Pressure dependence of superconductive transition temperature on $K_xFe_{2-y}Se_2$

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Abstract. We measured the electric resistance of $K_xFe_{2-y}Se_2$ which is said to have two SC dome. The measurement was performed at 1~14 GPa and at low temperature down to 1 K and the samples have different states of Fe-vacancy by annealing. From results of the measurements, we observed SC-like behaviour at over 11 GPa. The behaviour at higher temperature than $T_c$ indicates that there are two SC phases.

1. Introduction
Iron chalcogenides, $K_xFe_{2-y}Se_2$ receive attentions because they are high temperature superconductor (HTSC) whose transition temperature is over 30 K. It is well known that the $T_c$ decreases with applying pressure and disappear at around 10 GPa. Recently at over 11 GPa, another superconducting phase was discovered [1]. Its $T_c$ raises suddenly to 48 K and suddenly disappear at 13 GPa. The origins of these SC-phase are thought to be different [2]. In addition, the material has unique physical properties, mesoscopic phase separation. $K_xFe_{2-y}Se_2$ has two coexisting phase, one is superconducting phase and another is insulating one. [3]. The state of iron vacancies in $K_xFe_{2-y}Se_2$ as relation with this phase separation, and it can be changed by annealing and quenching [4]. The treatments also change physical property like electric resistance [5] [6] [7] [8]. $K_xFe_{2-y}Se_2$ show the hump in electric resistance [9]. The hump shifts to higher temperature region as annealing temperature is higher [5].

In this paper, we report the result of electric resistance measurement of $K_xFe_{2-y}Se_2$ under high pressure. We measure two samples, which were obtained by different heat-treatment, under high pressure to research how the states of samples can have effects on SC-phase at high pressure.

2. Experimental methods
Single-crystalline samples of $K_xFe_{2-y}Se_2$ were prepared by self-flux method using potassium [4]. The samples had different heat treatment. One was heated at 900°C for 12 hours and quenched at 550°C after the cooling down at the rate of 7 K/h (sample1). The other was also heated in same situation and cooled down to 700°C by 4 K/h, and then cooled down to room temperature by shut down the
furnace. Moreover this sample was annealed at 400°C and quenched at same temperature (sample2). Both samples made from $K_{0.8}Fe_2Se_2$, and precise compound of sample1 is measured as $K_{0.77}Fe_{1.68}Se_2$. We didn’t observed about sample2 precisely but it has almost same compound with sample1.

A non-magnetic diamond-anvil cell (DAC) was used to generate high pressure. The pressure was determined by the ruby fluorescence at room temperature. The samples were cleaved and put in the hole of the gasket with ruby as pressure marker and NaCl as a pressure medium. The diamonds 500 μm in diameter were used. Platinum and gold were used to make electrodes and the gold ones were placed in contact with the sample. The electrical resistance was measured with cooling down to 1 K. Between the stainless gasket and electrodes were insulated by cubic boron nitride (c-BN) powder mixed with epoxy resin. Four-probe method was used to measure the resistance but the results have extra resistance.

3. Result and Discussion

Figure 1 shows the temperature dependence of electric resistance of sample1 at various pressure. At 5.3 GPa, the minimum pressure in this run, the result shows positive slope and superconductive behavior. In other study, $K_xFe_{2-y}Se_2$ shows insulator region and metallic region, and $T$-dependence of $R$ shows a hump around 200 K at 5 GPa [9], [10]. In some study like reference [3], the quenching temperature have effects on the hump. The hung appears at higher temperature as the quenching temperature is higher. In our sample, the hump cannot be seen clearly. It seems that the hump went up to 300 K.

![Figure 1. Temperature dependence of electric resistance of sample1.](image)

As applying pressure, the value of resistance decreased, and $T$-dependence of $R$ was changed. The hump is seems to shift to higher $T$ with pressure. Over 10 GPa, the dependence became metallic. Similar behavior was observed in the study of [9], [10].
The result of Sample2 is shown in Figure 2. $T$-dependence of $R$ shows hump at 163 K and 2.7 GPa differently from Sample1. However the superconductive behavior can be observed. As applying pressure, the hump shifted to lower temperature and remained at 9.5 GPa. At 11.1 GPa, over 10 GPa, $T$-dependence of $R$ completely changed to metallic. These result indicated that the quenching temperature doesn’t have any effect on the metal transition at 10 GPa.

![Figure 2. Pressure dependence of electric resistance of sample2.](image)

$T$-dependence of $R$ at low temperature region is shown in Figure 3. In both Sample1 and Sample2, superconductivity could be observed. The superconductive region shift to low temperature as applying pressure. This behavior was observed other study [1], [9], [10], and in these study, superconductivity was disappear at 10 GPa. However in this study, the resistance decrease slightly at high pressure (inset of Figure 3). This slight decreasing can be seen over 10 GPa in both samples. This behavior has current dependence (Figure 4). From results of Figure 4, critical current is estimated as 5 A/cm$^2$. So this slight decreasing seems to come from superconductivity. In this region, resistance shows metallic

![Figure 3. Pressure dependence of electric resistance at low temperature region.](image)
behavior. It indicated that the origin of high pressure superconductivity is different from low pressure superconductivity.

We defined onset critical temperature of superconductivity $T_c$ by intersection of two line drown through linear region above transition and steep slope of the curve [1]. Pressure dependence of $T_c$ comparing to previous study is shown in Figure 5. Under 10 GPa region, there is no difference between two samples about $T_c$. In this study, $T_c$ didn’t go down to 0 K, and remained under 10 K differently from high temperature superconducting phase at high pressure region. It is indicated that differences of quenching temperature or $T$-dependence of $R$ don’t have an effect on SC-phase under 10 GPa, but that quenching temperature have some effect on SC-phase at high pressure.

![Figure 4. Temperature dependence of electric resistance of sample2 at 11 GPa and various current.](image)

![Figure 5. Pressure dependence of the $T_c$ comparing the result of this study to previous study.](image)
4. Conclusion
We measured electric resistance of two samples of K$_x$Fe$_{2-y}$Se$_2$ which were obtained by two type of heat treatment and their main difference is quenching temperature. Sample1 quenched at 550°C shows near metal behaviour in $T$-dependence of $R$, and Sample2 quenched at 400°C shows hump-behaviour. In spite of this difference, as applying pressure, $T_c$ of both two samples decreased and the samples became metal at 10 GPa. However over 10 GPa, some transition was observed under 10 K, instead of high temperature superconductivity at 48 K. This phenomena seems to be superconductivity whose origin is different from SC-phase at low pressure because of the metal behaviour over the transition temperature. We can get the possibility of SC-phase of iron selenide at over 10 GPa. We expected the $T_c$ increased at higher pressure. Changing the amount of Fe and K or changing process of heat-treatment may also induce HTSC-phase.

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