Journal of Power Sources 322 (2016) 169–178

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Energy impact of cathode drying and solvent recovery during lithium-ion battery manufacturing

Shabbir Ahmed*, Paul A. Nelson, Kevin G. Gallagher, Dennis W. Dees

Chemical Sciences and Engineering Division, Argonne National Laboratory, Bldg. 200, 9700 S. Cass Avenue, Argonne, IL 60439, USA

HIGHLIGHTS

- Energy for NMP vaporization is a small fraction of that needed for cathode drying.
- Large air flow is required to operate at low fraction of NMP flammability limits.
- Energy demand for cathode drying and NMP recovery because of large air flow.
- It requires ~10 kWh to evaporate and recover a kg of NMP.
- It requires ~420 kWh to evaporate and recover NMP for a 10 kWh LIB pack.

ARTICLE INFO

Article history:
Received 19 January 2016
Received in revised form
10 March 2016
Accepted 21 April 2016
Available online 14 May 2016

Keywords:
Lithium-ion battery
Manufacturing
Cathode solvent
Energy need
n-methyl pyrrolidone

ABSTRACT

Successful deployment of electric vehicles requires maturity of the manufacturing process to reduce the cost of the lithium ion battery (LIB) pack. Drying the coated cathode layer and subsequent recovery of the solvent for recycle is a vital step in the lithium ion battery manufacturing plant and offers significant potential for cost reduction. A spreadsheet model of the drying and recovery of the solvent, is used to study the energy demand of this step and its contribution towards the cost of the battery pack. The base case scenario indicates that the drying and recovery process imposes an energy demand of ~10 kWh per kg of the solvent n-methyl pyrrolidone (NMP), and is almost 45 times the heat needed to vaporize the NMP. For a plant producing 100 K battery packs per year for 10 kWh plug-in hybrid vehicles (PHEV), the energy demand is ~5900 kW and the process contributes $107 or 3.4% to the cost of the battery pack. The cost of drying and recovery is equivalent to $1.12 per kg of NMP recovered, saving $2.08 per kg in replacement purchase.

1. Introduction

The increasing deployment of electric vehicles is accompanied by a greater impetus towards reducing the cost of lithium ion batteries (LIB). While new chemistries can offer breakthroughs by introducing cheaper materials with higher capacity, safety, and durability, their manufacturing process [1,2] has to limit the cost of the final product (the pack) through process simplifications and optimization [3,4]. Industry continues to study each step in the battery manufacturing process and is seeking alternatives that can lead to faster production and lower cost. The work reported in this paper explores the cathode drying process in the manufacturing plant — its energy demand and contribution to the cost of the battery pack.

The cathode is composed of lithium metal oxide particles such as for example $LiMn_2O_4$, $Li_aNi_xMn_yCo_zO_2$, etc., carbon black conductive additive such as acetylene black, graphite, etc., and polymeric binder such as polyvinylidene difluoride (PVDF) typically in an approximate mass ratio of 94:3:3 in the dried state. Different kinds of solids are first dry mixed, and then mixed with a solution of the binder dissolved in the solvent, n-methyl pyrrolidone (NMP) [7], which is water soluble to form a slurry. The mixing condition has a strong impact on the slurry and ultimately the battery performance [8]. The slurry is coated on to both sides of the cathode current collector (i.e., aluminum foil) [9] using techniques such as the slot die coating heads. The alternate sides may be coated on a completely separate pass through the same coating line, at a second coating station installed after the first drying oven or nearly

---

* Corresponding author.
E-mail addresses: ahmeds@anl.gov (S. Ahmed), nelsonp@anl.gov (P.A. Nelson), kevin.gallagher@anl.gov (K.G. Gallagher), dees@anl.gov (D.W. Dees).

http://dx.doi.org/10.1016/j.jpowsour.2016.04.102
0378-7753/

simultaneously before entering the drying oven. The dried coating thicknesses are typically less than 80 μm. The electrode coats are interrupted at intervals where the current collectors will be slit (farther downstream) to match the length or width of the electrodes. The electrode-coated current collector is then passed through the dryer to evaporate the liquid content (NMP in the cathode, water in the anode). The dryer closely follows the coating station and stretches for several meters to ensure a drying rate that maintains the desired particle size, porosity (the porosity may be further reduced during calendaring downstream) and distribution. The coated sheet is slit at the intervals to produce the electrode layers which can be stacked (or rolled in the case of cylindrical cells) together with the separator layer. The cells are vacuum-assisted filled with electrolyte before sealing. The formation cycling process consists of charging and discharging the cells according to a manufacturer optimized protocol to facilitate the activation of the solid electrolyte interface, a critical layer needed for the cycle life of the cathode. The cells are then stored for a period at a certain state of charge to check for shelf life as part of the quality control process. The cells that pass the requirements are then ready for shipping.

Tagawa and Brodd [8] provide more details about the electrode processing. The performance of the cell is strongly influenced by the electrode preparation which includes the choice of additives, mixing ratios, mixing and dispersing techniques to produce the desired electrode structure [10], drying method and rate [3,4], and compacting to improve electronic conductivity [11]. Inconsistencies in the manufacturing of the electrode layers have a significant impact on the cell and module performance, resulting in lower or unavailable capacity, and capacity fade. Electrode thickness and porosity are important parameters for useful capacity [12] and internal resistance [13]; these characteristics require careful control during the slurry preparation [14], coating, drying [3,15,16], and calendering [17,18].

The NMP evaporated in the dryer is subsequently recovered and recycled, resulting in significant cost savings from the combination of avoided replacement cost and waste/emissions reduction. It is recognized that the cost savings and environmental benefits might be significantly greater if the use of solvents for the coatings could be avoided altogether by applying the coatings in a dry state [19]. However, that is not the subject of this study.

Argonne has a spreadsheet tool, BatPaC [1,5,6], that is used by researchers and organizations to design and estimate the cost of automotive LIB packs. It calculates the pack cost based on the contributions from a large number of processing steps. Petri et al. [20]. reviews and compares three models, including BatPaC; other lithium ion battery models include a cost model developed by TIAX [21] and a lifetime model by the National Energy Renewable Laboratory (NREL) [22].

The objective of this work is to study the energy demand and distribution for the drying of the cathode and the process for the recovery of the solvent (NMP), and to estimate the cost of this process. The spreadsheet model reported in this work is designed to review and update the estimates used in the BatPaC model, by looking more closely at mass and energy balances of the many unit operations in the process and follow through with more detailed estimates of the cost contributors. Results from this reported model will be used to update BatPaC input parameters, as needed.

2. Process model

The NMP recovery process studied here is for a battery manufacturing plant producing 100,000 packs per year of 60 kW, 10 kWh plug-in hybrid vehicle (PHEV) batteries, where each pack costs approximately $3100 ($310 per kWh). Over 4.1 million kg per year of NMP is used for depositing the cathode layers.

Consider the process as shown in Fig. 1, where the NMP in the cathode is evaporated in the dryer by exposure to flowing hot air. The air leaves the dryer at a specified temperature and the maximum allowable NMP concentration (140 °C and 1150 parts per million by volume (ppm) for the base case scenario). The NMP-laden hot air is first passed through an air-to-air heat exchanger – transferring part of the sensible heat to the incoming air, and then it is cooled in a chilled water condenser. The NMP, water, and higher hydrocarbons (degradation byproducts from the NMP processing) in the liquid condensate are separated in a distillation column. The cooled air from the condenser, containing a few ppm of NMP, is split into the recycle and purge streams. The purge stream is passed through a zeolite wheel where most of the NMP is adsorbed, followed by water scrubbing before it is sent out as exhaust.

Ambient air is drawn in as make-up and is heated to a temperature above 50 °C and passed through the zeolite wheel, thereby desorbing the trapped NMP, to regenerate the zeolite. The NMP containing make-up air is combined with the recycle stream from the condenser and passed through the air-to-air heat exchanger, preheating it on its way to the air heater. The recycled air is indirectly heated in the air heater and then fed into the cathode dryer. Indirect heating is advantageous in that it does not add any moisture (combustion product) to the dryer air. In an alternative system design, the recycled air can be blended with hot combustion air from a burner to eliminate the heat transfer surfaces in the indirect heater, but the combustion air will introduce moisture into the dryer air, increasing the loads of the condenser and the distillation column.

A base case set of process conditions was selected to estimate the energy and cost of operation of the drying and recovery process. Table 1 lists the input conditions used in the process model.

3. Results and discussion

The core necessity of this process is the removal of NMP from the cathode and to recover and recycle the same. The heat needed to vaporize the NMP from the cathode material is a moderate 130 kW, for the base case scenario. But the NMP has to be vaporized in a controlled manner to avoid damaging the morphology of the cathode coating, and the NMP vapor concentration in the gas phase has to be maintained far below the lower flammability limit, which is approximately 1.1% at 140 °C. This latter constraint imposes the requirement that a large amount of heated air be used for the drying process. Heating the large volume of air raises the heat load considerably.

For the process studied here, the heat load for the air heater, 3752 kW, is necessary to heat the 37 $m^3\ s^{-1}$ of air to elevate its temperature by 75 centigrade degrees. The specified air rate is needed to limit the NMP concentration in the dryer exit gas at 1150 ppm. The heat load would have been considerably higher without the air-to-air heat exchanger which recovers and transfers 2700 kW of heat from the dryer exit air to the heater inlet air. The air-to-air heat exchanger manages the heat transfer load with a surface area of ~6700 m^2. The other significant heat exchanger, the condenser has a higher heat transfer load of ~3500 kW, but because of the liquid water coolant the heat exchange is managed with a smaller surface area of ~3300 m^2. The cooling of the air, i.e., sensible heat removal, represents 93% of the condenser cooling load. The major heating and cooling loads of the components are shown in Table 2 and Fig. 1.

The energy demand for this process and set of input conditions is 1470 kW of electricity and 4381 kW of thermal energy, totaling to 5851 kW. This translates to 421 kWh per battery pack and 10.2 kWh