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Keywords: Electron diffraction, modulated structure, daliranite

MS25-P05

Application of laboratory X-ray diffraction equipment for Pair Distribution Function (PDF) analysis

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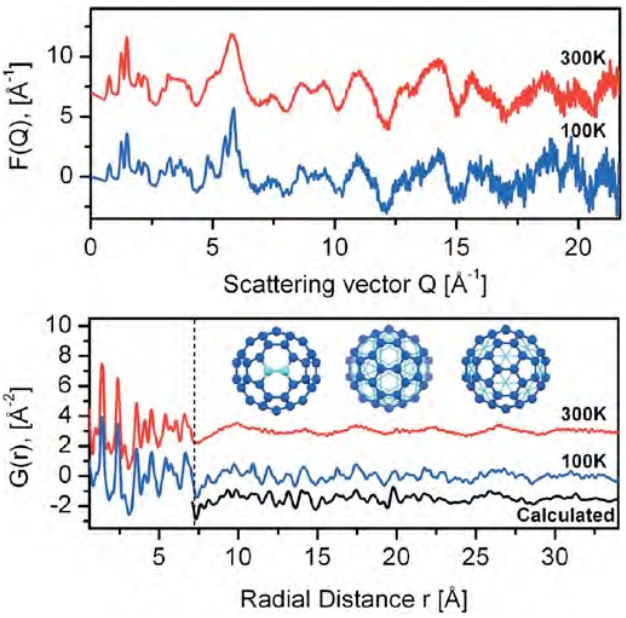
The increased interest in recent years regarding the properties and applications of nanomaterials has also created the need to characterize the structures of these materials. One of the most promising techniques to study nanostructures using X-ray diffraction is by using the total scattering (Bragg peaks and diffuse scattering) from the samples and the pair distribution function (PDF) analysis. The pair distribution function provides the probability of finding atoms separated by a certain distance. From experimental point of view a typical PDF analysis requires the use of intense high-energy X-ray radiation ($E \geq 15$ KeV) and a wide 2θ range. At present, synchrotron and neutron sources are the preferred choice for PDF analysis, but there is clearly an increasing need for a PDF solution based on laboratory diffraction equipment. Such a solution, though limited, will benefit areas where quick feedback about the materials properties is important and will allow the routine application of PDF analysis for materials characterization in university laboratories as well as industrial R&D departments.

After the initial feasibility studies regarding the use of standard laboratory diffraction equipment for PDF analysis [1,2] this application has been further developed to achieve improved data quality and to extend the range of materials, environmental conditions and geometrical configurations that can be used for PDF experiments. The recent introduction of detectors with improved efficiency for high-energy X-rays [3] has further enhanced the capabilities of laboratory diffractometers for total scattering experiments. This contribution presents several examples of laboratory PDF studies performed on different nanocrystalline and amorphous materials of scientific and technological interest (organic substances, oxides, metallic alloys, materials for battery applications, etc.) and demonstrates that PDF analysis with a laboratory diffractometer can be a valuable tool for structural characterization of nanomaterials.



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Keywords: Pair Distribution Function, Total scattering, Nanomaterials

MS25-P06

Ab initio structure determination from unindexed powder patterns by a global optimization approach using pattern comparison based on cross-correlation functions

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A method for *ab initio* structure determinations from powder diffraction data (SDPD) of organic compounds, which does not require prior indexing of the experimental pattern, has been developed. The global optimization approach is based on the direct fit of random trial structures to the powder data using an algorithm called FIDEL ("FIt with DEviating Lattice parameters"). It uses a similarity measure based on cross-correlation functions, which allows the comparison of simulated and experimental powder data even if the lattice parameters do not match [1,2].

The structure determination process starts with the molecular geometry of the compound and a global search space setup in various crystal symmetries. The lattice parameters, molecular position and orientation, and selected intramolecular degrees of freedom are fitted simultaneously in an elaborated multi-step procedure. The hierarchical search strategy developed is based on similarity screening, structure fitting and iterative adaptation of parameter ranges. It combines various methodologies in order to realize an efficient and effective exploration of the global search space. The standard procedure includes an adaptive mechanism for the balancing of computing time spent on the generation of random structures, similarity screening, fitting and clustering of structural models. The structure determination proceeds stepwise from e.g. more than 20 million trial structures to a small number of promising structure candidates that are subjected to an automated Rietveld refinement with TOPAS [3]. Finally, a user-controlled Rietveld refinement with TOPAS is performed.

By exploiting the full potential and versatility of the pattern comparison approach based on cross-correlation functions and supported by the integration of 3rd party components, a complete SDPD framework evolved, that covers almost all scenarios of finding crystal structures corresponding to given experimental data.

The global optimization approach presented is suitable for a wide range of "problematic" powder patterns, including powders of low crystallinity, non-phase-pure samples or disordered structures. While pushing SDPD to its limits, applications of the method deliver valuable insights into ambiguities arising from low-quality experimental data, where the "one powder – one structure" paradigm of SDPD is beginning to collapse. Even if the results do not converge into a single solution qualifying as a publication grade structure determination, the method can provide a set of structural models that match the powder as much as possible, for further analysis.

Examples of (metal-)organic compounds are shown, including the previously unknown structures of nanocrystalline