ENHANCED AB INITIO INDEXING OF TOF NEUTRON POWDER DIFFRACTION DATA USING MAXIMUM ENTROPY

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The first key step in the characterization of a new crystalline material by powder diffraction is the determination of its unit cell and possibly of the associated space symmetry (i.e., its 3D space group) using the so-called indexing procedure. This procedure is usually carried out with diffraction data obtained from synchrotron or laboratory X-rays rather than with electrons or neutrons. The recent development of high resolution powder diffractometers at increasingly more powerful Pulsed Neutron Sources such as POWGEN (ORNL, USA) and DREAM (ESS, SE) has prompted us to study the feasibility of the indexing process directly from the measured Time-of-Flight (TOF) diffractograms and without recourse to any complementary X-ray diffraction information.

As a first step, a wide range of plausible simulated noise-free TOF diffractograms were obtained using the working characteristics of TOF diffractometers around the world (HIPPO / LANSCE, POWGEN / SNS, POLARIS / ISIS) for compounds whose symmetry ranged from Cubic down to Triclinic. Our indexing attempts were always successful provided that the used a TOF range that allowed for the presence of the first observable Bragg peak (i.e., corresponding to the longest d-spacing). A non-trivial side-result of this first analysis is the mandatory need for a very careful computation of the Resolution Function [RF] / Point Spread Function [PSF] whenever the dynamic TOF range becomes large, such as in the case for the POWGEN instrument.

In real life measurements, a typical TOF diffractogram will feature broad and asymmetric Bragg peaks (associated with the back-to-back exponentials involved in the PSF), a non-negligible background, noisy data and a few detector banks (typically 4-6) covering distinct TOF / d-ranges. The new challenges associated with such TOF data involve: [1] merging information from distinct detector banks, [2] minimizing as much as possible the asymmetry effect due to the back-to-back exponentials and [3] coping with the noise in the data. As a second step, we show that the use of Maximum Entropy Regularization using the ubiquitous UK MEMSYS software solves all of these problems. The efficiency of our suggested procedure will be demonstrated using the 4-detector bank real TOF nickel data provided with the widely used GSAS software (Larson & Von Dreede).