A NiCrAl pressure cell up to 4.6 GPa and its application to cuprate and pnictide superconductors

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Abstract. A pressurizing trial was performed for a hybrid NiCrAl hybrid pressure cell and a pressure of 4.6 GPa was achieved under a steady load of 15.0 ton, which marks the highest pressure ever known for a piston-cylinder-type pressure cell. The pressure efficiency at 15.0 ton was 75% and the expansion of the inner diameter of the NiCrAl cylinder partially reached 5%. The pressure cell was applied to nuclear-magnetic-resonance experiments on cuprate and pnictide superconductors.

1. Introduction
A piston-cylinder-type pressure cell made from nonmagnetic materials has been attracted researchers in the field of solid state physics because its large sample space is available to a variety of measurements. However, the maximum pressure is at most 1.5 GPa and 2 GPa for a CuBe pressure cell and a Ti-alloy pressure cell, respectively [1, 2]. For a hybrid WC pressure cell, which is constructed by an inner WC cylinder and an outer CuBe sleeve, the maximum pressure reaches almost 3 GPa [3]. As post-WC cylinders, NiCrAl or MP35N cylinders have been paid much attention because of their high tensile and yield strengths [4-6]. NiCrAl contains fewer impurities than MP35N, and is suitable for magnetic measurements [6]. For a hybrid NiCrAl pressure cell with an outer CuBe sleeve, we achieved a pressure of 4.0 GPa under a steady load of 15.0 ton, which marks the highest pressure at present [7]. A steady load was imposed on the pressure cell directly from a conventional press, or from a press mounted on the top flange of a temperature-variable insert for a magnet. The trials for both cases gave almost the same results with respect to the maximum pressure and the pressure efficiency. The sample space available at 4.0 GPa was 4.4 mm diameter x 6.0 mm length. The expansion of the inner diameter of the NiCrAl cylinder was less than 3% and the pressure efficiency against load was more than 75% at a load of 15.0 ton.

The questions that remain unknown are what is the limit of the load and what is the maximum pressure? Higher pressure efficiency, and higher pressure as well, would be attained if frictions on the inner surface of the NiCrAl cylinder are reduced. We performed a pressurizing trial using a short Teflon capsule as a sample holder to reduce inner-surface frictions.

2. Experimental apparatus and Pressurizing trial
Figure 1 shows an illustration of the hybrid NiCrAl pressure cell. The conditions of the present pressurizing trial are almost the same with those in the previous trials [7]. The NiCrAl pressure cell...
itself is the same with one used in the previous trials: the length of the NiCrAl cylinder is 30.0 mm, and its inner and outer diameters are 6.0- and 16.0-mm, respectively. A CuBe sleeve with an outer diameter of 40.0 mm covers the NiCrAl cylinder, as shown in figure 1. In the present trial, we newly used a short Teflon capsule instead of a Teflon tube as a sample holder. A Teflon capsule of 6 mm outer diameter and 10 mm length and a CuBe sealing ring were attached on a CuBe plug, forming a liquid container. The initial volume of the sample space was 4.4 mm diameter × 6.0 mm length. A mixture of fluorinate liquids, Fluorinert FC-70 and FC-77, was used as a pressure-mediation liquid.

![Figure 1. Illustration of a hybrid NiCrAl pressure cell.](image1)

![Figure 2. Ruby fluorescence spectra. Allows indicate the R1 transition.](image2)

To perform Ruby fluorescence measurements, ruby powder was pasted on the top of the optical fiber of 0.25-mm outer diameter and they were inserted to the sample space through a hole drilled in the CuBe plug. Figure 2 shows the spectra of the R1 and R2 transitions measured at ambient pressure and at 16.0 ton. The shift of the R1 transition is $\Delta \lambda = 1.68$ nm under a load of 15.0 ton (see arrows in figure 2): the shift corresponds to 4.6 GPa from the equation estimated by Piermarini et al. [8],

$$P \text{ (GPa)} = 2.74 \Delta \lambda \text{ (nm)}.$$

The spectra were unchanged for further load application up to 16.0 ton, implying that the maximum pressure is 4.6 GPa. The R1 line under a load of 16.0 ton in Fig. 2 shifted to lower wavelength region when the pressure cell was clamped at 16.0 ton and then the load was removed from the pressure cell. The pressure dropped to 3.9 GPa after removing the load. Figure 3 shows the inner-diameter expansion of the NiCrAl cylinder. The expansion was measured by a micro meter for holes after removing the load. As illustrated in figure 3, the length of the sample space at 4.0 GPa was about 2- to 3-mm, while that at 4.6 GPa was about 0.5- to 1-mm. Although the sample space became very short in length at 4.6 GPa, this pressure cell still has a larger volume than uniaxial anvil cells such as modified Bridgeman anvils cell. The inner-diameter expansion partially reached 4% at 4.6 GPa and the maximum load which can be imposed on this pressure cell is 15.0- to 16.0-ton: however, the NiCrAl cylinder hardly broken down during the pressurizing process up to 16.0 ton. The efficiency defined as,
is 87% at 15.0 ton. A substantial improvement was seen in this trial because the efficiency was only 75% at 15.0 ton in the previous trials [8]. The main cause is the absence of frictions between the inner surface of the NiCrAl cylinder and the CuBe sealing ring, which was inserted between a WC piston and a Teflon tube in the previous trials. The CuBe ring moves longer distance during pressurizing process and thus it experiences much more frictions than the CuBe sealing ring between the plug and Teflon tube or capsule.

3. Application of the NiCrAl pressure cell
As discussed above, the sample space of this pressure cell is substantially large even at 4.6 GPa, therefore the pressure cell is applicable to various experimental techniques. One of the applications is the technique with a single coil. The tank circuit, constructed from the coil in the pressure cell and the trimmer outside the cell, is available for nuclear magnetic resonance (NMR) and ac susceptibility measurements. The frequency detuning of the tank circuit reflects ac susceptibility, and thus NMR and the frequency detuning can be performed under the same experimental conditions. We show some experimental results performed on a spin-ladder cuprate Sr2Ca12Cu24O41 and an iron-based pnictide LaFeAsO1-xFx.

Sr2Ca12Cu24O41 is a pressure-induced superconductor and its critical pressure is 3 GPa. Therefore, the NiCrAl pressure cell is a very useful apparatus to investigate the superconductivity. The inset of figure 4 shows the detuning of the NMR tank circuit, and the bending points represent the superconducting transition temperature ($T_c$). The main panel shows the temperature ($T$) dependence of the relaxation rate $1/T_1$ at $^{63}$Cu sites [9, 10]. Humps were observed below $T_c$ at both 3.5 and 3.8 GPa, suggesting that a full superconducting gap opens below $T_c$. The spin-gap behavior at high temperatures, which exhibits an activated $T$ dependence, is followed by the $T$-linear dependence just above $T_c$, implying that holon-spinon bound state is responsible for the formation of superconductivity.
The main panel of figure 5 shows $^{75}$As-NMR spectra for non-doped and 2.6% F-doped LaFeAsO$_{1-x}$F$_x$. For the non-doped samples, the antiferromagnetic (AF) phase transition occurs at around 135-140 K, and thus the signals at 137 K come from a paramagnetic (PM) state. The central signal ($I^z = -1/2 \leftrightarrow 1/2$) at around 46-48 kOe splits into two edges due to the nuclear quadrupole interaction. As seen from the spectra at 10.0 K, the two central edges at 46-48 kOe in a PM state broadened in a wide range owing to the internal field in the AF state. As shown in the inset of figure 5, the detuning for the 2.6% F-doped samples exhibited no anomaly to 1.4 K at ambient pressure, implying that the SC volume fraction is almost zero. The $T$ dependence of the detuning at 3.0 GPa showed a bending point at about 16-17 K, reflecting the superconducting (SC) volume fraction. For the 2.6% F-doped samples, two kinds of signals were observed at both ambient pressure and 3.0 GPa, indicating that AF domains and PM domains coexist at ambient pressure, while AF domains and SC domains coexist at 3.0 GPa [11]. F doping of 2.6% corresponds to light electron doping in the $x$-$T$ phase diagram. The observed phenomena can be understood when one assumes the phase diagram with AF and SC domes separated from each other at ambient pressure: in the case that the AF and PM states coexist at ambient pressure, the AF and SC states could coexist at 3.0 GPa, if the SC dome moves to a lightly F-doped region by pressure application [11, 12].

In the present work, we demonstrated that the NiCrAl-CuBe hybrid pressure cell is effective for NMR and ac susceptibility measurements at 3 to 4 GPa. The further measurements under a pressure up to 4.6 ton would be possible in future considering the present pressurizing trial.

**Figure 4.** Relaxation rate of $1/T_1$ at $^{65}$Cu sites. The inset indicates the NMR detuning.

**Figure 5.** $^{75}$As-NMR spectra for non-doped and 2.6% F-doped samples. The inset indicates the NMR detuning for 2.6% F-doped samples.
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