

Stephen John Angyal 1914–2012

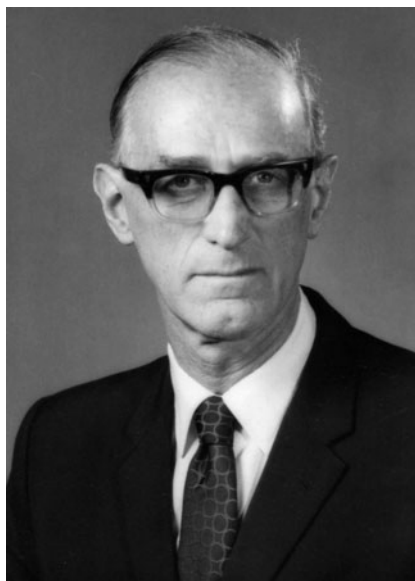
John D. Stevens

School of Chemistry, University of New South Wales, Sydney, NSW 2052, Australia

Stephen Angyal was born in Budapest, Hungary, on 21 November 1914 and died in Sydney on 14 May 2012. He had a distinguished career as an organic chemist as a lecturer in chemistry at Sydney University (1946–52), as an associate professor in organic chemistry at the New South Wales University of Technology (1953–9), and as Professor of Organic Chemistry at the University of New South Wales (1960–79) where he served as Head of School (1968–70) and Dean of the Faculty of Science (1970–9). He was internationally recognized for his contributions in the fields of inositol and carbohydrate chemistry, being appointed as the Haworth Memorial Lecturer of the Chemical Society, London, in 1980 and as the recipient of the Claude S. Hudson Award of the American Chemical Society in 1987. He was elected as a Fellow of the Australian Academy of Science in 1962 and as an External Member of the Hungarian Academy of Science in 1990 and his contributions to science in Australia were acknowledged in the award of Officer of the Order of the British Empire (OBE) in 1977.

Family Background

Stephen John Angyal was born in Budapest on 21 November 1914, the first child of Charles Engel and Marie Engel (née Szanto). His father, Professor Charles Engel, was born in a village in the North of Hungary in 1879. Charles studied in Budapest, received his medical degree in 1902 and became a highly respected specialist in syphilis. He published two books on the subject. Stephen married Helga Ellen Steininger in 1942. They had two children, Annette (b. 1944, now Associate Professor Annette Gero, a biochemist) and Robert (b. 1949, a barrister at law, married Abby Bloom, two daughters, Chloe and Claire). Helga came from Vienna where her father, Dr. Jacques Steininger practiced as a lawyer. Her mother was Augusta Steininger. As a keen reader of Literature, Charles told young Stephen about his books while they were on Sunday morning walks, introducing him to the works of one of his favourite authors, Somerset Maugham. Charles was an accomplished pianist and regularly played chamber music with several string players (including some of Hungary's now famous twentieth-century composers, who came to the house) Stephen often turned the pages for his father. These sessions introduced Stephen to nineteenth-century chamber works and laid the foundation for his life-long love of music. To speak several languages was important in Central Europe, and his parents engaged German



nannies, usually young country girls. Stephen spoke German before Hungarian. Later, for his last three summer vacations at school, he was sent to a Swiss institution to learn French, and he also took lessons in English at home in the afternoons. This early facility with several European languages was to stand him in good stead through his long life in academia.

Stephen had one brother, Charles Leslie Angell (1926–2007) who migrated to Sydney in

1947. After completing a MSc degree from the University of Sydney in 1951, Charles obtained a PhD degree in chemistry from Cambridge University in 1955. He worked as a Research Scientist with Union Carbide in New York. He was an expert in the field of infrared spectroscopy and was President of the Coblenz Society from 1969 to 1971. He is survived by his wife Margee, son Stephen and daughter Catharine.

Education

At ten years of age, Stephen enrolled in the prestigious high school of the Piarest order where he studied Latin and Greek, much literature and essentially no chemistry. During this time, Stephen joined the school's scout group where he learned to swim and also the art of skiing, two activities that he practiced with much enthusiasm until very late in life. He hung up his skis at the age of 92. After high school, he enrolled at the Royal Hungarian University of Science in Budapest where he studied chemistry, physics, mathematics, geology and mineralogy. For the PhD degree, Stephen moved to the other university in Budapest, the University for Technology and Engineering, where he enrolled under Professor Géza Zemplén, although most of the supervision was done by an assistant, Z. Csűrös. The research work on glucose derivatives which led to the award of the Doctors degree *Summa cum laude* in June 1937 did not spark the imagination of Stephen and he vowed never to work with carbohydrates again.

Prior to enrolling with Zemplén, Stephen changed his name from the German spelling (Engel) to the Hungarian version, Angyal. His formal education was completed by one year's military service, graduating from officers' school as a Lieutenant in the reserve.

Early Work

Stephen's first job after graduation was as a research chemist with Chinoin, one of Hungary's leading pharmaceutical manufacturers. Here he worked on the production of synthetic oestrogens and sulfathiazole. Knowledge of the latter would later lead to his first job when he arrived in Australia. With great foresight, Stephen obtained a landing permit for Australia, which he received on 3 July 1939. September 1939 saw the start of World War II and Stephen was called up for

army duty. Again, luck was with him as Hungary kept out of the conflict until November 1940 and Stephen was demobilized. He immediately made preparations for his emigration and left Hungary early in 1940, travelling to Milan and boarding in Genoa the passenger ship *Viminale*, which happened to be the last Italian ship to leave for Australia as, soon after, Italy entered the war on the Axis side.

After arriving in Sydney, Stephen met a fellow Hungarian, Dr. Andrew Ungar, with whom he started a small company, named Andrews Laboratories. Although at the time little came of this venture, the association was of considerable significance in later years. To keep in touch with the local chemistry community, Stephen used to visit the Chemistry Department of Sydney University where he met Dr. Frank Lions who told him of their synthesis of sulfathiazole as part of the war effort. Stephen noted that he had also made that drug, and after this was reported in a local newspaper he received a phone call from a Melbourne pharmaceutical company, Nicholas Pty Ltd, in which Stephen was offered a job as a research chemist. During the five years that Stephen worked in Melbourne, he became aware of a compound, inositol, thought at the time to be a member of the vitamin B group. He decided that as very little was known about its chemistry, it may be worth studying some day. In connection with the synthesis of a new sulfa drug, marfanil, Stephen noted an unusual reaction that had been discovered by the French chemist, Sommelet. He decided that this reaction warranted further study and the results led to a series of papers and a review.

A particularly happy event that had lifelong ramifications for Stephen was his meeting in Melbourne with a lovely young girl, Helga Steininger. Originally from Vienna, Helga had arrived from England six months earlier. She worked as a dress designer. They were married in Melbourne in February 1942 and remained an inseparable couple for the rest of Stephen's life.

Academic Appointments

In 1946, Stephen secured a lectureship in chemistry at Sydney University and here he started a study on the Sommelet reaction as well as the early work on inositols. During that year, he was admitted as an Associate of the Royal Australian

Chemical Institute (RACI) and later (1953) was elected as a fellow of the RACI. As a result of Australia being geographically isolated from the northern hemisphere countries where most chemical research is carried out, it was usual for Australian academics to be given a year's paid leave every seventh year to allow them to study overseas. During his sixth year at Sydney University, Stephen was invited by Alexander Todd, who was a Visiting Professor at the university, to spend his study leave at Cambridge University. As Todd was one of the trustees of the prestigious Nuffield Dominion Travelling Fellowship, he suggested that Stephen should apply for one which in due course he was awarded. During his stay in Cambridge, Stephen met an Australian chemist, John Mills. From their mutual interest in conformational analysis, they wrote a review³ for *Reviews of Pure and Applied Chemistry*. They later collaborated on several aspects of metal ion complexes.

In 1952, Stephen obtained a grant-in-aid from the Carnegie Corporation of New York. This allowed him to visit several institutions in the USA on his way back to Australia in February, 1953. Among the chemists that Stephen met were Hermann Fischer with whom he spent his next study leave and Ernest Eliel, a fellow contributor to the well known book, *Conformational Analysis*.⁴

While Stephen was on his study leave in Cambridge, an advertisement for an associate professor in organic chemistry appeared from the New South Wales University of Technology. He applied for that position and was successful, taking up the appointment in March 1953. In 1958, the name of the institution was changed to The University of New South Wales (UNSW hereafter) and Faculties of Arts and of Medicine were established in 1960 and 1961 respectively. In 1960, Stephen successfully applied for the newly created chair of organic chemistry and he became the first Professor of Organic Chemistry at UNSW.

Stephen was now in a position to start work on the inositols. Few aspects of cyclitol chemistry escaped the attention of the Angyal group. In over sixty papers, cyclic acetals, sulfonic esters, phosphoric esters, aminocyclitols and their deamination, 5-carbon cyclitols, acyl migration, tritium labelling, and selective protection were dealt with. Of the nine possible isomers

of inositol, only the common one, *myo*-inositol, which is widespread in nature, is available at a reasonable price from commercial sources. The preparation of acetal derivatives of inositols, in particular, acetone derivatives, was the key to the interconversion of the various isomers. Satisfactory acetonation of *myo*-inositol was achieved only after a detailed study⁵ of the reaction conditions. Two of the nine isomers had not been prepared at the beginning of this research. The first one, *neo*-inositol, was prepared⁶ from the tosylate of *levo*-inositol diacetal. The remaining isomer, *cis*-inositol, proved to be the most difficult to prepare. Initially isolated⁷ by chromatography over cellulose powder from the multitude of products formed by hydrogenation of hexahydroxybenzene, later prepared⁸ in 25% yield in seven steps from *epi*-inositol and finally obtained⁹ in 31% from hydrogenation of tetrahydroxyquinone using an improved catalyst and separation of products on an ion-exchange column.

A most significant result that emerged from these early studies involved the interaction between the inositol hydroxyl groups and sodium borate in aqueous solution.^{10,11} The products were shown to be tridentate boric esters. From the equilibrium constants determined for this reaction, the free-energy changes of complex formation were calculated. As the different non-bonded interactions determined the extent of complex formation, it was possible to calculate these various energies of interaction. Another reaction discovered at this time involved treatment of inositols with acetic acid containing sulfuric acid. For those inositols in which a *cis-trans* sequence of hydroxyl groups occurs, it was found¹² that epimerisation occurs at the middle carbon atom. A reasonable mechanism for this reaction was described. Again, non-bonded interaction energies could be calculated¹³ from the differences between the free energies of the epimers.

A detailed review on the cyclitols was written with Laurens Anderson¹⁴ at this time and the Royal Australian Chemical Institute recognized¹⁵ the research by the award of the H. G. Smith Memorial Medal in 1958.

With the values for interaction energies for inositols in hand, Stephen applied these values to the pyranose forms of reducing sugars in order to explain the α : β ratios for aqueous solutions¹⁶

and the ratios of reducing sugars and the 1,6-anhydrides in equilibrium¹⁷ and later extended this work to acyclic compounds.¹⁸ At this time Stephen published an article on the composition and conformations of sugar in solution.¹⁹ He regarded this as his most successful paper, as it was used in several universities as a teaching aid with every student being given a copy. A much more detailed review on conformational analysis as applied to carbohydrates was published in the well known text book.²⁰ Stephen was awarded the Archibold D. Ollé Prize by the Royal Australian Chemical Institute for this publication.

As part of a study leave in 1962, Stephen gave a lecture tour in Israel on the way to Paris where he spent six weeks at the Institute for the Chemistry of Natural Products. He then travelled to London to take up an appointment as temporary senior lecturer in D. H. R. Barton's department at the Imperial College of Science and Technology. During this study leave, Stephen attended the first International Symposium on Carbohydrate Chemistry (that meeting is now regarded as number II). Subsequently, he attended almost all of these biannual meetings and after his retirement from UNSW in 1979, he organized and hosted the tenth meeting which was held at UNSW in Sydney. Participants remember this meeting with much fondness. An historical account²¹ of these meetings was published in 2008.

Stephen's interest in carbohydrates was rekindled when he was invited to review the topic for *Annual Reviews of Biochemistry*.²² At this time, the award of a research grant enabled the purchase of a Varian A60 nuclear magnetic resonance (NMR) spectrometer. Now a method was available to him for the determination of the composition of many aldoses in aqueous solution using ¹H NMR spectroscopy. These studies were extended to ketoses²³ after a ¹³C NMR spectrometer became available. This work led to a comprehensive review²⁴ of sugar compositions in solution, followed by a supplementary review²⁵ seven years later. As a prelude to this experimental work, Stephen published a detailed paper²⁶ on the conformational free energies of aldopyranoses in solution in which the interaction energies mentioned above were applied to the reducing sugars.

Following up on electrophoresis studies by John Mills in which he showed that various

polyols complexed with a variety of salts, Stephen commenced a study,²⁷ using ¹H NMR spectroscopy, of the complexing of sugars with metal cations. This led to a significant series of papers which covered a diverse range of topics such as the modification of glycosidation using acidified methanol plus salts to give less readily obtained glycosides^{28–30} and dimethylacetals,³¹ and application of cation-exchange resins,^{32,33} commonly in the Ca²⁺ form, to effect separation of glycosides,³⁴ inositols,³⁵ and, in his last experimental paper, applied to the preparation of L-ribose.³⁶ These studies were the subject of several reviews^{37–40} and formed his topic for the Haworth Memorial Lecture,⁴¹ delivered on 31 March, 1980 and also for the C S Hudson Award Address.⁴² Support for the proposed structures of metal cation complexes deduced from NMR spectra was obtained by single-crystal X-ray diffraction studies.^{43–45}

In a detailed paper⁴⁶ on methyl aldofuranosides, Stephen extended his studies on conformations to five-membered ring compounds. In this paper, extensive ¹H NMR data were collected and the conformations deduced therefrom were correlated with optical rotation values.

In a short series of papers, the application of chromium trioxide in acetic acid was described in which O-methyl groups were converted⁴⁷ into O-formates. Acetylated methyl glycosides gave⁴⁸ acetylated methyl 5-hexuloseonates, acetylated acetals of alditols gave⁴⁹ derivatives of 3-hexuloses and cyclic methylene acetals gave cyclic carbonates.⁵⁰ Building upon the discovery by Koch and Stuart that the methyne hydrogen of secondary alcohols can be exchanged for deuterium using deuterated Raney nickel in deuterium oxide, Stephen applied the procedure to a variety of compounds, including inositols and inositol methyl ethers,⁵¹ methyl glycosides,⁵² and 1,6-anhydrohexoses.⁵³

Many research students and collaborators contributed over the years to the realization of the large body of experimental work recorded in Stephen's publications. His joint work with John Mills and Laurens Anderson documents important understanding of the inositols and the work of Dennis McHugh, to select just one of the many research contributors in Stephen's group, on the determination of interaction energies,⁵⁴ played a key role in predicting the conformations of polysubstituted six-membered ring systems.

As a world expert on inositols, Stephen was chosen to chair the IUPAC committee dealing with inositol nomenclature.⁵⁵

Stephen's role as a leading Australian chemist was recognized by the Royal Australian Chemical Institute when he was interviewed in 2011 as a 'living luminary' (*Chemistry in Australia*, March 2011).⁵⁶

The Andrews Lectures

During the course of the first International Symposium on the Chemistry of Natural Products which was held in Australia in 1960, Stephen decided to invite one of the participants to give a series of lectures over a two week period. Professor E. R. H. Jones accepted the invitation and became the first Andrews Lecturer. Funds for the lectures were provided by Dr. Andrew Ungar and the lectures were named after his company, Andrews Laboratories. Over the next fifty years, twenty six eminent organic chemists have been appointed as Andrews Lecturer, each one delivering three lectures, generally over one to two weeks. It was an essential part of the Lectureship that the lecturer should spend a period between lectures at UNSW and be available for discussions with staff and advanced students. Until his retirement in 1979, Stephen made all the arrangements for these lectureships with significant help from Helga for the social engagements. After he retired, management of the Andrews Lectures was passed to University of New South Wales Chemical Society and they continue to arrange biannual lectureships.

Administration

Stephen served as Head of the School of Chemistry at UNSW from 1968 to 1970. He was appointed as Dean of the Faculty of Science in 1970 and kept that position until he retired in 1979. Stephen enjoyed being involved in the organization of conferences. After the Natural Products meeting in 1966, the next major conference was the XXII International Congress of Pure and Applied Chemistry held at the University of Sydney in 1969. Stephen was the convener of the Social Program and Helga chaired the Ladies' Program Committee. Finally, after his retirement, Stephen organized the Xth International Symposium on Carbohydrate Chemistry, held in 1980.

Throughout his academic career, Stephen was a keen traveller. He was Visiting Professor at the University of California, Berkeley in 1957, Visiting Lecturer at the Imperial College of Science and Technology, London in 1962, Visiting Professor at the Universities of Oxford and Grenoble in 1968, Visiting Lecturer at ETH, Zurich in 1972 and Visiting Professor at the University of Grenoble in 1977. Besides attending the International Symposia on Carbohydrate Chemistry, Stephen attended several the B rgerstock Conferences on Stereochemistry and several of the Gordon Conferences on Carbohydrates.

Although his early papers were published in the *Journal of the Chemical Society* (London), Stephen later favoured the local publication, the *Australian Journal of Chemistry*. He served as a member and in turn, chairman, of its editorial board. Despite early reservations on the introduction of a journal devoted solely to carbohydrates, Stephen supported *Carbohydrate Research*, serving on the Editorial Board for many years and publishing more than forty papers there.

Stephen was elected as a Fellow of the Australian Academy of Science in 1962 (he served on the Council from 1967 to 1970), awarded the first UNSW DSc degree in 1964, having been admitted to the degree of MSc ad eundem gradum in 1941, appointed as the Haworth Memorial Lecturer of the Chemical Society, London, in 1980 and received the Claude S. Hudson Award of the American Chemical Society in 1987. In 1990, he was honoured by the Hungarian Academy of Science, being elected as an External Member. His contributions to science in Australia were acknowledged in the award of Officer of the Order of the British Empire (OBE) in 1977 and in 2001 he was awarded a Centenary Medal 'for service to Australian Society through science and stereochemistry'. More recently, his daughter Annette has endowed UNSW with the Angyal Medal, which is awarded each year to the top Honours student of the year in Chemistry, as a permanent memory of Stephen in the School of Chemistry at UNSW.

Outside Interests

Stephen enjoyed several interests besides chemistry. He had learned to ski while at high school and this became a life-long activity until his

early nineties. Not surprisingly, most of his study leaves were taken at places close to snow-fields and he also made full use of the skiing facilities in the Australian ski fields. Another life-long pleasure for Stephen was swimming, which he enjoyed year round in the mild climate of Sydney. Both Stephen and Helga were lovers of music. They regularly attended performances of the Sydney Symphony Orchestra and musica viva. Both were staunch supporters of the Australia Ensemble (resident at UNSW) for which Helga was a member of the Advisory Board for many years and it was rare for them to miss a performance.

Stephen died peacefully on 14 May 2012, aged 97. His beloved wife, Helga, after seventy years of marriage, died peacefully and unexpectedly six months later.

Acknowledgements

In preparing this obituary, I have drawn upon extensive biographical notes prepared by Stephen in 2000 and on an interview by Mr. David Salt on behalf of the Australian Academy of Science in 2003.

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A full bibliography of scientific publications by Stephen John Angyal is available online as Supplementary Material to this paper.

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