Transactions of the Materials Research Society of Japan. Vol. 20 1996 MRS-J. All rights reserved.

# Percolation Study in Evaporated Film of C60 Cluster

Y. Shionoiri, Y. Xu and Y. Ochiai

Dept. Materials Science, Chiba University, 1-33, Yayoi-cho, Inage-ku, Chiba, Chiba263, Japan

We have studied a critical transport phenomena in C60 evaporated films by means of the percolation analysis. It is related to a certain metal insulator transition in low dimensional system. In order to clarify the difference between a bulk or an inter-molecular transport in monolayer and surface transport of the small cluster of C60, we have measured in-situ electrical resistance of the film. Also, the cluster size in the film and the critical resistance have been analysed based on the percolation behaviors.

## **1.INTRODUCTION**

Recently, it has been interested the superconducting properties in doped C60<sup>1</sup>) and related electrical properties.<sup>2,3)</sup> We have studied a critical transport phenomena in C60 evaporated films. Especially, it is related to insulator transition а metal in low dimensional system. As for these interestings, it is nessesary to clarify their differences between an inter-molecular transport in monolayer and a bulk or surface transport of the small cluster of C60. Another interest is on a transport difference between C60 monolayer and cluster films. Such a analysis has been related to the morphological problems. Therefore, determination of the percolation threshold is very important to investigate such a difference in transport mechanisms between in the thin film of mono-layer molecules and in clusters of C60.

In this study, we discuss critical percolation behaviors in thin layer of C60 cluster and the estimation of the cluster size and the critical resistance of the percolation in electrical transport. The percolation behavior has been examined by means of computer simulation.

### 2.EXPERIMENTS

Our C60 cluster films were prepared by using a conventional evaporation method from high purity C60 powder. Used C60 powder was separated from C60 rich shoot. Such purification were performed by column chromatography technique and the final purity of our C60 powder was 99.9 percent or more.

The film was prepared by a conventional evaporation technique using a tungsten baskettype heater. The evaporation rate was controlled by varying of the heater power. The thickness of the film was monitored by a quartz oscillating method. In a vacuum pressure of  $10^{-6}$  torr or low, the evaporation of C60 was performed at room temperature on a thin guartz substrate on which two or four thin Gold electrodes previously were deposited in order to use for electrical measurements.<sup>3)</sup> The separation of the electrode was ranging from a few mm to 0.1 mm. At the same time in-situ electrical measurements were performed. Electrical resistances of the film during the evaporation were measured by means of the two terminal method using an electrometer or a source measure unit.

And after in-situ measurements, we have analyzed the surface morphologies of the film using SEM observation. The characteristics of

 $p_{dep}$  (10<sup>-6</sup>torr)  $\rho(10^7 \Omega \, \text{cm})^{**}$  $r_a(\mu m)^*$ P(%) sample  $d(\mu m)$ 3 6003 0.11 0.10 83 6.4 10 6005 0.01 8.0 7.4 1.2 6006 0.04

Table 1 Sample characteristics

\* Radius is estimated from the SEM observation.

\*\* Resistivity is defined at the transition point as shown in Fig.1.

three films measured are shown in Table 1.

# 3.RESULTS AND DISCUSSIONS

The result of an in-situ resistance measurement for the sample of 6003 is plotted in Fig. 1. We consider that the transition from the insulating to the metallic phase seems not to be so sharp as observed at low temperature because there exists a leakage current path on the substrate, the lead-through and/or others. The arrow indicated in the figure shows the metal insulator transition point. The higher resistance region at the left side of the dependence is probably attributed to such leakages that result from another nonmetallic conduction process or thermal smearing effect. Although we can not see a clear metallic dependence in Fig.1 since we stop the measurement at just appearing of metallic behavior indicated by the arrow in the case of 6003, the resistance after metallic transition in other two samples of 6005 and 6006 shows an almost constant or a weak decrease as the thickness increases. The critical thickness, d, at the transition is estimated and the critical resistivity value are also determined as listed in Table 1.

Here, the disorder structure of the conducting path of the film is modeled by random percolation networks in two dimensions. Therefore, we have used Monte-Carlo simulations for the percolation path in C60 evaporation film. The unit of the percolation path in the simulation was postulated to be a small square in our computer program. The critical percolation can be indicated by the ratio of the conducting area to the whole one. The ratio is also considered to be a certain percentage of the such conducting area to the total and to depend on the total number of the unit, N, in the whole area of the film. We can calculate the percentage of the critical percolation, P, for this case

$$P(\%) = 3/4(d/r_a)$$
 (1)

where d is the critical thickness of the thin film, r, is the average radius of the clusters of Here, we can deduce the the thin film. relation (1) as followings. N, which is also the number of the particles of the thin film can be calculated by the volume, V, of the film reagion between two electrodes and the one particle volume,  $(4/3)\pi r_a^3$  And V can be also calculated by the area, s, between two electrodes and d, where V=sd, then we can obtain the relation N=sd/( $(4/3)\pi r_3^3$ ). Since  $P=\sigma/s$  where  $\sigma$  is the total area of the number of the particles, we get  $\sigma = N\pi r_a^2$  then finally obtain the relation (1). The N dependence of computer simulation is shown in Fig. 2 and eventually has a constant value of 0.73.



Fig.1 Resistance vs thickness for the thin film of 6003. The critical thickness is determined at 1100A and is pointed by the arrow.



Fig.2 Percentage vs number of total units for the computer simulation of the percolation. The percentage has an almost constant value about 73 when the number is larger than 100.

If we apply the above 2-dimensional percolation model using a simple square unit for our experimantal result, the percolation transport can be discussed by use of the critical depth and resistance. We can also analysis the visible cluster size from the photograph of SEM observations. Such a distribution of the cluster of C60 molecule just after percolation transition is plotted in Fig.3 for the film of 6003. It is found that the measured percentage, 83%, based on the relation (1) almost agrees with the simulation value, 73%. Therefore, although we have no data on r<sub>a</sub> except for 6003, we can estimate the  $r_a$  value for other two samples using the percentage defined by the relation (1). If those are obtained, the r<sub>a</sub> must depend on the vaccum pressure. And, we can expect that the size of flying clusters during evaporation strongly



Fig.3 The distribution of the radius of the cluster for the thin film of 6003

depends on the vaccum pressure, p<sub>dep</sub>.

Next, we discuss the critical resistance at the transition previouly mentioned at Fig.1. From the result of Table 1, these resistivities for three films almost depend reversely on the cluster size and have higher values than single crystal value<sup>2,3</sup>. Considering this result, the electrical transport in C60 cluster is probably expected to come from a surface conduction of the cluster.

### **4.CONCLUSION**

We have observed the percolation properties using in-situ electrical transport measurements of the C60 cluster films. It is found that the critical behavior gives an estimation of the cluster size of the C60 monolayer film. Also, there exists a clear dependence of the evaporation pressure for the cluster size of C60.

#### REFERENCES

1. Y.Maruyama, T.Inabe, H.Ogata, Y.Achiba, K.Suzuki, K.Kikuchi and I.Ikemoto, Chemical Lett.,(1991) 1894.

2. T.Arai, Y.Murakami, H.Suematsu, K.Kikuchi, Y.Achiba, and I.Ikemoto, Solid State Commun., 84(1992)824.

3. Y.Ochiai, K.Yamamoto, H.Yamasaki, Y.Shionoiri, H.Ogata and Y.Maruyama, Fullerene Sci. Techn., 3(1995)79.