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Bridging the Gap Between Pouch and Coin Cell Electrochemical Performance in Lithium Metal Batteries

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In lithium metal battery research, coin cells (CC) are the most widely used laboratory instrument in academic settings. However, results thus obtained often don't translate into pouch cell (PC) performance, which is regarded as a more reliable indicator for commercial relevance. Using both experimental and computational results, we show here that the root cause lies in the pressure distribution in these two cell formats. CCs suffer from a severe pressure inhomogeneity due to the geometry of the wave spring used to apply pressure to the cell stack. Replacing the wave spring with an elastic rubber disc applies a laterally uniform force to the cell stack, resulting in a homogeneous pressure distribution. LillCu half cells and CullLiNi_{0.5}Mn_{0.3}Co_{0.2}O₂ anode-free full cells using the updated structure show performance metrics on par with chemically identical PCs while traditional CCs underperform. Our solution to this common problem retains the quick, easy fabrication of CCs while producing results comparable to the PC-level.

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Extensive research has been conducted to improve and utilize the lithium metal anode (LMA), with the promise to deliver batteries with energy densities beyond lithium-ion.¹ In common electrolytes, Li metal forms a mossy, dendritic morphology during deposition.^{2,3} The large surface area of this morphology increases the reactivity of the anode, leading to high rates of parasitic reactions, low coulombic efficiency (CE), and poor cycle life.²

Recently, the field has focused on forming and maintaining large, columnar Li deposits throughout cycling.⁴ This minimizes the Li surface area, limits side reactions, and prevents dendritic growth. Strategies such as electrolyte engineering,^{2,5} 3-dimensional anodes,⁶ or anode surface coatings⁷⁻⁹ have been studied. Interestingly, the addition of an external pressure on the cell stack has been shown to have a significant positive impact on Li morphology, especially in combination with other strategies.^{2,4,10–12} Computational results suggest that mechanical stress concentrates at dendrite tips, which drives Li creep to smooth potential dendrites.^{13,14} Moreover, extensions of the Butler-Volmer equation indicate that larger overpotentials are required to deposit Li in areas of high stress concentrations, making plating on dendritic growths unfavorable.¹⁵ However, sufficiently high current densities can overcome increased overpotentials and outpace Li creep, leading to dendritic growth. Thus, to a certain extent, the application of external pressure has a dual mechanical-electrochemical effect on lithium dendrite growth. Using this experimental and computational basis, it is reasonable to assume that external pressure is a critical variable in all lithium metal batteries (including those with 3-D anodes, surface coatings, etc), even if not explicitly studied.

Li metal batteries are generally studied either in coin cell-type batteries (CC),⁵ or pouch cell-type (PC) batteries.² CCs are made by enclosing the cell stack within a small stainless-steel case with a plastic sealing gasket. Stack pressure is roughly applied by adding stainless steel spacers and a ring-shaped wave spring to the cell stack. The CC is then crimped closed and cannot be reopened without destroying the cell. CCs are quick to fabricate and cheap, allowing researchers to efficiently create large datasets.¹⁶ However, CCs are inherently closed systems, making in situ measurements (e.g., pressure monitoring) difficult. Moreover, multiple reports show that CCs suffer from severe pressure inhomogeneities across the plane of the cell stack.^{5,12,17} Explanations of these results range

from deformations of the spacer⁵ or the case¹⁷ to the geometry of the wave spring.¹² While the root cause of these inhomogeneities remains elusive, they are likely detrimental to the performance of Li metal CCs.

PCs, on the other hand, are significantly more difficult to fabricate but allow for more variation in the exact cell structure and are considered more commercially relevant.¹⁸ In a PC, the cell stack is inserted into a laminated aluminum bag which is heat-sealed under vacuum. Once the PC is fabricated, a pressure sensor can be included in the cell stack, allowing for active monitoring of the pressure.² In contrast to CCs, the pressure distribution in PCs can be homogenized by adding rubber sheets and springs to the cell stack.¹⁹ Recent literature has reported that CCs produce inferior performance in comparison to chemically identical PCs, possibly due to the pressure inhomogeneity in CCs.²⁰

In this study, we utilize both experimental and computational results to bridge the gap between CC and PC performance in Li metal batteries. We show that traditional spring-based coin cells (SCC) suffer from severe heterogeneities in the stack pressure due to the geometry of the wave spring, which has been hypothesized previously.¹² The lack of material at the center of the spring results in a non-uniform applied force and, therefore, an inhomogeneous stack pressure. By replacing the wave spring with an elastic rubber disc, we create rubber-based coin cells (RCC). Since the rubber disc is continuous across its entire surface, the elastic force applied to the cell stack is evenly distributed, resulting in a pressure distribution comparable to that of PCs. We demonstrate the effectiveness of this structure in anode-free lithium metal batteries. Using the RCC structure, we can increase the capacity retention in CullLiNi_{0.5}Mn_{0.3}Co_{0.2}O₂ (NMC 532) anode-free full cells by 7%, delivering a performance comparable to PCs using identical chemistries. This simple yet effective cell structure retains the quick, easy nature of CCs while producing performance on-par with chemically identical PCs.

Methods

Materials.—Commercial NMC 532 cathodes with a loading of 1.78 mAh cm⁻² were purchased from the MTI corporation, punched into 13 mm diameter discs, and dried under vacuum at 100 °C overnight prior to use. 20 mm diameter discs of Celgard 2325 separator were punched and dried at 70 °C overnight. 250 μ m thick Li discs were purchased from China Lithium Energy Co. 1,1,2,2-tetrafluoroethyl-2,2,3,3-tetrafluoropropyl ether (TTE) was purchased



Figure 1. Diagrams of the (a) SCC, (b) RCC, and (c) PC. The stack pressure in the CCs can be roughly modified by adding or removing spacers. The pressure in the PC is directly measured by the load cell.

from TCI Chemicals, 1,2-dimethoxyethane (DME) was purchased from Gotion, and 1,3 dioxolane (DOL) was purchased from Sigma-Aldrich. All solvents were dried with molecular sieves for at least 3 d prior to use. Battery-grade lithium bis(fluorosulfonyl)imide (LiFSI) and Lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) were purchased from Gotion and TCI Chemicals, respectively, and used as acquired. Lithium nitrate (LiNO₃) was purchased from Sigma-Aldrich. The localized high-concentration electrolyte was prepared by combining 1:1.2:3 LiFSI:DME:TTE (molar ratio) and mixing until all solids were dissolved. Similarly, 1 M LiTFSI and 1 wt% LiNO₃ were dissolved in a 1:1 by volume DOL/DME solution to create the DOL/DME electrolyte.

Cell preparation.—Three, 1.3 mm thick silicone rubber sheets were purchased from McMaster-Carr (product number 1460N22). Three different Durometer hardness ratings were used, 50 A, 60 A, and 70 A, which corresponds to elastic moduli of 4.2, 5.8, and 10.7 MPa, as measured by an Instron 5965 Dual Column Testing System (Fig. S1). The rubber sheets were then cut into 15 mm diameter discs, wrapped in Cu foil, and dried overnight at 70 °C before being transferred to a glovebox. CR2032 CCs and PC Allaminate bags were purchased from the MTI corporation. Cu foil was cut into 15 mm discs and etched in 1 M H₂SO₄ for 30 s under sonication to remove surface contaminates. The Cu discs were then sonicated 3 times for 30 s in deionized water and sonicated 3 times for 30 s in acetone. The discs were then dried in a glovebox antechamber under vacuum for 30 min. All cell preparation was conducted in an argon filled glovebox (MTI) with oxygen and water contents <1.0 ppm. For full cells, the cell stack consisted of an NMC 532 cathode, a Celgard 2325 separator, 100 μ l of electrolyte, and a 15 mm diameter Cu foil disc. In addition to the cell stack, two 20 mm diameter Al foil discs and Al-clad positive CCs caps were used to minimize oxidative corrosion in CCs. LillCu half cells consisted of a 13 mm diameter, 250 μ m thick Li disc, a Celgard 2325 separator, 100 μ l of electrolyte, and a Cu foil disc.

In SCCs, two 15 mm diameter, 1 mm thick spacers and a singlelayer, 3 peak wave spring were added to the cell stack. The wave springs used in this study have an outer diameter of 15 mm and an inner diameter of 11 mm. For RCCs, one 1 mm thick spacer and a Cu-wrapped rubber disc were added. This maintains the total thickness of the cell. CC components were placed in stainless steel positive (Al-clad for full cells) and negative caps and were crimped shut with an MTI MSK-110 hydraulic crimper. The crimping pressure was recorded as 850 psi which, using the manufacturer provided piston area of 11.34 cm², returns 6,645 N of force applied to the cell. Normalized to the active material area of 1.3 cm², this is roughly 50 MPa.

PCs were fabricated by sandwiching the cell stack between two metal foil current collectors. Cu and Al foils were used for full cells and Cu foil was used for half cells. This assembly was placed in a 5×7 cm pouch bag and vacuum sealed shut. Sealed PCs were then



Figure 2. Pressure distribution measured by pressure papers with different sensitivity ranges inserted in the cell stack for the (a, d, g, j) SCC, (b, e, h, k) RCC, and (c, f, i, l) PC. A range of sensitivities is used to obtain a detailed pressure map, with the color distribution for each sensitivity shown in the right column. The SCC and RCC record the crimping pressure (50 MPa). The PC records the applied stack pressure (1 MPa). All scale bars are 2 mm.

placed between two silicone rubber rectangles, approximately the area of the PC, and compressed between two steel plates. The load cell and a third plate were used to monitor the stack pressure (1 MPa) applied to the PC. Diagrams for each cell structure are shown in Fig. 1.

Pressure paper tests.—Fujifilm Prescale pressure paper of various sensitivities was used to qualitatively measure the pressure distribution within each cell structure. The pressure paper replaced the cell stack in each cell structure and had a diameter of 13 mm, which is identical to the diameter of the electrodes used in this study. Each cell was fabricated normally then immediately disassembled to prevent over-exposure.

Finite element model.—A stationary 3-dimensional finite element model (FEM) was constructed in COMSOL Multiphysics 6.0 (COMSOL Corp., Stockholm, Sweden) using the structural mechanics module. Each model mimics the cell structures presented in Fig. 1, but in 3 dimensions. Images of the model geometries are shown in Fig. S2. The geometry of each cell component matches those used in experiment except for the PC plates, which were



Figure 3. (a-c) Horizontal cross sectional pressure distribution maps through the cell stack, (d-f) 1-dimensional pressure values across the diameter of the cell stack, (g-i) vertical cross sections through the FEM. Each dataset is presented in the order of SCC, RCC, PC. In order to reflect the pressure paper experiments as closely as possible, the applied pressure is 50 MPa for the CCs, and 1 MPa for the PC. For a direct comparison, Fig. S4 recreates this figure, but with 1 MPa applied pressure on all simulations. In the low pressure case, the pressure distribution in each structure are similar to that shown here.

reduced to 2×2 cm plates to minimize computational cost. Materials were formed into a union to prevent slippage.

Built-in material parameters were used for the stainless steel. Mechanical properties for the rubber were measured with an Instron 5965 Dual Column Testing System (Fig. S1) and matched with previous reports.²¹ The cell stack was modeled with a lumped mechanical model, which has been shown to be a good approximation.²² All materials are linear elastic materials.

A fixed constraint boundary condition was applied to the bottom boundary of the bottom cap or plate while a variable boundary load was applied to the top boundary of the top plate or cap. The applied pressure is normalized to the area of the cell stack. All other boundaries were set as free boundaries. A physics-controlled, normal sized mesh was used for the RCC and PC. For the SCC, a physicscontrolled extra coarse mech was applied to the caps and spring while a physics-controlled normal sized mesh was applied to the spacers and cell stack.

Cell cycling protocol.—Galvanostatic cycling tests for half and full cells were conducted using a Land battery tester. LillCu half cells were cycled first by plating 1.0 mAh cm⁻² at 0.5 mA cm⁻² onto the Cu substrate, then stripping to 1.0 V. The ratio of the charge capacity and the discharge capacity is the CE of the given cycle. Full cells were cycled for two formation cycles at C/10 in a voltage window of 2.8–4.3 V. After this, the cells were charged at C/3 to 4.3 V, then pinned at 4.3 V until the anodic current fell below C/20. The cells discharged at C/3 to 2.8 V. The capacity retention is calculated as the ratio of the *n*th cycle discharge capacity and the first post-formation cycle (cycle 3) capacity.

Scanning electron microscopy and optical imaging.—For scanning electron microscopy (SEM) studies of cross sections, half cells at the 1st, 21st, 41st, and 61st plating were used. The cells were gently disassembled with care taken to prevent accidental removal of plated Li. The plated Li was rinsed thoroughly with DME, imaged with a cell phone camera, then gently torn in half. Cross-sectional SEM images of the samples were taken using an FEI Apreo SEM. No clear signs of lithium in the separator were observed, which has been reported in other types of batteries.²³

Results and Discussion

Pressure inhomogeneity and its causes.-First, we confirmed the pressure uniformity measurements previously reported^{5,12,17} and modified a CC to achieve a uniform pressure distribution. Figure 1 shows diagrams of the three different cell structures studied, where Fig. 1a is a standard 2032 CC, Fig. 1b is a 2032 CC with the spring and one spacer replaced with a Cu-wrapped disc of rubber, and Fig. 1c is a standard PC. To provide a qualitative map of the pressure distribution, we replaced the active materials in each structure with pressure paper over a range of sensitivities (Fig. 2). The size and shape of the pressure paper matches that of the electrodes used later in this study. We then fabricated the cells normally by crimping the CCs at 50 MPa and applying a 1 MPa stack pressure to the PC. Cells were immediately disassembled to prevent over exposure of the pressure paper. Since pressure paper records the maximum applied pressure, CC measurements reflect the crimping pressure, while PC measurements record the stack pressure. In a real cell, CCs will relax after crimping, so the applied pressure during cycling will be significantly lower than the crimping pressure.

The SCC shows severe pressure inhomogeneities across all sensitivities. At the lowest sensitivity (Fig. 2a) the pressure at the edge of the SCC exceeds the maximum measurable pressure of 49 MPa while the pressure at the center of the SCC is below the minimum measurable pressure of 9.6 MPa. This trend holds for the



Figure 4. Comparison of the first cycle Li plating morphology and thickness in the (a-d) SCC, (e-h) RCC and (f-l) PC using the two different electrolytes. The SCC shows significant variation in both the morphology and thickness—over a 600% increase in thickness using the 1LiFSI: 1.2DME: 3TTE electrolyte. The PC and RCC plating thickness and morphologies are similar at both the edge and the center.



Figure 5. LillCu CE for each cell structure for 300 cycles. Li is plated for 1 mAh cm-2 at 0.5 mA cm-2 and is stripped to 1 V. 100 μ l of the 1LiFSI: 1.2DME: 3TTE electrolyte was used. In the first 60 cycles, the RCC and PC perform identically, while the SCC suffers from low CE. After cycle 60, all cells average 99.3% CE, which is identical to previous reports.⁵

medium-low and medium-high sensitivities (Figs. 2d, 2g). Only at the highest sensitivity is a pressure recorded at the center of the cell, which is estimated to be 0.5 MPa (Fig. 2j). This suggests that, even during a 50 MPa crimping, the pressure at the center of the SCC is about 0.5 MPa. After crimping, this value likely decreases further.

We hypothesize that it is the geometry of the wave spring specifically the lack of elastic material in the center of the spring, which causes this inhomogeneity. Therefore, we switched to the RCC structure by replacing the wave spring and one spacer with a uniform rubber disc. The rubber disc had a Young's Moduli of 5.9 MPa (Fig. S1). In this case, the low sensitivity pressure paper (Fig. 1b) records roughly 25 MPa during crimping, while all other sensitivities exceed the maximum measurable pressure (Figs. 1e, 1h, 1k). The pressure distribution in the RCC is uniform for all sensitivities. We repeated these tests using rubber discs with Young's moduli of 4.3 MPa and 10.8 MPa and found that the pressure distribution was uniform for all sensitivities, though the maximum measured pressure was slightly reduced in both cases (Figs. S1, S3). For the remainder of our experiments, we chose to use the 5.9 MPa rubber discs.

We compare this result with the PC. Here, the pressure paper only records significant pressure at the highest two sensitivities because only 1 MPa is applied in this case (Figs. 1i, 1l). Similar to the RCC, the pressure distribution in the PC is uniform, and is estimated at 0.5 MPa, slightly lower than the applied pressure. It is possible that deformation of the rubber leads to slight contact between the top and bottom rubber pieces outside of the area of the pressure paper, which would explain the reduced pressure.

To further our understanding, we created 3-D FEMs for each cell structure. For a fair comparison, we present two different datasets. Figure 3 shows the FEM where the pressure applied to each cell structure reflects the pressure applied to the pressure paper in Fig. 2 -50 MPa for the CCs, and 1 MPa for the PC. Figure S4 shows the FEM where the applied pressure is 1 MPa for all structures.

First, we compare our pressure paper results to horizontal cross sections through the active stack in our FEM (Figs. 3a-3c). In all cases, the FEM matches the pressure paper results in terms of both pressure distribution and absolute pressure. The SCC (Fig. 3a) shows a slight triangular shape, reflecting the three peaks of the wave spring. For further analysis, we plot the absolute pressure along a line across the diameter of the cell stack (Figs. 3d-3f). For the SCC (Fig. 3d), the pressure at the edge of the active stack reaches over 100 MPa, drops to almost -20 MPa before returning to about 5 MPa at the center of the cell. This pattern is repeated on the other half of the cell. Here, negative pressure indicates expansion rather than compression. This deviates slightly from the pressure paper experiments, which measure a small pressure in the center of the SCC during crimping. Assumptions made for the model such as perfectly elastic materials and a laterally uniform crimping pressure likely



Figure 6. Li growth measured by cross-sectional SEM over 60 cycles for (a) the SCC, (b) the RCC, and (c) the PC. Thickness measurements are taken at the 1st, 21st, 41st, and 61st plating. Optical images from the 61st plating are shown in the top right of each plot with markers showing the approximate measurement location. Measurement locations are the same for all samples. SEM images corresponding to these measurements can be found in the supporting information, Figs. S6–S9.

account for this difference. However, the overall trend of high pressure on the edge of the SCC and significantly lower pressure its center is consistent across both experiments. The RCC shows a bowed pressure distribution, ranging from 20 to 35 MPa (Fig. 3e). The PC shows a reasonably flat pressure distribution at about 0.5 MPa (Fig. 3f).

Finally, we investigate the root cause of the SCC pressure inhomogeneity by taking vertical cross-sections through the FEMs, shown in Figs. 3g-3i. The left edge of Fig. 3g shows that, in the SCC, pressure concentrates where the wave spring contacts the bottom spacer. At these three points of contact, large pressure concentrations create pressure inhomogeneities in the spacers, which are then transferred to the cell stack. Thus, the root cause of the pressure inhomogeneity in the active stack is the geometry of the wave spring. Changing to the RCC drastically improves the pressure distribution because the rubber applied a uniform force to the bottom spacer (Fig. 3h). Similarly, the PC shows a uniform pressure distribution (Fig. 3i). These results hold for lower applied pressure, as shown in Fig. S4.

Effect on Li plating morphology and coulombic efficiency.-Next, we investigated the effect of the cell structure, and therefore the pressure (in)homogeneity, on Li plating and CE in LillCu half cells. To demonstrate the chemistry independent nature of this phenomenon, we examined the first cycle plating in two different electrolytes. We used a localized high concentration electrolyte comprised of 1LiFSI: 1.2DME: 3TTE (molar ratio) known for dense, columnar Li deposits, and a 1 M LiTFSI + 1:1 DOL/DME (by volume) electrolyte with a 1 w% LiNO₃ additive known for spherical deposits.³ We plated 1 mAh cm⁻² of Li onto Cu and took cross-sectional SEM images at the edge and center of each cell with each electrolyte (Fig. 4). The effect of the inhomogeneous pressure in the SCC is immediately noticeable. At the high-pressure edge, the plated Li is compact and fits the expected morphology for both electrolytes (Figs. 4a and 4c). However, at the low-pressure center, the plated Li becomes significantly more dendritic, leading to a large increase in thickness-over 600% for the 1LiFSI: 1.2DME: 3TTE electrolyte (Fig. 4b) and 93% for the DOL/DME electrolyte (Fig. 4d). On the other hand, Li plated in the RCC or PC fits the expected morphology and thickness in both electrolytes and in both locations (Figs. 4e-4l). There are no obvious dendritic growths, and the maximum thickness variation is only 25% across all four samples. This is consistent with the well-known effects of pressure on Li morphology.²

Next, we examined the effect of the cell structure on a long-term LillCu CE experiment using the 1LiFSI: 1.2DME: 3TTE electrolyte, shown in Fig. 5. For each cycle, 1 mAh cm⁻² of Li is plated at 0.5 mA cm^{-2} then stripped to 1 V at 0.5 mA cm⁻². Interestingly, only

the first 60 cycles show deviation between the cell structures. The RCC and PC reach a steady-state within the first 30 cycles, while the SCC requires 50 cycles to stabilize. After stabilization, all three cells have an average CE of 99.3%, which is identical to previous reports for this electrolyte.⁵ This was contrary to our expectations based on the initial plating morphology of each cell structure.

To investigate this effect, we examined changes to the Li plating behavior in the first 60 cycles using thickness measurements via cross-sectional SEM (Fig. 6) and optical imaging (Fig. S5). The SEM images used to create Fig. 6 are shown in Figs. S6-S9. In Fig. 6a, the SCC shows large variations for all cycles, with a maximum of 640% for the 1st cycle, and a minimum of 283% for the 61st cycle. Residual solid electrolyte interface (SEI) build-up from previous cycles increases the Li thickness from 5.073 μ m to 11.88 μ m (left edge) and from 37.72 μ m to 45.06 μ m (center). Thickness variations for the RCC remain below 14% for the 1st, 41st, and 61st plating, with the average thickness increasing from 5.99 μ m (1st cycle) to 16.42 μ m (61st cycle) due to residual SEI accumulation. The 21st plating shows a variation of 138% and is considered a statistical outlier, possibly caused by localized surface roughness of the rubber creating small regions of low pressure. Similarly, thickness variations for the PC remain below 55% for all cycles with a general upward trend in the average thickness (6.85 μ m to 14.47 μ m) from the 1st to 61st plating.

Variations to the Li plating location do occur. For the RCC, PC, and high-pressure edges of the SCC, Li plates on top of the residual SEI in dense deposits (Fig. S10). This is consistent with previous reports for similar electrolytes.²⁴ In the low-pressure center of the SCC, however, Fig. S10d reveals that Li transitions from plating on top of the residual SEI to within it. Despite this change, the CE stabilizes to 99.3% after 60 cycles. The change in deposition location may be responsible for the apparently identical steady-state CE and warrants investigation, but is outside of the scope of this study.

Full cell capacity retention.—As a final comparison between the SCC, RCC, and PC, we fabricated CullNMC anode-free cells using the 1LiFSI: 1.2DME: 3TTE electrolyte and compared the capacity retention over 100 cycles (Fig. 7). The charge-discharge profiles (Figs. 7b–7d) clearly indicate that the cells lost capacity due to a loss of cyclable lithium, rather than impedance rise. As our hypothesis suggests, the RCC and the PC have similar performances, and both outperform the SCC due to the improved pressure distribution. Over 100 cycles, the RCC and PC retain 65.6% of their initial capacity, while the SCC retains 58.6%—an increase of 7%. All cells show apparent excellent capacity retention during the first 20 cycles. This is due to the inability of the NMC cathode to re-intercalate 100% of the Li extracted during the first charging step as a result of irreversible



Figure 7. (a) CullNMC anode-free full cell capacity retention for each structure and selected voltage profiles for the (b) SCC, (c) RCC, and (d) PC. Cells contain a 1.78 mAh cm⁻² NMC cathode and 100 μ l of the 1LiFSI: 1.2DME: 3TTE electrolyte). After two C/10 formation cycles, cells are charged at C/3 until 4.3 V, then pinned at 4.3 V until the anodic current drops below C/20. Cells are discharged at C/3 to 2.8 V. After 100 cycles, the PC and RCC retain 7% more capacity than the SCC.

structural changes to the cathode.²⁵ Thus, a "well" of Li remains on the anode and is gradually consumed during the first 20 cycles, leading to a high capacity retention. After 20 cycles, the cell capacity degradation reflects the consumption of active Li. We used the data from cycles 30 to 100 to calculate the CE for the 3 structures, which returned values of 99.4%, 99.4%, and 99.5%, for the SCC, RCC, and PC, respectively. These results are consistent with previous reports for this electrolyte.⁵ As with the LillCu half cells, the SCC still performs impressively given the wide pressure range found within the cell, possible due to the same residual SEI growth mechanism and changes to the Li plating location. However, the initial Li loss due to the lowpressure zone and subsequent dendritic growth in the first 60 cycles prevents the SCC from retaining as much capacity.

Conclusions

In this work, we bridge the gap between Li metal PC and CC performance via pressure homogenization. With both computational and experimental results, we have shown that the geometry of the wave spring used in the SCC creates an inhomogeneous pressure distribution in the active layers. As a result, lithium plated in the low-pressure center of the SCC is dendritic. Replacing the wave spring with a disc of rubber averts this problem and creates a homogeneous pressure distribution akin to PCs. Lithium plated in RCCs, and PCs is uniform across the entire surface. Improvements to the CE in LillCu half cells occur in the first 60 cycles, though all three cell structures stabilize to 99.3% CE thereafter. This may be due to a transition in the Li plating location from the above the residual SEI to within it. In CullNMC anode-free full cells, both the RCC and the PC perform similarly, retaining 7% more capacity than

the SCC after 100 cycles. This solution retains the low cost, quick nature of CCs and produces PC-level performance.

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Competing Interests

The authors declare no competing interests.

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