POSTER

A Reaction of Li-Graphite Intercalation Compounds under Gas Atmospheres and Their Characteristic Analysis

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1. Introduction

In putting the lithium secondary battery with high density and high output to the practical use the reversibility between the charge and discharge of the lithium intercalation compound anode is an increasingly important factor for battery mechanism[1]. Generally there are electrochemical and chemical methods to synthesize the Li-GICs. The chemical methods can be classified in two processes : Chemical Vapour Deposition and add-pressure method [2, 3]. In lithium intercalation compounds obtained by such methods, the reversibility between the intercalation and deintercalation process is an important factor for the battery mechanism.

In this research, we have synthesized the Li-GICs by the chemical method using temperature and pressure, and studied the effect of O_2 and N_2 on the deintercalation process during which such synthesized compounds decompose spontaneously. The properties of each decomposed compound obtained during such reaction processes were investigated using the X-ray diffraction method, UV/VIS spectrophotometric method and BET specific surface area measurement.

2. Experimental

Li-GICs were synthesized by the chemical method under temperature and pressure with lithium metal and natural graphite.

In order to know the reactivity of Li-GICs under the atmosphere of O_2 and N_2, the Li-GICs were made as minimal size as possible, and these compounds were oxidized under the condition of the ratio 2 : 8 of O_2 and N_2 for the slow reaction.

For studying the structural change of Li-GDICs during the deintercalation of lithium ion from the interlayer spaces of the graphite, the X-ray diffraction analysis was made with the Debye-Scherrer camera. In order to know the electron energy state of Li-GDICs, the UV/VIS spectrophotometer of PU 8700 Series(UNICAM, PU 8710/01, FALCAN-SCAN) was used with the diffuse reflectance accessary. The measurement was performed between the region 1.37-4.96 eV(Wavelength : 900-250 nm).

After calibration of the BET analyser, the BET specific surface area was measure.

3. Results and Discussion

The structural change which occurred during the decomposition process of the graphite intercalation compounds was investigated by the X-ray diffraction method and the results are shown in Figure 1. Figure 1 suggest that pure stage 1 graphite intercalation compounds were obtained initially. However, after contact with the gas for 1 hour, the pure stage 1 compound changed to a mixed stage 1 and 2 compound. Results obtained after 1 week showed almost the same features as that obtained after 1 hour.

According to the X-ray reflection analysis of the compound after two weeks, the peaks for stage 2 had reduced while those for stage 5 began to appear and most peaks for the original graphite appeared. After 3 weeks, it was observed that more peaks for stage 2 had reduced, while those for stage 5 had increased, and that the additional (G112) peaks for the original graphite appeared. According to the X-ray analysis for the compounds obtained after 4 weeks, the peaks for stage 1 disappeared completly, (3s007) peaks for stage 3 emerged and those for stage 5 became dominante. After 5 weeks, the same features as those after 4 weeks were observed. After 6 weeks, only the peaks for stage 5 were observed and all others had disappeared. After more than 6 weeks the features of both graphite peaks and intercalation peaks remained

the same as in X-ray diffraction peaks observed at 6 weeks. This final X-ray diffraction data includes stage 5 peaks and some, but not all, of the peaks for the original graphite. Therefore, we believe some Li-ions could not decompose from the interlayer. They remain in the space between the graphite layers and from the Li-residue compound.

The analysis results for the energy state of the Li-GDICs measured by the UV/VIS spectrophotometer are shown in Figure 2. According to Figure 2, there seems to be a relation between the energy state and the concentration of the intercalants : the clearer spectrum is observed and the R_{min} values are formed in the higher energy region when the concentration of the intercalants is high.

BET specific surface area values of Li-GDICs measured with the BET surface area analyzer(4200 Series, Beta Scientific Corporation) are shown in Figure 3. This indicates that the BET values for the compounds obtained between 1 and 3 weeks are relative similar to each other. For the compounds obtained from 4 weeks to 5 weeks, however, those BET values tend to increase, and the compounds from after 5 weeks show almost the constant BET values, with minor differences. From these facts we belive that the deintercalation reaction ended after 5 weeks.

4. Conclusion

From studying the structural change of Li-GDICs during the deintercalation of lithium ion with the X-ray diffraction method, we believe that some Li-ions which could not decompose from the interlayer still exisit between the graphite layers. From UV/VIS results, it can be seen that the clearer spectrum is observed and the R_{min} values are formed in the higher energy region with high concentration of the intercalants. From BET results, we can also believe that the deintercalation reaction ended after 5 weeks.

References

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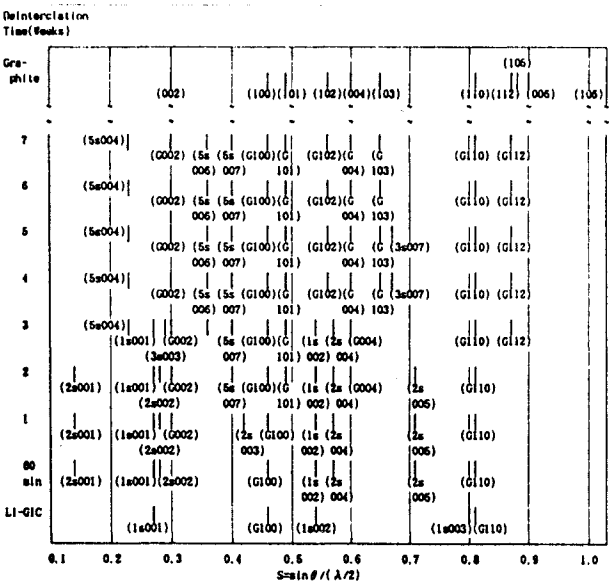

Figure 1. Development of (hkl) interference for the Lithium-Graphite Deintercalation Compounds. e 1s : stage 1, 2s : stage 2, 3s : stage 3, 4s : stage 4, 5s : stage 5, 6s : stage 6, 7s : stage 7, G : graphite

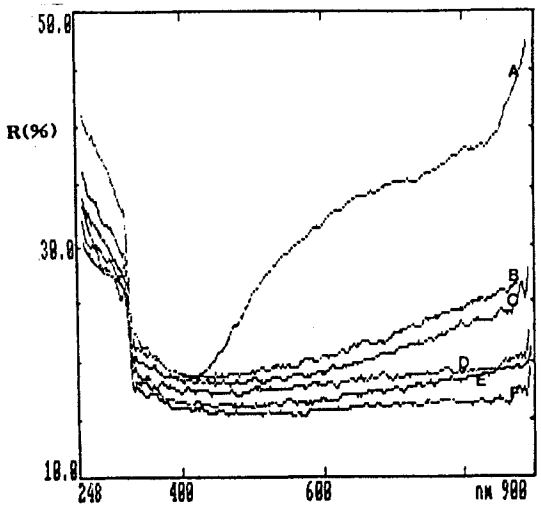

Figure 2. Evolution of UV/VIS reflectance spectra between 248-900nm for Li-GDICs ; A→Li-GDIC, B→Li-GDIC after 1 hour, C→Li-GDIC after 1st week, D→Li-GDIC after 2nd week, E→Li-GDIC after 3rd week, F→Li-GDIC after 4th week.

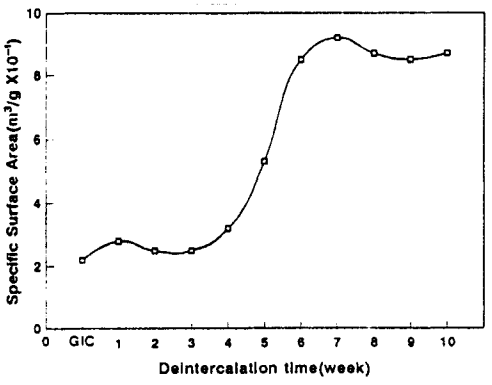

Figure 3. Specific Surface Area with Deintercalation time.