Laser heating in nano-polycrystalline diamond anvil cell

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Abstract. Laser heating in a diamond anvil cell equipped with nano-polycrystalline diamond anvils (NPDAC) was tested for the first time. NPDAC was found to provide better heating efficiency than the standard DAC with single crystal anvils. This is probably attributed to an order of magnitude lower thermal conductivity of the NPD. High-temperature generation exceeding 4500 K was observed during laser heating of hcp-iron sample at pressure of ~100 GPa. The present study demonstrates a promising potential of laser-heated NPDAC for generation of ultra-high temperature under multi-megabar pressure conditions equivalent to those of the Earth’s core.

1. Introduction

Nano-polycrystalline diamond (NPD) is a binder-less polycrystalline aggregate of nanodiamonds and is synthesized by direct conversion of graphite under static high-pressure and high-temperature (> 12 GPa, >2000°C) in a multi-anvil apparatus [1-3]. It has high hardness of up to 140 GPa (Knoop scale), which is equivalent to or even exceeding that of single crystal diamond (SCD), and no anisotropy of hardness [4-6]. The high hardness can be maintained even up to high temperature due to grain boundary blocking of micro-slip deformation [6]. These physical properties of NPD have attracted considerable attentions not only in industrial applications such as cutting and drilling tools [5], but also in scientific applications such as diamond anvil cell (DAC) [7] and related high-pressure generating apparatus [8]. It has been demonstrated that the use of DAC equipped with NPD anvils (NPDAC) successfully generated a pressure of 210 GPa, which is almost equivalent to the maximum pressure achievable using a standard DAC with SCD anvils (SCDAC), at room temperature, despite that the size of the former anvils is about 30% smaller than the latter’s [7]. Therefore, the use of larger NPD anvils and further optimization of the culet forms can potentially extend the current upper limit of static high-pressure generation.

Despite that NPD shows a dark yellow color to our eyes (Fig. 1) due to the presence of nitrogen impurity and lattice defects [9], it has high optical transparency (equivalent to that of SCD) in the near-infrared range. It should also be noted that NPD has a thermal conductivity of an order of magnitude lower than that of SCD, due to grain boundary scattering of phonons [10]. These
characteristics of NPD provide a positive outlook for laser heating in NPDAC to achieve very high
temperature and high pressure conditions, which is an essential requirement for further advancement
in deep Earth mineralogy. In this paper, we report for the first time the result of laser heating in
NPDAC and demonstrate its effectiveness for high-temperature generation at megabar pressure.

Figure 1. Photograph of nano-polycrystalline diamond anvils.

2. Methods

The NPD anvils (ϕ 3.5 mm × H 2.0, 300-µm flat culet) used in this study were fabricated from as-
sintered NPD blocks by rough laser-shaping [11] and subsequent mechanical polishing. Standard SCD
(natural type Ia) anvils with the same dimension were also prepared and used to compare the laser
heating efficiency in NPDAC and SCDAC. Pure iron (Fe) was used as a sample (target of laser
heating). A pelletized sample with a thickness of 20 µm was sandwiched between two pelletized disks
of an Al2O3 (corundum) pressure medium (10 µm in thickness) and loaded into a sample hole (ϕ 100 ×
H 40 µm) of a pre-indented Re gasket. The Al2O3 layers also serve as a thermal insulator. The
thickness of these pellets was carefully adjusted to minimize the influence of the volume difference of
the insulator and laser absorbent (i.e. sample in this case) on the heating efficiency. Pressure was
measured by the standard ruby fluorescence technique (up to 60 GPa) and the equation of state (EoS)
of Al2O3 [12].

Laser heating was carried out using a double-sided, dual-laser-heating system at BL10XU of
SPring-8, in which each of two fiber-laser beams (λ = 1070 nm) is independently shaped by passing
through an beam expander and a shaper to attain a flat-top energy distribution, and is focused onto
each side of the sample in a DAC. The use of the dual-laser system can simply double the laser energy
and provide more effective heating, compared to the conventional system where a single beam is split
into two beams (i.e. the laser energy is also half). The temperatures generated on the both sides of the
DAC were measured individually by in situ spectroradiometry, and a temperature value at each input
power was obtained by averaging the peak temperatures. For the measurements above 2500-3000 K,
neutral density filters and relevant calibration data were used. We used the same temperature
 calibration data for the laser heating both in NPDAC and SCDAC. Although NPD shows yellow color,
its high optical transmittance (80-97% of that of SCD) in the range of 600-800 nm [9], in which
observed spectral intensity profiles are actually analyzed for temperature measurement. We therefore
assume that the influence of the yellow color of NPD on the temperature calibration and measurement
is not so significant. Indeed, the radiation intensity profiles obtained through NPD and SCD anvils
during laser heating at ~2800 K appear to be almost identical (Fig. 2). Typical exposure time for a
single temperature measurement is 600 msec to 1 sec.

In situ X-ray diffraction (XRD) measurements were conducted at BL10XU beamline using a
monochromatic X-ray (λ = 0.41 Å) and an imaging plate (IP) detector and a CCD detector.
3. Results and Discussion

Laser heating of the sample in both a NPDAC and a SCDAC was conducted after compression to 80-85 GPa at room temperature. Continuous emission of light from the heating spot in the sample began to be observed when the laser power reached 20 W for the SCDAC and 24 W for the NPDAC, and the emission intensity gradually increased with increasing the laser power. The relations between the laser power and measured temperatures are plotted in Fig. 3. SCDAC appears to provide better heating efficiency than NPDAC below 40 W, but above this the temperature increase reached a plateau at around 2500 K. In contrast, in the case of NPDAC, the temperature increased almost linearly with increasing the laser power and eventually reached ~4500 K at 65 W. A slight stagnation of the temperature increase at 40-45 W is possibly a sign of melting of the hcp-iron sample, as previously reported [13]. The melting temperature of hcp-iron is estimated to be around 2700 K at pressure of approximately 100 GPa, which is slightly lower than but may be within error of the results of previous studies [13-15]. After heating, the pressures were found to increase to 97.7 GPa (in NPDAC) and 98.2 GPa (in SCDAC).

The results from these two heating experiments suggest that NPDAC provides better thermal sealing to the sample under laser heating (although the heating efficiency is not so high as the case with SCDAC at low laser powers < 40 W). This is probably related to the thermal conductivity of NPD which is an order of magnitude lower than that of SCD [10], although we do not think that the observed result is entirely attributed to the difference in such a physical characteristic. Since the plastic deformation (dislocation movement) of diamond is significantly accelerated with increasing temperature, the achievable pressure range using NPDAC may be limited at very high temperature conditions. However, it has been empirically demonstrated that compared with SCD, NPD maintain its high stiffness and resistance to plastic deformation up to higher temperature [6, 9] due to grain boundary blocking of micro-slip deformation. Therefore, we believe that the use of laser-heated NPDAC may potentially lead to generation of extreme pressures and temperatures comparable to the conditions of the Earth’s inner core (up to 360 GPa and 6000 K).

Fig. 4 shows an example of XRD profiles of the sample assembly in a NPDAC after laser heating at ca. 80 GPa. At first glance, strong diffraction peaks from the NPD anvils are only observed (Fig. 4a), but when magnified, peaks from the sample (hcp-Iron) and Al₂O₃ pressure medium are also visible (Fig. 4b). The diamond peaks are sharp but much broader than those from the sample and pressure medium. Since NPD anvils have a thickness (table to culet distance) of 2 mm and the incident X-ray

Figure 2. Radiation spectra obtained through NPD and SCD anvils during laser heating at ~2800 K.
transmits through this volume in the case of vertical diffraction geometry, reflections by diamond lattices occur at a wide range of detector-sample distances, resulting in the broadening in the two-theta range. Therefore, for in situ XRD observation through NPDAC, attention has to be paid to possible peak overlaps with these strong peaks from the anvils. Nevertheless, the quality of the diffraction peaks from the sample assembly appears to be good enough for phase identification, accurate pressure determination using EoS, melting determination, etc. The use of an X-ray-transparent gasket and radial diffraction technique is probably effective to improve the pattern quality. However, it is not applicable to the measurements under multi-megabar pressures, in which we can fully exploit the potential of the NPDAC.

![Figure 3](image1.png)

**Figure 3.** The relation between the input laser power and generated temperature during laser heating of hcp-iron in a NPDAC and in a SCDAC at ~100 GPa. The inlet shows microscopic images of the sample in the NPDAC (a) during and (b) after heating.

![Figure 4](image2.png)

**Figure 4.** X-ray diffraction profile collected through a NPDAC after laser heating at 80 GPa. (a) A whole profile (b) a magnified profile from (a), showing diffraction peaks from sample (hcp-Iron) and pressure medium (Cor).
4. Conclusions
We have, for the first time, tested laser heating in a diamond anvil cell equipped with nanopolycrystalline diamond anvils using fiber lasers. NPDAC was observed to provide higher heating efficiency than the standard DAC with single crystal diamond and led to high temperature generation exceeding 4500 K. The intrinsically low thermal conductivity of NPD anvils likely plays a major role in providing better thermal sealing to the sample under heating. In situ X-ray diffraction measurement through NPDAC is practically available for phase identification, pressure determination, etc., although attention needs to be paid to possible peak overlaps with the strong diffractions from the NPD anvils.

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References
[1] Irifune T, Kurio A, Sakamoto S, Inoue T and Sumiya H 2003 Nature 421 599
[2] Irifune T, Kurio A, Sakamoto S, Inoue T, Sumiya H and Funakoshi K 2004 Phys. Earth Planet. Inter. 143-144 593
[3] Sumiya H, Irifune T, Kurio A, Sakamoto S and Inoue T 2004 J. Mater. Sci. 39 445
[4] Sumiya H and Irifune T 2004 Diamond. Relat. Mater. 13 1771
[5] Sumiya H and Irifune T 2007 J. Mater. Res. 22 2345
[6] Sumiya H, Harano K and Irifune T 2008 Rev. Sci. Instrum. 79 056102
[7] Nakamoto Y, Sumiya H, Matsuoka T, Shimizu K, Irifune T and Ohishi Y 2007 Jpn. J. Appl. Phys. 46 L640
[8] Kunimoto T, Irifune T and Sumiya H 2008 High. Pressure Res. 28 237
[9] Sumiya H, Harano K, Kagi H and Irifune T 2009 (in press) Jpn. J. Appl. Phys. Optical characteristics of nano-polycrystalline diamond synthesized by direct conversion from graphite
[10] Sumiya H, Nakamoto Y, Shimizu T, Irifune T, Yagi T 2007 Spec. Issue Rev. High Press. Sci. Technol. 17 14
[11] Okuchi T, Ohfuji H, Odake S, Kagi H, Nagatomo S, Sugata M and Sumiya H 2009 Appl. Phys. A 96 833
[12] Dubrovinsky L S, Saxena S K and Lazor P 1998 Phys. Chem. Miner. 25 434
[13] Saxena S K, Shen G, Lauz P 1994 Science 264 405
[14] Shen G, Mao H-k, Hemley R J, Duffy T S and Rivers M L 1998 Geophys. Res. Lett. 25 373
[15] Boehler R 1993 Nature 363 534