Pressure generation to 125 GPa using a 6-8-2 type multianvil apparatus with nano-polycrystalline diamond anvils

T. Kunimoto¹ and T. Irifune¹
¹Geodynamics Research Center, Ehime University, Bunkyo-cho 2-5, Matsuyama, 790-8577, Japan
E-mail: kunimoto@sci.ehime-u.ac.jp

Abstract. Pressures to about 125 GPa at room temperature have been confirmed by synchrotron in situ X-ray observations, using a newly designed triple-stage (6-8-2) multianvil apparatus with nano-polycrystalline diamond (NPD) as the third-stage opposed anvils. Upon heating the sample at a fixed press load (500 tons), the pressure stayed virtually the same at temperatures to 1100 K, while it decreased to 83.7 GPa when the temperature was further increased to 1300 K. This pressure was maintained for 2 hours without notable decrease during the heating. The present 6-8-2 system can be used in studies of in situ X-ray observation of phase transitions under these pressures, which are far higher than those achieved in conventional Kawai-type multianvil 6-8 system.

1. Introduction

The pressures available in multianvil apparatus have long been limited to about 30 GPa using WC anvils (e.g., Ito and Takahashi, 1989 [1]; Irifune et al., 1992 [2]) and relatively small sintered diamond (SD) anvils of less than 10 mm edge length (e.g., Funamori et al., 1996 [3]). However, introduction of SD anvils with larger dimensions (14 mm edge length; Ito et al., 1998 [4]) combined with a newly designed DIA-type multianvil apparatus (SPEED-MkII; Katsura et al., 2004 [5]) and improvements in materials and designs for the cell assembly and gasket, dramatically expanded the pressure limit to 80-90 GPa in the last few years (e.g., Ito, 2007 [6]; Tange et al., 2008 [7]). Nevertheless, it has been difficult to achieve the pressures in Mbar region in the
conventional 6-8 type (or Kawai-type) multianvil apparatus.

Endo and Ito (1982) [8] firstly introduced the 6-8-2 system, where a pair of opposed third-stage SD anvils were inserted in the octahedral pressure medium as the pressure intensifiers. They successfully produced pressures about 100 GPa at room temperature, as confirmed by in situ X-ray observations using conventional X-ray source, but the pressure substantially dropped to less than 10 GPa upon increasing temperatures to 1300 K, presumably due to the softening of the SD anvils used as the third-stage anvils. Utsumi and Aoki (2003) [9] also introduced a 6-2 system using the similar concept to the 6-8-2 system, although the opposed SD anvils were placed in a cubic cell assembly operated in cubic-type apparatus. A similar pressure of ~90 GPa was recorded at room temperature, but the pressure also significantly dropped with increasing temperature.

Kunimoto et al. (2008) [10] introduced a new design for the 6-8-2 system, and tested various diamonds (SD, single crystal (SCD), and NPD) as the third-stage anvils, possessing the tapered rod shape with a diameter of 1.7 mm (0.8 mm on the culet surface) and 1.0 mm long. The pressures achieved with three kinds of anvils at the room temperature were essentially the same (~80 GPa) by using these anvils, which were maintained upon increasing temperature to about 800 K, where the pressures with SD and SCD anvils dramatically dropped down to 20 GPa at 1400 K. In contrast, the pressure remained virtually the same or even slightly increased up to about 1300 K when NPD anvils were used. The pressure then started to decrease above this temperature, but was remained above 50 GPa even at a temperature of 1400 K, demonstrating that NPD is superior to SD and SCD as anvils for high pressure generation at high temperature.

We have recently succeeded in producing larger NPD rods of up to ~7 mm in both diameter and length (Isobe et al. [11]), which are now easily processed with pulse laser (e.g. Okuchi et al., 2008 [12]). Then we designed a 6-8-2 system for practical use for in situ X-ray observations in Mbar region using larger NPD anvils. Here we report performance of the newly designed 6-8-2 system in pressure generation both at room temperature and high temperature using NPD anvils. Results of a preliminary experiment on in situ X-ray observation of a phase transition in Mbar regime are also shown.

### 2. Experimental

Figure 1 shows schematic illustrations for the cell assemblage used for the present 6-8-2 system. NPD was synthesized at 15 GPa, 2700 K using a 3000-ton multianvil press at Geodynamics Research Center, Ehime-University, and processed with a pulse laser.
laser and finished by polishing with diamond powders at SYNETEK Co. Ltd. Thus produced NPD anvil has a diameter of 2.8 mm and a length of 2.0 mm, with a beveled top surface of 0.8 mm in diameter. We adopted tungsten carbide cubes with an edge length of 14 mm and a truncated edge length (TEL) of 5.0 mm as the second-stage anvils, and used conventional preformed gaskets made of pyrophyllite.

A cylindrical heater made of hBN+TiB$_2$ was used, and the sample was directly put into a small hole (0.3 mm in diameter and 0.2 mm in length) in semi-sintered MgO disk (Fig. 1 (a), (b)), which was also used as the pressure reference. A W$_{97}$Re$_3$-W$_{75}$Re$_{25}$ thermocouple was used to measure the sample temperature, which was covered with MgO sleeves to avoid possible reactions with the heater. A cBN disk and amorphous B disks were used as “internal gasket” to confine high pressure in the sample placed in the MgO pressure medium (Fig. 1 (b), (c)). An LaCrO$_3$ thermal insulator and a WC electrode were arranged on the back of each third-stage NPD anvil.

In situ X-ray measurements were made using Kawai-type multianvil apparatus (SPEED-Mk.II) at BL04B1, SPring-8. White X-ray beam was collimated to a square with 50 $\mu$m in horizontal and 100 $\mu$m in vertical lengths for in situ X-ray diffraction measurements. Diffracted X-ray was acquired at 2$\theta$ of 6.0$^\circ$ with a Ge solid-state detector and a multi-channel analyser. A rutile-type GeO$_2$ powder synthesised at 5 GPa and 1500 K was used as the starting material. The pressure was determined from the unit-cell volume of the surrounding polycrystalline MgO using an equation of state by Jamieson et al. (1982) [13]. For comparison, pressures were calculated on the basis of other equation of states, which were recently proposed by Speziale et al. (2001) [14] and Tange et al. (2009) [15]. The press load was increased first to a target value (500 tons), and then the temperature was increased to 1000 K, fixing the press load. After having acquired the X-ray diffraction data, the temperature was further increased to 1300 K, where the diffraction measurement was conducted for 2 hours. The run was then quenched, and the sample was retrieved to the ambient condition for further inspections.
3. Results and discussion

In the present 6-8-2 system with NPD anvils, we were able to achieve pressures as high as 125.2 GPa (cf. 117.9 GPa with Speziale et al., 2001; 122.9 GPa with Tange et al., 2009) at room temperature, and a press load of 500 tons. The pressure stayed virtually the same or even slightly increased upon increasing temperature to 1000 K (125.7 GPa; 117.8 GPa with Speziale et al., 2001; 124.4 GPa with Tange et al., 2009) which is consistent with our earlier result with smaller NPD anvils (Kunimoto et al., 2008). X-ray diffraction data was acquired at this load for 10 minutes. The fluctuation of the temperature was less than 5 K during this period, and we confirmed the formation of pyrite-type GeO$_2$, consistent with Ono et al. (2003) [16].

Upon further increasing temperature, the pressure started to decrease at temperatures near 1100 K, and gradually dropped to 83.7 GPa (81.4 GPa with Speziale et al., 2001; 82.3 GPa with Tange et al., 2009) at 1300 K, where we acquired X-ray diffraction data for 2 hours, as shown in Fig. 2. Both the pressure and temperature did not change significantly during this period, having been within 0.5 GPa and 5 K, respectively. We
found pyrite-type GeO$_2$ was formed at this P, T, condition, which is close to but is actually higher in pressure than that on the phase boundary between $\alpha$-PbO$_2$ and pyrite structures determined using laser heated diamond anvil cell (Ono et al., 2003). Further details of the phase transition boundary determined by the present 6-8-2 system should appear elsewhere (Kunimoto et al., in preparation).

Figure 3 summarizes the pressure and temperature conditions achieved by various types of multianvil apparatus. We have substantially extended the pressures available in multianvil apparatus up to about 125 GPa, at high temperatures of ~1000 K. This is partly due to the superior hardiness of NPD, which maintains its hardness at temperatures substantially higher than those of single crystal diamonds and sintered diamonds with Co binders (Kunimoto et al., 2008). Further optimization of the cell assemblage suitable for the 6-8-2 system with NPD anvils will extend the pressure and temperature conditions achievable in multianvil apparatus, which should greatly contribute to the mineral physics studies for the entire region of the Earth’s mantle.

![Figure 2](https://example.com/figure2.png)

**Figure 2.** X-ray diffraction pattern at 83.7 GPa and 1300 K obtained using the present 6-8-2 MA system. G, pyrite-type GeO$_2$; M, MgO pressure marker; D, NPD anvil; W, W-Re thermocouple.
Figure 3. Pressure-temperature limits in various types of multianvil apparatus. KMA/WC, Kawai-type multianvil apparatus with tungsten carbide anvils (e.g. Ito and Takahashi, 1989 [1]; Irifune et al., 1992 [2]); KMA/SD, Kawai-type multianvil apparatus with sintered diamond anvils (e.g. Irifune et al., 2002 [17]; Ito, 2007; Tange et al., 2008 [7]); 6-2 MA, 6-2 type multianvil apparatus (Utsumi and Aoki, 2003 [9]); 6-8-2 MA, 6-8-2 type multianvil apparatus (Kunimoto et al., 2008 [10]; present study).

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