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A structure analysis on simulated powder diffraction pattern from a proposed moderator in J-SNS

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Abstract

A design of high-resolution powder diffractometer proposed for the Japan neutron scattering facility (tentatively called as J-SNS) is now in progress. At present, powder diffraction patterns are simulated using the pulse shape of the moderators of the candidate, and the Rietveld analysis is carried out for the simulation patterns. The desirable pulse characteristics are discussed.

1. Introduction

The choice of the moderator is one of the most important issues for every instrument. The desirable characteristics for the pulse shape are 'a high integrated intensity' and 'a symmetrical and a Gaussian-like peak profile'. Of course, these are contrary to each other. In the case of recent high-resolution time-of-flight (TOF) powder diffractometer with supermirror neutron guide, both high resolution and high intensity has been realized. However, the peak asymmetry and the tail cannot be overcome by any technique at present, and the situation would not be drastically changed in near future. To estimate the bad influence of the peak asymmetry and the tail, we are trying to simulate the diffraction pattern for each moderator as accurate as possible, and analyze the simulated diffraction pattern by the Rietveld method.

2. Analytical function for expression of the pulse shape of the moderators

To simulate the diffraction pattern, we tried to fit the given simulated pulse data of the moderators of the candidate. The data sets are listed below.

	Reflector type	Decoupling energy
(1) Coupled H_2 moderator with extended premoderator	Pb	–
(2) Decoupled H_2 moderator with extended premoderator	Pb	1eV
(3) Decoupled H_2 moderator	Be	0.3eV

(4) Decoupled H_2 moderator with extended premoderator (Poisoned moderator)	Pb	1eV

The 34 pulse data between 1 meV and 400 meV for each moderator were given. Examples of pulse data for the moderator of (2)-unpoisoned type and (4)-poisoned type are shown in Fig. 1(a) and 1(b).

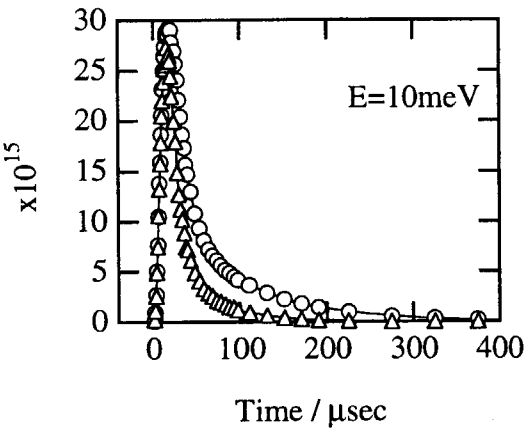

Fig. 1(a) Pulse data of E=10meV for the moderator (2) ; circle and (4) ; triangle.

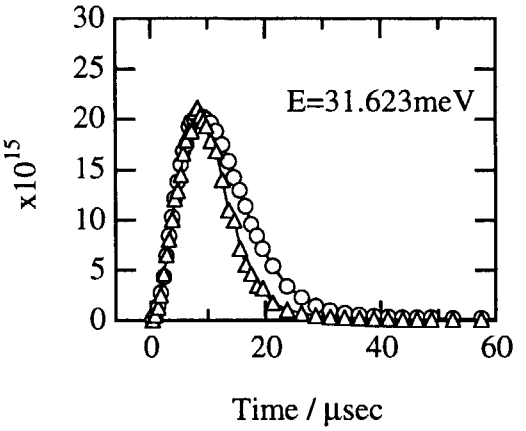

Fig. 1(b) Pulse data of E=31.623meV for the moderator (2) ; circle and (4) ; triangle.

To fit the pulse data for whole energy region, some profile shape functions are tried, such as Ikeda-Carpenter function [1], Cole-Windsor function [2], etc. After some trial and error, we finally chose Ikeda-Carpenter based function.

The definition of an original Ikeda-Carpenter function is as follows.

$$\phi(t) = \frac{\alpha}{2}(\alpha t)^2 \exp(-\alpha t) \quad (t > 0)$$

$$\Phi(t) = \int dt' \phi(t)\left[(1-R)\delta(t-t') + R\beta\theta(t-t')\exp\{-\beta(t-t')\}\right]$$

$$= \frac{\alpha}{2}\left[(1-R)(\alpha t)^2 \exp(-\alpha t) + R\frac{2\alpha^2\beta}{(\alpha-\beta)^3}\left\{\exp(-\beta t) - \exp(-\alpha t)\left[1 + (\alpha-\beta)t + \frac{1}{2}(\alpha-\beta)^2 t^2\right]\right\}\right]$$

This function (hereafter, written as $\Phi_2(t)$) fits well at low-energy region, but some discrepancies are found at high-energy region. To get better fit at high-energy region, we changed the equation of $\phi(t)$ expressed above to the next expression, and led the $\Phi(t)$ as follows.

$$\phi(t) = \frac{\alpha}{2}(\alpha t)^3 \exp(-\alpha t) \quad (t > 0)$$

$$\Phi(t) = \int dt' \phi(t)\left[(1-R)\delta(t-t') + R\beta\theta(t-t')\exp\{-\beta(t-t')\}\right]$$

$$= \frac{\alpha}{6}\left[(1-R)(\alpha t)^3 \exp(-\alpha t) + R\frac{6\alpha^3\beta}{(\alpha-\beta)^4}\left\{\exp(-\beta t) - \exp(-\alpha t)\left[1 + (\alpha-\beta)t + \frac{1}{2}(\alpha-\beta)^2 t^2 + \frac{1}{6}(\alpha-\beta)^3 t^3\right]\right\}\right]$$

This function (hereafter, written as $\Phi_3(t)$) fits well at high-energy region, but some discrepancies are found at low-energy region. Then, we combined these functions by

introducing an additional parameter, R'.

$$\Phi(t) = (1-R')\Phi_3(t) + R'\Phi_2(t)$$

After the careful fitting of the pulse data for the whole energy region, it was found that the energy dependence of the R' parameter is similar to the R parameter in Φ_3(t) and Φ_2(t), and can be substituted R for R'. Then, the final expression for a pulse shape is as follows.

$$\Phi(t) = (1-R)\Phi_3(t) + R\Phi_2(t)$$

$$= (1-R)\frac{\alpha}{6}\Big[(1-R)(\alpha t)^3\exp(-\alpha t) + R\frac{6\alpha^3\beta}{(\alpha-\beta)^4}\Big\{\exp(-\beta t) - \exp(-\alpha t)\Big[1+(\alpha-\beta)t + \frac{1}{2}(\alpha-\beta)^2t^2 + \frac{1}{6}(\alpha-\beta)^3t^3\Big]\Big\}\Big]$$

$$+ R\frac{\alpha}{2}\Big[(1-R)(\alpha t)^2\exp(-\alpha t) + R\frac{2\alpha^2\beta}{(\alpha-\beta)^3}\Big\{\exp(-\beta t) - \exp(-\alpha t)\Big[1+(\alpha-\beta)t + \frac{1}{2}(\alpha-\beta)^2t^2\Big]\Big\}\Big]$$

Figure 2(a) and 2(b) shows the examples of the single peak fitting using this function.

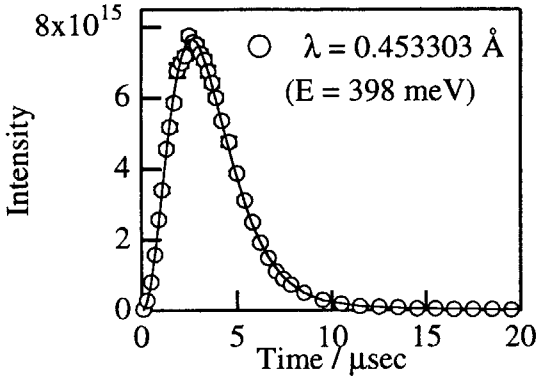

Fig. 2(a) A single peak fitting of E = 398 meV for the moderator (2).

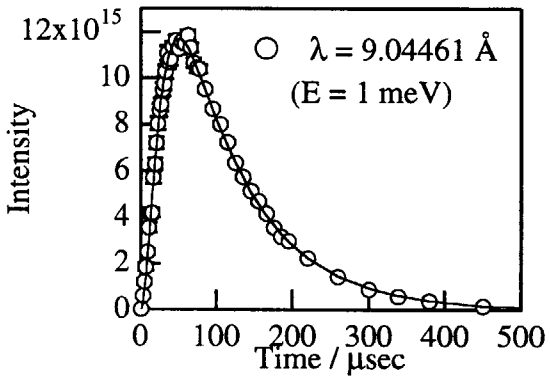

Fig. 2(b) A single peak fitting of E = 1 meV for the moderator (2).

Definitions of wavelength dependence of the parameters for original Ikeda-Carpenter function were changed to get better fitting on these parameters. Here, another forms of wavelength dependence were tentatively made.

$$\alpha(\lambda) = \alpha_1 + \alpha_2\exp(-\alpha_3\lambda)$$

$$\beta(\lambda) = \beta_1 + \beta_2\exp(-\beta_3\lambda^3)$$

$$R(\lambda) = \exp\left(-\frac{E}{E_0}\right) + R_1 + R_2\exp\left[-\frac{1}{2}\left(\frac{\lambda-R_3}{R_4}\right)^2\right]$$

Figure 3(a) and 3(b) shows the fitting result of the wavelength dependencies for the parameters α, β and R of the moderator (2).

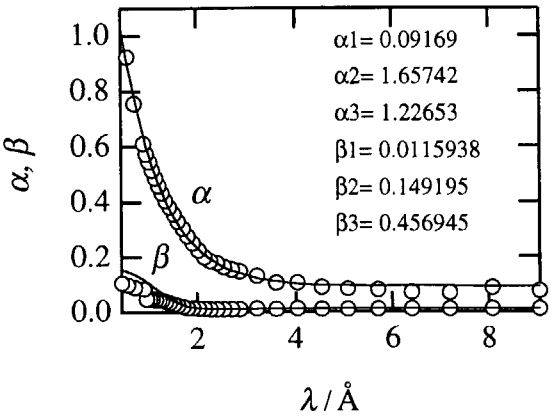

Fig. 3(a) Wavelength dependence of the parameter α and β for the moderator (2).

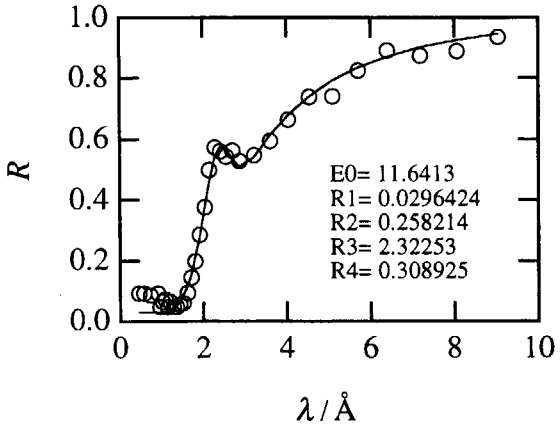

Fig. 3(b) Wavelength dependence of the parameter R for the moderator (2).

3. Peak asymmetry and tail of the pulse shape

The high-resolution powder diffractometer is one of the most severe instruments which performance is directly affected by the pulse shape. In profile analysis (including Rietveld refinement), a symmetrical profile shape is highly desired. A Gaussian-like tail is also hoped, instead of a Lorentzian-like or an exponential-like tail.

To calculate the full-width at half-maximum (FWHM) values and peak asymmetry factors, the pulse data were fitted again by a Cole-Windsor based function [3]. Figure 4(a) shows the FWHM values for moderator of (2)-unpoisoned type and (4)-poisoned type. The resolution ($\Delta t/t$) of poisoned moderator can be realized with unpoisoned moderator by putting instruments far away (for example, L_1=100 m → 150 m).

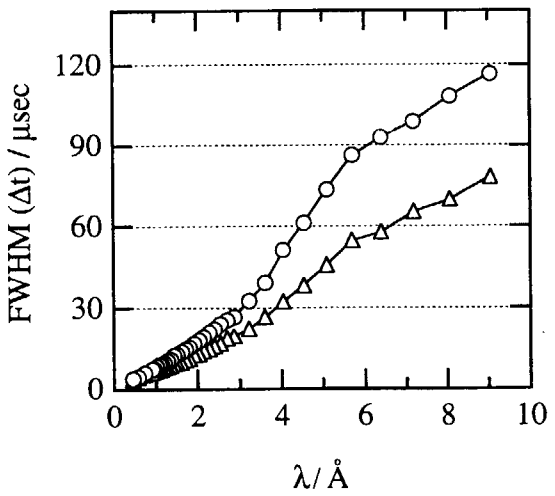

Fig. 4(a) The FHWM values for the moderator (2); circle and (4); triangle.

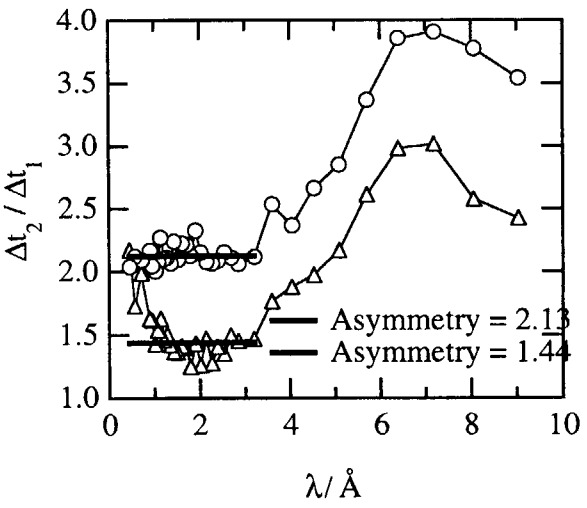

Fig. 4(b) The asymmetry values for the moderator (2); circle and (4); triangle.

Now, the peak asymmetry is defined by a ratio, $\Delta t_2/\Delta t_1$ (Δt_1 and Δt_2 are FWHM values for leading and tailing part, respectively), and those of two moderators are plotted in fig. 4(b). This figure show that peak asymmetry in slowing down region of the poisoned moderator is far better than that of the unpoisoned moderator.

4. The strategy for judgment for the proposed Moderators with designed diffractometer using simulation program, McStas.

To estimate the bad influence on structure analysis due to highly asymmetric long tail, Rietveld refinements are tried on the diffraction patterns simulated by McStas. The basic strategies are as follows.

Step 1 : Input initial structure parameters and calculate the structure factor for hkl's with Rietveld software.
Step 2 : Input the structure factor for hkl's and simulate the diffraction pattern with McStas.
Step 3 : Rietveld refinement on the simulated diffraction patterns.
Step 4 : Compare the initial parameters in the step 1 and refined parameters in the step 3.

Example : Crystal structure of $LiMn_2O_4$. (LT orthorhombic phase. Unit cell dimensions: 25 Å × 25 Å × 8 Å) [4]. Oxygen vacancy and/or interstitial atoms are artificially introduced in the step 1.

Figure 5(a) and 5(b) show the Rietveld refinement of 'real' and 'simulated' diffraction pattern of LT phase of $LiMn_2O_4$. To simulate the high-resolution diffraction pattern, the profile shape of the unpoisoned-type moderator (2) was applied. Any kinds of line broadening are not considered yet.

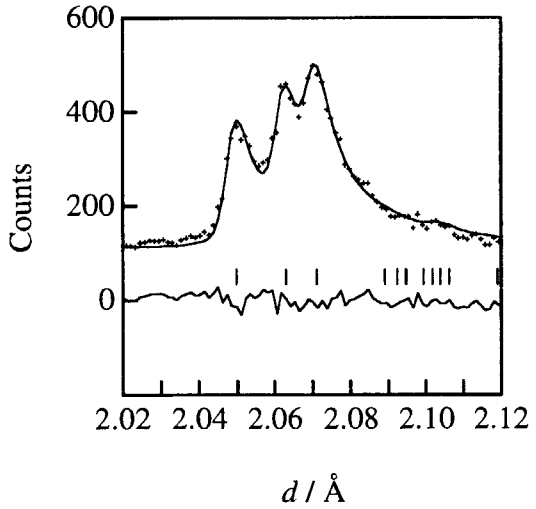

Fig. 5(a) A part of the Rietveld refinement pattern of $LiMn_2O_4$ at 250 K measured on Vega diffractometer ($\Delta d / d$ ~ 0.25 %). Observed intensity data are shown by crosses, and the solid line is the calculated intensity. Vertical markers below the diffraction patterns indicate positions of possible Bragg reflections. Differences between the observed and calculated intensities are plotted in the same scale.

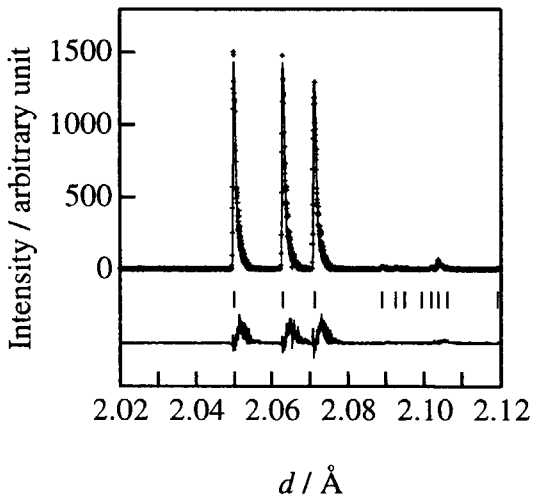

Fig. 5(b) A part of the Rietveld refinement pattern of LT phase of $LiMn_2O_4$ simulated by McStas (L_1 ~ 150 m, $\Delta d / d$ ~ 0.03 %). The profile shape of the unpoisoned moderator was applied in the program. Any particle size and/or strain broadening effects are not considered in the simulation.

At present, the statistics of the simulated diffraction patterns are poor, and the profile shape function used in the Rietveld refinement is not optimised for the simulated diffractometer. To make a decision for the choice of the moderator, these problems have to be improved as soon as possible.

5. Present status

Energy dependencies of candidate for moderator were analytically expressed.

Some simple powder diffractometer were constructed in McStas.
⇒ Make them more realistic. Bandwidth Choppers, Detector locus, time focusing, and so on.

Structure refinements using simulated patterns were just started.
⇒ Need good statistics. Profile shape function in Rietveld refinement program should be modified.

References

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