

STRUCTURE DETERMINATION OF TWO ORGANIC COMPOUNDS FROM LABORATORY X-RAY DATA, EMPLOYING THE XCELERATOR DETECTOR

C. A. Reiss¹ K. Goubitz²

¹Philips Analytical Lelyweg 1 ALMELO 7602 EA NETHERLANDS

²Laboratory for Crystallography, University of Amsterdam

The crystal structures of two organic compounds have been determined using laboratory X-ray powder diffraction, employing the revolutionary XCelerator detector. The XCelerator uses the RMTS (Real Time Multiple Strip) detection technology, which allows for a huge increase in recording speed, while maintaining the resolution of the traditional powder diffractometer. Both compounds were measured on a XPert Pro Alpha-1 diffractometer. The powders were prepared as flat samples as well as capillary samples and recorded using a focusing incident beam monochromator in the case of the flat samples and a hybrid monochromator in case of the capillary samples.

Both structures were solved using a grid-search technique [1]. Grid search techniques are based on the handling of molecular models in direct space. This procedure does not require all extracted intensities from a powder pattern. A small number of reflections from the low-angle region, where overlap is not so severe, are in general enough to locate the model. Moreover, the program also can handle groups of overlapping reflections. The program MRIA [2] was used for the structure determination and refinement.

References

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STRUCTURE DETERMINATION OF ANHYDROUS CAFFEINE

C₈H₁₀N₄O₂ FROM X-RAY POWDER DIFFRACTION DATA

F. Stowasser¹ C. Lehmann²

¹Bruker AXS GmbH Oestliche Rheinbrueckenstrasse 49 KARLSRUHE 76187 GERMANY ²Max-Planck-Institut fuer Kohleforschung, D-45470 Muehlheim, Germany

For a long time the anhydrous form of caffeine monohydrate C₈H₁₀N₄O₂ * H₂O was known but so far no solid state structure has been proposed. The compound is prepared by drying a powder sample of caffeine monohydrate at 60°C in air. So far it has not been possible to grow suitable single crystals for structure determination. However, the obtained powder sample proved to be of high quality and a structure determination from powder data has been carried out successfully.

The powder pattern could be indexed with monoclinic symmetry by using ITO. Several cells and space groups were found, but finally two cells and space groups were found to be useful for further work. The cell volume of 4450 Å³ is approx. 4.5 times larger than the cell of the monohydrate compound with one very long axis. The structure determination and Rietveld refinement were done using Topas. The calculation was carried out using the simulated annealing technique together with five rigid bodies for the description of the caffeine molecules. All calculations were done in real space directly against step intensities without preceding intensity extraction.

The final result consists of 5 individual molecules in the asymmetric unit giving a total amount of 20 molecules in the unit cell. The number of molecules per cell was supported by additional pycnometric density measurements. To our knowledge this structure determination represents one of the biggest ever solved structures using powder diffraction data and documents the effectiveness of real space structure determination methods.

Keywords: SIMULATED ANNEALING REAL SPACE STRUCTURE DETERMINATION

PATSEE - A VALUABLE TOOL FOR STRUCTURE DETERMINATION FROM POWDER DIFFRACTION DATA

S. Ruehl E. Egert

Goethe-Universität Institut fuer Organische Chemie Marie-Curie-Str. 11 FRANKFURT 60439 GERMANY

PATSEE is a molecular replacement program, which attempts to combine both Patterson and direct methods in order to locate a known fragment in the unit cell. The list of successful applications of PATSEE is very long and comprises organic and biological structures up to 500 non-hydrogen atoms as well as, sporadically, structures determined from powder diffraction data. The purpose of this work is the development of a suitable strategy for structure solution from powder data with PATSEE. We decided to approach this goal in three consecutive steps: First we wanted to know the accuracy and the minimum size of a fragment required as well as an estimation of how well the several figures of merit calculated in PATSEE distinguish between correct and false solutions. In order to reduce experimental and systematic errors, powder data were generated from single crystal intensities for a collection of known structures. Fragments were either taken from the Cambridge Structural Database or calculated with the molecular modelling program MOMO. The investigations indicate that a small accurate fragment distinguishes better from false solutions than a larger, less accurate fragment. In some cases it was possible to locate a fragment comprising less than 10 percent of the total scattering power. For the next step powder diffraction data were actually measured, but we restricted ourselves to known structures with up to 44 atoms. This provided a measure of how large a fragment - 50 to 60 percent of the total scattering power - is needed for successful structure solution (location and extension). At present, we are working on the last step: structure solution of unknown structures.

Keywords: STRUCTURE SOLUTION POWDER DIFFRACTION PATSEE

DIFFERENTIAL EVOLUTION AS A METHOD OF STRUCTURE SOLUTION FROM POWDER DIFFRACTION DATA

M. Tremayne C. C. Seaton

School of Chemical Sciences University of Birmingham Edgbaston BIRMINGHAM B15 2TT UK

The crystal structure determination of molecular materials from powder diffraction data is a rapidly expanding field, driven in recent years by the development of direct space methods of structure solution. These approach structure solution by generation of trial crystal structures, often based on the known connectivity of the material, and assessment of the fitness of each structure by comparison with the experimental data. Global optimization methods are then used to locate the global minimum corresponding to the structure solution.

In this paper we will compare results obtained using two global optimization techniques: a) a heuristic technique based on Monte Carlo sampling and b) a new evolutionary algorithm based on differential evolution (DE). DE is an evolutionary process that is both relatively simple and easy to implement, while offering robust searching of minima. In addition, the association of upper and lower bounds on elements used in the DE calculation allows the incorporation of molecular geometrical limits that enhance the efficiency of the search rather than disrupting the natural optimization pathways. The application of these methods will be illustrated by the structure solution of a range of systems including new polymorphic materials, structures in which the primary interest centres on intermolecular aggregation, organic cocrystals, macrocyclic complexes and inorganic-organic hybrid materials.

References

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