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For High-Pressure Experiments Using Total Scattering Spectrometer NOVA at J-PARC

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Abstract. To reveal hydrogen-metal interaction in metal hydride systems with high hydrogen density, we have constructed the high-pressure device for neutron diffraction at total scattering spectrometer NOVA in J-PARC. The device consists of the Paris-Edinburgh press, the alignment stages, the automatically regulated oil pump system and the temperature control system. By using this device, diffraction data are expected to be taken at pressures above 20 GPa over wide Q-range of 1-40Å⁻¹ with the resolution of ΔQ/Q~0.6 %, which enables us to determine hydrogen positions in high hydrogen-density materials in the real-space resolution of 0.16Å.

1. Introduction
Nowadays, the energy and environmental problems become important issues in this consuming society. Among several technologies, the fuel cell and hydrogen storage system are promising technology to solve these problems. To invent innovative hydrogen storage materials, fundamental understanding of the metal-hydrogen interaction in metal hydride systems is indispensable. Designing high hydrogen-density states by compression and the investigation of hydrogen positions in these states using neutron diffraction would be one of the most effective ways for that purpose. Now, high-pressure technique can generate pressures enough high to change the hydrogen density drastically [1] and the intense pulse neutron source is about to enable the neutron diffraction for a tiny sample at such pressures.

Neutron scattering experiments above 10 GPa have already done in foreign neutron facilities, such as ISIS[2-4], LANSCE[5], ILL[6], LLB [7], but those have not been so popular in Japan because of low neutron flux of Japanese neutron facilities. Recently constructed intense proton accelerator, Japan Proton Accelerator Research Complex (J-PARC) has changed the situation and now high-pressure neutron diffraction is ready to start at the Material and Life Science Experimental Facility (MLF) at J-PARC. Among several spectrometers already constructed in MLF/J-PARC, the total scattering spectrometer, NOVA has best performance in quick data acquisition. This character suits the high-pressure neutron experiments. In order to realize high-pressure neutron diffraction experiments at NOVA, we have constructed a high-pressure device.

2. Total scattering spectrometer NOVA
The NOVA is the total scattering spectrometer constructed at BL21 in MLF/J-PARC. The schematic image is shown in Fig. 1. It has five kinds of detector banks, which cover from a small to large scattering angle. As the results, the diffraction data can be acquired over wide Q-range from 0.01 Å⁻¹ to 100 Å⁻¹ with the minimum resolution of ΔQ/Q=0.35 %. Wide Q-range down to 0.01 Å⁻¹ for NOVA reveals density and chemical fluctuations in the large length of several hundred angstroms, simultaneously to that in the small length. This will help us investigating coexistence of crystalline and amorphous states, density fluctuation of hydrogen and chemical fluctuation in alloys. Furthermore, the spectrometer shows high performance in quick data acquisition, so it is expected to be useful not only for the conventional structure analysis of liquid and amorphous materials, but also for PDF analyses of crystalline materials and for time-resolved Rietveld analyses. This character also becomes a big advantage in high-pressure neutron experiments, where the tiny samples are used.

![Figure 1. Schematic image of the total diffractometer NOVA and the detector configuration.](image)

### 3. Selection of high-pressure device and the scattering geometry

Even though the neutron source becomes stronger in new pulse neutron sources, we cannot expect such intense flux like that in synchrotron radiation source. We practically need the sample with the size of more than a few cubic millimetres for crystal structure analyses and more than 10 mm³ for liquid and amorphous structure analyses. Taking this into account, we selected large volume hydraulic press as the high-pressure device. Among several kinds of large volume presses, a Paris Edinburgh (PE) press (VX4, max. load 200 ton) with tridonal anvils is appropriate because it can generate pressures of more than 10 GPa, it has relatively large sample volume (88 mm³ for 10GPa-range use and 40 mm³ for 20 GPa-range use) and it is enough compact to be installed in the vacuum chamber of NOVA. The PE press is already installed in many neutron and synchrotron facilities and the fruitful results are reported [6].

PE press uniaxially compresses the sample with two opposed anvils[2-4]. In general, there are two scattering geometries when using this device. The first one is the 90 degree scattering geometry where the beam is introduced along the compression axis and neutrons scattered toward 90 degree are detected. The other is the horizontal scattering geometry where the beam is introduced from the gap between anvils and neutrons scattered within the equatorial plane are detected. We adopted the former geometry in the system, because it can significantly reduce background from materials surrounding sample by limiting the gauge volume using fine incident and receiving collimators.

### 4. High-pressure assembly

The schematic image of our high-pressure system is shown in Fig. 2, and the block diagram is shown is Fig. 3. Our system consists of the PE press, a flange with several feedthroughs, a press adjustment system with 2 translational stages, rotation and tilt stages, an optical observation system with a CCD camera, a pressure control unit, a temperature control unit and an anvil cooling unit etc. The NOVA is usually operated under the vacuumed condition of about 10 Pa to reduce the background from air, so
the high-pressure device must be put in the vacuum chamber. Therefore, the press and the adjustment stages are hung from the flange (Fig. 2). Pressure and temperature is remotely controlled by the oil pump and heating unit, respectively. The change of the sample position by compression, especially the shift in the beam direction which causes the error in d-value, is monitored by CCD and is readjusted by a translational stage. For these purposes, many feedthroughs, such as the 200 MPa oil tube, power supply for heating, thermocouples to measure sample temperature, motor cables for adjustment stages, water tubes for cooling anvils, are attached to the flange.

The incident beam is introduced from the breech of PE press and the neutrons scattered toward 90 degree are detected with the 3He position sensitive detectors in the 90 degree bank (blue parts in the right of fig.1). To reduce the background, the incident beam is truncated into an appropriate size (typically 2-5 mm in diameter) using the incident collimator made of hexagonal boron nitride. The receiving collimators made of B4C rubber is also attached to the both side of the press to reduce the background from the sample surrounding materials. Furthermore, the direction of the incident collimator can be optimized by means of the tilt and rotation stages to optimize the neutron intensity at sample position.

By combining this system to NOVA, we can obtain diffraction data over 2θ angles of 90 plus/minus 5 degrees [8] and can access the Q-range of 1.0 - 40 Å⁻¹. This makes it possible to investigate structure of the materials in the d-range of 0.16 - 6.3 Å with the real-space resolution of about 0.16 Å. This wide accessible Q-range is expected to help us investigating not only the hydrogen positions in crystalline materials but also those in liquid and amorphous materials.
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[8] Although the 90 degree bank of NOVA originally covers 2θ angle of 90 plus/minus 17 degrees, the small gap between two anvils restricts the accessible 2θ range to 90 plus/minus 5 degrees in the high-pressure experiments.