A Structural Study of Fullerene Compounds by the Maximum Entropy Method

M. Takata, T. Masuda, E. Nishibori[A], N. Machida[A] and M. Sakata[A]*
Department of Material Science, Shimane University, Matsue, 690-8504, Japan
[A] Department of Applied Physics, Nagoya University, Nagoya, 464-8603, Japan

Fullerene compounds such as endohedral metallo-fullerenes or alkali-dope fullerenes shows various interesting and important physical properties like superconductivity. It would be therefore extremely interesting to know the electronic level structure of these compounds. The Maximum Entropy Method (MEM) is most appropriate method for the present purpose because it can reveal detailed electron density distributions of the crystalline materials directly from the experimental data without any structural model. However, in order to study the electronic level structure by MEM, very accurate integrated Bragg intensity data are required. It is very easy to measure not so accurate integrated Bragg intensities by a modern X-ray diffractometer but many cares has to be taken in to account to collect an reliable and accurate integrated Bragg intensity data, particularly at SPring-8 which is not stationary X-ray source and may be too strong for Bragg intensity measurement in a certain case. It is important to establish the method to collect very accurate integrated Bragg intensity data at SPring-8 for the present purpose. In this study, an experimental results by using a NIST standard material, that is CeO_2, are given.

The experiment were carried out at BL-02B1 by using IP as a detector. The CeO_2 specimen are sealed into 0.1mm silica glass capillary. The wavelength of incident X-ray was 0.5 A with 0.5mm beam size. The exposure time was 2.5 hrs. In this experimental arrangement, any in-stability of incident X-rays shall not give serious effects onto the measured Bragg intensities. This is a big advantage over the conventional point by point step scan. The maximum 2θ was about 60^0 which is not big. But because of very short wavelength of incident X-rays, the resolution in real space expressed by the minimum d-spacing is 0.5 A. Considering the resolution limit of CuKα, which is 0.77A, the resolution in real space of the present experimental arrangement is extremely good.

The drawback of using very short wavelength for the measurement of whole powder pattern is the fact that peak separations in diffraction space become much smaller. In other words, the differences of peak positions with respect to 2θ angle become small. It turned out that parallel beam of SPring-8 SR light overcomes such an apparent disadvantage, namely FWHM of CeO_2 Bragg peak is typically about 0.050 and shows virtually no diffraction angle dependence up to 60^0. There arises another problem due to very sharp peak. The distance between the specimen and IP was 260 mm and pixel size of IP was 100μm. This means 1 pixel correspond to 0.022^0, which is too coarse compared with FWHM. The R_I-factor of the Reitveld refinement for this data was 2.74%, which may not be good enough for MEM analysis.

In order to overcome coarse pixel problem, the second experiment was done, changing IP position by approximately 0.01^0. The results of the Reitveld refinement shows great improvement in the value of R_I -factor from 2.74 to 0.61% when the data of both experiments are used for the refinement as a combined data. In this work, it has been proved that the present experimental method is very powerful to collect an accurate integrated Bragg intensities of not only fullerene compounds but also many other materials.