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Temperature Dependence of Electrical Resistivity in Ternary Graphite Intercalation Compounds

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The in-plane electrical resistivity of non-exfoliated stage 2 and stage 5 K-FeCl₃ graphite intercalated compounds was measured at a wide range of temperatures by using the standard four probe technique. The data of stage 5 exhibited a conductive behavior which is in contrast to the same stage of an exfoliated sample. The electrical resistivity exhibits a metallic behavior similar to the same stages of binary compound. The data are well fitted by a quadratic T⁻² dependence.

1. Introduction

Most of the reported work in this field has been devoted to the binary GIC's, whereas ternary compounds had little attention. Ternary GIC's result from intercalating two distinct intercalant species in the host graphite galleries such as the donor-donor type K-Cs-GIC's [1] and the acceptor-acceptor type BiCl₃-ReCl₄, CoCl₂-FeCl₃, TiCl₃-TlBr₃ GIC's [2]. In this work the in-plane conductivity of the ternary compound K-FeCl₃ was measured at temperatures between 10⁰K and room temperature and the data were compared with the results reported for the binary compound FeCl₃ GIC's [3].

2. Experimental

The ternary GIC's were prepared from a successive intercalation process using highly oriented pyrolytic graphite. The standard four-probe technique was used to measure the change of the sample's voltages as the temperature was cooling from room to about 10⁰K by using a closed cycle refrigerator system. Details of the experimental work can be found in ref.[4].

3. Results and Discussion

The data shown on figure 1 are the temperature dependence of the in-plane electrical resistivity ρ normalized to the room temperature value ρ/ρ_{300K} of several stages of FeCl₃ - GIC's. In this work we are only interested in the results of stages 2 and 5 for which the ternary compounds were prepared. As shown in the figures the

resistivity exhibits a metallic behavior. The temperature dependence of resistivity in these samples indicates that there are two regions of interest; the first region extends from 300 K to about 130 K, while the second region starts below 130 K and continues down to a temperature of about 10⁰ K.

The resistivity of intercalated binary compounds, as shown in Fig. (1), exhibits the weak temperature dependence typical of acceptor compounds [5]. At high temperature, the dependence is linear indicating that an acoustical phonon mechanism may be responsible for the resistivity. At lower temperature, below approximately 130K, there is a transition regime where the behavior is less clearly defined. Finally, the resistivity begins to saturate at temperature between 40 K and 50 K. Stage 2 shows the change in slope at around 200K, this was previously noted by Pendry et al [6] for low stage number acceptor compounds. They explain this as due to a decoupling of the intercalant atoms from the carbon lattice resulting in a corresponding decrease in phonon mass.

In our previous work [4] on stages 2 and 5 ternary compounds (K-FeCl₃-GIC's), it was found that exfoliated samples of stage 2 are electrical conductors whereas stage 5 samples are electrical insulators. The new results in this work, besides the studies of the resistivity - temperature dependence and the fits of the data to different physical models, is that stage 5 non-exfoliated sample is an electrical conductor which is in contrast to the exfoliated stage 5. It

seems that although the intercalation process controls the electrical conduction in these compounds, the exfoliation mechanism preserves similar magnetic behavior, but prevents the transfer of charge carriers along the plane.

The temperature dependence of the in-plane resistivity data of stages 2 and 5 ternary compounds are shown in Fig.(2). It is clear that the rate of decrease of resistivity of stage 2 with decreasing temperature is faster than that of stage 5. The data exhibit the same metallic behavior, but at different strength, which was observed in the binary compounds. It seems that this behavior is a characteristic of the ternary compounds, Mareche et al [7] and McRae et al [8] found that ρ_a of various ternary - GIC's exhibit a metallic behavior throughout the measured temperature range. The in-plane resistivity data in this work were fitted to the quadratic equation:

$$\rho(T)=A+BT+CT^2$$

The results of the fits are shown by the continuous lines in Fig. (2), as shown in the figure the data fit well to the above model. In the high temperature region, the resistivity is linear with temperature, which is consistent with electron-phonon scattering mechanism. At lower temperatures, the temperature dependence of the resistivity is quadratic, which describes a carrier-carrier scattering mechanism [8]. This effect has a large temperature range.

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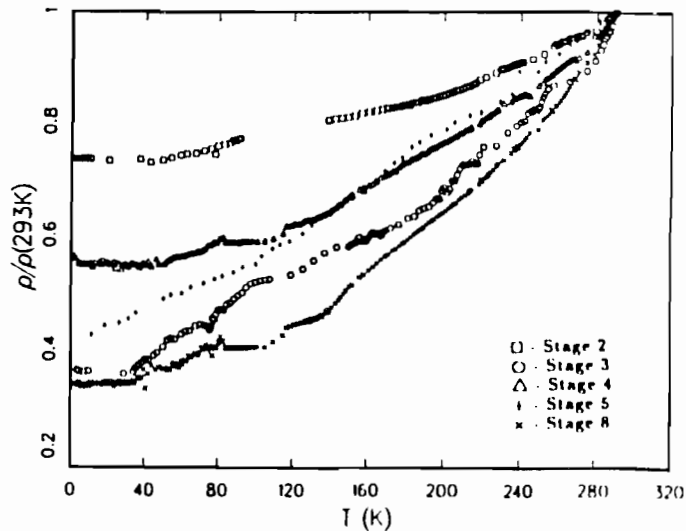


Fig. 1 -Normalized resistivity as a function of temperature for stages 2,3,4,5 and 8 FeCl₃ GIC

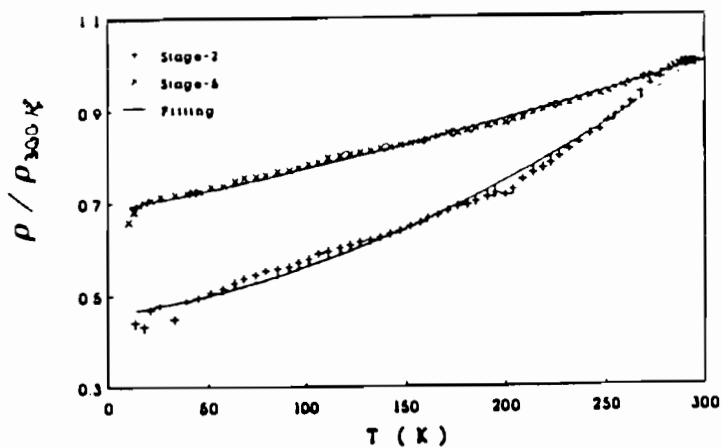


Fig. 2 -Temperature dependence of the in-plane resistivity ratio of stage 2 and stage 5 K-FeCl₃ GIC. Experimental data and the fits.