Investigations of the Exothermic Reactions of Natural Graphite Anode for Li-ion Batteries during Thermal Runaway

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Lithium ion rechargeable batteries possess high energy density and excellent power compared to other commercialized battery systems, which give them a significant market as power sources for portable electronics such as cellular phones, laptop computers, etc. However, their applications to electric vehicles (EV) and hybrid vehicles (HV) have been limited because these applications need better thermal stability to reduce the safety risks.[1-2]

It has been well documented that differential scanning calorimeter (DSC) and accelerating rate calorimeter (ARC) have been used to investigate the thermal stability of salts, electrolytes, cathodes, anodes, conductive materials, and binder additives.[3-10] The reactions of these individual components have also been correlated with the thermal runaway of full-cells.[3,4] In the DSC studies[3] of fully charged electrode materials ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$/natural graphite Mag-10, wt%: 64:36), more than 95% of the heat is generated by the anode from 80°C to 180°C; this heat is attributed to the decomposition of solid electrolyte interface (SEI) and the formation of a secondary SEI film. About 74% of the heat is generated by cathode from 180°C to 250°C; this heat is produced by the decomposition of the delithiated cathode and the combustion of the electrolyte with liberated O_2. Hence, the mechanisms of thermal runaway reactions for anode materials are useful in understanding the thermal behavior of Li-ion cells; this understanding can lead to the development of new materials with improved safety and stability.

Thermal behavior of fully or partial intercalated natural graphite Mag-10 was studied with DSC. Figure 1 shows the DSC curves of Mag-10 with different amount of intercalated Li. More intercalated lithium results in larger heat rate and more peaks, indicating that the graphite anode has the higher potential to undergo thermal runaway with a higher state of charge. This is the first time it has been noted that graphite loses its structure with more than 0.7 intercalated lithium when electrolyte is present at high temperature by detecting the existence of (*002*) graphite X-ray diffraction peak. A possible reaction mechanism for graphite thermal runaway will be presented and discussed in detail.

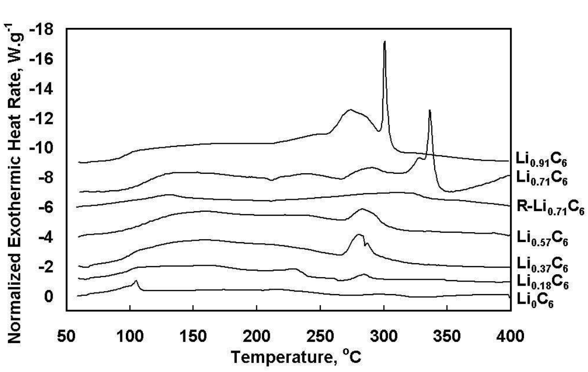

Fig. 1. DSC curves of Mag-10 samples with different amounts of intercalated lithium at a scan rate of 10°C.min^{-1}. R-$Li_{0.71}C_6$ represents a graphite anode rinsed with DMC and vacuum dried to remove the remaining salt and EC.

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