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## Considering Battery Degradation in Life Cycle Greenhouse Gas Emission Analysis of Electric Vehicles

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### Abstract

Lithium ion batteries (LIBs) are predominantly employed to power Electric Vehicles (EVs). Battery degradation can significantly affect battery performance and further influence the energy consumptions and life cycle greenhouse gas (GHG) emissions from EVs. However, currently battery degradation has not been taken into account in life cycle assessment (LCA) of EVs. In this paper, we report our research on considering battery degradation into life cycle GHG emissions of EVs, by analyzing a mid-size EV using a 24 kWh Lithium-Manganese-Oxide (LMO)-graphite battery. Both cycling loss and calendar loss in battery degradation are analyzed under average driving conditions of U.S. The model is validated with experimental testing data on Nissan Leaf from Argonne National Lab. In this study, the required battery replacement is also integrated into the LCA GHG emission model and results, by considering the cradle-to-gate GHG emissions from a battery pack. This model can improve the accuracy and reliability of life cycle GHG studies on EVs.

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### 1. Introduction

The transportation sector generates 28% of total Greenhouse Gas (GHG) emissions in the United States [1,2]. Electric vehicles (EVs) powered by lithium-ion batteries (LIBs) have been recognized as a clean alternative to conventional vehicles for reducing the GHG emissions from the transportation sector [2]. During the operation of EVs, LIBs undergo a sophisticated degradation process which leads to the material and structural change and capacity losses [3,4]. The battery capacity loss further affects the battery performance during the operations of EVs in two aspects: reduced driving range due to reduced available capacity, and decreased battery efficiency due to the increased resistance [4]. Consequently, the life cycle performance of EVs will be affected through the shortened

driving mileage, increased energy loss, and required battery replacement. However, past LCA studies of EVs tend to ignore the sophisticated battery degradation process by employing simplified assumptions on EV battery performance. For instance, Leuenberger and Frischknecht assumed the EV battery having the same lifetime as vehicles [5]; Hawkins et al. proposed one battery replacement over the entire life cycle of vehicles [6].

The LIBs on board EVs also generate GHGs during its life cycle. As reported, a LIB can release 57-85 kg CO<sub>2</sub> eq. per kg battery during the life cycle of EVs [7,8]. The ignorance of battery degradation may result in a significant variation in the life cycle emissions of EVs and cause problems in decision support and policy-making. To provide robust decision support, the battery degradation on board EVs needs to be scientifically

investigated and accurately taken into account to ensure the reliability of the LCA results.

This study is conducted to investigate the impacts of battery degradation on the life cycle GHG emissions from EVs, with a case study on a mid-sized battery EV using a 24 kWh Lithium-Manganese-Oxide (LMO)-graphite battery pack. The battery degradation model is developed to simulate battery capacity loss during EV operations in the United States based on the average U.S. geographical and operating conditions. The impacts of battery degradation on the subsequent EV operation have been explored based on traffic statistics in the United States. Finally, these impacts have been incorporated into a comparative life cycle GHG emissions analysis to illustrate the influence of battery degradation in the life cycle assessments of EVs.

**2. Methodology**

This study evaluates the EV battery degradation process in the United States and its influence on life cycle GHG emissions through the adoption of a battery degradation model. As one of the widely deployed EVs, Nissan Leaf with a 24 kWh Lithium-Manganese-Oxide (LiMn<sub>2</sub>O<sub>4</sub>)/Graphite battery has been selected as the representative mid-size EVs. The vehicle is assumed to operate in the United States based on its average geographical and operating conditions during a 10-year lifetime. The battery will reach its end of life for replacement upon 30% capacity loss based on Nissan’s recommendation [9]. The functional unit of the study is set as the amount of GHG emissions per mile driven from the EV during a 10-year lifetime in the United States.

*2.1. Battery degradation model*

According to numerous studies on Lithium-ion batteries, it is known that battery degradation is a complicated process including loss of active materials [10,11], solid-state-interphase (SEI) formation [12], resistance increase [13,14], and self-discharge [15]. Capacity loss is the loss of battery capacity, which is the result of battery degradation [10]. In this section, a comprehensive EV battery degradation model, composed of cycling loss and calendar loss, is presented to simulate the battery capacity fading during the EV’s operation on an annual basis. The degradation model is built on a 24 kWh LiMn<sub>2</sub>O<sub>4</sub>-Graphite battery as used in Nissan LEAF EV.

*2.1.1. Cycling loss model*

The cycling loss is the capacity fading of lithium-ion battery during its charge-discharge operations. The model employed in this study includes charge, energy transport, and mass transfer processes inside lithium-ion batteries [15]. Theoretically, there are two charge carriers inside a battery: electrons and lithium-ions. The charge balance of LiMn<sub>2</sub>O<sub>4</sub>/Graphite battery includes the charge transport processes in solid materials and liquid electrolyte, which can be formulated by the generic Ohm’s law,

$$\begin{cases} \text{Solid phase: } \nabla \cdot (-\sigma_j^{eff} \nabla \phi_{1,j}) = i_j^{tot} \\ \text{Liquid phase: } \nabla \cdot (-\kappa_j^{eff} \nabla \phi_{2,j}) + \frac{2RT(1-t_+^0)}{F} \nabla(-\kappa_j^{eff} \nabla(\ln c_j)) = i_j^{tot} \end{cases} \quad (1)$$

where,  $\sigma_j^{eff}$  and  $\kappa_j^{eff}$  are the effective conductivities in solid phase and liquid phase respectively;  $t_+^0$  is the transference number of lithium-ion;  $F$  is the Faraday’s constant;  $T$  is the temperature,  $t$  is the time;  $\phi_{1,j}$  and  $\phi_{2,j}$  are the potentials in solid phase and liquid phase of battery respectively;  $i_j^{tot}$  is the total current transferred between the solid electrodes particles and the electrolyte solution.

The mass transfer of lithium-ions in the battery contains the transport processes on solid electrode particles and in liquid electrolyte solution, which can be formulated by the Fick’s second law:

$$\begin{cases} \text{Electrode particle: } \frac{\partial c_j^s}{\partial t} = D_j^s \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \frac{\partial c_j^s}{\partial r}) \\ \text{Electrolyte solution: } \varepsilon_j \frac{\partial c_j}{\partial t} = \frac{\partial}{\partial x} (D_j^{eff} \frac{\partial c_j}{\partial x}) + \frac{(1-t_+^0)(i_j^F + i_j^s)}{F} \end{cases} \quad (2)$$

where,  $c_j^s$  is the lithium-ion concentration;  $D_j^s$  is its diffusion coefficient in solid materials;  $r$  is the reaction rate;  $\varepsilon_j$  is the electrode porosity;  $D_j^{eff}$  is the effective diffusion coefficient [1].

Besides the effect of environmental temperature, the massive heat production from battery cells by charging/discharging operation can also significantly intensify the battery capacity fading process. To take the influence of both environmental temperature and battery internal heat production into consideration, we applied the Fourier’s law to model the battery global heat production and transport,

$$\rho c_p \frac{\partial T}{\partial t} + \nabla \cdot (-\lambda \nabla T) = \sum_i Q_i, \quad \sum_i Q_i = Q_{rxn} + Q_{rev} + Q_{ohm} \quad (3)$$

where,  $\rho$  is the density;  $c_p$  is the specific heat capacity;  $\lambda$  is the heat conductivity;  $\sum_i Q_i$  is the heat source term, which is composed of total reaction heat generation  $Q_{rxn}$ , total reversible heat production  $Q_{rev}$ , and total Ohmic heat production  $Q_{ohm}$ . Those heat source terms can be formulated by [1],

$$Q_{rxn} = (i_j^F + i_j^s)(\phi_{1,j} - \phi_{2,j} - U_j - \frac{(i_j^F + i_j^s)}{S_j} R_f) \quad (4)$$

$$Q_{rev} = (i_j^F + i_j^s) T \frac{\partial U_j}{\partial T} \quad (5)$$

$$Q_{ohm} = \sigma_{eff} (\frac{\partial \phi_1}{\partial x})^2 + \kappa_{eff} (\frac{\partial \phi_2}{\partial x})^2 + \frac{2\kappa_{eff} RT}{F} (1-t_+^0) \frac{\partial(\ln c)}{\partial x} \frac{\partial \phi_2}{\partial x} \quad (6)$$

where,  $i^F$  is the Faradaic current in the battery;  $i_j^s$  is the electrolyte reduction current;  $U_j$  is the battery open circuit potential;  $R_f$  is the total SEI film resistance;  $S_j$  is the specific active interfacial area;  $c$  is the local lithium-ion concentration. Besides, the battery charging-discharging induced solid-electrolyte interphase generation and LiMn<sub>2</sub>O<sub>4</sub> active material

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