Modeling and simulation of heat of mixing in lithium ion batteries

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Heat generation is a major safety concern in the design and development of lithium ion batteries (LIBs) for large scale applications, such as electric vehicles. The total heat generation in LIBs includes entropic heat, enthalpy, reaction heat, and heat of mixing (1-3). The heat of mixing will be released during relaxation of Li ion concentration gradient. For instance, after the drivers turn off their vehicles, the generation of entropy, enthalpy and reaction heat in LIBs will stop, but the heat of mixing is still being generated. Thomas and Newman derived methods to compute heat of mixing in LIB cells and investigated the heat of mixing on a Li|LiPF6 in ethylene carbonate:dimethyl carbonate|$LiAl_{0.2}Mn_{1.8}O_{4-\delta}F_{0.2}$ cell (4). The objective of this study is to investigate the influence of heat of mixing on the LIBs with different materials, porosities, particle sizes, and charge/discharge rate and to understand whether it is necessary to consider heat of mixing during the design and development of LIBs.

In this study, a mathematical model was built to simulate heat generation of LIBs using COMSOL Multiphysics. The LIB model was based on Newman's model. $LiCoO_2$ was applied as the cathode materials, and LiC_6 was applied as the anode material. The results of heat of mixing were compared with the other heat sources to investigate the weight of heat of mixing in the total heat generation. Table 1 shows the heat of mixing, irreversible heat, and reversible heat in anode and cathode electrodes at 5 min during a 2 C discharge process. As shown in Table 1, the heat of mixing in cathode is smaller than the heat of mixing in anode, mainly due to the lower Li ion diffusivity and larger particle size of LiC_6. The heat of mixing is not as much as the irreversible heat and reversible heat, but it cannot be neglected for this operating condition. The heat of mixing in different LIB cells and under different operating conditions will be reported.

The mathematical model:
Mathematical model equations:

$$Q = I(V - U^{avg}) + IT\frac{\partial U^{avg}}{\partial T} + \sum_k \Delta H_k^{avg} r_k + \int \sum_j \sum_i (\overline{H_{\iota j}} - \overline{H_{\iota j}^{avg}})\frac{\partial c_{ij}}{\partial t}dv$$

$$Q_{mixing} = \frac{\partial}{\partial t}[\frac{1}{2}\cdot\frac{\partial H_s}{\partial C_s}\int(C_s - C_{s,\infty})^2 dv]$$

$$\frac{\partial H_s}{\partial C_s} = -F\frac{\partial U_H}{\partial C_s}$$

	Cathode($LiCoO_2$)	Anode(LiC_6)
Heat of mixing	50 J/m^2	25 J/ m^2
Irreversible heat	800 J/m^2	2500 J/m^2

Table 1. Comparison of the heat of mixing within active material particles with irreversible heat sources during a 1C discharge process.