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Pressure generation to 65 GPa in Kawai-type multi-anvil apparatus with tungsten carbide anvils

We have expanded the pressure ranges at room and high temperatures generated in a Kawai-type multi-anvil apparatus (KMA) using tungsten carbide (WC) anvils with a high hardness of Hv = 2700 and a Young's modulus of 660 GPa. At room temperature, a pressure of 64 GPa, which is the highest pressure generated with KMA using WC anvils in the world, was achieved using 1°-tapered anvils with a 1.5-mm truncation. Pressures of 48-50 GPa were generated at high temperatures of 1600 to 2000 K, which are also higher than previously achieved. Tapered anvils make wide anvil gaps enabling efficient X-ray diffraction. The present pressure generation technique can be used for studying the upper part of the Earth's lower mantle down to 1200 km depth without sintered diamond anvils.

Keywords: Kawai-type multi-anvil apparatus; tungsten carbide anvil; sintered diamond anvil; synchrotron; lower mantle

1. Introduction

The Kawai-type multi-anvil apparatus (KMA) is one of the most popular high-pressure apparatus in the field of deep-Earth science, which adopts the 6-8-type double-stage compression system [1]. The KMA is one type of multi-anvil apparatus which is characterized by the double-stage compression such that an octahedral pressure medium is squeezed by truncated corners of eight cubic anvils which are compressed by six outer anvils. Main advantages of KMAs are relatively large volumes, high pressures and homogeneous *P-T* distributions in samples. For example, KMAs have by ~1000 times larger sample volumes than diamond anvil cells under similar pressure conditions. Because of these advantages, KMAs have been especially used for precisely determining mantle mineralogy and measuring physical properties of minerals.

Second-stage anvils of KMAs are usually made of tungsten carbide (WC), but sintered diamond (SD) is sometimes employed to generate higher pressures. KMAs with conventional WC anvils, whose hardness and Young's modulus are about Hv = 1800 and 560 GPa, respectively, can generate pressures up to 25 GPa [e.g. 2], corresponding to the uppermost part of the lower mantle down to 700 km depth. On the other hand, pressures up to 60 GPa were routinely generated by using SD anvils [e.g. 3-

5], and pressures over 100 GPa were attained by using the latest techniques [6]. However, SD anvils are much more expensive than WC anvils. Moreover, special manufacturing techniques for processing anvils and parts of pressure cells are necessary for successful high-pressure and -temperature generation. As a result, SD anvils can be used by only limited research groups [7,8]. For these reasons, it is desired to expand the pressure range of KMAs with WC anvils.

Recently, pressures over 40 GPa were achieved at both room temperature and high temperature of 2000 K even using WC anvils in combination with sophisticated high-pressure techniques [9]. The most essential point in this high-pressure generation is use of hard WC, TF05 produced by Fuji Die Co. Ltd., whose hardness and Young's modulus are Hv = 2200 and 610 GPa, respectively. Another important point is tapering of the second-stage anvils, which allows higher pressure generation by making more stress concentration to anvil top [9]. In addition to Ishii et al. [9], pressures close to 50 GPa were generated by using even harder WC anvils without anvil tapering (TJS01, Hv = 2700 and Young's modulus = 660 GPa), which were newly developed also by Fuji Die Co., Ltd. [10]. However, Kunimoto et al. [10] conducted the pressure generation test only at room temperature. For application to deep-Earth science, simultaneous generation of high-pressure and high-temperature conditions is crucial. Additionally, Kunimoto et al. [10] conducted the pressure generation tests at press loads only up to 10 MN in a DIA-type guide block system. Their pressure generation curve, however, indicated potential to generate even higher pressures at higher press load. Therefore, it is worthwhile to attempt generation of higher pressures even at ambient temperature. We also consider that there is a room to generate higher pressures more efficiently than Kunimoto et al. [10] by adopting the anvil tapering.

In this study, we attempted higher-pressure generation using DIA-type KMAs with TJS01 flat and tapered anvils up to a press load of 15 MN at temperatures of 300-2000 K. We compared pressure efficiencies between the flat and tapered second-stage anvils and discussed usefulness of anvil tapering.

2. Experimental methods

High-pressure-temperature experiments were conducted using KMAs, "SPEED-1500" [11] and "SPEED-*Mk.II*" [12] installed in the beam line BL04B1, at the synchrotron radiation facility SPring-8, Japan, which adopt the DIA-type guide block system and have a maximum press load of 15 MN. As mentioned above, the second-

stage anvils were made of TJS01 produced by Fuji Die Co., Ltd. Samples were compressed using the flat and 1º-tapered anvils with 1.0-mm and 1.5-mm truncations with SPEED-1500 and SPEED-*Mk.II*, respectively. We adopted the geometry of the tapered anvil used in Ishii et al. [9]. Although optimization of the taper angle may be important for high-pressure generation, it was empirically fixed to 1 degree in this study. A detailed design of the tapered anvil was described in Ishii et al. [9]. The reason why Kunimoto et al. [10] tested only to 10 MN is use of relatively small (14-mm edge length) second-stage anvils. This small anvil size limited applied press loads to avoid serious damage of first-stage anvils. We applied higher press loads up to 15 MN by employing the second-stage anvils with a 26-mm edge length.

Generated pressures were determined from the unit cell volume of Au by energy dispersive X-ray diffraction with the equation of state by Tsuchiya [13]. White X-ray beams were collimated to 50 μm horizontally and 100 μm vertically. Diffraction angles (2θ) were fixed to 6 and 8º for SPEED-1500 and SPEED-*Mk.II*, respectively. Diffracted X-rays were collected for 180-900 s using a germanium solid-state detector (SSD) in an energy range up to ca. 160 keV calibrated using fluorescence of Cu, Mo, Ag, Ta, Pt, Au and Pd. Samples were oscillated around the vertical axis between 0 and 8º in the case of SPEED-*Mk.II* to suppress effects of grain growth [12]. Errors in pressures were typically 0.1-0.3 GPa and 0.5 GPa at most.

Figure 1 shows cross sections of cell assemblies for the flat and tapered WC anvils. Cr_2O_3-doped MgO octahedra with 4.1-mm and 5.7-mm edge lengths were used as pressure media, combining with the flat and tapered anvils, respectively. Figure 1a illustrates a cell assembly for the flat anvils. A starting material was a mixed powder of $CaSnO_3$ perovskite (pv) and Au with a weight ratio of 8:1. $CaSnO_3$ pv was synthesized by heating a mixture of $CaCO_3$ and SnO_2 with a mole ratio of 1.03:1 at 1300 K and ambient pressure for 24 h after decarbonation. A cylindrical TiB_2 + BN + AlN composite heater was put in the pressure medium, and electrically connected with two anvils using Re electrodes. The sample was located at the center of the furnace. $LaCrO_3$ discs were put at the both ends of the furnace for thermal insulation. The furnace and electrodes were filled with diamond powder. A cell assembly in Figure 1b has no furnace and was used for pressure generation at room temperature with the tapered anvils. An Au foil in MgO powder was put at the center part of the pressure medium for measurement of pressure. Dense Al_2O_3 rods were placed at the both sides of the MgO + Au part. Figure 1c shows a cell assembly for pressure generation at high temperatures

with the tapered anvils. A sintered mixture of MgO and 10 wt.% of Au was synthesized at 2 GPa and 1300 K for 1 hour using a KMA, and was placed between dense Al_2O_3 rods in a Mo foil heater. A $LaCrO_3$ sleeve was placed around the heater for thermal insulation. Mo electrodes were placed on the both ends of the heater. Al_2O_3 X-ray windows of 0.5 mm in diameter were set along the X-ray path outside of the furnace (Figure. 1c-2). Temperature was measured using a W97%Re3%–W75%Re25% thermocouple. Pyrophyllite gaskets were put around the pressure medium. For the flat-anvil experiment, gaskets were baked at 1083 K for better pressure generation.

We conducted experiments with each cell assembly in the following way. In S3052, which is the flat-anvil experiment with the assembly of Figure 1a, the press load applied to the sample was increased at a constant rate. Compression was stopped every 2-3 MN, and the sample was heated to 1100-1300 K, and then cooled to ambient temperature for relaxation of stress from the pressure medium and the gaskets. Diffraction patterns of the sample were taken before, during and after the heating at each press load. Heating was made at press loads up to 9 MN. In M1873, which is the ambient-temperature tapered-anvil experiment with the assembly of Figure 1b, the sample was compressed at a constant rate, and sample diffraction patterns were taken at intervals of 1-3 MN up to 15 MN. In M1904, which is the high-temperature tapered-anvil experiment with the assembly of Figure 1c, the sample was compressed to 15 MN with collecting diffraction patterns at intervals of 1-3 MN. At the highest press load, heating was conducted to 2000 K at a rate of 100 K/min with collecting diffraction patterns every 200-300 K. Another run (M1890) was also conducted using the assembly of Figure 1c. However, we tested high-pressure generation only at room temperature in the same way as M1904 because the temperature measurement with thermocouple was failed during compression.

3. Results and discussion

Table 1 summarizes experimental conditions of each run and results at 15 MN and 300 K.

3.1. High-pressure generation with flat anvils

Figure 2 shows results of the pressure generation test with flat anvils. At room temperature, a pressure of 52.2 GPa was generated at a press load of 9 MN. A pressure of 54.0 GPa was achieved at 1100 K and the press load. In a heating-cooling cycle at

each press load, pressures increase and decrease by heating and cooling, respectively, due to thermal pressure. At press load above 9 MN, an increase rate of pressure generation becomes worse, and a pressure of only 53.6 GPa was generated even at the maximum press load of 15 MN and room temperature. Although sample pressures were expected to increase by heating, the sample was not heated because the anvil gap was already closed to 50 μm at 9 MN, and we were afraid of complete closure of the anvil gap by heating at higher press load.

3.2 High-pressure generations with tapered anvils

Figure 3 summarizes results of the pressure generation tests with the tapered anvils at room temperature. In all runs, we succeeded in generating pressures higher than 60 GPa at the maximum load of 15 MN. In M1873, the highest pressure of 64.3 GPa was achieved with the cell assembly for room temperature (Figure 1b). In addition to usage of the hard tapered anvils, generation of such a high pressure may be due to a replacement of large volume of MgO pressure medium to alumina, which has higher bulk modulus than MgO, as discussed in Ishii et al. [9]. The pressure efficiency in M1873 is higher than those of the others especially below 12 MN. This is probably because the pressure medium for M1873 (Figure 1b) is replaced by less kinds of cell parts than that of the other runs (Figure 1c). Therefore, the internal filling rate for the cell assembly of M1873 would be higher, which may enhance the pressure efficiency because applied force is transmitted to the sample more efficiently. Figure 3 also shows pressures generated using TF05 tapered anvils with a hardness of Hv = 2200[9] for comparison. At 15 MN, pressure generated with the TJS01 tapered anvils (64 GPa) is by more than 20 GPa higher than that with the TF05 ones (43 GPa). This result clearly shows use of second-stage anvil with high hardness is very useful for pressure generation in KMA as discussed in previous studies (e.g. [9,10]). We also indicate usefulness of anvil tapering for pressure generation. The present results show the pressure generation with the tapered anvils up to 6 MN is relatively high or almost the same efficiency in comparison with that of the flat anvils, even though the tapered anvils adopted a larger anvil truncation (1.5 mm) than the flat ones (1.0 mm). Ishii et al. [9] compared pressure efficiencies between flat and tapered anvils with 1.5-mm truncation and showed anvil tapering enhances pressure efficiency even at 1 MN. Therefore, the present pressure efficiency difference was probably filled by anvil tapering. The pressure generation with the tapered anvils of 54-57 GPa was also by 2-5

GPa higher than that with the flat anvils at 9 MN. Additionally, the pressure-increasing rate with the flat anvils was only 0.2 GPa/MN at press loads above 9 MN, whereas that with tapered anvils was 1.5 GPa/MN. Thus, we have confirmed that the technique of anvil tapering is very effective for high-pressure generation especially at high press-load.

In M1904, the sample was heated to 2000 K at a press load of 15 MN (Figure 4). Compared to the flat anvil experiment (Figure 2), pressure continuously decreased during heating to 2000 K. This is probably because of the less support for confining pressure caused by wider anvil gap than that of flat anvil. Although sample pressures drastically decreased with increasing temperature, pressures over 50 GPa were kept up to 1600 K, and it was still 48.0 GPa at 2000 K. The reason for the drastic pressure drop could be due to high heat flow from the heater through the Al_2O_3 X-ray window, which has a thermal conductivity (~7 W/m·K at 1100 K and 1atm) higher than the $LaCrO_3$ thermal insulator (~2 W/m·K at 1100 K and 1 atm) [14]. We need to improve the high-temperature generation technique to suppress the large pressure drop. Nevertheless, we emphasize that the maximum pressure generated using KMAs with the WC anvils was 44 GPa at high temperatures of 2000 K, which was achieved by the present author's previous work [9]. Since generated pressure by an assembly with X-ray windows can generate much lower pressure than that without X-ray windows [9], it is expected that much higher pressures than 48 GPa can be generated at temperatures around 2000 K by using the cell assembly without X-ray windows.

It is noted that the tapered geometry held an anvil gap of ~200 μm even at 15 MN (Figure 5b), which allows efficient collection of X-ray diffraction at such high press loads. Kunimoto et al. [10] measured surface undulation of 1.5-mm truncation anvils, which was caused by plastic deformation during compression. They showed that the hinder parts at ~3 mm backward from the truncation are raised by 10-20 μm with the depression (40-80 μm) near the truncated surface, depending on hardness of second-stage WC anvil and applied press load. Under load, such undulation should become larger because of elastic deformation. Figures 6a and 6b, respectively, show schematic cross sections of flat and tapered anvils deformed by stress from confining pressure. As shown in Figure 6a, the surface deformation in the flat-anvil experiment would prevent a part of X-ray from going through the anvil gap. On the other hand, the tapered anvil geometry can avoid the closure because the backside of the truncation is originally lower than the anvil top.

In the present experiments, blow-outs occurred every time during decompression and all anvils were broken. Although one of the reasons would be because of too-short decompression time (2-3 hours) due to limited time in synchrotron experiment, this may be able to be suppressed by further optimization of cell assembly, gasket size and anvil geometry.

As a conclusion, our high-pressure generation technique will allow us studying material properties under conditions corresponding to the upper part of the lower mantle (1200 km) without the SD anvils. Especially, this technique will facilitate investigation of phase relations, physical and chemical properties in lower mantle minerals such as bridgmanite (e.g. elasticity, diffusivities of each atom and spin state of Fe), which will improve models of structure and dynamics in the lower mantle. It will be also expected to be applied for synthesis of novel material under extremely high pressure.

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Table 1. Experimental conditions and results at 15 MN and 300 K.

Run no.	KMA	OEL/TEL	Anvil geometry	Cell assembly	*P* (GPa)
S3052	SPEED-1500	4.1/1.0	flat	Fig. 1a	53.6
M1873	SPEED-Mk.II	5.7/1.5	tapered	Fig. 1b	64.3
M1890	SPEED-Mk.II	5.7/1.5	tapered	Fig. 1c	63.3
M1904	SPEED-Mk.II	5.7/1.5	tapered	Fig. 1c	62.4

KMA, Kawai-type multi-anvil apparatus; OEL, octahedral edge length of pressure medium; TEL, truncated edge length of second-stage anvil.

Figure captions

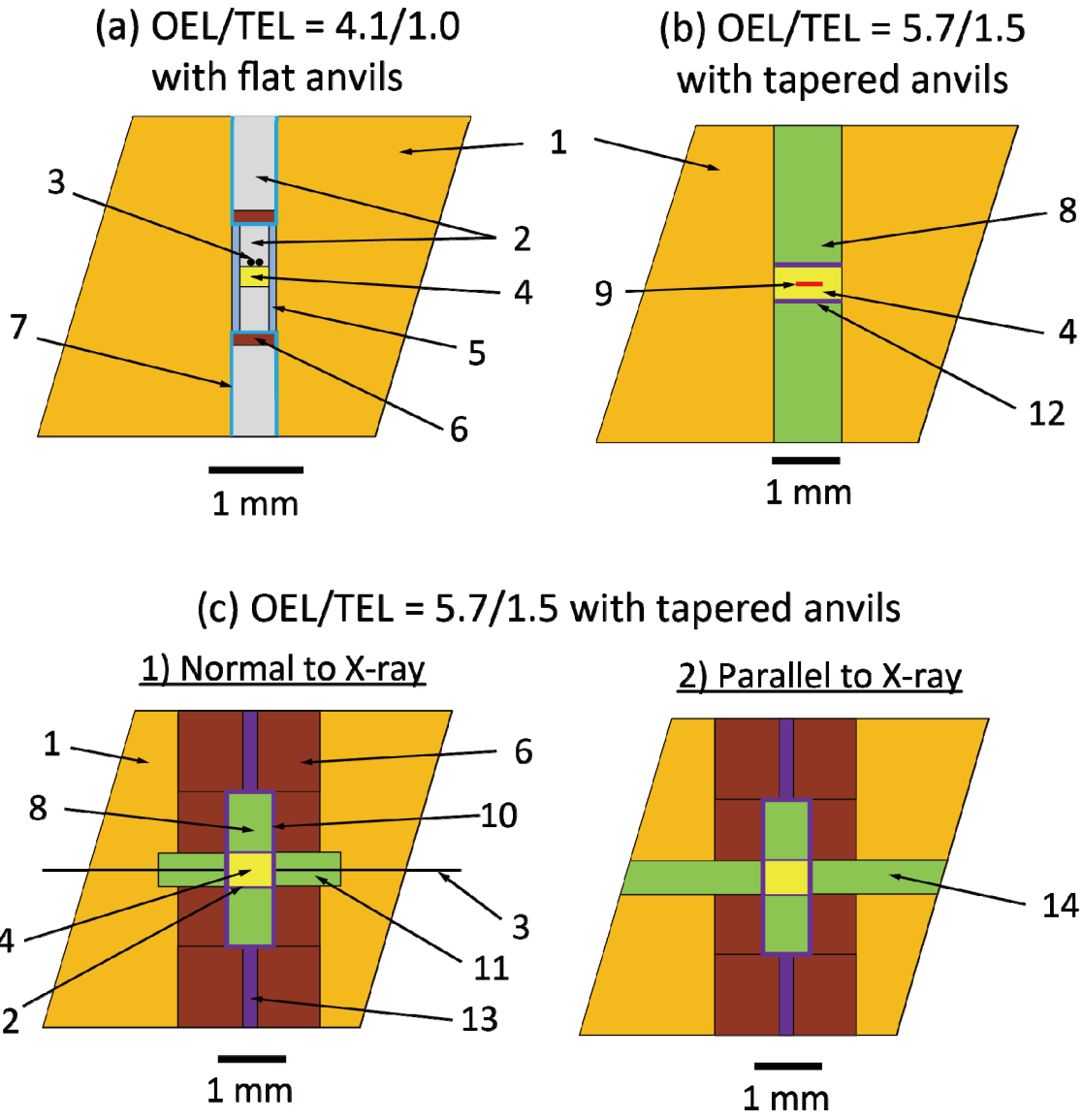
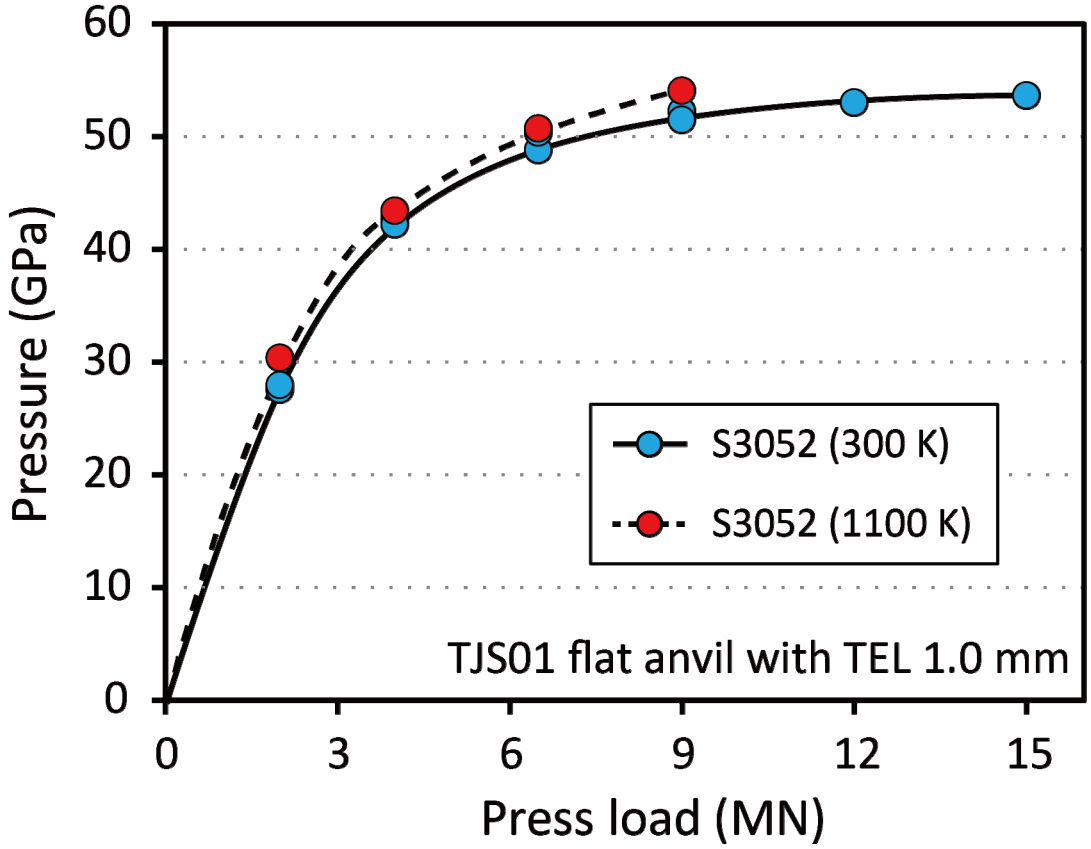


Figure 1. Cross sections of cell assemblies combined with (a) flat anvils with 1.0-mm truncation, (b) and (c) 1°-tapered anvils with 1.5-mm truncation for pressure generation tests at room temperature and high temperature, respectively. 1, 5 wt.% Cr_2O_3-doped MgO pressure medium; 2, diamond powder; 3, W97%Re3%-W75%Re25% thermocouple; 4, sample; 5, TiB_2+BN+AlN composite heater; 6, $LaCrO_3$ thermal insulator; 7, Re electrode; 8, dense alumina; 9, Au foil; 10, Mo heater; 11, dense alumina electrical insulator for the thermocouple; 12, Mo disc; 13, Mo electrode; 14, dense alumina X-ray window. OEL, octahedral edge length of pressure medium; TEL,

318 truncated edge length of second-stage anvil.



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320 Figure 2. Results of a pressure generation test (S3052) in the KMA using TJS01 flat
321 anvils with 1.0-mm truncation. Pressures at 300 K were measured before and after
322 heating up to 9 MN.

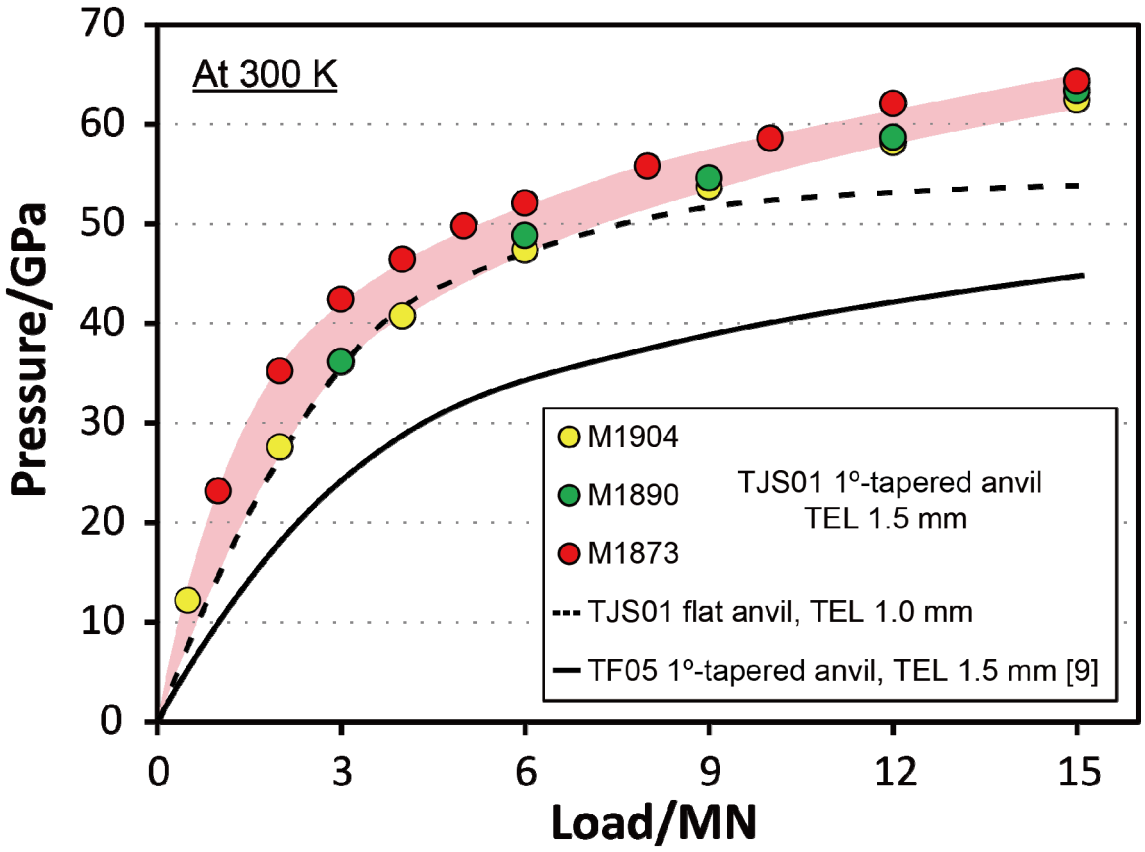

Figure 3. Results of pressure generation tests in the KMA using TJS01 tapered anvil with 1.5-mm truncation. Band-like curve indicates a region of pressure generation with the TJS01 tapered anvils. Dashed curve represents pressure generation using the TJS01 flat anvils with 1.0-mm truncation in the present study (see Figure 2). Solid curve represents pressure generation using the TF05 1°-tapered anvils with 1.5-mm truncation reported by Ishii et al. [9]. TEL, truncated edge length.

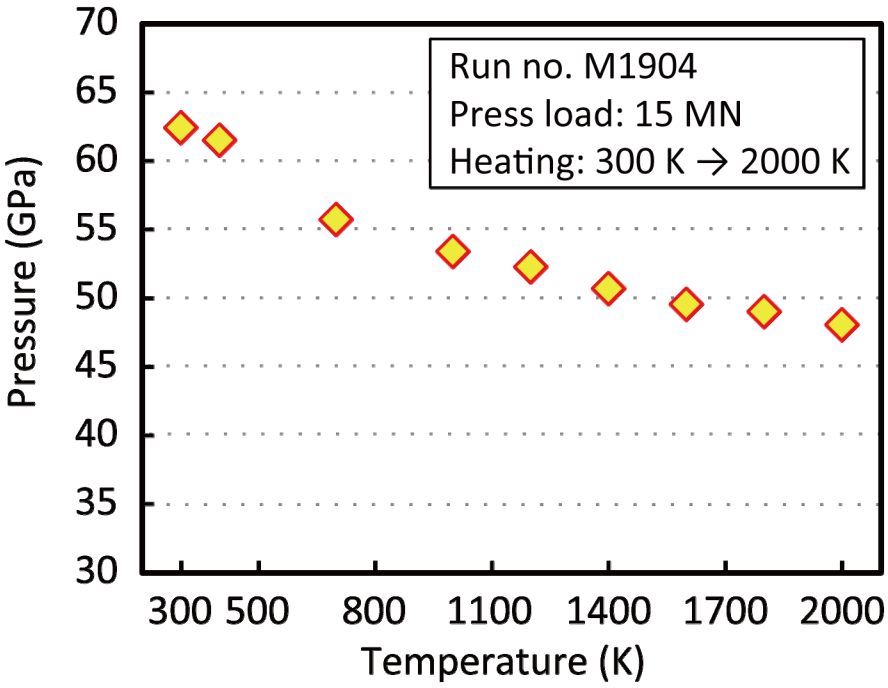

Figure 4. Pressure generations in the KMA with TJS01 tapered anvils during heating up to 2000 K in M1904.

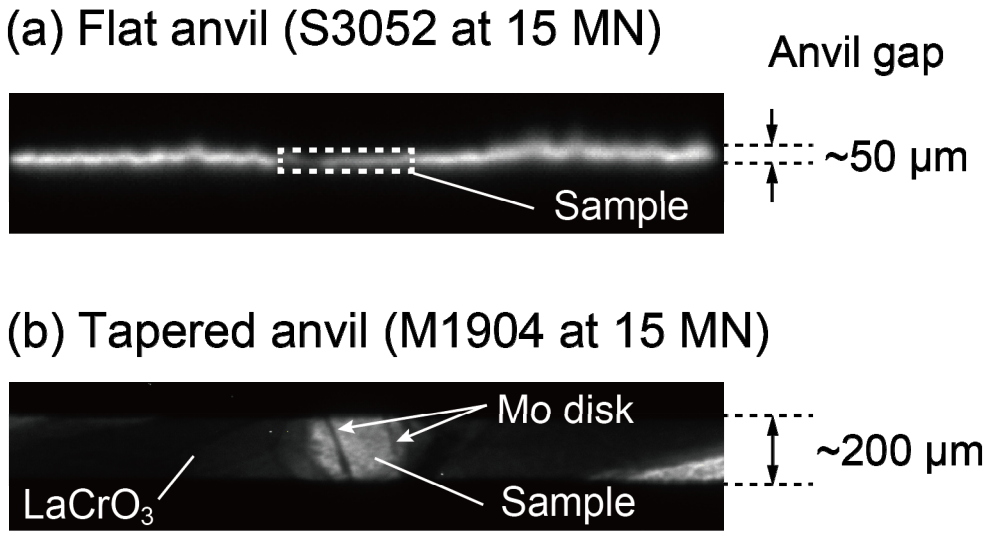

Figure 5. X-ray radiographic images of anvil gaps for (a) the flat and (b) tapered anvils at a press load of 15 MN and room temperature.

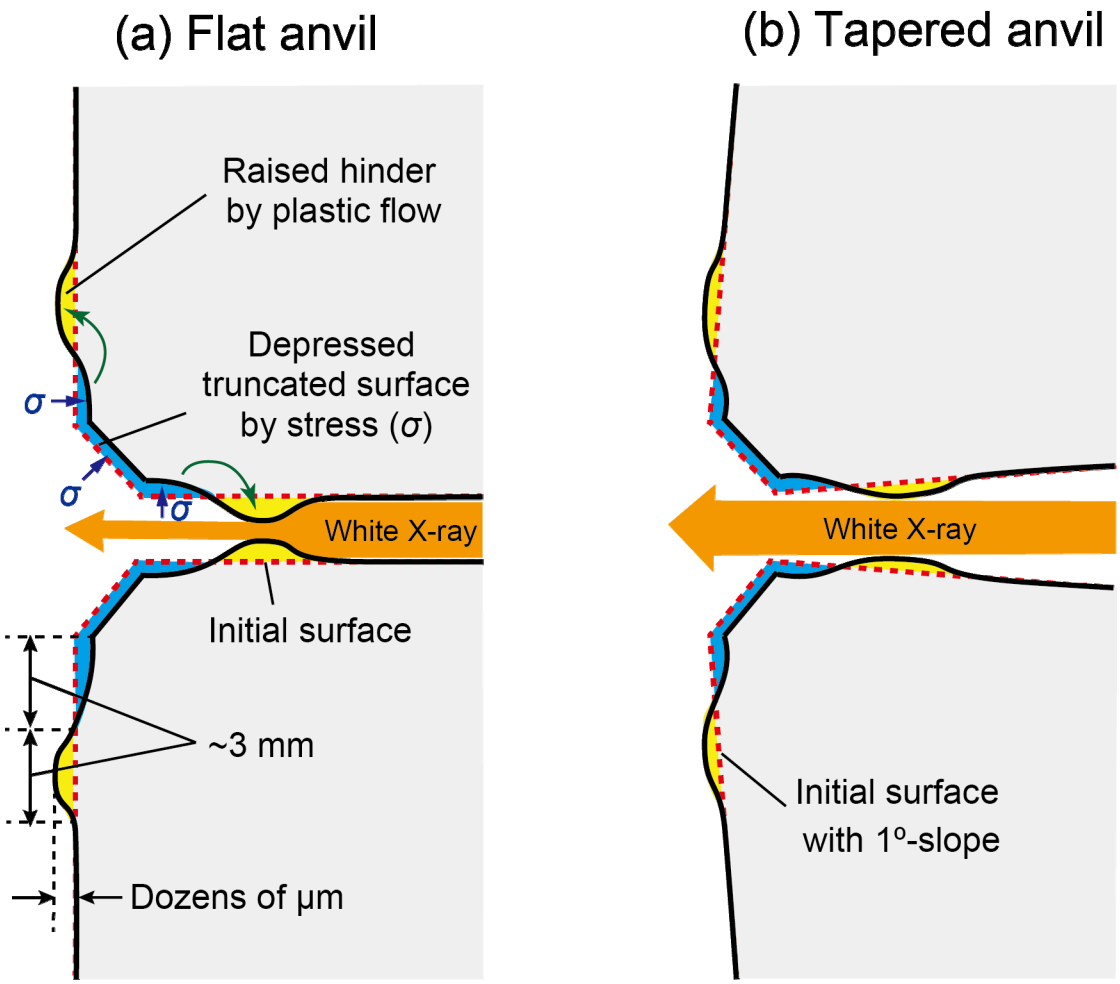

Figure 6. Deformations of (a) flat and (b) tapered anvil surfaces caused by stress from confining pressure. Dashed and solid lines represent initial and deformed surfaces, respectively. Small blue arrows indicate the simplified stress (σ) from confining pressure. The deformation of anvil top raises the hinder part by dozens of micrometers due to the plastic flow (green arrows).