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Lionel A. K. Staveley 11 November 1914–8 February 1996



# Lionel Staveley: The Man and The Scientist

Lionel Alfred Kilby Staveley, who died on 8 February 1996, was a leading figure of worldwide reputation in Chemical Thermodynamics and one of the great experimentalists of his age. In a career spanning over 40 years he made outstanding contributions to the development of experimental thermodynamics and the advancement of theory. He was born in Stamford (Lincolnshire) on 11 November 1914 and received his education at Stamford School and Trinity College, Oxford. In Oxford he came under the influence of the redoubtable C. N. Hinshelwood (Chemistry Nobel Prize winner, 1956), who was his tutor at Trinity and with whom he published his first papers (on reaction kinetics) while still an undergraduate. However, his very first paper appeared in 1933 — when Staveley was barely eighteen! — the result of a vacation project carried out in Cambridge with E. A. Moelwyn-Hughes. After obtaining First Class Honours in Chemistry in 1936, he went to Munich, Germany, to work for a Ph.D. degree in low temperature thermodynamics under K. Clusius. Those were troubled times and Staveley had to cut his stay short and return to England after a year because of the impending war. While in Munich, he carried out experiments on the effect of isotopic composition on the triple-point of cryogenic liquids, including argon and xenon. (He would return to some of these problems some 40 years later.) The first papers on thermodynamics (with Clusius) were published in 1941 but, because by then Britain was at war with Germany, Staveley's name was only listed as that of a collaborator, responsible for the experimental part of the work! Study grants were small then, and Staveley used to take his evening meals at the main railway station in Munich because of their good value for money.

At the end of 1938, he went back to Oxford to take up a lectureship in the Faculty of Physical Sciences, and at the same time was elected as Fellow and Tutor at New College where he stayed until his retirement in 1982. His research during World War II reflected British war needs. As a part of a team under the tutelage of Hinshelwood, he worked on the adsorption properties of respirator charcoals and later, for the Ministry of Supply, on the combustion kinetics and thermochemistry of mixtures of an oxidizing and an oxidizable solid. Already in this work, he demonstrated great experimental ingenuity and a sound grasp of theory — two hallmarks of all his future research. His Munich period may have been short (1937–38), but it set him on a path for which he would receive worldwide recognition — the thermodynamic investigation of systems composed of simple condensed gases. Since many of these substances have also both ordered and disordered crystalline forms, his later interest in the subject of disorder in solids was also then established.

From Clusius, Staveley learned that if one measured suitable physical properties accurately enough, one might come across some new, important qualitative truth. He was proud of the fact that by having worked under Clusius he fitted into some sort of genealogical sequence going back to Nernst, the founding father of low-temperature physics and chemistry (Clusius had been a pupil of Eucken who, in turn, had studied with Nernst). But his true idol in the field of thermodynamics was W. F. Giauque (another Chemistry Nobel Prize winner, 1949) whose dedication to accuracy, complete exploration of all the detailed aspects of a problem, and ruthless rejection of approximation wherever exactness and precision could be achieved, were legendary. However, as Staveley often pointed out, precision should simply be a means to an end, not an end in itself. Hinshelwood, Clusius, Giauque — these were the giants on whose shoulders Staveley stood. Being a very tall man himself, he was able to see — and did see — very far!

With the help of a Royal Society grant (£13) Staveley set up a laboratory in the Abbot's Kitchen (the old Oxford chemical laboratory adjoining the University Museum, modeled on the kitchen at Glastonbury Abbey), long before he moved to more spacious premises on the first floor (east) of the Inorganic Chemistry Laboratory in South Parks Road. He later became a Doctor of Science of Oxford University and Reader in Inorganic Chemistry, and often acted as a much respected and efficient Head of Department. He was an Oxford don in the traditional mould, utterly dedicated to university and college activities. It was often a very strenuous life, spent coping with administrative duties, tutoring and lecturing undergraduates on the whole spectrum of inorganic and physical chemistry, while at the same time pursuing first-class research in the whole gamut of thermodynamics. He was an inspiring teacher. His lectures were meticulously prepared and he had a knack of presenting the most difficult or tricky topic in a rigorous and yet very clear way. His grasp of detail was phenomenal and he could be an exacting supervisor. But he was also patient and compassionate with his students. Whenever there was a problem, he was always ready to help with his wisdom, a sparkling remark, a humorous quote. He took a daily interest in the progress of the work, even finding the time, at the end of a long and tiring day, to stay on until he felt that we were in complete control of the technical difficulties. I will never forget, soon after I arrived in Oxford, how he sat with me at the bench to introduce me to the pleasures (and burns!) of glassblowing! As it has often been said, he projected the aura of "a man of Edwardian dignity", self-effacing and yet capable of arousing the fiercest loyalties.

Two essential features stand out in Staveley's scientific research: first, the simplicity and elegance of his techniques and apparatuses (which he designed himself), and then the quality and accuracy of the results, which have withstood the test of time. The solutions that he devised to cope with some complex experimental problems were often ingenious, but they were also clearly based on the fundamental principles of physics and thermodynamics, and as such they served to deepen the students' understanding of physical laws and of the properties of matter. Everything was built in the university workshops, sometimes on a shoestring budget. His research group was never very large, but he inspired such loyalty that everyone worked hard and no-one wanted to fail him. In 1968 he received (jointly with W. B. Streett) the Russell

B. Scott Memorial Award of the American Chemical Engineering Society for the outstanding papers of 1967 in Cryogenic Engineering Research.

Staveley's work was not restricted, however, to low temperature work. In the 40s and 50s he did important studies on hydration and hydrogen bonding (solubility of water in various solvent mixtures and volume relations in dilute solutions of primary alcohols in non-polar solvents, etc). There was also important work on supercooling and homogeneous nucleation in liquids, and he published some fundamental papers on the thermodynamics of complex formation in solution (several polyaminocarboxylic acids with metal ions), etc.

Staveley's research on solids was chiefly concerned with order-disorder problems (positional, orientational and magnetic disorder) in a variety of substances: hydrates, clathrates, mixed ammonium halide systems, charge-transfer complexes, metal tetra-alkyls, plastic crystals, layered compounds, etc. (more than 120 compounds at the latest count!). He showed great imagination and originality in the selection of systems and, ahead of his time, often foresaw their ultimate importance. In some cases {Hg(NH<sub>3</sub>)<sub>2</sub>X<sub>2</sub>, where X = Cl or Br} the compounds were chosen because they illuminated the so-called "dimer problem", *i.e.*, the number of ways in which dimers may be placed on a lattice so that all of the sites are occupied exactly once; in other cases (clathrates), because they provided a good model for the lattice theory of liquids. Most of these studies were pursued through adiabatic calorimetry, from T = 1 K to T = 600 K. He measured a whole range of properties, from heat capacities to transition temperatures and the changes of enthalpy, entropy and Gibbs energy associated with those transitions; these studies were often supplemented by dilatometric determinations of volume changes through transitions, and the measurement of adiabatic and isothermal compressibilities, thermal expansion coefficients, even electric conductivity. He also made important contributions to the understanding of the complex phenomenon of hysteresis in phase transitions in solids. Later he would publish the definitive account of the subject, Disorder in Crystals (Clarendon Press, 1978), with his former pupil Neville Parsonage.

He was fond of saying that "theoreticians, like artists, need their models" and thus felt impelled to provide the community of theoreticians with good, reliable results for model systems on which they could test their theories. The problem is that for the theory to work, the model has to be simple, and the simpler the molecular system, the more complex gets its practical realization. But Staveley had an easy rapport with theoreticians, leaving out, in their discussions, "the sordid details of the experiment". His work on simple liquid mixtures (mixtures of condensed gases) was prompted by the conflicting predictions of two current theories in the 1950s — the cell theory (later Average Potential Model) developed by Prigogine (Chemistry Nobel Prize winner, 1977), and the Conformal Solution Theory of Longuet–Higgins. Prigogine sent one of his collaborators (Victor Mathot) from Brussels to Oxford to work on a system simple enough to provide a good test for the theory. For practical reasons the choice fell on the CO + CH<sub>4</sub> system, and the results vindicated the cell model. A large majority of the simple systems investigated during the next 25 years originated in Staveley's laboratories, where he measured the three major excess

functions ( $G^{E}$ ,  $H^{E}$  and  $V^{E}$ ) and also the equation of state over wide ranges of temperature and pressure (up to 4000 bar). When, in the mid-1970s, Keith Gubbins (in collaboration with Chris Gray) was developing his perturbation theory for non-spherical molecules, he gravitated naturally towards Staveley, the only experimentalist who could provide him with the necessary measurements. Thus followed a series of (now) classical papers investigating the role of non-central forces and polarity in the thermodynamic properties of mixtures.

Staveley was for several years a member of the IUPAC Commission on Physicochemical Measurements and Standards, serving as its chairman from 1963–67 and in that capacity he edited an important report on *Characterisation of Chemical Purity* — *Organic Compounds*, published in 1971. In Britain he was honoured with the Chemical Society Award for Thermodynamics and Electrochemistry, and he was also the tutelary figure of many Dense Fluids and Mixtures Groups, and Experimental Thermodynamics Conferences. B. J. Alder, the father of computer simulation, said that Staveley was his ideal type of scientist. When he looked for experimental data on really interesting systems, he invariably found that Staveley had obtained them, and that the quality was always good. John Prausnitz once told me, with a mixture of admiration and envy, that Staveley's research projects were those that he himself would like to have tackled, if only he could have found the corresponding financial support (at the time, Britain had a much freer academic environment than the U.S.A., as far as research was concerned).

His connections with America began in 1962 when he spent a sabbatical term as Cherwell Visiting Fellow at Pennsylvania State University. There were also extended visits to the University of Michigan (where his good friend Edgar Westrum Jr. had built another world-class calorimetric laboratory), to the (then) National Bureau of Standards in Boulder and Cornell University (where Bill Streett and Keith Gubbins had been appointed to chairs in chemical engineering).

Probably his most lasting influence has been felt in Portugal where his disciples occupy top academic positions in five universities. Staveley was a frequent visitor to Portugal to give courses and seminars at various graduate schools, discuss doctoral theses and advise and collaborate on several research projects. For his services to chemical education and research in Portugal, he received the Special Award of the Portuguese Chemical Society. He was also a corresponding member of the Lisbon Academy of Sciences, and a *honoris causa* Doctor of the Technical University of Lisbon. There is also a Lionel Staveley Lecture as part of the International Conferences on Thermodynamics of Non-Electrolyte Solutions (a joint-organization of the Chemical Societies of France, Spain and Portugal). He gave the first Staveley Lecture at a conference in Lisbon in 1982 (other Staveley lecturers have been K. E. Gubbins, C. J. Wormald and E. U. Franck). When, in 1983, he delivered the second Lennard-Jones Lecture at the University of Hull, Brian Smith, in his introduction said that Lionel was fortunate indeed to have given both the Staveley and Lennard-Jones lectures, for Lennard-Jones would never be able to give either of them.

Staveley's career was one of great integrity, moulded by intellectual curiosity, in total independence, sometimes even against fashion. I remember his observation,

after I had written my D. Phil. thesis, that there was enough material there for two independent papers. He seemed to be slightly disappointed that not everything could be fitted into a single paper! For him "less was more", a lesson that I have not forgotten. Lionel Staveley inspired and shaped the careers of many of us, so much so that when he retired in 1982 almost 200 former students and collaborators turned up from all over the world to pay tribute, in a Symposium in his honour, to his extraordinary achievements. He was genuinely surprised that so many came, some from great distances to take part in it. He then generously donated his equipment and apparatuses to former students and collaborators in Britain, Canada and Portugal. A good part of this equipment is still in use at the Thermodynamic Laboratories of Instituto Superior Técnico, in Lisbon. In fact, most of the current work on isotopic mixtures in Lisbon is carried out in an apparatus that he designed and built but never used. He also donated his scientific books (including some rare items) and papers to the Instituto Superior Técnico library, as well as some drawings (real works of art!) of complex pieces of equipment (calorimeters, a cell for the study of vapour pressure isotope effect) which he had designed.

After his retirement he carried on with his scientific collaborations with some of his former students. I remember, a couple of years ago, how pleased he was when another paper appeared exactly 60 years after he had published the first one. In all, there have been over 180 scientific papers, with at least another one in the pipeline, due to appear in the coming months. His work lives on, and 63 years in print is an amazing achievement, almost on a par with that of Joel Hildebrand!

Lionel Staveley had many interests besides science and thermodynamics. He was a very cultured man, with a love and knowledge of music and literature. Once, when he caught me reading Jane Austen in the laboratory to pass away the long waiting hours for the system to reach equilibrium, he surprised me by reciting long chunks from Pride and Prejudice that he had learnt by heart. He was also an expert gardener, serving in the Garden Committee of his College. (At the time of his retirement his friends and colleagues gave him a glasshouse for his home garden, as a farewell present.) A devoted husband and father, Lionel Staveley was a man with an almost unlimited capacity for friendship. He and his wife Joyce always made us feel welcome and their hospitality and kindness towards visitors, especially those coming from abroad, was proverbial. I also treasure the countless trips I made with them to manor houses and stately homes, chosen usually for their artistic merit or literary associations. His last years were marred by debilitating arthritis which he bore with great stoicism, attended by the loving care of his wife, Joyce, and the help of their daughter Rosalyn and son John (their youngest son, Anthony, had moved to France after marrying). It is often said that behind a great man there stands a great woman, and this was also true of Lionel and Joyce Staveley who were blessed with a happy marriage lasting 56 years.

An important era in experimental thermodynamics has come to an end. Lionel Staveley's work reached standards of excellence which will rarely be matched, let alone surpassed. We will not see his like again.

I am grateful to all the contributors to the Lionel Staveley Retirement Meeting (*Rev. Port. Quim.* 1983, 25, 65), in particular to Professors Douglas Everett and Roger Linford, as well as to Dr Peter Dickens, for their personal reminiscences about Lionel Staveley, my beloved teacher and mentor.

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JORGE C. G. CALADO

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