Pople, Sir John Anthony

(1925–2004)

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Sir John Anthony Pople (1925–2004)

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Pople, Sir John Anthony (1925–2004), mathematician and theoretical chemist, was born on 31 October 1925 at the War Memorial Hospital, Burnham-on-Sea, Somerset, the elder son (there were no daughters) of (Herbert) Keith Pople, who owned and managed the principal clothing shop for men in Burnham, and his wife, Mary Frances, *née* Jones, who came from a farming background in Shropshire, Somerset, and Wiltshire. He attended Bristol grammar school from 1936 to 1943. The journey to school consisted of a two-mile bicycle ride, twenty-five miles by train, and one mile on foot. During the war he and his brother, Donald, would wend their way to school around burning buildings and unexploded bombs. Some classes were held in damp concrete shelters under the playing fields. Nevertheless the teaching was excellent and Pople entered Trinity College, Cambridge, as a scholar in 1943. While most young men of his age were inducted into the armed services, a small group of students in mathematics, science, and medicine was permitted to attend university before taking part in wartime research projects. It was intended that degree courses would be completed in two years and followed by secondment to a government research establishment. Pople completed part two of the mathematical tripos as the European war was ending in May 1945; he was then a senior scholar of Trinity.

Pople spent a year with the Bristol Aeroplane Company before returning to Cambridge in 1947 to take part three of the tripos, concentrating on theoretical physics. He joined the department of theoretical chemistry in 1948 as a research student under the supervision of Sir John Lennard-Jones. Lennard-Jones, or L-J as he was affectionately known, had made significant contributions to statistical mechanics and to the quantum mechanics of molecules, before becoming the wartime chief superintendent of armament research at Woolwich and then director-general of scientific research at the Ministry of Supply. Pople received his PhD in 1951 for a thesis entitled 'Lone pair orbitals'. He was an author of several papers in the *Proceedings of the Royal Society* under the serial title 'The molecular orbital theory of chemical valency', started by Lennard-Jones in 1949. He rapidly began writing papers on his own and his paper entitled 'Molecular association in liquids: a theory of the structure of water' (1951) was influential. He was Smith prizeman in 1950 and a fellow of Trinity from 1951. Meanwhile from 1948 he had been learning to play the piano; his teacher was Joy Cynthia Bowers (1924–2002), daughter of George Bowers. After a long courtship they were married on 22 September 1952 in Great Saint Mary's Church, Cambridge. They lived initially in the converted kitchen of a Tudor house in Thriplow and later in a modern house in west Cambridge. They had a daughter and three sons and enjoyed a stable and loving relationship.

In 1954 Pople was appointed to a lectureship in mathematics at Cambridge, so his teaching was in mathematics and his research in theoretical chemistry. Nuclear magnetic resonance was rapidly emerging as an important tool in chemistry, and Pople joined W. G. Schneider and H. J. Bernstein at the National Research Council of Canada in Ottawa in the summers of 1956 and 1957, leading to their influential book *High-Resolution Nuclear Magnetic Resonance Spectroscopy* (1959). In 1958 Pople left Cambridge to become head of a new division of basic physics at the National Physical Laboratory in Teddington. It was a comparatively fallow time for him, so in 1961–2 he went to the Carnegie Institute of Technology in Pittsburgh as Ford visiting professor. He and his wife and young family enjoyed travelling in eastern USA. It was during this year that he decided to seek a position with more time for research. After considering a number of offers, he returned to Pittsburgh in 1964 as Carnegie professor of chemical physics. The Carnegie Institute merged with the Mellon Institute in 1967 to become Carnegie Mellon University, and Pople became the John Christian Warner professor of natural science.

On arrival in Pittsburgh, Pople resolved to return to the fundamental problems of the electronic structure of molecules. The rapid evolution of high-speed computers had greatly enhanced the prospects for developing quantitative 'model chemistries'. Pople's idea of a model chemistry was based on Schrödinger's equation and involved a precisely formulated mathematical procedure; it would be tested against known chemical data to determine its reliability and could be used to make predictions and solve disagreements. His research group in Pittsburgh quickly began extending the semi-empirical Pariser–Parr–Pople theory of planar unsaturated hydrocarbons, first put forward in 1953, culminating in the book *Approximate Molecular Orbital Theory* by Pople and D. L. Beveridge (1970). Semi-empirical model chemistries are an economical theoretical approach to solving Schrödinger's equation but have serious shortcomings and were later superseded by advances in density functional theory.

In the late 1960s Pople saw a natural progression from the semi-empirical to the *ab initio*. He greatly improved the efficiency of existing computer programs and set about developing efficient basis sets. He made use of Gaussian basis functions, as originally suggested by S. F. Boys in Cambridge in 1950. Pople made these advances in *ab initio* quantum chemistry widely available in 1970 through the Gaussian 70 programme which was distributed at low cost through the quantum chemistry program exchange of the University of Indiana. The code was fast and user-friendly, and rapidly became the market leader. This phase of the development of Hartree–Fock single-electron quantum chemistry was later described in the book *Ab-initio Molecular Orbital Theory* (1986) by W. J. Hehre, L. Radom, P. von Rague Schleyer, and Pople.

The main deficiency of Hartree–Fock theory is the neglect of correlation in the motion of electrons of opposite spin, leading to serious errors in computing bond dissociation energies and other properties. Most practical procedures for incorporating correlation start with the Hartree–Fock determinant and form linear combinations with other determinants that involve excited electron configurations. Pople made much use of the 1934 perturbation theory of Møller and Plesset, denoted as MP*n* when terminated at order *n*. The MP1 energy is identical to Hartree–Fock, and MP2 is the simplest perturbative approach to the correlation problem. This provided a useful and practical improvement to Hartree–Fock and became widely used. The MP2, MP3, and MP4 approximations were developed by Pople's group and incorporated into the Gaussian programme from 1976 to 1980. Other approaches to the correlation problem

investigated by Pople and others included configuration interaction and the coupled cluster method, in which excited electronic configurations are incorporated into the trial wave function in the Schrödinger equation. If all possible substitutions are included the method is called 'full configuration interaction', giving the solution of Schrödinger's equation for the basis set chosen. Unfortunately full configuration interaction can be implemented for small basis sets only.

Pople's G1 and G2 model chemistries of 1989 and 1991 provided a general procedure for predicting molecular energies. In his last version, G3 in 1998, the mean absolute energy deviation for a data set of 299 energies was 1.02 kilocalories per mole (it was 1.48 in G2) and the largest error in a heat of formation was 4.0 kilocalories per mole.

In the late 1980s the density-functional theory approach to the electronic structure of molecules formulated by Walter Kohn was rapidly gaining popularity following the introduction of much improved functionals. Pople was initially antagonistic to density-functional theory because it did not readily conform to his notion of systematic development of a model chemistry, but he embraced it actively in his last years at Carnegie Mellon.

By 1981 all four of the Pople children had left home in Pittsburgh, so John and Joy Pople moved to Illinois to be near their daughter and her young family. He continued to run his research group in Pittsburgh, commuting frequently and communicating by telephone and modem. He was an adjunct professor of chemistry at Northwestern University in Evanston, Illinois, from 1986 until his retirement from Carnegie Mellon in 1993, and was appointed trustees professor in 1993. He continued his research there until shortly before his death.

The Royal Swedish Academy of Sciences awarded the 1998 Nobel prize for chemistry jointly to Kohn, for his development of the density-functional theory, and Pople, for his development of computational methods in quantum chemistry. The award was widely acclaimed. While *ab initio* methods were already being successfully applied to small molecules when he transferred to the field from semi-empirical theory in 1969, he proceeded to publish results of computations on a wide range of molecules using well-defined models. His user-friendly Gaussian programme and its ready availability at modest cost meant that many chemists could perform informative calculations and be aware of the uncertainty in the results. He successfully popularized *ab initio* quantum chemical calculations. Many other honours were bestowed upon him. He was awarded the Marlow medal of the Faraday Society in 1958 and elected a fellow of the Royal Society in 1961. He was a founding member of the International Academy of Quantum Molecular Science in 1964 and served as president from 1997 to 2000. He received the Irving Langmuir award of the American Chemical Society in 1970 and its Linus Pauling award in 1977. He was elected a fellow of the American Academy of Arts and Sciences in 1971 and a foreign associate of the National Academy of Sciences in 1977. He was awarded the Wolf prize in 1992 at a ceremony in the Israeli Knesset. He was elected an honorary fellow of Trinity College, Cambridge, in 1999 and in 2001 received an honorary doctorate from the University of Cambridge. In 2002 he was awarded the Copley medal of the Royal Society, the premier award of the society (he received the Davy medal in 1980). He was knighted KBE in the new year's honours list of 2003.

Pople was a gifted teacher and a very successful lecturer, but it was his strong focus on research that was most admired by his students and colleagues. He travelled widely, particularly to Australia, New Zealand, Israel, and Germany. He was the founding director of Gaussian Incorporated of Pittsburgh in 1987. Gaussian Inc. took over the copyright ownership of future Gaussian programmes from Carnegie Mellon University after a long and costly court case. Nevertheless Pople failed to maintain control of the company and in 1993 his interest was bought out. A rival company, Q-Chem. Inc., was founded by Pople's co-workers in Pittsburgh in 1993, but he was prevented from being involved until 1999 by a six-year non-competition agreement with Gaussian Inc. Thereafter Pople was an influential director of Q-Chem, where his daughter, Hilary, also worked, as the company accountant. He died at his home in Chicago on 15 March 2004 of cancer of the colon. He was survived by his four children. A memorial service was held at the First United Methodist Church of Evanston, Illinois, on 24 March. A computer room at Bristol grammar school, endowed by his brother, was named the Pople room in his honour.

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Likenesses

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