Neutron powder diffraction under high pressure at J-PARC

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A B S T R A C T
It is expected that high-pressure material science and the investigation of the Earth’s interior will progress greatly using the high-flux pulse neutrons of J-PARC. In this article, we introduce our plans for in situ neutron powder diffraction experiments under high pressure at J-PARC. The use of three different types of high-pressure devices is planned; a Paris–Edinburgh cell, a new opposed-anvil cell with a nano-polycrystalline diamond, and a cubic anvil high-pressure apparatus. These devices will be brought to the neutron powder diffraction beamlines to conduct a “day-one” high-pressure experiment. For the next stage of research, we propose construction of a dedicated beamline for high-pressure material science. Its conceptual designs are also introduced here.

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1. Introduction
Application of high pressure causes decrease of inter-atomic distances, and atomic interactions may change. As a result, high pressure often induces drastic changes in structure, physical properties, chemical reactions, etc., and sometimes remarkable phase transitions may occur. Thus, the research on materials under high pressure is important as an academic science, and it is also a powerful tool to produce exotic new materials. In addition, high-pressure research has another important application related to Earth and planetary science, because the Earth’s interior is a world of high pressure.

Some information on phenomena under high pressures may be obtained by the samples recovered to the ambient conditions. In order to characterize the phenomena more precisely, however, in situ observation is indispensable, and in situ diffraction experiments under high pressure can play a key role in obtaining structural information on various materials. For X-ray diffraction studies, there are many dedicated high-pressure beamlines at the synchrotron radiation sources in the world, and many satisfactory results have previously been obtained.

It is expected that further outstanding high-pressure researches which are difficult to conduct with X-rays will be made with neutrons. Studies of crystal structure of hydrogen-bearing materials, order–disorder transitions of minerals, structure of light element liquids at high pressure, etc. are among such research topics. Generally, in situ diffraction experiments under high pressure have many difficulties. First, the diffraction signal is weak because the sample is very small and is surrounded by the pressure medium. Secondly, the opening window for diffraction is limited by the shape of high-pressure devices, and thus it is difficult to cover the wide angle range needed for the angular dispersion method. High fluxes of neutrons and time-of-flight (TOF) measurements are expected to overcome these difficulties. This is the main reason why high-pressure scientists have been eager to use J-PARC.

So far, ISIS, in the United Kingdom, has been at the forefront of high-pressure pulse neutron study [1]; it has a dedicated high-pressure beamline, and many significant papers have been published. At SNS, in the United States, the high-pressure research project SNAP (Spallation Neutrons and Pressure) is about to start [2]. Here, we present our plans for in situ neutron powder diffraction experiments under high pressure at J-PARC.

2. High-pressure devices
As the first step, we are planning to make new high-pressure devices suitable for neutron diffraction, and bring them to the conventional beamlines. A toroid-type high-pressure apparatus with a so-called “Paris-Edinburgh press” is a standard high-pressure device for neutron study [3]. It can generate pressures up
to 10 GPa on samples with volumes of ~100 mm$^3$, and pressures up to 30 GPa with its special design. We will make further improvements of this device particularly for stable high temperature generation for experiments at J-PARC.

In order to expand the pressure region, a new high-pressure device is being developed in Japan. Several years ago, Irifune et al. [4] succeeded in synthesizing a nano-polycrystalline diamond (NPD) by direct conversion of graphite at high pressures and high temperatures. This NPD is a solid chunk made of nano-meter sized diamonds that is transparent and has a yellowish color (Fig. 1(a)). To our surprise, its Koop hardness is higher than that of single crystal diamond (Fig. 1(b)). Unlike the single crystal diamond, NPD has no cleavages, and potentially larger size chunks can be produced. Hence, NPD is a very promising anvil material for high pressure generation. Diamond anvil cell (DAC) is widely used in X-ray experiments, but its sample volume is extremely small ($\sim 10^{-3}$ mm$^3$), rendering it useless for neutron diffraction. In our new device, single crystal diamonds in DAC are replaced by NPD, with which we intend to generate pressures up to 60 GPa on sample volumes of ~1 mm$^3$. Fig. 2 is a schematic drawing of the NPD high-pressure device mounted on a neutron powder diffractometer. Since NPD is transparent, optical observation and/or Raman scattering measurements can also be made simultaneously.

Another original Japanese high-pressure device is a cubic anvil apparatus combined with a hydraulic press (Fig. 3). In this device, a cubic pressure medium is compressed from three directions, and thus fairly isotropic pressure is obtained. Pressures up to 20 GPa on sample volumes of 10 mm$^3$ can be generated, and very stable high temperatures, up to 2000 K, can simultaneously be generated by a small furnace placed in the pressure medium. For the powder neutron diffraction experiment, an incident neutron beam passes through the vertical anvil gaps and irradiates the sample in the pressure medium. Diffracted neutrons go through the other anvil gaps at a 90° direction and are collected by the detectors on the 90° bank. It is expected that the use of sintered cubic boron nitride as an anvil material will greatly decrease the contaminating signal coming from the environment (anvil, pressure medium). The cubic anvil apparatus usually requires a large hydraulic press, but recently we were successful in reducing its size and producing a clamp type “palm cubic anvil cell.” Fig. 4 is a schematic drawing of the cell (80 mm in diameter and 85 mm in height). High pressure generation up to 7.7 GPa has been confirmed at room temperature, and the clamp cell was successfully refrigerated to 0.57 K with a $^3$He refrigerator [5].

These high-pressure devices will be brought to the neutron powder diffraction beamlines. We plan to conduct our “day-one” high-pressure experiment on the engineering materials diffractometer.

3. High-pressure dedicated beamline

The Japanese high-pressure-science community has proposed construction of a dedicated beamline for high-pressure material science in the next stage of research. Fig. 5 shows a schematic drawing of the beamline. L1 (the distance from the neutron source to the sample) and L2 (the distance from sample to detectors) are 24 and 1 m, respectively, and two disc choppers are placed at 7.3 and 10.15 m from the source, which enable a wavelength range $0.45 < \lambda < 13.40$ Å. The $d$-spacing ranges from 0.3 to 9.5 Å at the scattering angle 90°.

This beamline has two significant characteristics. One is the installation of a large-sized hydraulic press that can apply forces of ~1000 ton to the sample. Such large presses were installed on the synchrotron radiation beamlines at SPring-8 and APS, and...
have produced many successful results that are difficult to obtain with compact high-pressure devices. We are investigating the specifications of large presses suitable for pulse neutron diffraction of the high-pressure beamline at J-PARC.

The other significant characteristic is the use of a focusing device. Focusing is crucial to high-pressure diffraction experiments because it is necessary to focus the neutron beam spatially on the tiny samples to increase the neutron flux and reduce the background noise. We have investigated four types of supermirror guides (straight, linearly tapered, parabolic, and elliptical) by Monte Carlo simulation with regard to our requirements: (i) focus thermal neutrons 0.5–10 Å in wavelength, (ii) produce a focal spot less than 1 mm, (iii) keep beam divergence low and (iv) use the reflection from a multi-layer (3Qc) super-mirror guide. We have concluded that an elliptically shaped guide mirror is the best candidate, taking all these factors into consideration. The details of the simulation results are described in another article in this issue [6].

Our proposal for the high-pressure and high-temperature material science beamline was accepted by the J-PARC committee. We are now making an effort to obtain a grant for its construction.

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References

[1] [http://www.isis.rl.ac.uk/Crystallography/hipr/].