Hessisches Kompetenzzentrum für Hochleistungsrechnen

Development and Implementation of New Methods for Crystal Structure Determination from Powder Diffraction Data



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Introduction

A new method for the determination of organic crystal structures from powder diffraction data has been developed. The method is based on a new algorithm using cross-correlation functions.

The preferable method for the determination of crystal structures of organic compounds uses diffraction data of a single crystal. In many cases, however, crystallization of a single crystal of sufficient size fails or the characterization of a powder is required, and only powder diffraction data is available. Important examples are industrial organic pigments or pharmaceutical formulations. Crystal structure determinations from powder data generally require indexing of the powder diffraction data as a first step, which relates peak positions in the powder diagram to crystal symmetry and lattice parameters. In practice, however, a considerable percentage of the powder diagrams resists reliable indexing. The main reasons are insufficient crystallinity resulting in broad peaks, or mixtures containing other phases disturbing the indexing procedure. Without prior indexing, most methods for structure solution cannot be applied and the structures remain undetermined.

Results

A new method has been developed, which is also suitable for unindexed powder data, powder diagrams of very low quality

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and powder diagrams of non-phase-pure samples. The program FIDEL (FIt with DEviating Lattice parameters) [1] implements this method, which is based on a similarity measure using the cross- and auto-correlation functions of a simulated and an experimental powder pattern [2]. The molecular geometry and relevant degrees of freedom are modelled based on quantum-mechanical [3] and force-field calculations [4] as well as chemical and crystallographic experience. The lattice parameters, molecular position, molecular orientation and selected intramolecular degrees of freedom are optimized until the similarity measure reaches a maximum. The final stage of a crystal structure characterization based on powder data is a Rietveld refinement. FIDEL provides crystal structure candidates suited for validation and refinement using the Rietveld method. The program can perform an automatic follow-up Rietveld refinement process using TOPAS [5].

The developed method has shown to be very powerful. The method was used to determine various crystal structures of organic compounds from laboratory X-ray powder data, including the crystal structure of pharmaceutical compounds and organic pigments. This has been shown for structures initially derived from isostructural compounds (e.g. a solvate, hydrate or chemical derivative), or from crystal data measured at a different temperature or pressure. Accuracy, robustness and performance of the approach implemented in FIDEL also allows structure determination by screening of a large number of structure candidates, e.g. from crystal structure predictions by standard force-field methods. FIDEL is capable of indentifying favourable structure candidates with a correct crystal symmetry and molecular packing and can fit them to the experimental data sufficiently accurate for Rietveld refinement.

Outlook

Recent development focussed on making the systematic structure determination from diffraction data of very poor quality feasible, even if only the chemical composition and some probability of certain space group symmetries is known.

Reference

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