Special Synergy between Electrolyte Additives and Positive Electrode Surface Coating to Enhance the Performance of Li[Ni_{0.6}Mn_{0.2}Co_{0.2}]O_{2}/Graphite Cells

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1) Uncoated NMC622 – compared to Al_{2}O_{3}-coated NMC622 - performs quite poorly in a standard electrolyte and;
2) Under identical conditions Al_{2}O_{3}-coated NMC performs quite well, still retaining >85% of 1 C capacity after 1000 cycles.

Hence Al-based materials, especially Al_{2}O_{3}, appear to be a good choice for surface coatings on positive electrode materials. In this study, the impact of an Al_{2}O_{3} coating on NMC622 on the cell performance has been studied in comparison with uncoated NMC622.

Electrolyte additives are known to form a modified SEI at electrode surfaces. The modified/improved SEI can improve calendar life, cycle life and yield better safety as well. Ma et al. studied...
the effect of some electrolyte additive blends on the performance of Li[Ni0.8Mn0.2Co0.2]O2 (NMC442)/graphite cells.12 They showed that the additive blend 2 wt% prop-1-ene-1,3-sultone (PES) + 1 wt% methylene methyl disulfonate (MMD) + 0 wt% tris (trimethylsilyl) phosphate (TTPS) (PES 211) helped suppress impedance growth and improve capacity retention when NMC442/graphite cells were cycled between 3.0 V and 4.4 V. Nie et al. showed that 1 wt% PBF improved the electrochemical properties of both NMC111/graphite and NMC442/graphite cells.13 In this study, the following electrolyte additive blends were selected for comparison: 1 M LiPF6 ethylene carbonate:ethyl methyl carbonate (3:7 by weight) = control, 2 wt% vinyl carbonate (VC) = control, 2 wt% prop-1-ene-1,3-sultone (PES) = control, 2 wt% prop-1-ene-1,3-sultone (PES) + 1 wt% methylene methyl disulfonate (MMD) = control, 2 wt% trimethylsilyl phosphate (TTPS) in control (PES 211) and 1 wt% pyridine boron trifluoride (PBF) in control.

Using additive blends along with positive electrode surface coatings should improve electrochemical properties. Xia et al. showed that NMC442/graphite cells with a 3 wt% LaPO4-coating on the surface of the NMC442 resulted in reduced electrolyte oxidation and better capacity retention, when cycled to 4.5 V when certain electrolytes were used.14 They also showed that for those NMC442/graphite cells, the benefits of additives were greater than those of the LaPO4 surface coating. However, Song et al. showed that a 3 wt% coating of LaPO4 on Li[Ni0.8Mn0.2Co0.2]O2 (NMC532) was highly effective at improving charge-discharge capacity retention in the absence of additives.15 Apparently, the electrochemical properties depend on the nature of the coating, the presence or absence of additives and also on different experimental conditions.

Nelson et al. reported the importance of impedance growth caused by electrolyte oxidation.3 They studied the LaPO4-coated NMC442/graphite cells using simultaneous cycling and electrochemical impedance spectroscopy (EIS) experiments in the presence of control electrolyte + 2 wt% prop-1-ene-1,3-sultone (PES) + 2 wt% 1,3,2-dioxathiolane-2,2-dioxide (DTD) + 2 wt% tris (trimethyl-silyl) phosphate (TTPS), called PES222. They found severe impedance growth in coated cells when aggressively cycled above 4.4 V. In their studies, the behavior of coated and uncoated cells differed with changes in the upper cutoff voltages. Hence the cumulative effect of a particular combination of a positive electrode material, a surface coating and an additive blend at a particular upper cutoff voltage is unique and can only be determined from experimental observations. This article explores the effectiveness of an Al2O3 coating in presence of selected electrolyte additives in NMC622/graphite cells.

**Experimental**

The control electrolyte was prepared by dissolving 1 M lithium hexafluorophosphate (LiPF6) (BASE, ≪20 ppm HF) in a solvent mixture containing 3:7 wt% ratio of ethylene carbonate and ethyl methyl carbonate (EC/EMC from BASF, 99.99%). Desired amounts (1 or 2 wt%) of one or more selected additives (VC from BASF, 99.99%; PES from Lianchuang Pharmaceutical, 98.2%; PBF from 3 M, ≻99%) were added to the control electrolyte to obtain the additive blended electrolytes. The electrolyte with 2% PES + 1% MMDs (from Tinci Materials Technology, ≻98.7%) + 1% TTPS (Sigma-Aldrich, ≻95%) is referred to as “PES 211”. Figure S1 shows the chemical structures of the electrolyte additives used in this work.

To prepare uncoated NMC622, lithium carbonate and a mixed Ni-Mn-Co oxo hydroxide were homogeneously blended in a vertical single-shaft mixer by a dry powder mixing process. The blend ratio was targeted to obtain Li1.01(Ni0.6Mn0.2Co0.2)0.99O2, which was verified by inductively coupled plasma (ICP) mass spectroscopy. The mixture was then sintered in a tunnel furnace in dry air at 900 °C for approximately 10 h. After sintering, the sample was milled in a grinding machine to a mean particle size of around 12 micrometers (μm). Al2O3-coated NMC622 was prepared from the uncoated NMC622 according to the procedures outlined in PCT patent application PCT/IB16/050257.

Uncoated and coated NMC622/graphite pouch cells of 402035 size with respective capacities of 200 and 220 mAh were used in this study. The cells with NMC622 positive electrode material that was coated with 0.2 wt% of Al2O3 are referred to as coated-NMC 622 cells. Both types of cells were balanced for 4.45 V operation. The uncoated and coated positive electrodes comprised of 92:4:4% by weight of active material, PVDF binder and carbon black, respectively. The negative electrodes were made from artificial graphite, carbon black and CMC/SBR binder in a 96:2:2 weight ratio. The dry pouch cells were assembled in a dry room at Umicore, vacuum sealed without any electrolyte and then shipped to our laboratory in Canada for electrolyte filling and testing.

**Electrolyte filling.**—The pouch cells were opened and dried at 80 °C under vacuum for about 14 h to remove residual water before filling with electrolyte. The cells were then taken into an argon-filled glove box for electrolyte filling and subsequent vacuum sealing. The argon used in the glove box was ultra high purity grade (Praxair UHP grade 99.999% purity) that was further treated by the glove box purification system to reduce water and oxygen levels below 1 ppm. 0.75 mL (0.90 g) of the electrolyte was poured into the pouch cells and they were vacuum-sealed at −90 kPa (gauge pressure) using a compact vacuum sealer (MSK-115A, MTI Corp.). After the electrolyte filling and sealing, the volume of the cell was measured using Archimedes’ principle as explained in the upcoming section.

**Formation steps.**—Formation step 1: The cells were held at 1.5 V at 40 °C for 24 h to ensure sufficient wetting. Then, they were charged to 3.5 V at C/20 at 40 °C. After formation step 1, the volume of the cell was measured again. The difference in the volumes measured before and after formation gives the volume of any gas generated. Then, the cells were transferred into the glove box, opened to release any gas generated and vacuum sealed again.

Formation step 2: In this step, the sealed cells were charged again from 3.5 V at C/20 to 4.4 V, held at 4.4 V for 1 h and then discharged to 3.8 V. After reaching 3.8 V, the volume of the cells were measured again, the cells were transferred into the glove box, degassed and vacuum sealed again. After the two degassing processes, electrochemical impedance spectroscopy (EIS) testing was conducted at 10 °C with the cells at 3.8 V.

**EIS measurement.**—Impedance spectra were measured on the pouch cells before and after cycling at 3.8 V at a temperature of 10 ± 0.1 °C using Biologic VMP-3 system. Alternating current (AC) impedance spectra were collected with ten points per decade from 100 kHz to 10 mHz with a signal amplitude of 10 mV.

**Ultra high precision charger (UHPC) experiment.**—Selected cells were cycled using the ultra high precision charger (UHPC) at Dalhousie University between 2.8 and 4.4 V at 40 ± 0.1 °C using currents corresponding to C/20 for 16 cycles.

**Cycle-store test.**—Figure 2 shows the protocol used for cycle-store testing. Initially, the pouch cells were kept in temperature boxes at 40 ± 0.1 °C. The cells were first cycled between 4.4 V and 2.8 V twice at a rate of C/20 and then at C/5 for the subsequent cycles up to a desired number of cycles. During the C/5 rate cycling, the cells were left at open circuit for 24 h at the top of every charge (4.4 V), while their potentials were recorded.

**Gas measurement.**—The gas volume (Ex-situ) of the cells were measured after formation steps and cycling17 with Archimedes’ principle. First, the sealed pouch cells were suspended under a Shimadzu balance (AUY200D) using a fine wire hook. The cells were immersed in a beaker of de-ionized “nanopure” water (18.2 MΩ) maintained at 20 ± 1 °C. The weight of the cell was measured while immersed before and after gas generation. The difference in the weights (Δm) of the cell before and after gas generation is equivalent to the volume change (ΔV) caused by change in the buoyant force. The weight
change, $\Delta m$, is related to the volume change, $\Delta V$, through the density of the fluid ($\rho$) as shown in Equation 1.

$$\Delta V = -\frac{\Delta m}{\rho}$$  \[1\]

Results and Discussion

Figure 3 shows the voltage-specific capacity profiles (V vs Q) of half-cells made from coated (red solid line) and uncoated (black dotted line) NMC622 active materials. Coin cells were made from coated or uncoated NMC622 positive electrodes and Li negative/reference electrodes. 1 M LiPF$_6$ in 1:2 (w:w) ethylene carbonate: diethyl carbonate (EC:DEC) mixture was used as the electrolyte for the coin cell study. The cells were cycled between 3.0 V and different upper cutoff potentials: 4.2 V, 4.4 V or 4.6 V vs Li/Li$^+$. Figure 3 shows that the specific capacity increased as the upper cutoff potential increased reaching a maximum of $\sim$250 mAh/g for the first charge to 4.6 V. The reversible capacity was 225 mAh/g for the cells tested to 4.6 V.

The difference in measured specific capacities between coated and uncoated NMC622 cells were negligible, as expected based on the small weight fraction (0.2% of Al$_2$O$_3$) in the coating.

Figure 4 shows the data collected from the UHPC experiments (2.8–4.4 V at C/20 and 40°C) on coated NMC622/graphite pouch cells with the selected electrolyte additive blends. In Figure 4, the legends corresponding to black dots, blue squares, red triangles, green crosses and magenta triangles represent the electrolyte additives - control, 2% VC, 2% PES, PES211 and 1% PBF respectively. Figure 4a shows the coulombic efficiency (CE) versus cycle number for the cells with the studied additives. Cells with the additive combinations showed better CE than those of the cells with control electrolyte. Cells with the additive PES211 exhibited the best CE (0.9986 at the 16th cycle) suggesting that cells with PES211 would have the longest lifetime. A CE of 0.9986 at cycle 16 under these test conditions is the best ever measured in this laboratory on any NMC/graphite cell.

Figure 4b shows the discharge capacity versus cycle number for the first 16 cycles. Cells with PES211 showed the best capacity retention. Figure 4c shows the charge end point capacity versus cycle number for the first 16 cycles. For cells with all the studied additives, the charge end point capacity increased with increasing cycle number indicating electrolyte oxidation. The charge end point capacity slippage in cells with the additive blends was less than that of cells with control electrolyte suggesting that additive blends help mitigate electrolyte oxidation at the positive electrode interface. Cells with the additive blend PES211 exhibited the lowest increase in charge end point capacity. Figure 4d shows the voltage difference between the average charge and discharge voltage ($\Delta V$) versus cycle number for the first 16 cycles. $\Delta V$ decreased during the first 5 cycles and became almost constant after that for all cells.

Figure S2 shows the data for the first 16 cycles collected from the UHPC experiments (2.8–4.4 V at C/20 and 40°C) on uncoated NMC622/graphite pouch cells with the selected electrolyte additive blends. The trends in the UHPC data corresponding to uncoated NMC622 cells were almost similar to those of cells with the other additive blends. The trends in the UHPC data corresponding to uncoated NMC622 cells were almost

![Figure 2](https://example.com/figure2.png)

Figure 2. Schematic of the testing method used for the cycle-store test.

![Figure 4](https://example.com/figure4.png)

Figure 4. Data collected on the ultra-high precision charger (UHPC) (a) Coulombic efficiency (CE), (b) discharge capacity, (c) Charge end point capacity, and (d) $\Delta V$ versus cycle number for Al$_2$O$_3$-coated NMC622/graphite cells with selected electrolyte additives as indicated.
Figure 5. Coulombic efficiency (CE) versus cycle number for Al₂O₃-coated and uncoated NMC622/graphite cells with some selected electrolyte additives.

same as those of the coated NMC622 cells. However, it is difficult to compare the CE measurements on the coated and uncoated cells, respectively, from Figures 4a and S2a. Figure 5 shows the CE versus cycle number in a format that allows easy comparison between coated and uncoated NMC622 cells with the same electrolyte additives. Figure 5 shows that the CE of coated NMC622 cells were always better that of the corresponding uncoated cells. However for cells with PES211, the difference in the CE between coated and uncoated cells is very small suggesting that the use of PES211 outweighs the advantage brought by the Al₂O₃ coating. In fact, the uncoated NMC622 cells with PES211 have a higher CE than the coated cells with any of the other electrolytes.

Figure 6 shows a summary of UHPC data presented in Figures 4 and S2. Figure 6a shows 5 sets of comparative histograms of coulombic inefficiency (CIE), which is equal to 1-CE, of the cells corresponding to the studied additive blends. The CIE reported in Figure 6a was the average of the CIE of the last 5 cycles. In all the 5 sets of histograms, which correspond to the 5 selected additives, the CIE of the coated cells (red) are better than that of the uncoated cells (blue) indicating the positive role played by the Al₂O₃ coating in reducing the parasitic reactions. Upon comparing the effect of the additive blends, PES-containing additive outperformed all others, especially PES211.

Figure 6b shows the charge end point capacity slippage data corresponding to all the additive blends. The charge end point capacity slippage per cycle (mAh/cycle) was determined by calculating the slope of the best fit line to the final five points (11-15 cycles) of the charge end point capacity versus cycle number graphs (Figures 4c and S2c). Once again, all the cells with coated NMC 622, irrespective of the additive blends, had smaller (better) charge end point capacity slippage than those of the uncoated cells. In the presence of the electrolyte additive combinations, the charge end point capacity slippage per cycle was lower than that of the control electrolyte. Cells with PES211 exhibited the least charge end point capacity slippage per cycle among all the additives.

Figure 6c shows the increase in ΔV per cycle corresponding to cells with all the additive blends. ΔV per cycle was determined by calculating the slope of the best fit line to the final five points (11-15 cycles) of ΔV vs. cycle number graphs (Figures 4d and S2d). Figure 6c shows that the increase in ΔV (impedance growth) for both coated and uncoated cells is very small during the UHPC cycling irrespective of the nature of the additive blends. The distinction between the increase in ΔV between cells with coated and uncoated NMC622 is negligible.

Figure 6d shows the discharge capacity loss per cycle (mAh/cycle), which was determined by calculating the slope of a best fit line to the final five points (11-15 cycles) of the discharge capacity vs. cycle number graphs in Figures 4b and S2b. The discharge capacity loss per cycle for coated cells tested with control, 2% VC and 1% PBF was clearly lower (better) than that of uncoated cells. However, when PES-based additives were used, the discharge capacity loss per cycle was almost same for coated and uncoated cells and the capacity loss per cycle was smaller than for cells with the other additive blends.

Figures 7a and 7b show the impedance spectra of the coated NMC622 cells after formation and after UHPC experiments (16 cycles) respectively. The diameter of the semicircle represents the sum of the charge-transfer resistances, Rct, of the positive and negative electrodes. After the formation step, the impedance of the coated cells tested with any additive blend was higher than that of the control electrolyte. However, after the UHPC experiments, PES-based additives helped lower the impedance compared to that measured after formation whereas other additives did not affect the impedance
Figure 6. Summary of the high precision data including: (a) Coulombic inefficiency (CIE) average from the last five cycles; (b) The charge end point capacity slippage was calculated from the slope of a best fit line to the final five data points (cycles 11-15) of the charge end point capacity versus cycle number graph; (c) Slope of $\Delta V$ versus cycle number (cycles 11-15); (d) The discharge capacity loss was calculated from the slope of a best fit line to the final five points (cycles 11-15) of the discharge capacity versus cycle number for both coated and uncoated NMC622/graphite cells with different electrolyte additives as indicated.

Figure 7. Impedance spectra for (a) coated NMC622/graphite cells after formation; (b) coated NMC622/graphite cells after UHPC cycling; (c) uncoated NMC622/graphite cells after formation and (d) uncoated NMC622/graphite cells after UHPC cycling.

Figure 8. Volume of gas evolved during: (a) formation step 1; (b) formation step 2; (c) after UHPC cycling at 40.0 ± 0.1°C for both coated and uncoated NMC622/graphite cells with various electrolyte additives as indicated.
gas evolution after formation step 2 is very small except in the cells having 2% VC. This gas is primarily CO₂ and is created at the positive electrode. Figure 8c shows that none of cells exhibited any significant gas evolution after UHPC cycling. Overall, Figure 8 suggests that the evolved gas volume is almost same for both coated and uncoated NMC622/graphite cells after each step.

Figure 9 shows the discharge capacity of the NMC622/graphite cells measured between 2.8 and 4.4 V for about 150 cycles using the cycle-store protocol (See Figure 2). Figures 9a, 9b, 9c, 9d and 9e correspond to the cells containing control electrolyte (black), 2% VC (blue), 2% PES (red), PES211 (green) and 1% PBF (magenta) respectively. In each of the panels in Figure 9, the closed and open circles correspond to the coated and uncoated NMC622/graphite cells respectively. In all cases, the cells with coated NMC622 had better capacity retention than the uncoated ones indicating the advantage of the Al₂O₃ coating. Comparing the effect of additive blends, those cells containing either control electrolyte or 2% VC exhibited poor capacity retention with cells showing <70% capacity before 70 cycles. On the other hand, the cells with PES-containing additives and 1% PBF exhibited much better capacity retention especially in cells with coated NMC622. In particular, the capacity retention of cells with both PES211 and coated NMC622 was the best (>80%) even after 160 cycles.

Figure 10 shows a comparison between the coulometric efficiency results in Figure 5 and the long-term cycling results in Figure 9. Figure 10a shows the CE measured at cycle 16 from Figure 5 and Figure 10b shows the time (in months) for the cell capacity to reach 90% of the initial capacity during the long-term cycling protocol applied in Figure 9. The close correlation between Figures 10a and 10b is obvious which points to the value of coulomnic efficiency as a predictor of cell lifetime as has been proposed before. In addition, Figure 10 shows that uncoated NMC622/graphite cells with PES211 are virtually as good or better than coated NMC622/graphite cells with any other electrolyte. This really shows the effectiveness of the PES211 blend.

Figure 11 shows a summary of the voltage drop (V_{drop}) of the tested cells during the storage portion of the cycle-store testing between 2.8 and 4.4 V at 40°C. Figure 2 shows the typical voltage relaxation during the storage periods. Parasitic reactions at the positive electrode such as electrolyte oxidation are responsible for the long time (>1 h) voltage relaxation while direct current (DC) cell resistance is the source of the short time (<15 min.) voltage relaxation. Figure 11 does not distinguish between these two contributions. Different legends in Figure 11 have been used to indicate V_{drop} measured after a certain number of cycles: 1, 15, 35, 70, 90 and 130. V_{drop} is generally stable for all cells over the first 35 cycles except for cells with control electrolyte. Cells with coated NMC622 that use PES211 have the smallest and most stable V_{drop} of all cells. Cells with uncoated NMC622 that use PES211 have the next smallest and next most stable V_{drop} of all cells. These results are even more indicative of the stability of the PES211 electrolyte system in combination with coated NMC622. Realize that those cells have been exposed to higher potentials than the others cells (smaller V_{drop}) during the storage periods which would accelerate electrolyte oxidation during the storage period if the additive system had not been effective.

Figure 12 shows the impedance spectra collected after cycle-store testing between 2.8 V and 4.4 V at 40°C. Figures 12a and Figure 12b show the impedance spectra of the cells with coated and uncoated NMC622 respectively. The impedance of the coated NMC622 cells were much smaller than the corresponding cells with uncoated NMC622 implying that the Al₂O₃ surface coating was effective in
Figure 10. a) Coulombic efficiency measured at cycle 16 from Figure 5 for the coated (red bars) and uncoated (blue bars) NMC622/graphite cells; b) number of months of testing for the cells to reach 90% of initial capacity during the charge-store testing described by Figure 9 using the protocol shown in Figure 2. The electrolytes labeled A, B and C are 2% PES, PES211 and 1% PBF, respectively. The cells tested in Figures 9 and 10b are duplicate cells of those tested in Figure 5.

Figure 11. Summary of the voltage drop (V\textsubscript{drop}) for both coated and uncoated NMC 622/graphite cells with selected electrolyte additives during “cycle-store” testing between 2.8 and 4.4 V at 40\textdegree C after the cycles indicated.

Figure 12. Impedance spectra collected after “cycle-store” testing between 2.8 and 4.4 V at 40\textdegree C for (a) NMC622 coated/graphite cells and (b) NMC622 uncoated/graphite cells. The spectra were collected after the number of charge discharge cycles displayed in Figure 9. The spectra were collected at 10\textdegree C with the cells at 3.8 V.

controlling the impedance. The impedance of the uncoated cells was quite small when tested with PES-containing additives especially PES 211. A change in the scale of the graph is required to observe the changes in the cells with 2% PES and PES211. To this end, Figure 13 shows the impedance spectra collected after formation, after UHPC cycling and after cycle-store testing for the cells tested with the PES211 additive. Both uncoated and coated NMC622/graphite cells show relatively large impedance after formation, which decreases after the UHPC cycling and cycle-store test. The results in Figures 12 and 13 indicate that PES211 could control impedance growth even in the absence of an Al\textsubscript{2}O\textsubscript{3} coating. However, the development of the low frequency lobe in the impedance spectrum of the uncoated NMC622/graphite cells is indicative of developing problems with the positive electrode/electrolyte interface. Ma et al. showed that the impedance of NMC442/graphite cells with PES211 decreased slightly during 500 cycles. This result is in good agreement with the results found here for NMC622/graphite cells tested with PES211. Clearly impedance growth can be impacted by selective surface coating or additives or the combination of both.

Figure 14 shows comparative histograms for the gas volume and the impedance measured after cycle-store testing. Figure 14a shows that coated NMC622 cells with 2% PES, PES211 and 1% PBF produced more gas than cells with the other additives (control and 2% VC). Figure 14a shows that NMC622/graphite cells with PES211 produced the most gas, however, these cells were tested for longer periods of time and were exposed to a higher potential during the storage periods (see discussion surrounding Figure 11) than control cells and cells with 2% VC. Coated NMC622 cells with PES211 produced less gas after 120 cycles than uncoated NMC622 cells with PES211 after only 70 cycles. This again, points to the value of the coating when
Figure 14. (a) Gas evolution and (b) $R_{ct}$ collected after “cycle-store” testing between 2.8 and 4.4 V at 40 °C for both coated and uncoated NMC622/graphite pouch cells. The numbers above the bars indicate the cycle number after which the measurements were made.

PES211 is used. However, cells with 1% PBF showed the opposite trend, pointing to the complexities of the coating/additive interaction which will, frankly speaking, take years to understand. Figure 14b shows a summary of the impedance data collected after cycle-store test, which were introduced in Figure 12. Figure 14b once again reiterates that PES-containing additives can suppress impedance growth much better than the other additives.

Conclusions

The electrochemical performances from continuous cycling tests such as discharge capacity, coulombic efficiency, voltage drop and impedance growth were systematically investigated on Al$_2$O$_3$-coated and uncoated NMC622/graphite pouch cells using five different electrolytes: control, 2% VC, 2% PES, 2% PES, and uncoated NMC622/graphite pouch cells. The numbers above the bars indicate the cycle number after which the measurements were made.

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