

SOLVENT-FREE ELECTRODE MANUFACTURING FOR LI-ION BATTERIES

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Abstract. Cathodes for Li-Ion batteries, mainly applied in consumer products and electric vehicles, are currently manufactured using the toxic solvent n-methyl pyrrolidone (NMP). For processing this, solvent recovery and explosion preventing equipment is necessary. Therefore, a high amount of energy is needed, which causes ~10 % of energy demand and ~14.5 % of costs in cell manufacturing. Avoiding solvents in the electrode manufacturing process reduces the environmental impact of Li-Ion battery production significantly. Therefore, this work [1] focuses on realizing a solvent-free electrode manufacturing process. LiNi_{0.6}Mn_{0.2}Co_{0.2}O₂ (NMC622) cathodes were successfully manufactured on pilot scale with a loading of 4.0 mAhcm⁻² to 6.9 mAhcm⁻². The practicability of the manufacturing process was investigated by comparing electrochemical results to those of NMP-based cathodes. Specific discharge capacities at 0.5C (10th cycle) of 137 mAh/g in coin cells and up to 163 mAh/g in pouch cells were achieved. The capacity retention after 50 cycles and the discharge capacity at different C-rates are comparable to NMP-based cathodes and therefore proof the potential of solvent-free manufacturing.

Keywords: Li-Ion battery, sustainable manufacturing, solvent-free electrode production

1 MOTIVATION

The production of cathodes for Li-Ion Batteries is generally based on the toxic solvent n-methyl pyrrolidone (NMP), necessary for processing the binder material polyvinylidene fluoride (PVDF). There are economical and ecological aspects, such as high unit costs compared to other solvents, flammable and toxic vapours during manufacturing and greenhouse gas emissions due to the energy intensive drying process [5, 8]. Solvent-free manufacturing decreases the energy usage by 42 kWh per kWh capacity, which corresponds to approximately 10 % of overall energy demand in cell production [2]. The use of NMP causes up to 14.5 % of overall cell manufacturing costs [7].

NMP/PVDF can be substituted by deionized water and water soluble binder materials. Slurry agglomeration, low wettability and corrosion of the current collector or active material, especially when focusing on high-nickel NMC, are challenges of water-based processing. These can be solved via pre-processing steps and including additives [3, 4, 6]. This and the nevertheless necessary drying step lead to a reduction of the ecological and economical advantages of water-based processing.

Therefore, solvent-free manufacturing of electrodes seems the mean of choice for future battery manufacturing [2]. The main advantages are:

- reducing cost and environmental impact of battery manufacturing
- time and energy consuming drying step can be avoided

2 METHODS

Different solvent-free manufacturing processes in the literature were considered and the method with the least environmental impact was selected for further work. During pilot tests basic manufacturing parameters were analysed, a suitable binder was tested and the material composition was specified. The characteristics of the solvent-free produced cathodes were investigated using mechanical and as well electrochemical tests, e.g. electrode drop tests and conductivity measurements. The impact of the mixing process, the binder and the conductive additive on the electrode characteristics were analysed in 4 test series. Based on the experience gained, a coating equipment was developed and integrated into an existing roll-to-roll machine. The production process was used for manufacturing cathodes for pouch cells. To prove the feasibility of the manufacturing process, electrochemical results of solvent-free cathodes in full cells were investigated.

2.1 ELECTRODE PRODUCTION

Compared to other solvent-free manufacturing processes like extrusion-based processes, no additional components, which have to be removed after coating, are needed for the proposed dry coating process. Therefore, the demand of materials and energy are brought towards a minimum, which leads to a significant reduction of environmental impact in electrode production. The solvent-free manufacturing process is described according to Figure 1.

Formulation and Mixing: $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ NMC622 (active material) by BASF, PVDF (Binder) Kynar HSV 900 by ARKEMA and carbon black super C65 (conductive additive) by IMERYS are combined in a ratio of 90:6:4 wt%. The mixing was done using a ball milling process in batches of several mg.

Dry Coating: The materials were coated on an aluminum current collector in an area of 160 mm x 100 mm. The initial layer height was adjusted between 300 μm to 550 μm .

Calendaring: In a final manufacturing step the loose materials is compressed to a third of the initial thickness in a roller press heated to 150°C.

Coil up: The electrode is coiled up and ready for further processing. The achieved areal capacity is 4.0 mAhcm⁻² to 6.9 mAhcm⁻², calculated on a basis of 180 mAhg⁻¹ for NMC 622.

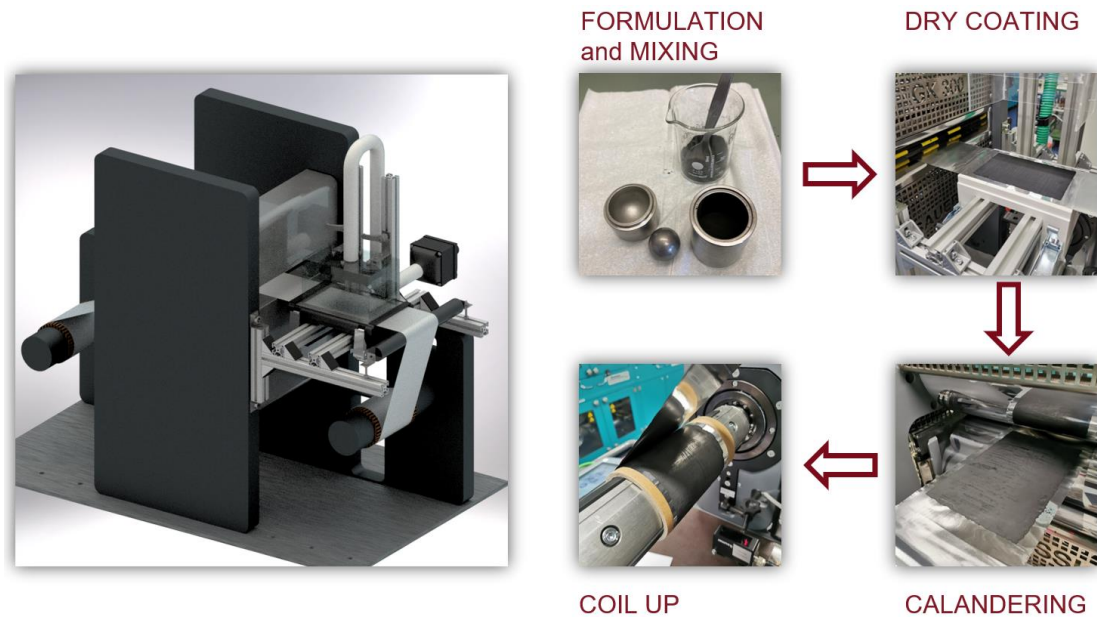


Figure 1. Solvent-free manufacturing process using pilot scale equipment and newly developed coating device [1]

2.2 ELECTRODE PROPERTIES AND CELL ASSEMBLY

Dry manufactured cathodes in coin cell and pouch cell format and as well microscope captures of solvent-free cathodes and NMP-based cathodes are shown in Figure 2.

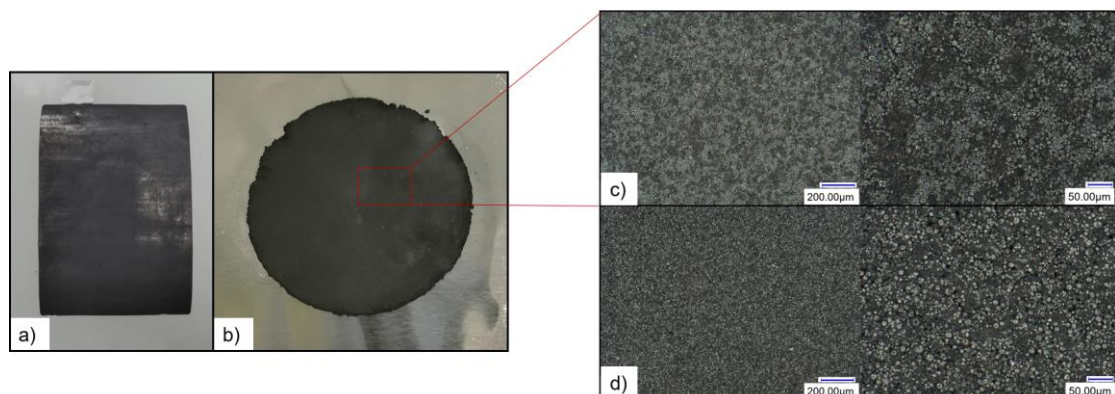


Figure 2. Images of dry processed cathodes a) for pouch cells and b) for coin cells, Images of c) solvent-free processed and d) NMP-based cathodes taken via optical microscope in 200x and 500x magnification [1]

From optical inspection, the proposed dry mixing and coating process causes a slightly more inhomogeneous distribution of active material (grey, spheric shapes) and conductive additive (dark black, contourless). Still, no obvious negative effects on the electrode structure or the overall electrochemical performance was found. The binder material is well mixed with the other components, as no binder agglomerates (white) can be seen. The mechanical properties of the produced electrode coatings were suitable for assembling coin cells and pouch cells.

3 RESULTS AND DISCUSSION

Electrochemical tests of solvent-free and NMP-based cathodes were carried out in full cells with a water-based processed graphite anode. A CELGARD 2320 separator was used and the electrolyte consisted of 1M lithium hexafluorophosphate (LiPF₆) in ethylene carbonate and ethyl methyl carbonate (EC:EMC, weight ratio of 3:7) with 2 wt% vinylene carbonate (VC).

Figure 3 shows the corresponding discharge capacities in coin cells and pouch cells. The results in coin cells are shown in proportion to the discharge capacity in the 1st cycle. At C-rates below 0.5C the capacity loss at increasing c-rates is comparable for both manufacturing methods. At 0.5C and 1C, solvent-free cathodes achieve a higher discharge capacity, which indicates an improved rate capability compared to NMP-based cathodes. The capacity loss after 50 cycles is on the same level. In 260 mAh pouch cells solvent-free NMC622 cathodes achieved 162 mAhg⁻¹ at 0.5C.

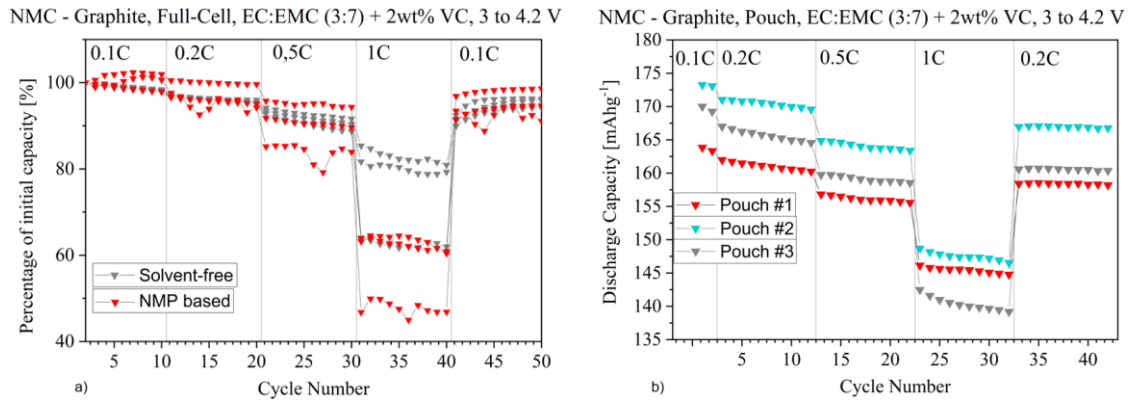


Figure 3. a) Constant current and constant voltage (CCCV) rate capability test at 0.1C, 0.2C, 0.5C and 1C for 3.0 - 4.2 V of solvent-free cathodes and NMP-based cathodes in coin cells, same anode (graphite) b) CCCV rate capability test at 0.2C, 0.5C and 1C for 3.0 - 4.2 V of solvent-free cathodes in pouch cells, full cell with 260 mAh [1]

4 CONCLUSION

The electrochemical data of solvent-free cathodes in coin and as well pouch cells show that the rate capability and the capacity retention are equivalent to conventionally manufactured cathodes. In NMC622 – graphite full cells discharge capacities at C/2 (10th cycle) of up to 137 mAh/g in coin cells (5.7 mAhcm⁻²) and up to 163 mAh/g in pouch cells (3.9 mAhcm⁻²) were achieved. The results show that solvent-free manufacturing of cathodes leads to reduced economical and ecological impact without impairing the electrochemical properties.

5 ACKNOWLEDGEMENT

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