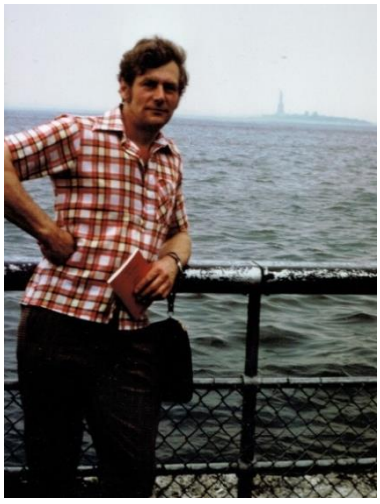
In memory of J.I. (Ian) Langford

Aug 12th, 1935 – Nov 17th, 2013



At N.Y., after "Accuracy in Powder Diffraction"
Gaithersburg MD (USA), 11-15.06.1979

The EPDIC community is honoured to entitle MicroSymposium 7 on "Diffraction line profile analysis, stress, strain and texture" (EPDIC 14, Aarhus, Monday 16th, June 2014 at 10:00 hours) to Ian Langford, a leading figure in the development of modern powder diffraction. All participants are kindly invited to attend the brief commemoration ceremony at the MS7 opening.

John Ian Langford was born in Chingford, Essex, U.K., on 12 August 1935. It was at the Viriamu Jones Laboratory in the Department of Physics, University College, Cardiff, that he started his academic career as a Research Associate. Initially, the main purpose of his appointment was to design and construct a system for the programmed control of an X-ray powder diffractometer and for the automatic recording and storage of diffraction data in digital form, the forerunner of the modern Automatic Powder Diffractometer. He was also employed to obtain high quality data for the Powder Diffraction File (PDF). Ian became interested in the theory and application of powder diffraction in materials science and has been since engaged in this field. His contribution has mainly been concerned with the development of high resolution diffractometry with conventional and synchrotron X-ray sources, with improving the reliability and accuracy of parameters derived from diffraction data and developing procedures for studying the microstructural properties of materials routinely. During his time at Cardiff he collaborated with Professor A.J.C. Wilson in the development of a new technique for analyzing diffraction line profiles, based on their variance (second central moment), for characterizing microstructural properties – the size and shape and crystallites and 'mistakes' of crystallization, dislocations, etc. This work, together with the design of the Cardiff powder diffraction system, was the subject of a thesis for which he was awarded the degree of PhD by the University of Wales in 1965.

In 1966, Ian moved to the Department of Physics, University of Birmingham, with Professor Wilson. He was responsible for the installation of a high precision X-ray powder diffractometer. Early work included the development of an improved technique and associated software for determining precise and accurate dimensions of the unit cell for any crystal system, taking into account both random and systematic errors. Its potential was demonstrated in a variety of mineralogical and other applications. A major contribution to procedures employed in powder diffraction was his development of the variance method from a theoretical concept to a technique used in studies of structural defects. A consideration of various systematic errors led to greatly improved accuracy in measurements of crystallite size and microstrains.

One the most important developments, to which Ian has contributed substantially, is the modeling of complete, and often complex, powder diffraction patterns by using suitable analytical functions. A wealth of information could be obtained for most crystalline materials, whereas detailed studies of lattice imperfections were previously restricted to substances with relatively simple diffraction patterns. A starting point for this work was his introduction in 1978 of the Voigt function. This turned out as the most convenient function which allows a straightforward separation of the instrumental and sample contributions and at the same time describes the form of the peaks adequately. The use of the Voigt function was the subject of an invited lecture at the Symposium on Accuracy in Powder Diffraction, Gaithersburg, 1979. This work was continued in association with Drs Th.H. de Keijser, E.J. Mittemeijer and coworkers, Delft University.

The "Dream Team" of Line Profile Analysis at "Accuracy in Powder Diffraction", Gaithersburg MD (USA), 11-15.06.1979. Left to right: E.J. Mittemeijer, J.I. Langford, Th.H. de Keijser, D. Louër

An important and novel contribution to line-broadening theory and practice has been his interpretation of direction-dependent line breadths in terms of the mean dimensions and shape of crystallites. Much of this work was carried out in collaboration with the University of Rennes, where Ian has been *Professeur Invité* (several times for about twenty years), in close collaboration with Dr D. Louër. Initially the theory was applied to high symmetry materials, notably ZnO samples, prepared in various ways, of interest in studies at Rennes on solid-state reactions, and the results showed excellent agreement with theoretical predictions. While accurate methods for line-broadening analysis could only be applied to diffraction patterns with

well resolved lines, the use of flexible analytic functions, e.g. the Voigt and pseudo-Voigt, opened the door to the fitting of a suitable model to the total diffraction pattern. The pattern decomposition approach gave the possibility to extend line broadening analysis to more line profiles. The development of a program for total pattern fitting was undertaken, in 1985, in collaboration with Drs E.J. Sonneveld and J.W. Visser from Delft University. Moreover, the consideration of line broadening due to microstructural effects in the Rietveld method was the subject of contacts with the Laboratory of Metallurgy at Delft University, Drs E.J. Mittemeijer, R. Delhez and Th.H. de Keijser, and the Rennes laboratory. A chapter based on this work is included in the book on the Rietveld method (IUCr-OUP, 1993).

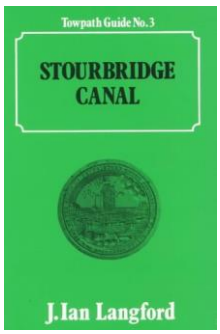
In the second half of the 90s Ian became interested in the effect of a crystallite size distribution in line profile analysis. A fruitful collaboration was undertaken with Professor P. Scardi, Trento University, who was working on a new pattern modeling approach, based on models of microstructural effects. The effect of crystallite size distribution was pointed out in the study of a nanocrystalline cerium oxide, with negligible microstrains, prepared at Rennes. The collaboration Birmingham, Rennes and Trento resulted in a noteworthy article published in 2000. In the course of his research, Ian has been involved in studies using synchrotron X-rays and he contributed to the characterization and some developments of the HRPD at Daresbury (UK), in collaboration with R.J. Cernik and coworkers.

In 2001, Ian received the BCA Industrial Group Award for his work on line profile analysis. He has been a member of the Joint Committee on Powder Diffraction Standards (JCPDS) and its successor, the International Centre for Diffraction Data (ICDD), since 1981. In 1986, Ian was invited to serve in the Commission on Powder Diffraction (CPD), formed at the 14th General Assembly of the IUCr in Perth, Australia, and was elected Secretary until 1990, then ordinary member from 1990 to 1993. Ian was very active in meetings and schools organized or sponsored by the CPD. He was invited to contribute to sections on powder diffraction in Vol. C of the International Tables for Crystallography (2006).

Meeting of the IUCr Commission on Powder Diffraction at Chester (UK)

Beyond science…

Ian's interests and passions extended well beyond powder diffraction and science. For one, Ian was a good expert and author of Guides on Towpath:



Towpath Guide No. 1: *Staffordshire & Worcestershire Canal*, pp 276,
Cambridge: Goose & Sons, 1974
Towpath Guide No. 3: *Stourbridge Canal*, pp 59,
Birmingham: Lapal Publications, 1992
He was editor and joint author of *The Dudley and Stourbridge Canals*, pp 46, Birmingham: Lapal Publications, 1980.

Ian was also a great fan, and
very fond of his *Morris Minor Traveller*

In the recent years, he had an interest for water mills in Shropshire. His study was recently published: *Granite millstones of Shropshire and adjoining counties* (Wind and Water Mills, 30, 2-31, 2011).

… and whenever possible: hiking !
Here is Ian on 2005, walking along the north coast of Brittany

Milestones in modern powder diffraction: a selection of Ian's main contributions

Ian Langford in his lab with the Picker diffractometer at Birmingham in about 1975
(picture taken by the Physics Dept. Photographer, David James)

It is not an easy task to select the most relevant papers in a collection of more than 150 contributions to powder diffraction which mark so many significant advancements of the discipline in the years between 1960 and 2000. The following selection is not exhaustive and necessarily subjective: most of Ian's production is still easily available on the Web, where search engines report papers and citations, still numerous today, even after 14 years since Ian's retirement.

Early times: development of the Automatic Powder Diffractometer

- Langford, J.I., "Counter diffractometer: the effect of axial divergence on the breadth of powder lines", *J. Sci. Instrum.* 39 (1962) 515-516.
- Langford, J.I. & Wilson, A.J.C. "Counter diffractometer: the effect of specimen transparency on the intensity, position and breadth of X-ray powder diffraction lines", *J. Sci. Instrum*. **39** (1962) 581-585.

Breakthroughs in powder diffraction: the variance method

- Langford, J.I., "Variance as a measure of line broadening: particle-size determination", *Nature* 207 (1965) 966-967.
- Langford, J.I., "The variance and other measures of line broadening in powder diffractometry I. Practical considerations", *J. Appl. Crystallogr.* 1 (1968) 48-59; "II. Determination of particle size", *ibidem*, 131-138.

Breakthroughs in powder diffraction: introducing the Voigt function in line profile fitting

- Langford, J.I., "A rapid method for analysing the breadths of diffraction and spectral lines using the Voigt function", *J. Appl. Crystallogr.* 11 (1978) 10-14.

Basic reviews on Line Profile Analysis: the Scherrer formula

- Langford, J.I. & Wilson, A.J.C., "Scherrer after Sixty Years: a survey and some new results in the determination of crystallite size", *J. Appl. Crystallogr.* 11 (1978) 102-113.

Line profile analysis for the study of crystallite shape and size

- Langford, J.I. & Louër D., "Diffraction line profiles and Scherrer constants for materials with cylindrical crystallites", *J. Appl. Crystallogr.* 15 (1982) 20-26.
- Louër, D., Auffrédic, J.P., Langford, J.I., Ciosmak, D. & Niepce, J.C., "A precise determination of the shape, size and distribution of size of crystallites in zinc oxide by X-ray line broadening analysis", *J. Appl. Crystallogr.* 16 (1983) 183-191.

Toward total pattern fitting and full understanding of Line Profile Analysis

- Langford, J.I., Louër, D., Sonneveld, E.J. & Visser, J.W., "Applications of Total Pattern Fitting to a study of crystallite size and strain in powder zinc oxide", *Powder Diffraction* 1 (1986) 211-221.
- Langford, J.I., Delhez, R., de Keijser, Th.H. & Mittemeijer, E. J., "Profile analysis for microcrystalline properties by the Fourier and other methods", *Austral. J. Phys.* 41 (1988) 173-187.
- Delhez, R., de Keijser, Th.H., Mittemeijer, E. J. & Langford, J.I., "Size and strain parameters from peak profiles: sense and nonsense", *Austral. J. Phys.* 41 (1988) 213-227.
- Langford, J.I., "The use of the Voigt function in determining microstructural properties from diffraction data by means of pattern decomposition", *Accuracy in Powder Diffraction II*, ed. E. Prince and J.K. Stalick, NIST Spec. Pub. No. 846 (Gaithersburg MA: US dept of Commerce) pp. 110-126, 1992.

Line Profile Analysis with the advent of synchrotron radiation

- Langford, J.I., Cernik, R. J. & Louër, D., "The breadth and shape of instrumental line profiles in high resolution powder diffraction", *J. Appl. Crystallogr.* 24 (1991) 913-919.

Major reviews on powder diffraction

- Langford, J.I., "Some applications of pattern fitting to powder diffraction data"; in Recent Advances in X-ray Characterization of Materials, pp 185-211, ed. P. Krishna. *Progress in Crystal Growth and Characterization 14*. Oxford: Pergamon Press, 1987.
- Delhez, R., de Keijser, Th. H., Langford, J.I., Louër, D., Mittemeijer, E.J. & Sonneveld, E.J., "Crystal imperfection broadening and peak shape in the Rietveld Method"; in The Rietveld Method, pp 132-166, ed. R.A. Young. Oxford: IUCr/OUP, 1993, 2nd ed. 1995.
- Langford, J.I. & Louër, D., "Powder Diffraction", *Rep. Prog. Phys.* 59 (1996) 131-234.

Memories of Ian's mentor

- Langford, J.I., "A.J.C. Wilson, FRS, 1914-1995", *Acta Crystallogr.* A52 (1996) 7-10; Abrahams, S.C. & Langford, J.I., "A.J.C. Wilson", *Physics Today* 122-124, March 1996.

Powder diffraction: industrial applications and reference literature

- Langford, J.I., "Line profiles and sample microstructure"; in Industrial Applications of X-ray Diffraction, ed. F.H. Chung & D.K. Smith, chapter 33 (751-775). New York: Dekker, 1999.
- Parrish, W. & Langford, J.I., "Powder and related techniques: X-ray techniques", in International Tables for Crystallography, Vol. C, ed. A.J.C. Wilson & E. Prince, Section 2.3 (42-79). Dordrecht: Kluwer, 1999; "X-ray diffraction methods: polycrystalline", ibidem, Section 5.2 (487-499).
- Langford, J.I., "Use of pattern decomposition or simulation to study microstructure: theoretical considerations", in Defect and Microstructure Analysis by Diffraction, ed. R.L. Snyder, J. Fiala & H.J. Bunge, chapter 5 (59-81). Oxford: IUCr/OUP, 1999.

Laying the foundations of modern Powder Diffraction

- Langford, J.I., Louër, D. and Scardi, P., "Effect of a crystallite size distribution on X-ray diffraction line profiles and whole powder pattern fitting", *J. Appl. Crystallogr.* 33 (2000) 964-974.