Fabrication of 4GPa Class Non Magnetic High Pressure Clamp Cell

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We fabricated the piston cylinder type hybrid high-pressure clamp cell by using a home-made Ni-40Cr-3.5Al-0.005B alloy. It has been found that a maximum working pressure could be raised constantly up to 4 GPa at T = 2K without any trouble.

1. Introduction

In the field of solid state physics, there have been many investigations on the electric and magnetic properties of the transition metals and its alloys, and of the rare earth compounds. These properties are very sensitive to their electronic states which are generally controlled by external pressure, alloying, high magnetic field and so on. Among them, pressure which causes the change in the electronic state through the continuous change of volume is the most important parameter for understanding of the materials science. Thus many experimental and theoretical works are carried out as a function of pressure. Especially, the recent advances in the field of optical experiments at high-pressure are remarkable. In fact DAC makes it possible to perform many kind of measurements in very tiny samples less than 10^{-3} mm^3. Also DAC becomes one of the powerful tools for qualitative evaluations of the magnetic and electric properties at high-pressure and low temperature. However we still need large size samples more than $1mm^3$ for quantitative evaluations in the experiments under pressure of electrical resistivity, magnetic susceptibility, specific heat, NMR etc.

The development of a high-pressure apparatus with a large sample space for use at low temperature and in high magnetic field is an important subject for clarifying of the materials properties. In fact, many piston-cylinder type high-pressure clamp cells made of Cu-Be alloy are used as the apparatus which satisfies the required conditions. However the maximum pressure is at most 2GPa in the cell of Cu-Be alloy. Except for Cu-Be alloy there are a few non-magnetic materials with a high strength, which are suitable to the high-pressure apparatus at the extreme conditions of high magnetic field and low temperature.

In the present circumstances, Ni-Cr-Al (Russian alloy) and MP35N alloys are materials widely expected to achieve the breakthrough of the solid state physics as the piston-cylinder type high-pressure clamp cell. Their applications to the high pressure apparatus which successfully generated the pressure over 3GPa are introduced by Eremets[1] and Walker[2]. There is the advantage in Ni-Cr-Al alloy as the high pressure apparatus at the extreme conditions from the following reasons: The magnetic susceptibility of Ni-Cr-Al alloy is one order smaller than that of MP35N at low temperature. And the processing of Ni-Cr-Al alloy for the high-pressure cell is very easy compared with that of MP35N. However the disadvantage in the Ni-Cr-Al alloy is in the difficulty to get the alloy outside the Russian Federation. Thus we started the preparation of Ni-Cr-Al alloy and fabricated the piston cylinder type pressure clamp cell by using the home-made Ni-Cr-Al alloy. This is the report on our challenge of the fabrication of the high-pressure cell and its characterization.

2. Experimental

2-1 Preparation of alloy

The alloy was melted from weighed amounts of Ni (99.9%), Cr (99.5+%), Al (99.999%) and Ni-15.5%B alloy using an induction furnace and the melt was casted under an Ar atmosphere. The contents of the constituent were 40.0 wt% Cr, 3.5 wt% Al and 50ppmB. The balance was Ni. We modified the Russian alloy a little by the addition of about 50 ppm B in order to improve forging process. A casted ingot of about 7 kg was hot worked to rods with various diameters, 12 mm to 27mm in the forging process. Before each working, the ingot was reheated to 1200˚C. The working ratio was estimated to be 90-95% from the reduction in area which was obtained from the change in cross section before and after the hot working. After the hot working, the alloy was finally annealed at 1150˚C for 30 min in air and quenched into water. The average grain size was estimated to be 10μm after this heat-treatment. The alloy rod with this small grain size was deformed easily more than 50% at room temperature. So alloy rods with diameters less than 10mm were prepared by using a swaging machine. After the swaging, the alloy rods were also heat-treated at the same condition as described above.

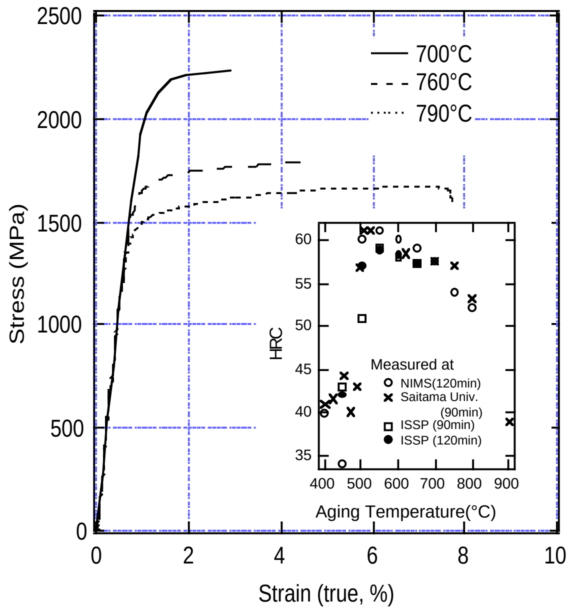

Fig.1 Stress-strain curves of Ni-Cr-Al alloy aged at 700, 760 and 790°C for 2 hours. The inset shows the change in the HRC hardness measured at the several institutes as a function of aging temperature. The aging times are given by round brackets.

2-2 Mechanical and magnetic properties of alloy

We used the heat-treated Ni-Cr-Al alloy as a starting material for the fabrication of high-pressure cell, of which tensile test and hardness measurement were performed at room temperature as a function of aging temperature, 400˚C through 900˚C. Both samples of tensile test and hardness measurement were prepared from the starting material.

A tensile test was carried out at room temperature for samples with the gauge section of 3.5mm dia. x 25 mm length after aging. Yield strength and ultimate tensile strength (UTS) of the starting materials were about 500 MPa and 800-900MPa, respectively. Figure 1 shows the stress-strain curves for samples aged at between 700˚C and 790˚C.

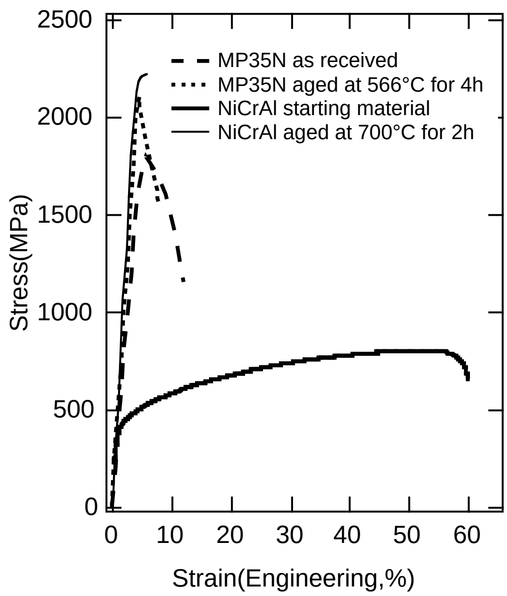

Fig.2 Comparison of stress-strain curves for MP35N and Ni-Cr-Al alloys.

The alloy aged at 700˚C for 2 hours showed the true strain of about 2%, and the UTS of over 2.2 GPa. This value was almost the same as that of aged MP35N.

A hardness measurement was performed by samples with disk shape aged in air at the various temperatures from 400˚C to 900˚C. Aging temperature dependence of hardness is given in the inset of Fig. 1. The hardness of alloys suddenly increases around 500˚C to the maximum hardness about 60 HRC, and then the hardness decreases with increasing aging temperature. The change in the strength of Ni-Cr-Al alloy is due to the precipitation of γ' (Ni_3Al) phase. Because the γ' phase dissolves into the γ (fcc Ni) matrix above 800˚C, the hardness decreases with increasing temperature above 800˚C. These mechanical property of Ni-Cr-Al alloy is similar to that shown in a previous report[3].

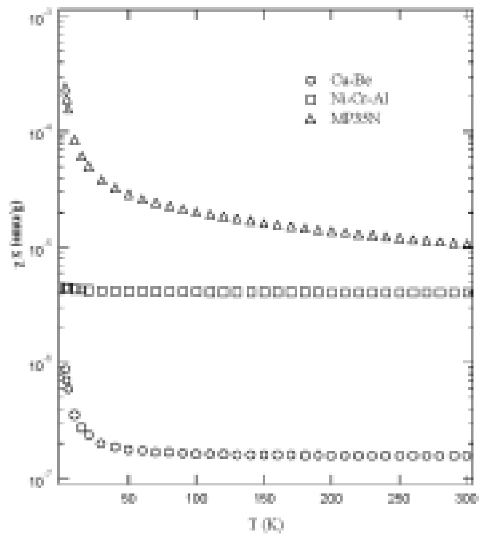

Fig.3 Temperature dependent susceptibility of MP35N alloy, Ni-Cr-Al alloy and Cu-Be alloy.

From these results, we decided the aging of 700˚C for 2 hours as the optimum condition in order to fabricate the hybrid high-pressure clamp cell consisting of Ni-Cr-Al inner cylinder and Cu-Be outer cylinder.

Here we mention the comparison of strengths of Ni-Cr-Al and MP35N alloys. Fig.2 shows their stress-strain curves. As indicated in this figure, the yield strength of the starting material of Ni-Cr-Al alloy is about one thirds of that in work-strengthened MP35N (as received). However the maximum strength reaches almost the same level in both of aged alloys, though the aging condition is different each other, for Ni-Cr-Al aged at 700˚C for 2h and for MP35N aged at 566˚C for 4h. Of course, annealed MP35N is very soft, but the strength is not increased so much by the aging process only. This is typically different to the mechanical property of Ni-Cr-Al alloy.

We measured the magnetic susceptibilities of aged Ni-Cr-Al alloy, MP35N and Cu-Be alloy using SQUID magnetometer (Quantum Design). Figure 3 shows the temperature dependence of their magnetic susceptibilities. The magnetic susceptibility of Ni-Cr-Al alloy is less than 4 x10^{-6} emu/g and no ferromagnetic ordering is observed at least down to 2 K. The absolute value is almost ten times lager than that of Cu-Be alloy, but less than one tenth of that in MP35N at low temperature. The susceptibility of MP35N have large temperature dependence, while temperature dependence of Ni-Cr-Al is not so much. Thus the Ni-Cr-Al seems to be better material for high pressure cell at high magnetic field and low temperature.

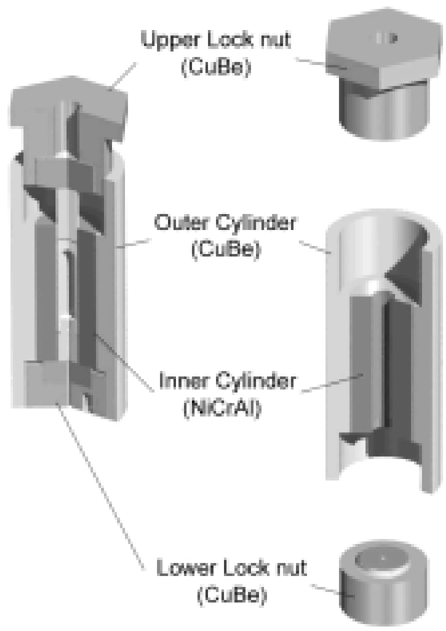

Fig.4 Scheme of the hybrid piston-cylinder type high-pressure cell.

3. Design of high pressure cell

We designed a two layer cylinder. Basic idea of the cell is similar to the former designed hybrid cells [1,2]. That is, the inside Ni-Cr-Al alloy and outside Cu-Be alloy cylinders were tapered and forced into each other by a press. A nonmagnetic WC was used as the piston. A schematic drawing of the hydrostatic cell is shown in Fig. 4. The outer diameters of the inner and outer cylinders are 15, 25 mm, respectively, and the tapered angle is about 0.2˚. We made several types of high pressure cell having different inner diameters of 4, 5 and 6 mm, whose length is 35 mm. The total length including screws for upper and lower locknuts is 63 mm.

The machining process of the hybrid clamp cell is as follows: First we make the tapered inner cylinder from the starting material of Ni-Cr-Al alloy and the tapered outer cylinder from the fully hardened Cu-Be alloy. In this case, the inner diameter of the Cu-Be outer cylinder is designed to be about 1% smaller than that of the outer diameter of the Ni-Cr-Al inner cylinder. Second the Ni-Cr-Al inner cylinder is aged at 700˚C for 2 hours. The size of the inner cylinder decreases about 0.3% after the ageing. Third both cylinders are joined together by a press. After shrink-fitting in this step, a radial stress remained at the internal wall is roughly estimated to be 1GPa. Finally the joined cylinder is machined again along the drawing, for example, thread cutting for the part of locknuts in the Cu-Be cylinder, adjustment of the inner wall of Ni-Cr-Al cylinder by polishing, and so forth.

In the case of the machining of MP35N, we must use the as-received MP35N. The hardness is very high level for machining as expected from Fig 2. Thus it is easily understood that the machining of MP35N as received is very difficult compared with that of the starting material of Ni-Cr-Al alloy. Therefore the machining cost becomes very expensive for MP35N.

As indicated in Fig.1, there is another advantage of the mechanical properties in the aged Ni-Cr-Al alloy. That is, the strength and plasticity change depending on the aging condition. Therefore it is possible to achieve the higher pressure if we can replace the Cu-Be outer cylinder to the Ni-Cr-Al one aged at around 800˚C, which shows the higher strength and suitable plasticity compared with that of the fully hardened Cu-Be alloy. Actually this project is in progressing. The details will be reported elsewhere.

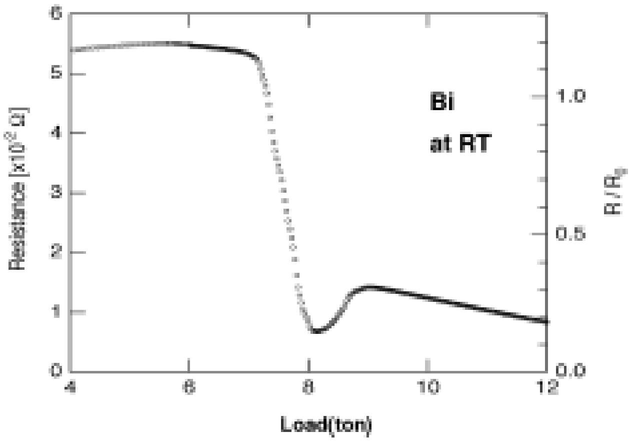

Fig. 5 The change in electrical resistance of Bi measured at room temperature as a function of the external load.

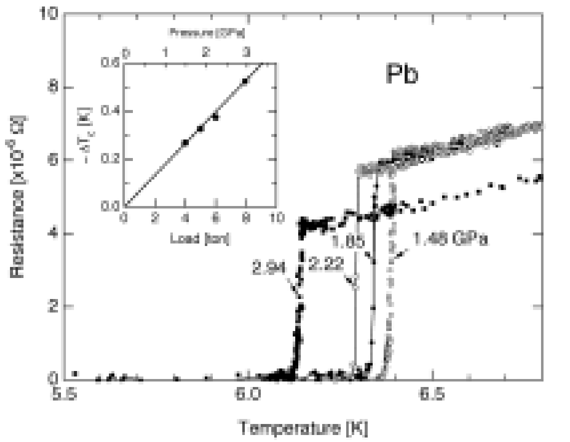

Fig.6 The electrical resistance of Pb. The generated pressures were decided from the pressure dependence of T_C. The inset shows that the efficiency of pressure generation is almost constant in this pressure region.

4. Test of pressure generation

The sample space was sealed by Teflon cell technique [4]. A mixture of Fluorinert FC70:FC77=1:1 is used as a pressure transmitting fluid. In this test we used the pressure cell with the inner diameter of 5mm. The pressure calibration at room temperature(RT) and at around liquid helium temperatures were performed from the change in electrical resistance due to the pressure induced phase transition in Bi and the change in superconducting transition temperature $T_C(P)$ in Pb which have well known pressure dependence.

Fig. 5 shows the electrical resistivity of Bi at RT as a function of external load. The generated pressure at RT was estimated from the phase transitions of $Bi_{I\text{-}II}$ and $Bi_{II\text{-}III}$. The temperature dependent resistivity of Pb is shown in Fig. 6. The generated pressure at low temperatures was estimated from the pressure dependence of superconducting transition temperature. As shown in the inset of Fig.6, we can obtain the linear relation between the external load at RT and the generated pressure at low temperature. Finally we show all of data in Fig. 7, which are used for the pressure calibration in this experimental series, as the relationship of the external load with the generated pressure. The data point determined from phase transition of $Tl_{II\text{-}III}$ is also added in this figure.

From this result, we can roughly estimate 71% as the efficiency of pressure generation. In addition, it is interesting to notice that the change of pressure in this clamp cell is very small during varying temperature, thought the influence of thermal expansion is expected to be different for every parts of the cell. That is, the external load is always clamped at RT. The pressures of phase transition for Bi and Tl are given from the phase diagrams at RT, while the pressures from T_C of Pb is, of course, determined at low temperature. However these data almost sit on a single line. At present we could not explain why the pressure change is so small for the temperature change. We must confirm this problem precisely in the whole temperature range hereafter. Also we are going toward higher pressure to know where the limit is in this system.

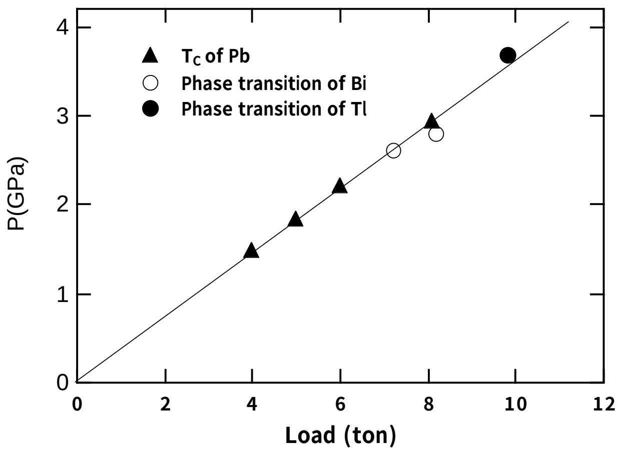

Fig.7 The relationship between the external load and the generated pressure. See text for details of the data points.

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