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Versatile IBARAKI materials design neutron diffractometer at J-PARC

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Abstract. Ibaraki prefecture, the local government of the area for J-PARC site, has decided to build a versatile powder diffractometer (IBARAKI Materials Design Diffractometer) to promote industrial applications for neutron beams in J-PARC. This diffractometer is designed to be a high throughput one enabling materials scientists to use it like the chemical analytical instruments in their material development processes. It covers in d range $0.18 < d$ (Å) < 5 with $\Delta d/d = 0.16\%$ at the high resolution scattering detector bank, and covers $5 < d$ (Å) < 800 with gradually changing resolution. Typical measuring time to obtain a 'Rietveld-quality' data is several minutes for the sample size of laboratory X-ray diffractometer. To promote industrial application, a utilization system for this diffractometer is required. We will establish a support system for both academic and industrial users who are willing to use neutrons but have not been familiar with neutron diffraction.

Introduction

The use of the neutron powder diffraction (NPD) showed a great progress after the high-T_c superconducting oxide boom, since NPD played an important role in the structural clarification of superconductors. Recently, the NPD users are spread from physics and chemistry fields to materials development fields, since many material scientists and industrial users have found that the NPD is effective to solve their research problems. The Ibaraki prefecture, one of local governments in Japan, decided to build a high throughput diffractometer (IBARAKI Materials Design Diffractometer) to promote industrial applications for neutron beams in the J-PARC. In this article, we describe this high throughput neutron powder diffractometer in detail.

Structure evaluation system by neutron powder diffractometer

High quality X-ray powder diffractometers (XRD) are coming into market, and users are trying to extract more structural information from XRD data. Most of the XRD users are actually dealing with materials consisting of both heavy and light elements which are difficult to detect by X-ray: cement, lithium-ion batteries, fuel cell materials, hydrogen absorbing materials, ferroelectric materials, many kinds of oxides, etc. Nevertheless, they never think to use neutrons, because there are a lot of difficulties moving from XRD to NPD, for example limited knowledge, hard access, slow to start actual experiments, amount of sample, limited opportunity, etc. This situation would be altered if we could establish a system of structural evaluation for IBARAKI Materials Design Diffractometer in J-PARC with easy access, quick start, accepting several tens of thousand experiments in a year. Users can use this diffractometer for crystal structure analysis on their own materials Design-Synthesis-Characterization cycle (DSC cycle)(Figure 1). Because the function of materials is often related with its structure, the structure information is utilized to optimize the function of materials. Impact on novel functional materials development would be expected.

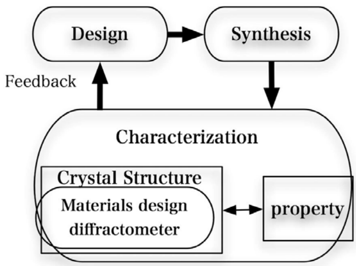

Figure 1. New materials DSC (Design-Synthesis-Characterization) cycle using neutron diffractometer.

Specification for IBARAKI Materials Design Diffractometer

The roles for neutron diffraction in materials science are (1) to do structural analyses of newly developed materials, (2) to clarify the correlation between structures and properties (functions), and (3) to clarify the relation between structural changes and improvements of functions especially for the practical materials. To carry out such purposes, a diffractometer with super high resolution is not required. The matching among intermediate resolution around $\Delta d/d = 0.15\%$, high intensity and wide d coverage is more necessary. For example, some excellent works have been done by Sirius ($\Delta d/d = 0.1\%$ for back scattering bank) diffractometer in KENS Facility [1] for lithium-ion batteries [2], fuel cell materials [3], hydrogen absorbing materials [4] and ferroelectric materials [5].

Table 1. The resolution for back scattering bank and Q range for all detector bank in IBARAKI Materials Design Diffractometer and another TOF versatile type powder diffractometers.

Instrument	$\Delta d/d$	Q_{max}(Å^{-1})	Q_{min}(Å^{-1})	
IBARAKI Materials Design Diffractometer	0.16%	35 70 35	0.015 0.015 0.007	Normal mode High-*Q* mode Long-*d* mode
GEM (ISIS)	0.25%	55.0	0.04	
Vega (KENS)	0.25%	12.6	0.34	
Sirius (KENS)	0.12%	31.4	1.80	
GPPD (IPNS) [9]	0.24%	38.65	0.96	
SEPD (IPNS) [10]	0.34%	15.3	0.20	

IBARAKI Materials Design Diffractometer is planned to be a high throughput diffractometer with intermediate resolution; it is designed to have wider *Q* range than GEM[6] of ISIS and Vega[7] of KENS with slightly worth resolution than that of Sirius of KENS [8] (Table 1). This Diffractometer is designed to look at a decoupled-poisoned liquid hydrogen moderator (36 mm, off-centered), and to have the incident flight path (L1) of 26.5 m with a T_0-chopper, three wavelength selection disk-choppers and straight neutron guides with the total length of 14.0 m. The detailed design of the neutron guide was reported in another paper [11]. The schematic 3-D view and the instrumental parameters of this diffractometer are listed in Figure 2 and Table 2, respectively. There are four detector banks including a low angle and a small angle scattering detector bank. The angular coverage of each detector bank is also shown in Table 2. The rotation speeds for the T_0-chopper and disk-choppers are the same with the pulse repetition rate 25Hz for the most applications. In this case, the diffractometer covers $0.18 < d$(Å) < 2.5 with $\Delta d/d = 0.16\%$, and covers $2.5 < d$(Å) < 400 with gradually changing resolution. When we select 50Hz for the T_0 rotation speed, we can access d_{min} =0.09 Å ($Q_{max} < 70$ Å^{-1}) with 0.16% resolution. It should be very effective for PDF analysis of crystalline samples. When the speed for wavelength selection disk-choppers is reduced to 12.5Hz, we can access a wider *d*-range, $0.18 < d$(Å) < 5.0 with $\Delta d/d = 0.16\%$, and $5.0 < d$(Å) < 800 with gradually changing resolution.

A typical measuring time for 'Rietveld-quality' data is several minutes for the sample size of laboratory X-ray: a few hundred milligrams. Therefore, it enables materials scientists to use this diffractometer like the chemical analytical instruments in their materials development process.

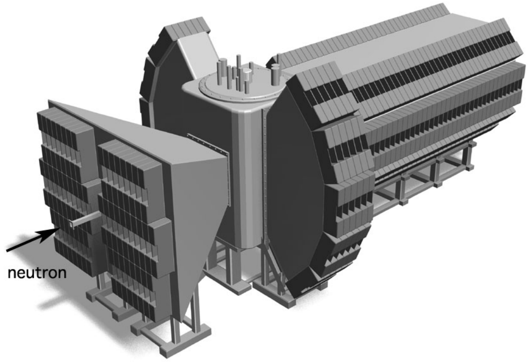

Figure 2. Schematic view of IBARAKI Materials Design Diffractometer. High-resolution bank, special environment bank (90 degree bank), can be seen from left to right. Low angle bank and small angle detector bank, which are not shown in figure, are situated in the low angle vacuum chamber (right hand of the figure).

Table 2. Instrumental parameters of IBARAKI Materials Design Diffractometer. L2 is the scattered flight path.

L1	26.5m
Position of T_0 Chopper	10.53 m
Position for Disk choppers (Wavelength-selection choppers)	7.5 m (double) 11.25 m (single) 18.75 m (single)
2θ and L2 for high-resolution bank	$150^\circ \leq 2\theta \leq 175^\circ$ 2.0-2.3 m
2θ and L2 for special environment bank	$80^\circ \leq 2\theta \leq 100^\circ$ 1.5 m
2θ and L2 for low angle bank	$10^\circ \leq 2\theta \leq 40^\circ$ 1.2-4.5 m
2θ and L2 for small angle bank	$0.7^\circ \leq 2\theta \leq 5^\circ$ 4.5 m

This diffractometer accepts various kind of apparatus to meet users demands: temperatures, pressures, magnetic fields, chemical cells etc. It also will have a special stage for combinatorial chemistry. The concept of the combinatorial chemistry robot is the combination of the automatic sample changer robot, synthesis robot and analysis system. The combinatorial chemistry robot selects some composition from libraries, and begins *in situ* neuron diffraction during synthesis process. After measurement, the analysis is automatically done by data analysis system. The obtained structural data, for example bond lengths, will be examined by the global optimization to select the next synthesis condition and will be fed back for new material designing.

The utilization of IBARAKI Materials Design Diffractometer for wide user access

Development of a utilization system of IBARAKI Materials Design Diffractometer is the most important factor for promoting potential users to become actual users. We need to establish a support system for both academic and industrial users who are willing to use neutrons but have not been familiar with neutron diffraction. For example, tutorial courses should routinely be held, easy access to IBARAKI Materials Design Diffractometer should be introduced followed by quick experiments and output. The details of this system have been discussed with the prefectural government. In addition, since several tens of thousand experiments in a year will be carried out, the samples handling would be a big problem. We have to prepare special large amounts of sample holders, and the sample databases, which are relevant with user's information, safety information, experimental data information, etc. This will be especially useful for those who will use IBARAKI Materials Design Diffractometer like chemical analyzers in their materials development processes. They may visit the facility and carry out experiments by themselves, or may not come to the facility and ask for mail-in measurements. Some users also would like to use remote access route to the facility. A software package consisting of combination of several powder-diffraction software, structural databases and visualization software should be easily utilized for the materials structural studies (Figure 3). Most of the know-how in data pre-processes and analysis software themselves make NPD analyses difficult to conduct. They depend on the kinds of studies. For example, the know-how for structural study of metals is different with those for hydrogen absorbing metals. The know-how data can be implemented so that users do not have to worry about it. This software package should be also able to deal with XRD data because users prefer single platform. Results of theoretical calculation are also directly compared with visualized results on the same platform.

Current status for construction of the instrument

The designing work of IBARAKI Materials Design Diffractometer has almost finished. The beamline shielding will be installed in the beginning of 2007. Total 14m of super mirror guide was already delivered and will be installed in the middle of 2007. The construction of IBARAKI Materials Design Diffractometer will be completed in the beginning of 2008, as one of day-one instruments for J-PARC. The preparation for software package as describe above is also in progress.

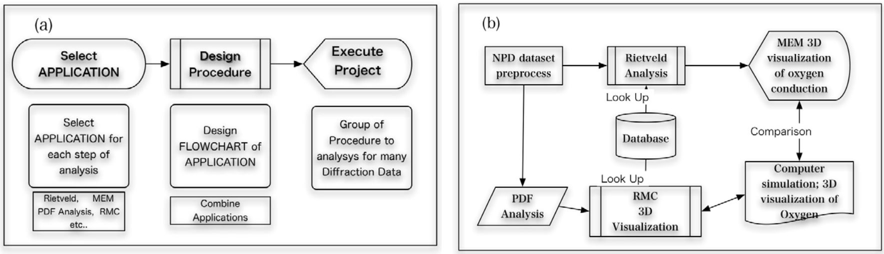

Figure 3. Concept (a) and example (b) of data analysis software package for Materials Design Diffractometer.

Conclusion

IBARAKI Materials Design Diffractometer with four detector banks design for wide Q range coverage. This means that it is not only the high throughput powder diffractometer, but also a SANS (nano structure) and a total scattering instrument (local structure). We will also prepare for the analysis software package. The wide Q coverage and the short measuring time for this diffractometer and software package will lead to the holistic understanding of materials structure and their function for materials development.

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