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Super High Resolution Powder Diffractometer at J-PARC

Shuki TORII*, Masao YONEMURA, Teguh Yulius SURYA PANCA PUTRA, Junrong ZHANG, Ping MIAO,
Takashi MUROYA, Ryoko TOMIYASU, Takahiro MORISHIMA, Setsuo SATO, Hidenori SAGEHASHI,
Yukio NODA[1] and Takashi KAMIYAMA

High Energy Accelerator Research Organization, Tokai-mura, Naka-gun 319-1106

[1]Tohoku University, Aoba-ku, Sendai 980-8577

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Super High Resolution Powder Diffractometer, SuperHRPD, is located at about 100 m from a thin side of a decoupled poisoned moderator at the Materials and Life Science Experimental Facility in the Japan Proton Accelerator Research Complex. The first neutron was produced successfully from a spallation neutron source in J-PARC in the end of May of 2008, and SuperHRPD has achieved the world best resolution $\Delta d/d$ = 0.035% in June using the existing *Sirius*[1] chamber. In the summer of 2009, a new SuperHRPD chamber was installed aiming at increasing the detector solid angle and expanding *d*-range / *Q*-range, with at most 1500 one-dimensional ^{3}He position-sensitive detectors of 1/2 inch in diameter. The on-beam commissioning of the new SuperHRPD was started in the autumn of 2009.

KEYWORDS: J-PARC, Powder diffractometer, Time-of-flight, Neutron

1. Introduction

In 1980s, the Japanese history of high resolution neutron powder diffractometers (NPD) using time-of-flight (TOF) neutron was started from the construction of HRP[2] at the Neutron Science Laboratory (KENS) of the High Energy Accelerator Research Organization (KEK). HRP had been operated successfully since 1983, with resolution of $\Delta d/d$ = 0.2 ~ 0.3 % and with a reasonable counting rate. After that, the diffractometer VEGA[3], being superior in resolution and counting rate, was constructed in 1993, with $\Delta d/d$ = 0.2 % at backward bank detectors. VEGA was active in the field of structure analysis of high temperature superconductors and electric battery materials. Demand for an upgraded NPD experiment, however, came to exceed the ability of VEGA, and we built super high resolution powder diffractometer *Sirius* in the autumn of 1996. *Sirius* was the world's first TOF NPD with supermirror guide tube whose resolution was $\Delta d/d \approx 0.1$ %. VEGA and *Sirius* were kept operating until the shutdown of KENS in 2005, and then Japanese TOF NPD moved the stage to J-PARC.

Recently, the two powder diffractometers of SuperHRPD and iMATERIA[4] have been installed in the MLF of J-PARC. iMATERIA was funded by Ibaraki Prefecture and was designed as a versatile NPD with the best resolution of *Sirius*. SuperHRPD was funded by KEK and was designed so as to achieve the best resolution of synchrotron X-ray powder diffraction, $\Delta d/d$ = 0.03 %. The high resolution of SuperHRPD will enable us to analyze more complex structures, which have not been solved before, and to detect slight structural changes that have so far been disregarded, thereby possibly resulting in new science being progressed.

2. Super High Resolution Powder Diffractometer, SuperHRPD

2.1 Moderator

The lower repetition rate of 25 Hz than that of ISIS TS-I (50 Hz) and SNS (60 Hz) is advantageous for NPD at J-PARC where a wide dynamical range is attainable with limited loss of neutrons. In order to achieve the designed resolution $\Delta d/d$ = 0.03 %, it was necessary to develop high resolution moderator[5]. The high resolution moderator was also required to produce more symmetrical pulses with less tails for accurate crystal structure analyses. We have chosen a decoupled poisoned hydrogen moderator (Fig. 1); hydrogen moderator is better than water moderator at an energy region lower than 100 meV because narrow and symmetrical pulse region (slowing down region) extends to longer-wavelength range. Demands on the signal to noise ratio (*S*/*N*) above 100 meV to be better than 1.5–2 orders of magnitude lead to the preferred decoupling energy of 1 eV with Ag-In-Cd alloy (AIC)[5,6,7]. As for the poisoning material, on the other hand, the choice of Cd results in a shift of the symmetrical region to a shorter wavelength region, and narrower and more symmetrical Δt at $\lambda > 1$ Å will be attained by selecting 20 mm depth in average (25 mm at the deepest). Although poisoning energy is larger in Cd, peak intensity is not so different. Detailed comparison between designed performance and experimental results is being examined.

2.2 Beamline design & suppermirror guide

SuperHRPD consists of tapered iron collimators in biological shields, 82.6 m supermirror guide tube with $m = 3$ starting at 7.145 m, disk choppers, beamline monitors, slits, a sample chamber with the sample position at 94.2 m, detectors, various neutron shields and beamstop. To install this long flight path instrument, the construction of a beamline building (MLF SuperHRPD BL building) with 4 m (H) × 3 m (W) × 50 m (L) and a SuperHRPD experimental hall (MLF SuperHRPD building) with 7 m (H) × 10 m (W) × 13 m (L) was started in July and completed by the end of December, 2007. The design, installation and alignment of the guide tube system were carefully executed because of an expected ground settlement and earthquake. It is noted that elliptical guide was examined but not adopted because the unequal sinking might deteriorate neutron transmission.

The supermirror guide tube is composed of a 31.245 m curved guide part (5.55 m, 25.695 m with 0.11 m gap for the second disk chopper at 12.75 m) with a cross section of 25 mm (W) × 75 mm (H), and a 51.4 m straight guide part with a cross section of 25 mm (W) × 55 mm (H) between the sample position and the moderator. The radius of curvature of the curved part is 5 km. The guide tube cross sections are different to minimize the effect of unequal subsidence of the MLF main building and that of the beamline building: with the present guide tube design, 10 % loss of intensity at most by the 10 mm difference in height due to subsidence[8].

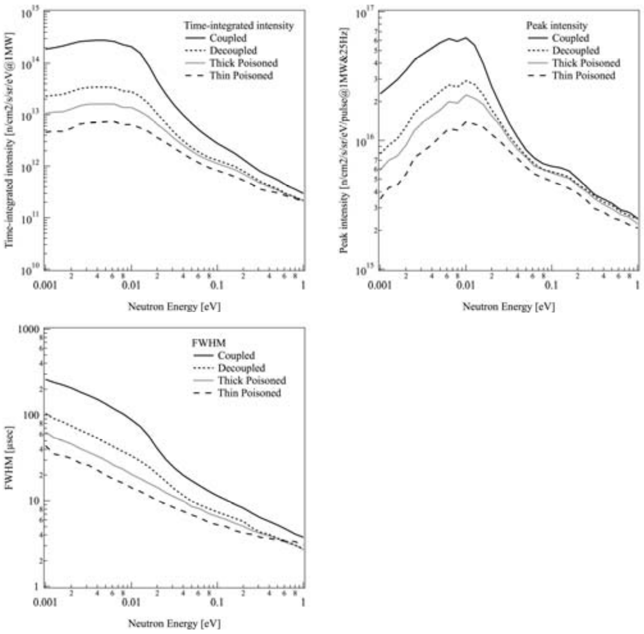

Fig. 1. The calculated pulse characteristics of MLF moderators. For high resolution realization, we selected a thin poisoned decoupled moderator.

2.3 Disk choppers

The flight path as long as 100 m causes frame overlap of pulsed neutron. Under 25 Hz operations, only a wavelength window of about 1.6 Å can be utilized. Therefore, in SuperHRPD, a 5 Hz mode of 1/5 thinned-out operation is adopted to be the basic TOF duration, resulting in a wavelength window of about 8.0 Å. SuperHRPD can realize various operation modes by the combination of two disk choppers. The two disk choppers were designed so as to prevent frame overlap and to utilize the wide wavelength. One is a single disk at 7.1 m with an opening angle of 31.2°, and the other is a counter-rotating double disk chopper at 12.75 m with 94.9°.

2.4 SuperHRPD diffractometer

We proceeded with the construction project of SuperHRPD in two phases in order to accept neutron beam and start the general user program as early as possible. In the "Phase-I", we concentrated on beamline components including guide tubes, shielding, choppers, *etc.* and started up quickly by using the existing *Sirius* chamber and detectors.

In the "Phase-II" starting in the summer of 2009, after the careful R&D studies on windows and chamber designing, we installed a new SuperHRPD chamber (Fig. 2). The new chamber was developed so as to improve *S*/*N* and to achieve better resolution as well as intensity. It was manufactured by a small and medium-sized enterprise group, JSS (the J-PARC Support Study Group), in Ibaraki prefecture. The new chamber consists of a vacuum sample chamber with capacity of about 1 m^3, and gas-filled scattering banks around it. In the design concept of a new chamber, the detector solid angle was increased, *d*-range / *Q*-range was expanded, and also choices of high intensity mode and high resolution mode were implemented by varying incident collimations. To cover this large detector solid angle, about 1500 one-dimensional ^{3}He position-sensitive detectors (PSDs) of 1/2 inch in diameter can be installed in the backward bank, 90 degree bank, and low-angle bank; at present, 320, 192 and 192 PSD(s) from the old *Sirius* diffractometer are installed, respectively. In future, high resolution detector will replace parts of detectors in the backward bank. The 'on-beam commissioning' of all detectors at the new SuperHRPD was completed in autumn, 2009, and general user program restarted with this new chamber.

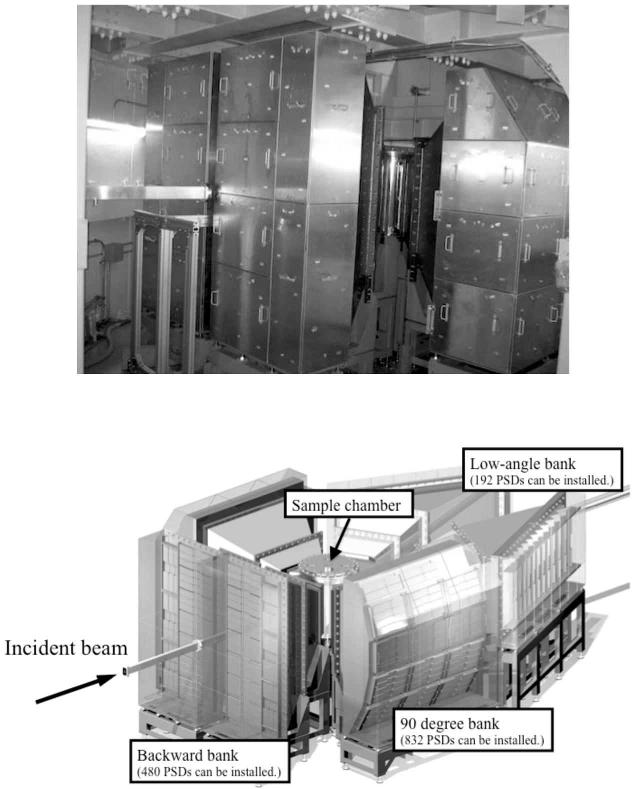

Fig. 2. A photograph (upper figure) and a 3D-image (lower figure) of new SuperHRPD chamber, which was installed in the summer of 2009. About 1500 PSD detectors can be installed in the SuperHRPD diffractometer.

Table I. Instrumental parameters for SuperHRPD.

Moderator	Poisoned decoupled hydrogen moderator
Primary flight path L_1	94.2 m
Curved guide	31.245 m (m = 3, r = 5 km)
Straight guide	51.4 m (m = 3)
Position for disk choppers	7.1 m (single), 12.75 m (double)
backward bank	
2θ	$150° \leq 2\theta \leq 175°$
L_2	2.0 - 2.3 m
d-range	0.3 - 4.0 Å
Resolution $\Delta d/d$	0.03 - 0.15 %
90 degree bank	
2θ	$60° \leq 2\theta \leq 120°$
L_2	2.0 - 2.3 m
d-range	0.4 - 7.5 Å
Resolution $\Delta d/d$	0.4 - 0.7 %
low-angle bank	
2θ	$10° \leq 2\theta \leq 40°$
L_2	2.0 - 4.5 m
d-range	0.6 - 45 Å
Resolution $\Delta d/d$	0.7 - 3.0 %

L_2 is the scattered flight path.

2.5 Sample environment

As for sample environments, the setup of the auto sample changer for 10 samples has been completed and linked with the data acquisition system, so the usual room temperature measurements are fully automated. Low temperature measurement environments are also advanced, and the setup of two cryostats is progressing now; a 5 K refrigerator under operation will be connected with data collection system immediately and a 1 K refrigerator will be introduced soon. The high temperature measurement using a vanadium electric furnace is also available up to 1000°C. In the future, installations of other sample environments such as a tension test machine, a high pressure cell and a high field magnet, *etc*. are planned.

3. Commissioning Report of SuperHRPD

The first powder diffraction pattern of an iron steel block (both α-Fe and γ-Fe are included) was recorded on May 31, 2008, with 500 shots of neutrons (0.3 Hz, 25 mins). Although optimization of a beam slit system still was not completed, the Bragg peaks of α-phase and γ-phase can be clearly separated (Fig. 3).

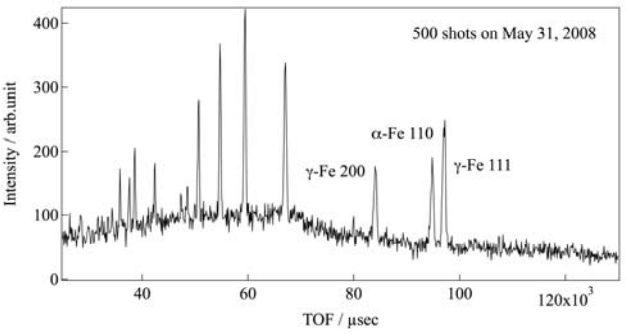

Fig. 3. The first diffraction pattern of J-PARC/MLF: iron steel block.

On June 21, 2008, we had succeeded in achieving the best resolution among all NPD in the world, and a part of the detector had achieved $\Delta d/d$ = 0.035 %. Figure 4 shows a comparison of the Bragg peaks measured by SuperHRPD and the *Sirius* diffractometer at KENS; the FWHM shows a three-fold improvement in SuperHRPD and the tails of the Bragg peaks observed in *Sirius* were not observed in SuperHRPD, resulting in a 10-fold improvement in the 1/10-width with very symmetrical peak profile. This success was achieved by the long-term R&D work for the high resolution decoupled poisoned moderator by the moderator group to attain the desired resolution within the 100 m flight path. This characteristic is very effective for both precise crystal structure analyses of complicated materials with large unit cells and the detection of tiny structural distortions. It is emphasized that development of the high resolution moderator with less tails in the profile is successful.

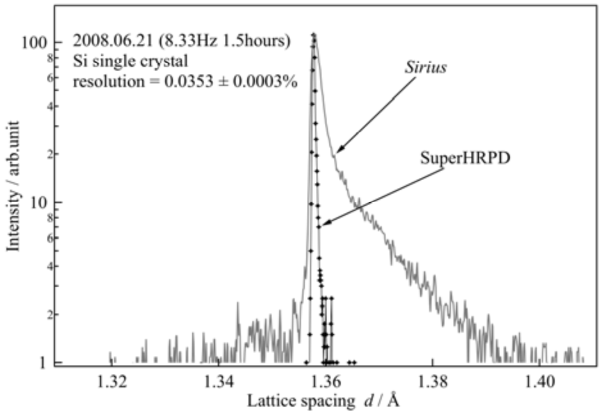

Fig. 4. Si (400) reflection peak measured by SuperHRPD. For accurate crystal structure analyses, the symmetrical pulses with less tails are quite important.

The comparison between Si single crystal data and simulation results in the several energies were carried out and shown in Fig. 5. These simulations were calculated by Monte Carlo simulation code McStas[9]. These Peak profiles have been reproduced to reflect the simulation results for each wavelength.

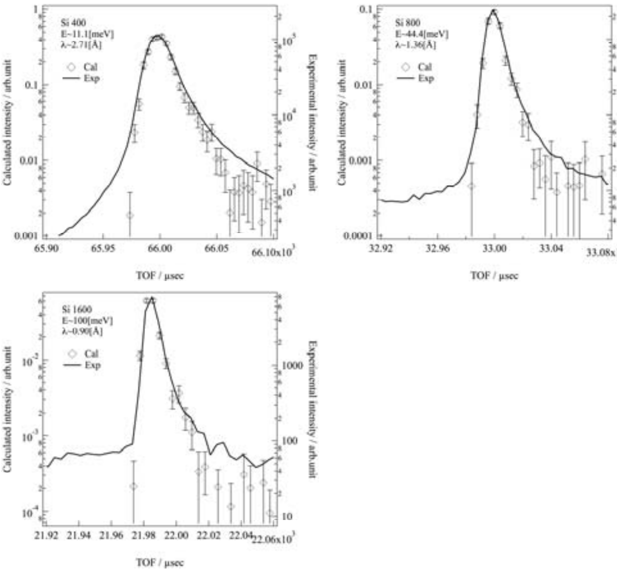

Fig. 5. The comparison between Si single crystal data and simulation results.

We measured some sealed standard samples filled with powder to check the "time focusing" procedure, as well as to verify the capability of SuperHRPD as a high resolution NPD. In Fig. 6, the diffraction pattern of Si powder (NIST SRM 640c) and its Rietveld analysis result with Z-Rietveld$^{10)}$ are shown. Z-Rietveld is Rietveld analysis software and one of the powder diffraction data analysis software suits, Z-Code, which has been developed by us. As the result, an excellent fitting was obtained, and the chi-square value of the fitting was as low as 1.30.

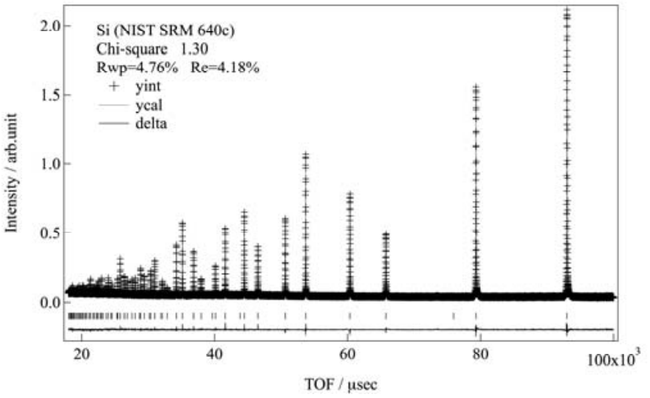

Fig. 6. The Rietveld analysis result of Si powder using Z-Code.

Although we are still using old PSDs of the *Sirius* diffractometer with 1 cm pixel size, it is worth to evaluate the present resolution in the high resolution mode with the diamond powder sample. Figure 7 shows a detector area map of backward bank, the center of the counter map is the detector area where scatter angle is bigger. Figure 8 shows a detector area dependence of the resolution. It was shown that the resolution Δ*d*/*d* better than 0.06 % was realizable by limiting the detector area as well as using sample holder of diameter 3 mm.

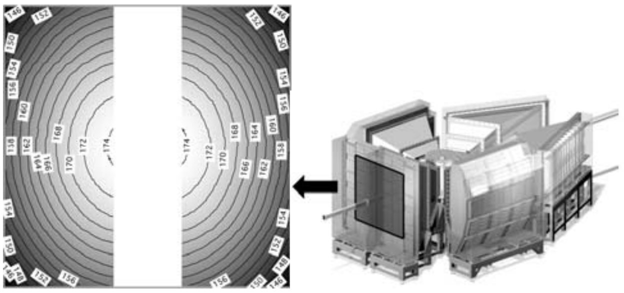

Fig. 7. The scattering angle 2*θ* area map of backward bank detector.

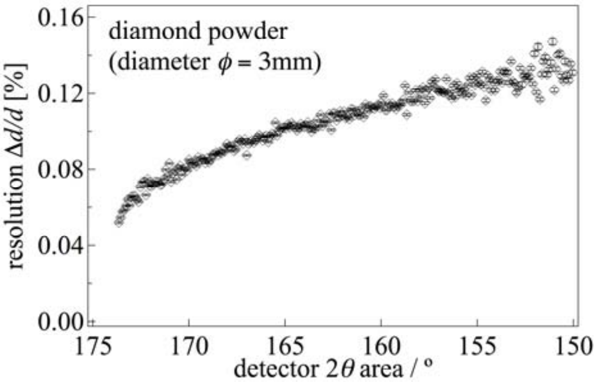

Fig. 8. A detector 2*θ* area dependence of the resolution. The horizontal axis shows the scattering angle 2*θ* of backward bank detector.

4. Conclusion

SuperHRPD has achieved the world best resolution Δ*d*/*d* = 0.035 %, indicated the superior performance of the original design. We have completed the installation of a new SuperHRPD vacuum scattering chamber and started on-beam commissioning. The general user programs have been started and more than 40 proposals have been carried out so far. The development of the sample environments such as a 1 K cryostat and a high field magnet are advanced, which is expected in the future development for new science.

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