Highlighting the Importance of Full-cell Testing for High Performance Anode Materials Comprising Li Alloying Nanowires

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Abstract

Herein, the electrochemical performance of directly grown Ge nanowire anodes in full-cell Li-ion configurations (using lithium cobalt oxide cathodes) are examined. The impacts of voltage window, anode/cathode balancing and anode preconditioning are assessed. The cells had a useable upper cut-off of 3.9 V, with a higher voltage cut-off of 4.2 V shown by SEM analysis to lead to Li plating on the anode surface. The rate performance of Ge NW anodes was shown to be boosted within full-cells compared to half-cells, meaning that existing studies may underestimate the rate performance of alloying mode anode materials if they are only based on half-cell investigations. The capacity retention of the full-cells is lower compared to equivalent half-cells due to progressive consumption of cyclable Li. This phenomenon is demonstrated using a parallel anode and cathode delithiation approach that could be extended to other full-cell systems. The findings stress the importance of testing promising anode materials within full-cell configurations, to identify specific capacity fade mechanisms that are not relevant to half-cells and aid the development of higher energy density storage systems.

Keywords: Li-ion batteries, full-cells, nanowires, alloying materials, germanium

Introduction

Li-ion batteries (LIBs) are the dominant energy storage systems underpinning electric vehicles and portable handheld devices due to their higher energy densities (Wh kg⁻¹) compared to other established battery chemistries.¹⁻⁵ For this dominance to continue, active material improvements are sought to facilitate improved energy density and high-rate performance.⁶,⁷
In terms of the anode composition, graphite continues to be the primary active material of choice due to its impressive cycling stability, low irreversible capacity losses and competitive cost. However, the specific capacity of graphite is a modest 372 mAh g\(^{-1}\), which has motivated research into higher capacity anode materials. The ability to store more Li per unit mass (i.e. higher gravimetric capacity) at the anode has been facilitated by conversion and alloying materials, which undergo lithiation differently to the intercalation mode associated with graphite. While conversion mode and alloying materials both offer potential for capacities that are multiples of those achievable with graphite, they are not without issues. Conversion materials offer high initial capacities, but with capacity fade profiles that are typically unsuited to practical devices. The performance hurdles associated with alloying mode materials (e.g. Si, Ge and Sn) are primarily related to the large volumetric contraction /expansion caused by lithiation/delithiation which can lead to significant material pulverisation. However, optimisation of these alloying materials through nanostructuring has been shown to facilitate 100s and even 1000s of cycles with minimal capacity fade.

A particularly interesting set of nanomaterials for LIBs are Li-alloying nanowires (NWs) that can be directly grown from current collector substrates. This material arrangement removes the requirement for conductive additives and binders, increasing the gravimetric efficiency of the anode and offering potential for major full-cell (FC) energy density improvements. A variety of NW geometries (including pure Si, pure Ge, Si/Ge alloy, branched NWs, coated NWs) have been illustrated as high specific capacity materials. Significantly, new binder-free alloying material architectures have also recently been shown to be compatible with higher areal capacities required for practical devices. The preparation of alloying materials in NW form has been shown to allow mitigation of the extended cycling related material pulverisation typically associated with bulk forms of these materials. As a result, NWs have also been extensively studied as model
systems for fundamental lithiation processes. For example, Liu et al. demonstrated reversible nano-pore formation within Ge NWs during in-situ TEM lithiation and examined this phenomenon as a function of repeated lithiation/delithiation. Significant material restructuring has also been exhibited at an electrode-scale with the progressive formation of a porous Ge nano-filament network across the electrode surface observed for Ge NW based electrodes. This porous network formation was seen as a key facilitator of long-term cycling as it ensures that the active material remained contacted to the current collector, thus limiting delamination and pulverisation.

To date, the majority of studies investigating the electrochemical properties of high capacity alloying NW anode materials have been undertaken in half-cell (HC) geometries (i.e. versus a Li counter/reference electrode). While these approaches allow attainable specific capacities and fundamental lithiation mechanisms to be examined, the excess Li source afforded by the bulk Li counter electrode does not truly reflect the finite source of Li ions associated with typical cathodes used in conventional FCs. Within FCs, the balancing of cathode (positive) and anode (negative) capacities (P:N ratio) is of critical importance for practical applications as any excess cathode requirement is heavily penalized from a weight perspective, potentially leading to a negation of the weight saving benefit of the advanced anode. This is of paramount importance for alloying materials, which are known to undergo large irreversible processes (particularly during early cycles) related to SEI formation, that limit the initial coulombic efficiencies (CEs) for alloying materials in HCs. While this is not a major issue for HCs, this ‘lost’ Li associated with SEI formation must be factored in to FCs when considering the anode/cathode capacity balancing. As a result, prelithiation protocols have been examined by a number of research groups, to attain anodes with preformed SEI layers or sacrificial Li that can lead to higher initial CE values within a FC. However, continued SEI formation (beyond the prelithiation phase) during FC operation will lead to a
decrease in cyclable Li, leading to faster capacity fade in FCs compared to their HC analogues.

Despite the immense potential of Ge and Si materials, in-depth studies assessing practical FCs requirements for these materials are limited. While a number of studies have examined FCs containing Si and Ge based anodes, the influence of anode mass loading (and related cathode/anode balancing), FC voltage windows, preconditioning cycles, full-cell performance and Li inventory depletion have not been thoroughly examined.

In this report we examine the performance of Ge NW based FCs vs fixed capacity lithium cobalt oxide (LCO) commercial cathodes (i.e. each cathode electrode used had the same capacity). Initially, the impact of cell voltage window and electrolyte composition on determining performance/capacity retention were assessed for different P:N ratio FCs. We show that capacity balanced anode/cathode full cells (i.e. P:N 1) show marked capacity fade compared to their analogous HCs and also compared to FC with a larger capacity excess (P:N >2). By examining the cyclable Li content as a function of cycle number (through separate HC delithiation of the anode and cathode as a function of cycle number), Li inventory losses could be tracked as a function of cycle number and initial P:N ratio. Prelithiation of the anodes was examined for P:N 1 cells, with the optimal number of preconditioning cycles found to be 50. While this figure is not practical for commercial applications, it illustrates that SEI formation needs to be examined within FCs rather than HCs to fully understand the formation of self-limiting stable SEI layers. This study aims to stimulate additional studies into FC pairings for promising alloying mode anode materials, to examine FC specific capacity fade issues not typically seen in conventional HCs and move high capacity anode materials closer to practical uptake.

Materials and Methods

Anode Fabrication
Full cells consisted of geometrically matched circular anode and cathode electrodes with areas of 0.64 cm². The Ge NW anodes were produced via our previously published rapid hotplate growth method. Briefly, this involved thermal evaporation of a 2 nm Cu catalyst layer onto stainless steel current collector substrates. Following this, the substrates were placed on a hotplate at 430 °C, covered with a stainless steel heat sink confiner through which 25-75 μl (depending on the desired anode loading) of diphenylgermane (Gelest) was injected. The substrates were removed from the heat after 5 minutes, allowed to cool and weighed to determine the mass of Ge NWs grown. This mass was used to determine the applied currents for testing with 1C taken as 1384 mA g⁻¹.

Cell Assembly

All specific capacities are given in terms of the anode mass for the various tests. The LCO cathodes were purchased as electrode tapes from NEI corporation and had an average active mass of 6.2 mg cm⁻², corresponding to a capacity of 868 μAh per cm² or 555.52 μAh for the area used in our full cells (0.64 cm²). The use of a cathode of fixed capacity (i.e. each cathode had a capacity of 555.52 μAh in every test, based on a theoretical capacity of 140 mAh g⁻¹) allowed the influence of various parameters such as anode mass loading (P:N capacity ratio), electrolyte composition and testing protocol to be accurately assessed. The capacity ratio for the cathode anode P:N is quoted for each FC test (these values are ± 0.1). Full cell assembly was conducted using two electrode Swagelok type cells with the anode as counter and reference. Celgard separators impregnated with a carbonate based electrolyte (1.0 M lithium hexafluorophosphate (LiPF₆) solution in ethylene carbonate and diethyl carbonate, battery grade Aldrich) with or without 3% or 10% by weight vinylene carbonate (97 % Aldrich). The full-cells were tested over two voltage ranges i) ‘STD limits’ 3.9 V - 2.8 V and ii) wider limits ‘4.1- 2.5 V’.
For Li inventory tests, FCs were prepared as above. Following the predetermined number of cycles, the anode and cathode were removed. In all cases, the FCs were disassembled immediately once the cell had reached a charged state (at 3.9 V) to avoid self-discharge. The recovered electrodes (cathode and anode) were assembled in fresh HCs vs Li and the remaining Li was extracted by cycling the HCs at C/10 to 1.5 V and 4.2 V for the Ge anode and LCO cathode respectively.

Results and Discussion

Figure 1: a) Voltage profiles for Ge anode and LCO cathode in HC configurations. b) DQ/DV plot for Ge anode in HC showing the primary lithiation and delithiation features for the first cycles. c) Initial FC voltage profile for charge and discharge of Ge/LCO FC cycled between 3.9 V and 2.8 V. d) DQ/DV profiles for LCO cathode within a HC (blue) and Ge/LCO FC (red) illustrating the potential shift between charge and discharge processes. e) Voltage profiles for charge and discharge of Ge/LCO FC (P:N 2) cycled between 4.1 V and 2.4 V with the cut-off for Li plating shown with the green dotted line. f) DQ/DV plot for e) illustrating the sharp Li plating feature shown in e).

To determine the appropriate voltage window for the Ge/LCO full cells, HCs of the individual components were initially investigated. The first charge and discharge profiles for the anode and cathode are presented in Figure 1a) with all voltages vs Li/Li⁺, illustrating the typical cycling ranges for the anode and cathode HCs. The primary lithiation for the Ge anode occurs
below 0.375 V, with delithiation starting at >0.5 V as shown in the differential capacity plot (Figure 1b). FC lithiation/delithiation was investigated within a voltage range of 3.9 V to 2.8 V (with the Ge anode as reference/counter electrode). The major charging plateau occurs at > 3.5 V, with an average discharge voltage of ~ 3.5 V. Comparing the DQ/DV profiles of the HC cathode and FC (Figure 1d), it can be seen that there is a voltage shift of approximately -0.3 V for corresponding lithiation/delithiation processed related to the cathode. To investigate the impact of a higher voltage cut-off for charge and the potential occurrence of Li plating within the full cell-architecture, a FC with a P:N of >2 was cycled between 4.1 and 2.5 V. From the initial charge profiles, it can be seen that the charge up to 3.9 V is similar to that seen in Figure 1c, however, an additional process occurs above this value that can be attributed to Li plating on the anode that is of critical importance for practical FCs.55 The differential capacity plot (Figure 1 f) illustrates the extremely sharp peak at ~3.95 V that is attributed to Li plating. It can also be seen that this process is at least partially reversible as illustrated by the broad peak between 4.1 to 3.9 V on the discharge profile. Comparative SEM analysis of (P:N 2) full-cells after a single charge to 3.9 V and 4.2 V showed clear evidence of Li plating for the latter (Figure S1). The occurrence of Li plating is particularly prevalent in the case of high P:N cells and its impact on longer term cycling performance is examined within Figure 2.
Figure 2: Extended charge/discharge testing for Ge/LCO FCs at a C/2 rate for P:N =2 (blue), P:N= 1.5 (Grey) and P:N= 1 (Red). a),b),c) correspond to FCs cycled in the standard potential window (3.9 V - 2.8 V) with electrolyte containing no VC, 3% VC and 10% VC (by weight) respectively. d),e),f) are FCs cycled using a wider potential window (4.1 V - 2.4 V) with electrolyte containing no VC, 3% VC and 10% VC respectively.

The cycling stability of FCs was compared as a function of the electrolyte composition, cycling range and P:N ratio. It is worth stating that the use of a P:N ratio of >2 is certainly not suitable for a practical FC due to dilution of the overall cell energy density, however, it does allow the impact of SEI formation and cathode excess on performance and capacity fade mechanisms to be examined in detail. The point at which the incorporation of excess LCO into the FC arrangement offsets the gravimetric savings for Ge is at a P:N ratio of approximately 1.3. The capacities are given as a function of anode active mass as an indicator of effective anode utilization. In Figure 2 a), FCs with different P:N and an electrolyte containing no VC additive were examined. It can clearly be seen that the FC capacity rapidly decreases for the P:N 1 and 1.5 cells. From previous studies on Ge NWs, the addition of VC to the electrolyte is critical for long term cycling stability. This factor explains the poorer performance compared to the VC containing cells (even for the high P:N ratio cell). The addition of 3 % VC to the electrolyte had a large impact on the cycling stability (Figure 2b), with the P:N 2 cell illustrating 88 % capacity retention after 100 cycles. At lower P:N ratio, the cycling performance is not as stable.
This is partially due to the Ge anodes, which show diminished capacity retention particularly at higher mass loadings (i.e. P:N 1 equivalent anodes). This phenomenon has been studied in detail and improved through current collector modifications and will be presented elsewhere. The capacity retention of the FCs in Figure 2b is contrasted with HC s with similar masses in SI Figure S2 to illustrate their respective performance. In the case of the P:N 1.5 FC, the capacity retention was 63% (compared to a 91% capacity retention for the HC) and the P:N 1 FC maintained just 27% of its capacity (compared to 55% for the half cell). Using a higher VC content within the electrolyte (10%) was not seen to significantly affect the performance (Figure 2c).

The impact of cycling within a wider voltage window was seen to be dominated by Li plating for high P:N FCs. In Figure 2d), the P:N >2 cells had an initial charge capacity in excess of 4000 mAh g⁻¹, well beyond the theoretical capacity of Ge, illustrating that a significant portion of the capacity was due to Li plating. This cell maintained a gradually diminishing capacity in excess of 1000 mAh g⁻¹ before catastrophic failure at cycle ~30. The lower P:N cells without VC also had lower long term cycling stability compared to the VC containing tests. In Figure 2e) the 3% VC containing P:N >2 cell again showed a large Li plating contribution. The cycling stability of the P:N 1.5 and 1 tests maintained 36% and 55% of their initial discharge capacities after 100 cycles. No obvious performance difference was noted with additional VC (10% weight content of the additive in the electrolyte) (Figure 2f), as also seen for the tests with narrower potential limits. The specific capacity retention is illustrated for these on the same Y axis scale in Figure S3 for clarity.
Figure 3: DQ/DV plots for 3% VC test with standard limit a) P:N 1, b) P:N 1.5, c) P:N >2 and wider limit tests d) P:N 1, e) P:N 1.5, f) P:N >2.

To further assess the electrochemical processes occurring for the various P:N ratio FCs, DQ/DV plots for the 3% VC tests were examined (Figure 3a P:N 1, Figure 3b P:N 1.5 & Figure 3c P:N 2). The locations of the primary cell charge and discharge features were similar for the three cells cycled between 3.9-2.8 V. The initial anode lithiation peak was found to occur at 3.6 V. The initial anode delithiation occurred at 3.75 V with the peak more defined for the higher P:N cells. Comparing the P:N 1 cell cycled between 4.1-2.5 V (Figure 3d) to the P:N 1 cell cycled between 3.9 V to 2.8 V, it can be seen that no substantial additional processes occur despite the elevated upper voltage cutoff. This is a strong indicator that the lack of cathode excess means that lithium plating is far less likely. Thus, for a perfectly matched Ge/LCO FC, an upper cutoff of 4.0-4.1 V is suitable. Even for a P:N of 1.5, there is evidence of Li plating in the 1st, 2nd and 5th cycles as suggested by the peaks at >4 V (Figure 3e). These processes are far more pronounced for the 3% VC P:N 2 cell and continue until the 50th cycle given the large excess of Li within the cathode (Figure 3f).
Figure 4: Capacity recovery tests for anodes cycled in FCs for 50 cycles before disassembly/reassembly of the anode within HCs for 100 cycles for anodes comprising a) P:N 1.5 and b) P:N 3. All tests are at C/2 based on the anode masses.

Given that the capacity of FCs fades faster than corresponding HCs over extended cycling, it is important to determine the mechanisms underpinning this effect. FC tests with P:N 1.5 (Figure 4a), small cathode excess) and P:N 3 (Figure 4 b), very large cathode excess) were tested for 50 cycles between 3.9 V and 2.8 V (to remove any contribution from Li plating). Following the 50 cycles, the anode was extracted from the FC and reassembled within a HC with fresh electrolyte and vs a Li counter/reference electrode. The FC capacities gradually decreased from 1000 mAh g\(^{-1}\) to < 750 mAh g\(^{-1}\) for the P:N 1.5 test (Figure 4a) over this period. The capacity of the anode immediately recovered to 1000 mAh g\(^{-1}\) when tested in the HC and maintained a noticeably more stable capacity retention profile compared to the previous FC cycling. For the P:N 3 FC (Figure 4b), the initial FC capacities were higher than P:N 1.5, and the capacity fade was much more gradual with the FC capacity still being > 1000 mAh g\(^{-1}\) after the 50 cycles. Despite this, testing in the HC still led to a slight increase in the capacity compared to the previous FC and the capacity after 100 HC cycles was higher than at the end of the FC testing. Taken in conjunction, these tests illustrate that HCs are not a good indicator of practical capacities for alloying materials. Even in the case of the P:N 3 FC tests (where a large Li reservoir is present), the measured capacities were lower than within the
This may be due to the 3.9 V upper voltage cutoff (required to prevent Li plating) preventing a complete lithiation/utilization of the anode. Furthermore, the capacity fade seen in the FC is not due to anode material degradation (as illustrated by the recovery within the FC), illustrating the importance of parallel HC and FC testing to identify the origins of capacity fade.

To monitor the impact of Li inventory depletion in light of the findings in Figure 4, where FC capacity fade was found to be largely independent of material degradation, a method was developed to extract cyclable Li from the anode and cathode for fixed P:N tests. Holtstiege et al. recently developed a three electrode cell configuration that allowed Li consuming and non-Li consuming processes to be differentiated by extracting the remaining Li from the cathode using a Li electrode. In our method (schematically depicted in Figure 5 a), the total cyclable inventory can be assessed for different numbers of cycles as the anode and cathode are

Figure 5: a) Schematic of Li inventory testing approach. b) P:N 2 Li inventory test showing FC capacity, extracted (remaining) cathode capacity, anode extracted capacity and ‘cyclable’ Li based on the sum of the cathode and anode extracted capacities. c) P:N 1 Li inventory test. Example voltage profiles of 10th cycle of P:N 2 test d) FC voltage profile e) Ge Anode delithiation and f) LCO cathode delithiation.
delithiated in separate half-cells after a given number of full-cell cycles. Thus, the total cyclable Li inventory can be measured by summing the Li extracted from the anode and cathode (assuming that there is a cathode excess present). Using this method, it is necessary to prepare different FCs for each number of cycles desired, necessitating fine control over the P:N ratios. Voltage profiles for the 10th full cell cycle for the P:N 2 ratio and subsequent anode and cathode delithiation voltage profiles are presented in Figure 5 c,d and e respectively.

In Figure 5 b), the P:N 2 Li inventory was tracked for the 1st, 5th, 10th, 25th, 50th and 100th cycle. For the cathode extracted trend (red), it can be seen that excess cyclable Li remains in the cathode after even 100 FC cycles. From an initial excess cathode capacity of ~300 µAh after the first cycle, ~150 µAh remains after 100 cycles. The cyclable Li profile experiences an overall decrease from ~450 µAh to below ~250 µAh after the 100 cycles. For a FC with a P:N ratio of 1 (i.e. anode/cathode matched) it can be seen that any excess cyclable Li from the cathode is almost entirely depleted by 10 cycles. Interestingly, after the first cycle, there is substantial capacity remaining in the cathode (~150 µAh), suggesting that complete lithiation of the anode and SEI related Li depletion occur gradually in the first ten cycles. By the 25th cycle, zero excess capacity can be extracted from the cathode and thus the remaining FC performance over extended cycles (as seen in Figure 2c) is related to all of the available Li cycling between the anode and cathode. Any further cyclable Li losses (to irreversible processes) will thus necessarily involve a loss of FC capacity.
Electrochemical preconditioning (i.e. cycling the anode in a HC prior to assembly in a FC) has been adopted as a means of reducing the Li inventory depleting impact of SEI formation within FCs.\textsuperscript{47, 56} The process is particularly important for alloying mode materials that undergo significant structural changes over a number of cycles, as the exposure of fresh surface to the electrolyte can lead to persistent new SEI formation. To assess the impact of electrochemical preconditioning on resultant FC performance, different numbers of HC conditioning cycles were performed on P:N 1 equivalent anodes as depicted schematically in Figure 6 a). The electrochemical performance shown in Figure 6 b) and summarized in Table 1 show that conditioning the anodes in HCs led to marked capacity enhancements after 200 total cycles. Even 5 conditioning cycles was sufficient to improve the FC capacity retention to 336 mAh g\textsuperscript{-1} after 200 total cycles compared to just 301 mAh g\textsuperscript{-1} for the unconditioned FC after only 100 cycles. The optimum number of conditioning cycles was found to be 50, with the resultant capacity value of 437 mAh g\textsuperscript{-1} corresponding to 86 % of the corresponding Ge NW HC. Interestingly, this coincides with the point at which the Ge NWs have formed a morphologically stable network.\textsuperscript{18} While this number of conditioning cycles is not practical, it highlights that the restructuring of alloying mode anodes and associated SEI formation cannot
be considered as a straightforward issue to be overcome. An ideal alloying material for FCs needs to reach a stable morphology (and thus limit continuing SEI formation) within the lowest number of lithiation/delithiation cycles possible. After 100 cycles the Ge NWs became more prone to delamination when the half-cell was disassembled, hence the reduction in capacity seen for this FC test (note the large capacity drop-off upon assembly within the FC due to active material loss). It should also be noted that the performance of higher mass loading Ge NWs can be enhanced through the use of a textured Cu current collector, meaning that there is scope to enhance the performance of P:N 1 tests in future by removal of the capacity fade effect seen at higher loading (Figure S4).57

<table>
<thead>
<tr>
<th>Half-Cell Capacity</th>
<th>511 mAh g⁻¹ (after 200 cycles)</th>
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<tbody>
<tr>
<td>Full-Cell Capacity (No Conditioning)</td>
<td>301 mAh g⁻¹ (after 100 cycles)</td>
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<tr>
<td>5 Cycles Conditioning</td>
<td>336 mAh g⁻¹ (after 200 cycles)</td>
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<tr>
<td>25 Cycles Conditioning</td>
<td>345 mAh g⁻¹ (after 200 cycles)</td>
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<tr>
<td>50 Cycles Conditioning</td>
<td>437 mAh g⁻¹ (after 200 cycles)</td>
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<tr>
<td>100 Cycles Conditioning</td>
<td>162 mAh g⁻¹ (after 200 cycles)</td>
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Table 1: P:N 1 capacity retention summary. Comparison of corresponding weight half-cell capacity with unconditioned and conditioned cells.

Figure 7: Rate capability tests comparisons of HCs and FCs with comparable anode massess corresponding to P:N ratios of 2, a) and 1, b).
Rate capability testing (Figure 7) showed that FCs outperformed HCs with equivalent masses and that the performance gap was particularly pronounced at higher rates. For example, at 10C the specific capacity of the P:N 2 HC was only ~30% of the FC value, while the P:N 1 HC capacity was only ~25% of the comparative FC. Following the faster rate cycling, the capacities of the HC and FC were well matched upon reverting to a C/10 rate. The improved rate performance of anode materials within full-cells has previously been noted for alloying materials and may be attributed to limited high rate delithiation of the lithium counter electrode within half-cells.\textsuperscript{30} In order to further examine the superior rate capability of the full cell configuration, electrochemical impedance spectroscopy measurements were performed on full and half cells. The resulting spectra are shown in Figure S5. The spectra indicated a significantly higher charge transfer resistance in the case of the half cell (~ 400 Ω) than the full cell (< 100 Ω). This was attributed to a greater resistance of the Li/electrolyte interface than that of the LCO/electrolyte interface, and is consistent with the superior high rate performance of the full cell. This finding suggests that conventional RCT testing in HCs that dominates the literature may be an underestimation of the real rate performance of promising anode (and cathode materials). For the advancement of novel electrode materials, it is thus critical to assess their performance within FC configurations.

**Conclusions:**

In depth investigations of Ge NW/LCO FCs as a function of voltage window, electrolyte composition, capacity excess and preconditioning have been used to identify FC specific capacity fade mechanisms for this promising anode material. In contrast to analogous HCs, capacity fade was hastened due to cyclable Li losses related to continuous SEI formation caused by material restructuring. Li inventory testing was examined using a parallel anode and cathode delithiation approach within separate half-cells, allowing the consumption of cyclable
Li to be tracked. Despite the diminished capacity retention for the FCs (particularly for capacity matched cells), the performance at high rates was significantly boosted compared to analogous HCs. Additionally, preconditioning of the anodes was found to enhance the long-term cycling stability compared to standard full-cells. The findings suggest that significant additional focus is required for novel FC pairings of anodes and cathode materials to move beyond the more superficial electrochemical understanding afforded by solely testing novel materials in HCs.

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