

Oral Contributions

[MS29 - 03] Guidelines for solving structures of polycrystalline materials using charge flipping

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In order to obtain structural information from X-ray powder diffraction data (XPD), both the phase and the overlap problem need to be solved. Structure determination methods developed for single-crystal diffraction data can be used, but their success depends on how (or if) the overlap problem is treated, and on the quality of the data. Direct-space methods, on the other hand, overcome both problems by using chemical knowledge. Although these methods also work on low resolution data, their success depends on the accuracy of the initial model. In this work, some of these points will be addressed: (a) importance of high quality data, (b) application of *ab initio* structure determination methods, and (c) bringing the benefits of *ab initio* and direct-space methods together to solve increasingly complex structures. (a) The XPD data used here were measured at the Swiss Light Source, using a Mythen detector [1]. The accuracy of such data is generally suitable for *ab initio* structure determination. (b) The charge flipping algorithm [2] implemented in the program *Superflip* [3] was used for structure determination for two reasons. (1) Charge flipping does not require symmetry information (often ambiguous in XPD data), and (2) the program has been adapted to deal with overlapping reflections [4]. First, the input parameters for the powder charge flipping algorithm were optimized using a known structure, and then they were applied to a set of unknown structures. Several factors that were expected to affect the success rate were evaluated: the degree of overlap in the XPD pattern, the types of atoms, the types of symmetry, the number of atoms in the unit cell, and the type of structural motif. It

was found that light atom structures, crystallizing in non-centrosymmetric space groups were more difficult (or impossible) to solve. Additionally, a large number of atoms in the unit cell or irregular structural motifs appeared to cause further problems. Surprisingly, the degree of overlap was only found to be problematic in extreme cases. In order to access these complex structures, a modified approach was developed [5]. (c) The modification exploited the fact that approximate (though incorrect) structures obtained from direct-space optimization could be used to generate non-random starting phase sets for charge flipping. Such phase sets, coupled with re-optimized input parameters were used to solve a series of organic structures of different complexities. The success of the approach was found to be limited only by the number of atoms in the unit cell. However, solution evaluation appeared to be problematic. The solution evaluation tools implemented in *Superflip* (convergence R value, symmetry R value) were not found to be useful for the modified approach. Therefore, two cluster analyses, log-likelihood functions and evaluation based on entropy calculations were investigated. Of these, entropy was found to be most promising. The end result of these tests can be summarized in a flowchart that indicates which approach is most suitable for a specific problem, depending upon the structural motif, the types of atoms, the symmetry, and the number of atoms. Optimal tools for solution evaluation are also suggested.

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