EFFECT OF PARTICLE SIZE ON THE SYNTHESIS OF COPPER CHLORIDE-GRAPHITE INTERCALATION COMPOUNDS

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Introduction

Graphite intercalation compounds (GICs) of copper chloride and other metal chlorides have received great attention because they possess relatively high electrical conductivity and are rather stable in air. Inagaki and Wang [1] reported an effect of particle size on the intercalation. They found that the amount of the coexisting graphite in the $CuCl_2$-GICs increased, very remarkably in the graphite with small particle size but not appreciably in that with larger particle size. In this study, the effect of particle size on the amount of intercalation was investigated. the copper concentration in GICs was measured quantitatively using EPMA.

Experimental

Graphite powders with different particle size were used as host materials. $CuCl_2$-GICs were synthesized using a gas phase reaction. For the quantitative measurement of intercalant amount inside the graphite host, EPMA was used. An electron beam with a diameter of 200 μm was used in the analysis.

Results and Discussion

Fig.1 shows the XRD patterns of $CuCl_2$-GICs prepared at different temperatures for 48 hrs. The average particle size of the graphite powders is 15.2 μm. At low intercalation temperature (250°C), no stage structure was found (Fig.2(b)). When the reaction temperature was raised to 360°C, weak and broad peaks of stage 2 structure appeared (Fig. 2 (c)). A strong and sharp (002) peak of graphite can be found, indicating that only a small portion of the graphite particles forms GICs. Both the stage 1 and stage 2 structures were found at the intercalation temperature of 420℃ (Fig. 2(d)). However, the peak intensity of stage 1 structure is much stronger than that of stage 2. The XRD pattern for $CuCl_2$-GICs prepared at 480℃ was similar to that at 420℃. Only stage 2 structure was observed at temperature of 540℃.

Fig.2 shows the copper concentration in the GICs as a function of intercalation temperature. The copper concentration increases with rising temperature up to 420 ℃, then the concentration remains relatively constant. Rüdorff [2] explained the relatively high stability of $FeCl_3$-graphite flakes based on the assumption that the intercalant near the edges of the flakes deintercalates and the empty galleries collapse in the periphery. The intercalated flakes were then sealed by themselves and the interior was protected from a further attack by water molecules. In this study, the copper concentration was measured after the sample was washed in water and dried in the oven. The intercalant at the surface of GIC particle can be washed out easily. As a result, the concentration measured represents the remaining stable concentration after washing. Since the graphite powders have only a mean particle size of 15μm, they can be fully intercalated easily. On the other hand, since the particle is small, the portion of intercalants which will be washed out is more appreciably than that for larger particles. Consequently, despite the fact that the amount of intercalation at the higher temperature should be larger, the intercalants tend to be washed out and a relatively constant amount of intercalants remains.

Fig.3 shows the XRD patterns of $CuCl_2$-GICs prepared at 480°C for 48 hrs using graphite powders with different particle size. EPMA measurements of copper concentration in $CuCl_2$-GICs (Fig.4) correlate well with results of x-ray diffraction. Higher copper concentration was found for $CuCl_2$-GIC samples using graphite powders of 75 and 150μm when compared with graphite powders of 15μm. This result is reasonable since the portion of intercalants which will be washed out is less for bigger particles. However, the copper concentration decreased as the particle size increased to 180 μm. This is because that the intercalants could not be intercalated into the center portion of the particles as the particles become larger under present processing conditions.

Conclusions

Particle size of graphite host has a significant effect on the amount of intercalation. The degree of influence is related to the processing conditions.

References

1. Inagaki M and Wang ZD. Synthesis of cupric chloride-graphite intercalation compounds by the molten salt method. Synth Met 1987;20:1-8.
2. Walter J. The decomposition of $TaCl_5$-graphite flakes by long-time exposure in water and air. Synth Met 1997;89:39-45.

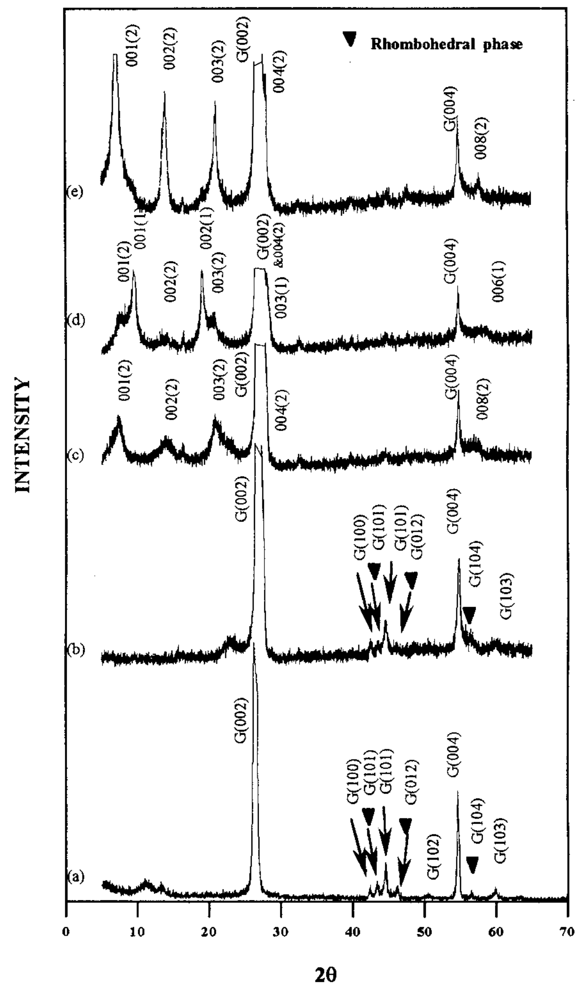

Fig. 1 XRD patterns of $CuCl_2$-GICs prepared at different temperatures for 48 hours : (a) before intercalation, (b) 250°C, (c) 360°C, (d) 420°C, and (e) 540°C.

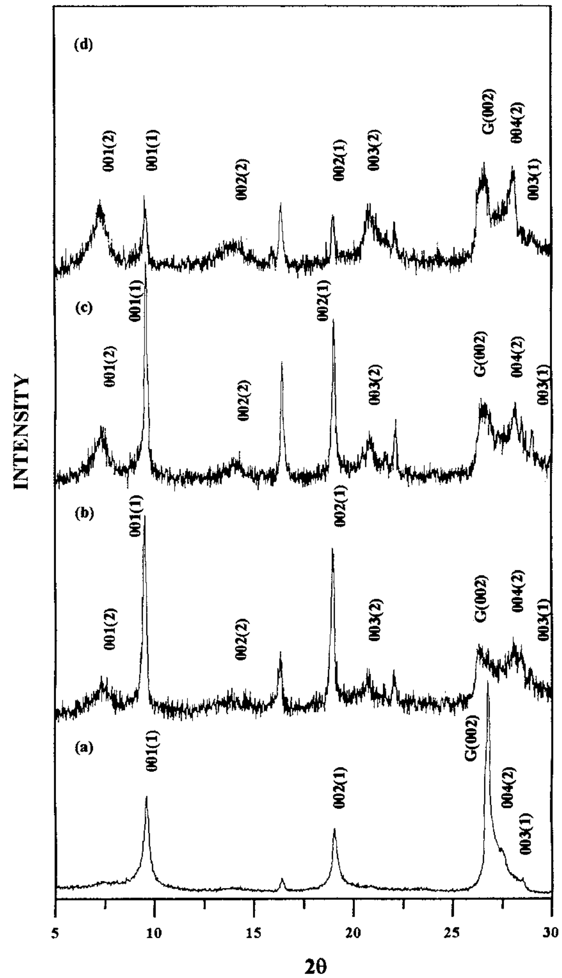

Fig. 3 XRD patterns of $CuCl_2$-GICs prepared at 480°C for 48 hours using graphite powders with different particle size : (a) 15.2 μm , (b) 75 μm, (c) 150 μm, and (d) 180μm

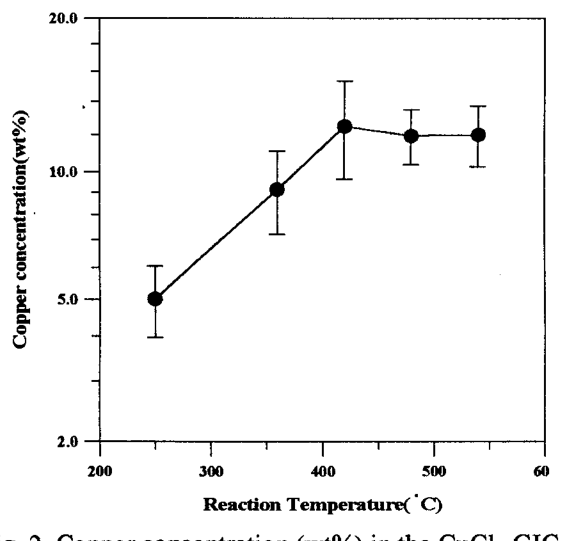

Fig. 2 Copper concentration (wt%) in the $CuCl_2$-GICs as a function of reaction temperature.

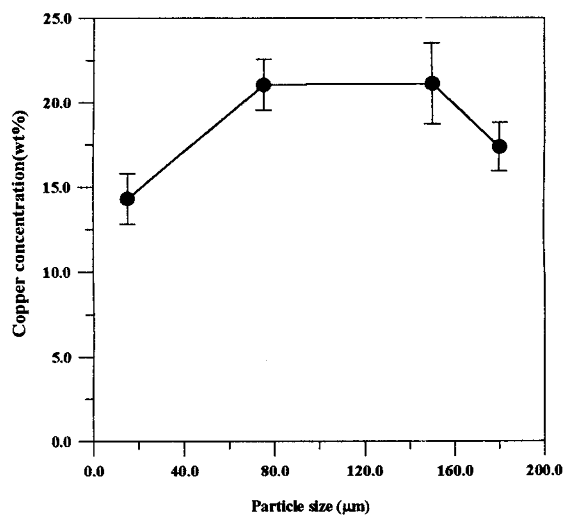

Fig. 4 Copper concentration (wt%) in the $CuCl_2$-GICs as a function of particle size.