Powder-Diffraction-Based Structural Comparison for Crystal Structure Prediction Under Pressure

A. Otero-de-la-Roza[1]

[1]*Departament of Analytical and Physical Chemistry and MALTA Consolider Team, School of Chemistry, University of Oviedo, 33006 Oviedo, Spain*

oteroalberto@uniovi.es

Crystal structure prediction (CSP) is a computational method for the prediction of the thermodynamical stable phase (or the energy landscape) from the stoichiometry or the molecular diagram of a compound. CSP is a hard problem because it involves a global search over the space of all possible structures, and it is computationally very demanding. It would be beneficial to be able to use experimental information, such as powder X-ray diffraction (XRPD) patterns, to inform and speed up the CSP search for the stable structure. This is particularly important in the field of high-pressure experimental X-ray diffraction experiments, where the powder data available is generally not of enough quality to allow the unequivocal determination of high-pressure crystal structures. To assist CSP, it is necessary to compare calculated structures with experimental patterns but, for achieving this, two problems need to be solved: i) lack of vibrational effects (and errors from the DFT method) may cause deformations in the unit cell relative to experiment, shifting peak positions, and ii) experimental patterns have peak shapes and widths that are directly not calculable from the structure. In this work, we present a crystal similarity index (VCGPWDF) for comparing structures with experimental patterns that allows for lattice deformations of the former [1]. VCGPWDF is simple, efficient, fully automatic, and does not require indexing of the pattern. It gives meaningful results even if the experimental pattern is of very low quality. The development of VCGPWDF opens the door for XRPD-assisted CSP searches aimed at the solution of structures under high pressure.

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