2004 did little to change Malcolm’s zeal for research, and he continued his work as an emeritus research professor from that time, typically in collaboration (figure 4). An example from 2006 shows an excursion into molecular immunology with a study to test the use of carbon nanotubes for vaccine or drug delivery with the result that carbon nanotubes indeed activate human complement via both classical and alternative pathways (35). Another biomedical result, from 2010, (36) involved investigation of filled-and-functionalized carbon nanotubes in vivo. Covalent functionalization of single-walled carbon nanotubes filled with I-125 radiolabelled NaI converted them into useful radioprobes. Intravenous administration of these nanotubes in mice was tracked in vivo by single-photon emission computer tomography. Specific accumulation in the lung was detected in an ultrasensitive imaging protocol and organ-specific therapeutics, and diagnostics were suggested as plausible future developments of the work. In addition to all the biomaterials work mentioned above,
organometallics and pedagogy were still a major part of his scientific work. For example a 2014 review with Ged Parkin showed how the CBC method is useful in teaching inorganic chemistry (38).

Malcolm Green died on 24 July 2020, aged 84, and is survived by Jennifer, their three children—Russell, who works in IT for Deutsche Bank, Sophie, who is a lawyer for Exxon-Mobil, and Matthew, who works in IT for Spotify—and eight grandchildren. His passing was regretted by all who knew him, but he leaves an indelible imprint, not only on science, but also on his many scientific progeny.

**MAJOR HONOURS AND AWARDS**

1972 Corday–Morgan Prize of the Chemical Society  
1977 Chemical Society Award in Transition Metal Chemistry  
1982 Tilden Prize and Lectureship of the Royal Society of Chemistry  
1984 American Chemical Society Inorganic Chemistry Award  
1985 Elected a Fellow of the Royal Society  
1988 Sir Edward Frankland Prize and Lectureship of the Royal Society of Chemistry
1992 Karl Ziegler Prize of the Gesellschaft Deutscher Chemiker
1995 Davy Medal of the Royal Society
1997 American Chemical Society award in Organometallic Chemistry
2000 Sir Geoffrey Wilkinson Medal and Prize of the Royal Society of Chemistry
2015 European Association for Chemical and Molecular Sciences’ European Prize for Organometallic Chemistry

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AUTHOR PROFILE

Robert Crabtree

Robert Crabtree FRS is Whitehead Professor of Chemistry at Yale and an alumnus of the Malcolm Green research group, where he did his first year of research in 1969/70. He holds DSc degrees from the University of Bath and the University of Sussex; he has been an ACS and RSC organometallic chemistry awardee, Bailar (Illinois), Sabatier (Toulouse), Williams (Oxford), Dow (Berkeley) and Mond (RSC) lecturer, has chaired the ACS Inorganic Division, is the author of an organometallic textbook, and was awarded the Yale Postdoctoral Mentoring Prize and the Franco-American Prize of the French and American Chemical Societies. He reversed catalytic alkene hydrogenation to obtain alkane dehydrogenation and saw C–H hydroxylation, all with iridium complexes. This was followed by work on H₂ complexation, dihydrogen bonding, and catalysis for green and energy chemistry, notably water oxidation. He is a fellow of the American Academy of Arts and Sciences and a member of the US National Academy of Sciences.

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