

# The Effect of the Heat Transfer Coefficient on Li-ion Battery Failure

Jacob Faulkner<sup>1</sup>, Wenhua H. Zhu<sup>1</sup>, Paul Dimick<sup>2</sup>, Kylie Minor<sup>2</sup>, & Bruce J. Tatarchuk<sup>1</sup>

<sup>1</sup> Department of Chemical Engineering,  
Auburn University  
Auburn, AL, USA, 36849  
jzf0072@auburn.edu

<sup>2</sup> Intramicon, Inc.  
368 Industry Dr.  
Auburn, AL, USA, 36831

## Abstract

*Transferring heat away from a Li-ion battery under stress could prevent a failure event from ever occurring. As a Li-ion battery starts to retain heat, self-heating reactions begin to occur inside the battery. Once the battery heats to its thermal runaway onset temperature, failure is practically unstoppable. In an inert-adiabatic reactor setup, a failing battery has the capability to produce about 5 kJ/Wh of thermal energy in a very short time period [1]. It is necessary to implement a cooling structure if these batteries are to be used in tightly packed battery packs. This cooling structure must be able to prevent failure propagation and compensate for heat generated during anticipated cycling. A cooling structure with an overall heat transfer coefficient (HTC) of at least  $24 \text{ W m}^{-2} \text{ K}^{-1}$  could have the ability to keep a battery from undergoing thermal runaway. This is due to the heat transferred away from the battery outweighing the heat evolved during self-heating reactions. HTC's of 300 and  $1000 \text{ W m}^{-2} \text{ K}^{-1}$  are able to prevent failure from heating rates of  $0.41 \text{ }^\circ\text{C/s}$  and  $2.26 \text{ }^\circ\text{C/s}$ , respectively. The  $1000 \text{ W m}^{-2} \text{ K}^{-1}$  HTC allows for the stopping of thermal runaway once thermal runaway has been initiated. Knowing how Li-ion batteries interact with a cooling structure is important for battery pack design considerations.*

## Keywords

Heat transfer modeling; isokinetic modeling; thermal runaway; microfibrinous metal mesh; phase change materials.

## Introduction

Li-ion batteries under thermal stress are prone to catastrophic failure [1]–[4]. This includes cycling batteries at high charge/discharge rates which is done during pulse power applications. Putting Li-ion batteries under these charge/discharge rates also creates a thermal stress on the battery due to the resistive heating within the battery. If sufficient heating occurs, the battery will reach its thermal runaway (TR) onset temperature where stopping TR is impractical. Before this onset temperature is achieved, self-heating reactions occur within the battery [3,5]. Self-heating reactions aid in bringing the battery to its TR onset temperature. Therefore, it is imperative to dissipate heat from the battery canister.

There are multiple methods for removing heat from Li-ion batteries and they mainly depend on the battery application. Some methods include liquid cooling [6], air cooling [7], or cold plates. Intramicon uses a cooling structure with active and passive cooling capabilities [8]. The passive cooling

includes a porous microfibrinous mesh (MFM) with a void fraction of 0.88. This MFM is created by sintering metal fibers to one another to create better solid-solid joint contact. This increases the thermal conductivity of the MFM. To fill the void space, a phase change material (PCM) is melted into the void space. The PCM is then solidified to create an MFM-PCM matrix. The MFM acts as the thermal diffuser, and the PCM acts as the thermal sink. Active cooling is provided by flowing cooling water through the passive cooling structure.

If a cooling structure proves to be ineffective, the battery will release a large amount of energy. This amount of energy depends on the kinetics of the reactions and the interaction between the battery and the cooling structure. This paper will study the interactions between a battery under thermal stress and its cooling structure.

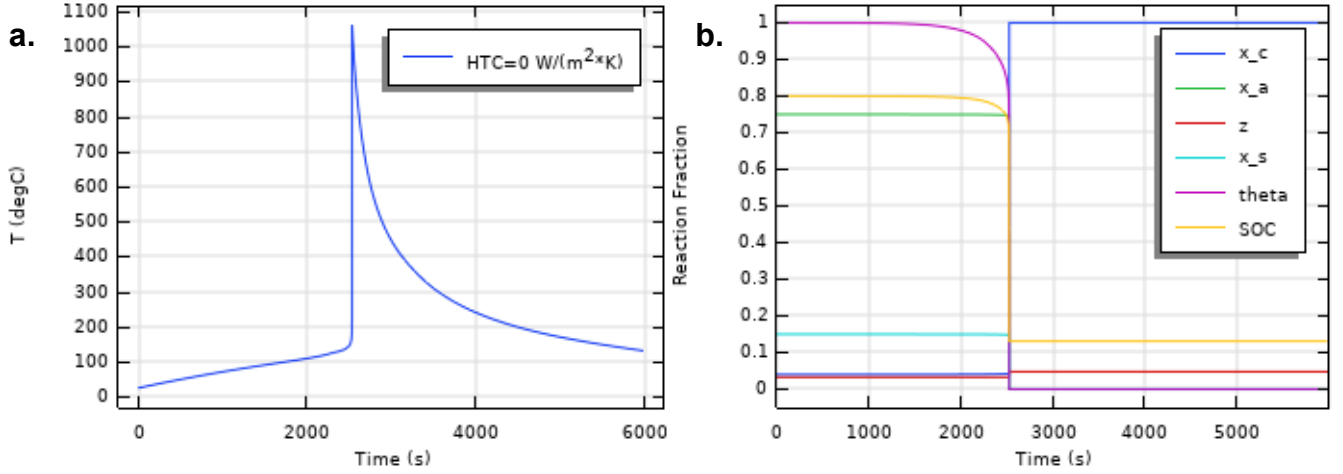
## Model

A kinetic model is composed of reaction kinetics coupled with an energy balance. The kinetic model is performed in COMSOL 6.1 as a zero-dimensional model. The focus of this paper will be the energy balance between the battery and its surroundings. All the reaction kinetics/energy balance information comes from [9]. These reactions can be seen in Table 1.

**Table 1. Reaction Kinetics for Li-ion Battery Failure [9].**

$$\begin{aligned} \frac{dx_c}{dt} &= -A_c x_c \exp\left(\frac{-E_c}{k_b T}\right) \\ \frac{dx_a}{dt} &= -A_a x_a \exp\left(\frac{-E_a}{k_b T}\right) \\ \frac{dx_s}{dt} &= -A_s x_s \exp\left(\frac{-z - E_s}{z_o k_b T}\right) \\ \frac{d\theta_e}{dt} &= -A_e \theta_e \exp\left(\frac{-E_e}{k_b T}\right) \\ \frac{dz}{dt} &= A_a x_s \exp\left(\frac{-z - E_s}{z_o k_b T}\right) \\ \frac{dSoC}{dt} &= -A_{ec} x_a (1 - x_c) \exp\left(\frac{-E_{ec}}{k_b T}\right) + \left(\frac{dx_c}{dt} - \frac{dx_a}{dt}\right) SoC \end{aligned}$$

$x_c$  is the fraction of lithium remaining in the cathode,  $x_a$  is the fraction of lithium in the anode,  $x_s$  is the fraction of lithium within the solid electrolyte interface (SEI),  $\theta_e$  is the fraction of electrolyte in the liquid phase,  $z$  is the thickness of the SEI layer, and SOC is the state of charge of the battery. The reaction kinetic data for each reaction ( $E_i$  and  $A_i$ ) is fitted by [9] from tests done by [2] on a 1.5 Ah Nickel



**Figure 1.** Adiabatic Failure of 1.5 Ah NMC Battery. a) Temperature Profile b) Reaction Fraction Profile.

Manganese Cobalt (NMC) cell. Initial conditions are also provided in [9].

The energy balance that [9] uses is shown in Equation 1:

$$V_{cell} \rho_{jr} c_{p,jr} \frac{\partial T}{\partial t} = -Ah_{conv}(T - T_{amb}) - A\varepsilon\sigma(T^4 - T_{amb}^4) + \sum_i \dot{Q}_i \quad (1)$$

where  $V_{cell}$  is the volume of the battery cell in  $m^3$ ,  $\rho_{jr}$  is the density of the jelly roll in  $kg/m^3$ ,  $c_{p,jr}$  is the specific heat capacity of the jelly roll in  $J kg^{-1} K^{-1}$ ,  $A$  is the surface area of the battery exposed to the cooling environment, in  $m^2$ ,  $h_{conv}$  is the heat transfer coefficient (HTC) that corresponds to natural convection occurring outside of the failing cell, in  $W m^{-2} K^{-1}$ ,  $T_{amb}$  is the ambient temperature surrounding the failing battery, in  $K$ ,  $\varepsilon$  is the surface emissivity, unitless, and  $\sigma$  is the Stefan-Boltzmann constant, in  $W m^{-2} K^{-4}$ . The summation term on the far right considers the energy introduced by the degradation reactions, boiling, and heaters.

$$\sum_i \dot{Q}_i = \dot{Q}_{rxn} + \dot{Q}_{boil} + \dot{Q}_{ej} + \dot{Q}_{heater} \quad (2)$$

$\dot{Q}_{rxn}$  is the heat produced by TR reactions,  $\dot{Q}_{boil}$  is the heat removed by the boiling of electrolyte,  $\dot{Q}_{ej}$  is the energy removed by the ejecta leaving the battery canister, and  $\dot{Q}_{heater}$  is the energy provided by the heater to the battery canister to induce failure. The parameter that this study is interested in is the  $h_{conv}$  term.

## Methods

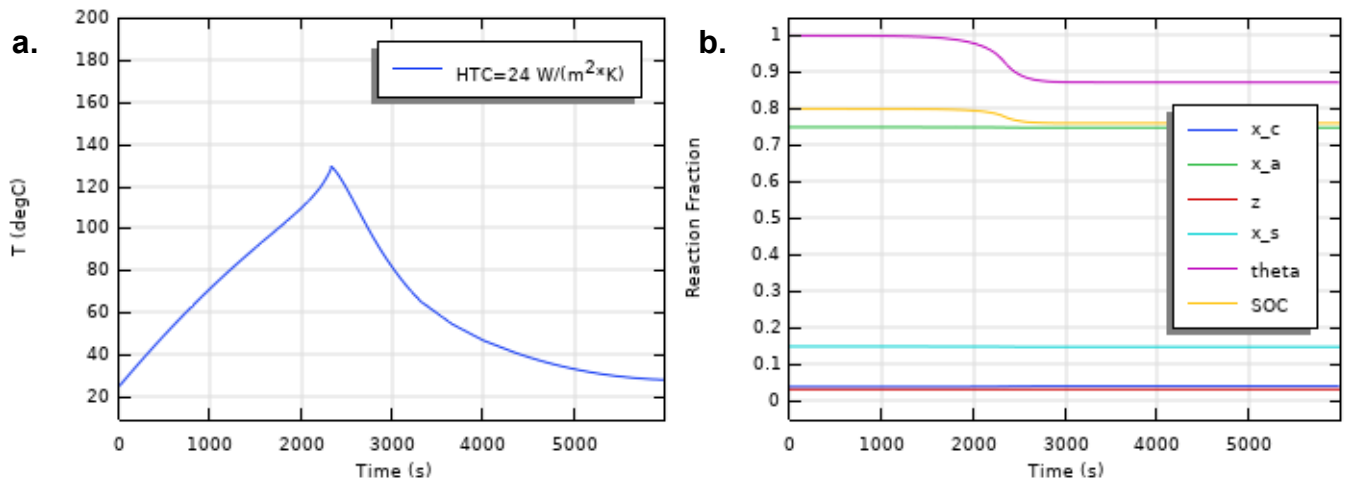
A simulated electric heater is used to force the modeled battery into thermal runaway. This heater term is represented by  $\dot{Q}_{heater}$  in the energy balance (Equation 2). The heater stays on during the simulation until self-heating is observed. Under normal heater operating conditions, the battery temperature increases at  $0.036 \text{ } ^\circ\text{C/s}$ . In this case, self-heating is defined as any temperature increase above  $0.036 \text{ } ^\circ\text{C/s}$ .

Once self-heating is observed, the heater is turned off, and the HTC is applied to the energy balance. The HTC is parameterized to determine the minimum required HTC to keep TR from occurring once self-heating is observed. The same method can be applied when considering the HTC necessary for stopping TR once the onset temperature is achieved.

## Results and Discussion

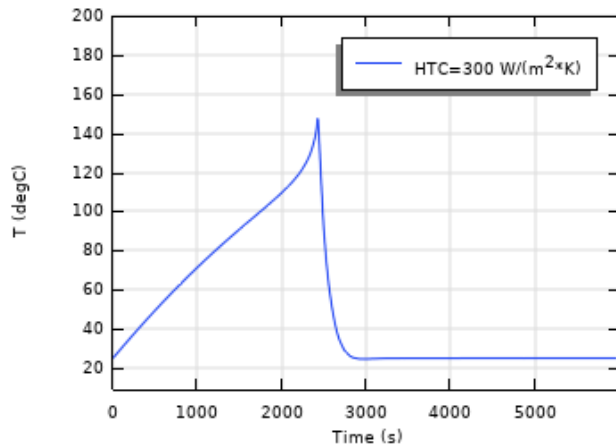
First, the failure of the battery must be understood at adiabatic-like conditions. This will be the baseline for all other calculations. In the adiabatic failure kinetic model, the heater initially changes battery surface temperature at  $0.036 \text{ } ^\circ\text{C/s}$ . The battery experiences initial self-heating at  $102\text{-}103 \text{ } ^\circ\text{C}$  because this is when the temperature change deviates from its steady state value of  $0.036 \text{ } ^\circ\text{C/s}$ . The model represents this initial self-heating as an electrochemical heat source because SOC is the first reaction fraction to change in the reaction profile in Figure 1b [9]. Furthermore, this initial heating is partially diminished by the endothermic boiling of the electrolyte because  $\theta_e$  is also changing during the initial self-heating of the battery. The TR onset temperature is hard to define, but it is usually based on the amount of temperature change occurring within the battery [2]. In this case, the onset temperature is set at around  $165 \text{ } ^\circ\text{C}$  where the temperature change of the battery surface is greater than or equal to  $2 \text{ } ^\circ\text{C/s}$ . Figure 1b shows that all the reactions are proceeding to a significant extent. This should be kept in mind when considering the reaction profiles in the applied HTC models. It is evident that TR occurred within the battery because it reaches a max temperature of about  $1050 \text{ } ^\circ\text{C}$ . A failure of this magnitude is dangerous to battery surroundings, especially in the case where batteries are tightly packed.

With this understanding of adiabatic failure, it is necessary to cool batteries to keep them from failing. This is where the HTC comes into consideration. An overall HTC of  $24 \text{ } W m^{-2} K^{-1}$  is able to keep the self-heating reactions from driving



**Figure 2.** Non-Adiabatic Failure Scenario ( $24 \text{ W m}^{-2} \text{ K}^{-1}$ ) a) Temperature Profile b) Reaction Fraction Profile.

the battery to TR. This HTC is applied once the battery is self-heating at almost triple the original heater rate,  $0.1 \text{ }^\circ\text{C/s}$ . The battery surface temperature is  $129 \text{ }^\circ\text{C}$  when the HTC is applied to the kinetic model. Notice in Figure 2b that the exothermic reactions do not go to completion, but they are halted by the dissipation of heat from the battery canister. Heat is dissipated from the battery canister faster than the electrochemical reactions can produce heat. The other HTC scenarios will undergo similar reaction profiles, so they will not be provided in this document.

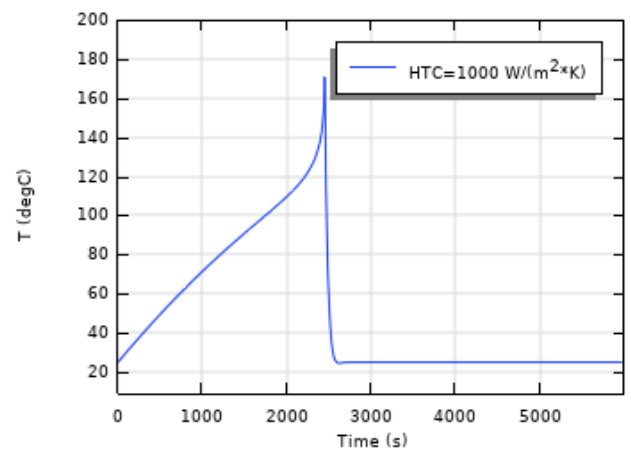


**Figure 3.** Non-Adiabatic Failure Scenario ( $300 \text{ W m}^{-2} \text{ K}^{-1}$ ).

Figure 3 shows that applying a  $300 \text{ W m}^{-2} \text{ K}^{-1}$  HTC allows for the battery surface to be heated to  $147 \text{ }^\circ\text{C}$  without experiencing TR. A temperature change of  $0.41 \text{ }^\circ\text{C/s}$  is able to be overcome by this HTC. Reaction progression will also be limited by the applied HTC.

Applying a  $1000 \text{ W m}^{-2} \text{ K}^{-1}$  allows for the TR onset temperature to be reached without proceeding to failure. The maximum temperature change of the battery is  $2.26 \text{ }^\circ\text{C/s}$  at  $169 \text{ }^\circ\text{C}$ . However, the HTC can supply sufficient cooling to the battery. Figure 4 demonstrates the cooling of the battery under these circumstances. This may seem impractical when

it comes to cooling capabilities, but this compares to the upper limits of forced convection of air, gases, and vapors [10]. This cooling scenario allows for a greater extent of reaction without allowing the battery to experience TR.



**Figure 4.** Non-Adiabatic Failure Scenario ( $1000 \text{ W m}^{-2} \text{ K}^{-1}$ ).

### Future Work

This work has provided the general methodology for interfacing cooling structure technology with battery kinetic modeling. If the overall heat transfer coefficient of the desired cooling structure is known, then the ability to push battery performance can be determined. This means that batteries can be cycled at higher charge and discharge rates without fear of thermal stress being the initiator of TR. With the Intramicron developed cooling structure, the overall heat transfer coefficient can be observed experimentally and applied to the kinetic model to determine how hot the battery can get without undergoing TR from thermal stress. Li-ion battery failure varies not only from cell chemistry to cell chemistry, but it varies between cells of the same chemistry and even the same batch. Therefore, it is imperative that batteries are tested within their respective cooling structures before operation. Doing this will help validate the modeling effort and further ensure safe cycling of Li-ion batteries.

The HTC's discussed in this paper apply for a 1.5 Ah cylindrical NMC cell; however, a similar methodology can be used for other battery chemistries provided the kinetic data is provided. Different failure methods should also be considered. The failure method described in this paper is thermally initiated failure not failure due to internal short circuit or other failure mechanisms. Other failure mechanisms could require different cooling capabilities.

### Acknowledgements

The authors would like to thank the Office of Naval Research for funding our research and development efforts. Work is done under Navy Contract N6449819D4025, Task Order 1 N6449819F4256.

### Conclusions

A lumped kinetic model provides the ability for batteries to communicate with battery cooling structures. This study shows that as a HTC is applied to a Li-ion battery under thermal stress, heat is taken away from the battery to keep the battery from reaching TR. TR is prevented at multiple self-heating rates:

1. Applying a HTC of  $24 \text{ W m}^{-2} \text{ K}^{-1}$  once a self-heating rate of  $0.1 \text{ }^\circ\text{C/s}$  is observed quenches TR.
2. Applying a HTC of  $300 \text{ W m}^{-2} \text{ K}^{-1}$  in a more pronounced self-heating region of  $0.41 \text{ }^\circ\text{C/s}$  quenches TR.
3. Applying a HTC of  $1000 \text{ W m}^{-2} \text{ K}^{-1}$  once the TR onset temperature has been achieved at a self-heating rate of  $2.26 \text{ }^\circ\text{C/s}$  quenches TR.

With effective cooling structures applied to Li-ion batteries, batteries can be pushed to their operating limits. This means that batteries can be cycled faster and can operate under pulse power applications.

### References

- [1] NASA, NREL, and Electrochemical Innovation Lab, "Battery Failure Databank." NASA, NREL, Electrochemical Innovation Lab, Oct. 2020. [Online]. Available: <https://www.nrel.gov/transportation/battery-failure.html>
- [2] A. W. Golubkov *et al.*, "Thermal-runaway experiments on consumer Li-ion batteries with metal-oxide and olivin-type cathodes," *RSC Adv.*, vol. 4, no. 7, pp. 3633–3642, 2014, doi: 10.1039/C3RA45748F.
- [3] A. W. Golubkov *et al.*, "Thermal runaway of commercial 18650 Li-ion batteries with LFP and NCA cathodes – impact of state of charge and overcharge," *RSC Adv.*, vol. 5, no. 70, pp. 57171–57186, 2015, doi: 10.1039/C5RA05897J.
- [4] F. Larsson, P. Andersson, P. Blomqvist, and B.-E. Mellander, "Toxic fluoride gas emissions from lithium-ion battery fires," *Sci. Rep.*, vol. 7, no. 1, Art. no. 1, Aug. 2017, doi: 10.1038/s41598-017-09784-z.
- [5] Q. Wang, B. Mao, S. I. Stolarov, and J. Sun, "A review of lithium ion battery failure mechanisms and fire prevention strategies," *Prog. Energy Combust. Sci.*, vol. 73, pp. 95–131, Jul. 2019, doi: 10.1016/j.pecs.2019.03.002.
- [6] Y. Ding, H. Ji, M. Wei, and R. Liu, "Effect of liquid cooling system structure on lithium-ion battery pack temperature fields," *Int. J. Heat Mass Transf.*, vol. 183, p. 122178, Feb. 2022, doi: 10.1016/j.ijheatmasstransfer.2021.122178.
- [7] P. T. Coman, E. C. Darcy, and R. E. White, "Simplified Thermal Runaway Model for Assisting the Design of a Novel Safe Li-Ion Battery Pack," *J. Electrochem. Soc.*, vol. 169, no. 4, p. 040516, Apr. 2022, doi: 10.1149/1945-7111/ac62bd.
- [8] W. Zhu, Y. Hongyun, P. Dimick, and B. Tatarchuk, "A Metal Microfibrous Enhanced Cooling Structure for Heat Rejection and Temperature Control of High Power Mid-Form Li-ion Battery Packs," presented at the 48th Power Sources Conference, Denver, Colorado, Jun. 11, 2018.
- [9] P. T. Coman, S. Rayman, and R. E. White, "A lumped model of venting during thermal runaway in a cylindrical Lithium Cobalt Oxide lithium-ion cell," *J. Power Sources*, vol. 307, pp. 56–62, Mar. 2016, doi: 10.1016/j.jpowsour.2015.12.088.
- [10] C. J. Geankoplis, *Transport processes and separation process principles*, Fifth edition. in Prentice Hall international series in the physical and chemical engineering sciences. Boston, MA: Prentice Hall, 2018.