

Pouch-Cell Architecture Downscaled to Coin Cells for Electrochemical Characterization of Bilateral Electrodes**

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Research groups all over the world are testing new innovative electrodes for batteries aimed at being the next generation of energy-storage devices. To date, the lab-scale coin cells, which house two one-sided electrodes, remain the most common tool to test new battery electrodes. However, many novel electrodes are bilateral by design. In addition, many new electrodes, such as silicon- and sulfur-based, enable large areal capacities that might not be properly tested with any single counter electrode. Here, we suggest a technique to assemble coin cells that incorporate two unilateral electrodes with a bilateral electrode sandwiched between them. This method requires no unique equipment, and can help laboratories with limited resources to test their electrodes under realistic conditions that can better display the capabilities of their research products.

Lithium-ion batteries are currently the most popular choice for portable energy-storage devices, with intercalation and conversion chemistry that has not changed markedly since its first commercialization 30 years ago.^[1] Nevertheless, new technologies are being developed worldwide with the purpose of breaking through as the next generation of battery materials and configurations.^[2,3] While the common coin cell is still the preferred tool for electrochemical testing of new electrode materials,^[4–6] better characterization of batteries for practical applications requires new ideas on how to modify cells to provide more relevant and detailed results. Coin and Swagelok cells can be fitted with a reference electrode in order to differentiate between the processes on the working and counter electrodes.^[7–9] Cells can also be modified to allow for in-situ characterization by methods such as SIMS,^[10] NMR^[11] or AFM.^[12]

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New materials and synthesis methods are another crucial part of the advancement process of LIBs. In addition to the production of new active materials that are integrated in existing technologies, some groups suggest preparation of whole, high active-material loaded electrodes with the use of various methods. Leveau et al. fabricated silicon nano-trees by chemical vapor deposition with 2.5 mgScm^{-2} .^[13] Wang et al. also created a binder-free silicon-based electrode by electro-spinning a silicon/carbon composite with over 2 mgScm^{-2} .^[14] In the field of cathode development, Yang et al.^[15] impregnated carbon papers with CoS_3 and achieved sulfur loadings of 10 mgScm^{-2} and Yuan et al. fabricated sulfur-based cathodes by filtration of sulfur/carbon nanotube suspensions with up to 17.3 mgScm^{-2} .^[16]

These technologies that are being developed need to be electrochemically characterized, first in half-cells (versus lithium counter electrodes), as well as in full-cells, versus commercial counter electrodes, that show the true applicability of the new electrodes. To that end, the electrodes should be tested in lab-scale cells as thoroughly as possible before testing them in larger cells such as jellyroll batteries and pouch cells.^[1] This issue could be challenging in the case of bilateral electrodes, i.e., electrodes which are synthesized with their active materials on both sides of the substrate. That it because concentration gradients could form in the electrodes' structure^[17] that are different between single-sided and bilateral cells. High-loading electrodes will also be a challenge in that counter electrodes may not be available to provide a well-balanced cell.^[18] Several studies have discussed the importance of adjusting the testing protocols when scaling up experimental electrodes from coin cells to pouch cells.^[19,20] However, said studies did not account for electrodes that are inherently bilateral, where the gap between the two systems would inevitably be more substantial. Moreover, most of the abovementioned methods produce electrodes smaller than A4 paper, so conservation of materials may also be a crucial factor.

For these reasons, we propose a simple method for assembling coin cells in an architecture which simulates pouch cells without the need for any special equipment or devices. This method is presented here for 2032 cells in the configuration of cathode/anode/cathode (C-A-C) but could be used in the reverse order for cathode research.

In order to assemble a bilateral cell, one only needs two additional separators beyond conventional coin cells, a thin copper strip to connect the anode to the current collector and an aluminum strip to connect the cathodes. These metal strips should be about 0.2 cm by 1.8 cm, and any standard electrode

current collector will do. The scheme of the cell is presented in Figure 1. Before assembly of the cell, the only recommended step is to reduce the size of two separators by about 2–3 mm from one side, as shown in the figure. For the assembly of 2032 cells, we recommend electrodes 12 mm in diameter, although other sizes may also be used. The steps of assembly are as follows:

- Place the 1.8 cm-long aluminum strip in the cell base beneath the SS disc and press it to the side of the base.

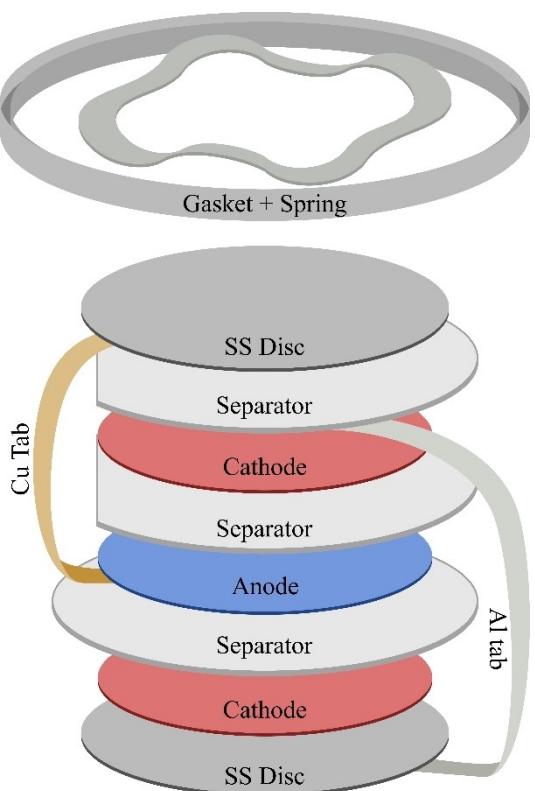


Figure 1. Scheme of the coin cell with a bilateral anode and two cathodes.

- Place a 12 mm-wide cathode on the SS disc, active surface facing upwards, and a 19 mm-wide separator on it.
- Place the 1.8 cm-long copper strip on the separator in the opposite direction from the aluminum foil, and the 12 mm-wide anode on top of it.
- Place a cut 19 mm-wide separator on the anode with the straight, cut side of the separator facing the copper strip.
- Place the second 12 mm-wide cathode at the center of the stack, active surface facing down. Fold the aluminum strip down to rest on the back of the cathode.
- Place the third 19 mm-wide separator in the same position as the second, and fold down the copper strip. It is important at this point to ensure that the copper strip is folded as close as possible to the cut line of the separators, so that the strip goes “inside” the cell gasket. Lay the SS disc on the folded copper strip.
- Add electrolyte, the gasket, the spring, the lid and seal the cell.

As proof of the efficacy of this configuration, we have tested cells with silicon nanowire anodes (SiNW) grown on a stainless-steel mesh.^[21] These anodes are grown in a single-step chemical-vapor-deposition process and as such, are by default, bilateral. The SEM images and scheme in Figure 2 show planar and cross-sectional views of these SiNW electrodes. As a result of their high surface area, silicon loadings of 1–5 mg cm⁻² have been demonstrated, with capacities of up to 15 mAh cm⁻² and 3000 mAh gSi⁻¹.^[22] For these anodes regular one-sided commercial cathodes would not be able to provide the capacity required to test a well-balanced cell, so a pouch-like cell would be optimal for small-scale tests of these anodes. On the other hand, lab-scale synthesis processes often produce limited amounts of materials which must be carefully utilized, and actual pouch cells can be wasteful and require different assembly equipment.

For these reasons, a coin-cell configuration with a “pouch-like” stack could be beneficial. The cycle life of a cell with such an anode, containing approximately 2.7 mgSi cm⁻², is shown in

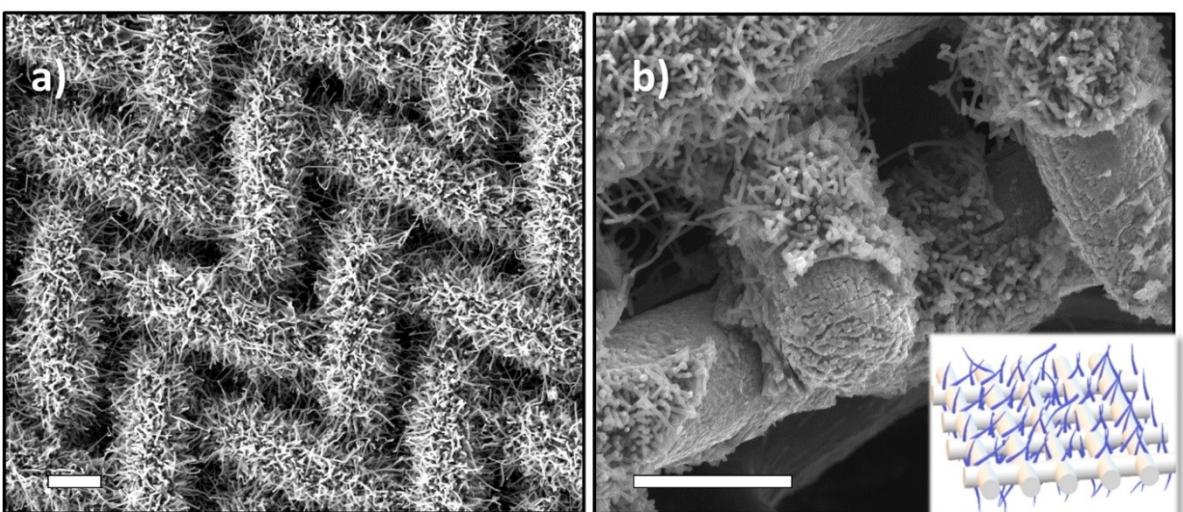


Figure 2. SEM images of the bilateral SiNW anodes: a) planar view, b) cross section of the edge; inset shows a general scheme of the anode. Scale bar: 25 μ m.

Figure 3. Before assembly, the anode was coated on one side with a drop of carbon nanotube suspension (TUBALL BATT, OCSiAl) for the sake of better electrical conductivity between the stainless-steel substrate of the anode and the copper strip. For different bilateral electrodes, other possibilities for electric contact might be masking the edge of the electrodes during synthesis to bare the metal substrate, cutting in advance discs with "tails" or threading a thin Cu wire through the electrode with a needle. The anode is sandwiched between two commercial NMC 622 cathodes (Targray) with a reversible capacity of approximately 2 mAh cm^{-2} each. The cell was tested at 2.8–4.1 V, displaying capacities of $1200 \text{ mAh gSi}^{-1}$ at C/3 and 800 mAh gSi^{-1} at 1C. The cell presented a capacity-fade rate of 0.3%/cycle at 1C. A reference cell was also tested with an anode containing 2.1 mgSi cm^{-2} and a single NMC cathode (C-A). The common practice of pairing the anode with a single cathode limits the anode's practical capacity that can reach up to 8 mAh cm^{-2} . Our C-A-C design results in a more well-balanced cell, while still providing more than three times the gravimetric capacity ($\sim 1200 \text{ mAh gSi}^{-1}$) of commercial graphite anodes.^[1]

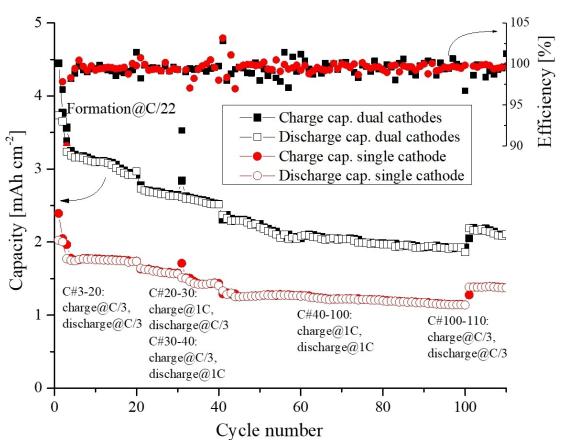


Figure 3. Cycle life of a bilateral coin cell with a SiNW anode and two NMC cathodes (C-A-C stack, black curves) versus a cell with a SiNW anode and a single NMC cathode (C-A stack, red curves), cycled at 2.8–4.1 V

Given that, we prevent lithium deposition on the anode by continuing to ensure anodic excess. The bilateral C-A-C cell displayed an initial geometric capacity of $3.8/2 = 1.9 \text{ mAh cm}^{-2}$, using most of the cathodes' theoretical capacity of 2 mAh cm^{-2} each. The performance of the cell when charged at 1C rates, surpasses even that of the 1/3AAA jellyroll cells presented in our previous work (Ref. [22]), with 2.7 mAh cm^{-2} in the coin cell versus 2.5 mAh cm^{-2} in the 1/3AAA cell. The reference C-A cell displayed 2 mAh cm^{-2} , meaning it was also limited by the cathode, and provided upon cycling about 50–60% of the total capacity of the C-A-C cell. Figure 4a shows charge/discharge plots of selected cycles of the C-A-C cell and the reference C-A cell. The voltage profile of both cells are similar, indicating the same phase transformations occur at both cells. It is worth mentioning the slightly lower overpotential buildup (approximately 30 mV) in the normalized dQ/dV^{-1} plots of the cell comprising two cathodes (Figure 4b).

Electrochemical impedance spectroscopy (EIS) is a convenient in-operando method for evaluation of the impedance components in the cell at various points of its cycle life. We present in Figure 5 EIS spectra of the bilateral and reference cells after the 20th and 110th charges. The intercept of the first semicircle with the X-axis, which represents the resistance of the electrolyte of the cell (R_{bulk}), is 2 ohms, close to the R_{bulk} in regular coin cells using the same SiNW anode. This proves that the unique connections of the electrodes do not hinder the current flow into and out of the electrodes. The diameter of the semicircles represents the sum of the SEI and charge-transfer resistances in the SiNW anode and NMC cathodes.^[22,23] Since the cathode surface area in the bilateral cell is twice that of the typical cell with one cathode, the capacitance of the second semicircle is more than double that of the reference cell. This confirms that this impedance originates primarily from the cathodes, whereas the first semicircle mostly originates from the natural evolution of the SEI on silicon anodes during cycling.^[22]

In summary, we have developed a simple method of coin-cell assembly for the electrochemical characterization of bilateral electrodes. The use of this method is exemplified in SiNW anodes encased between two commercial cathodes. It

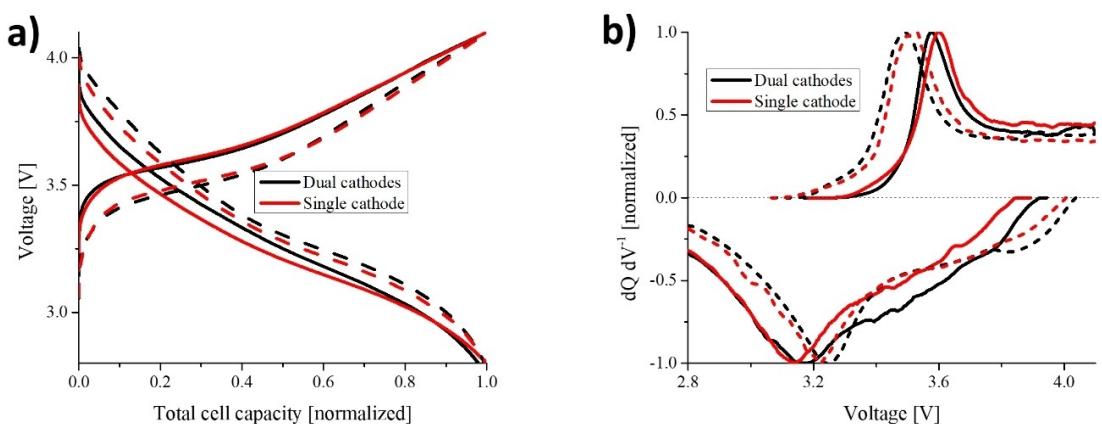


Figure 4. a) Voltage profiles and b) dQ/dV^{-1} plots of the bilateral (C-A-C) and reference (C-A) cells at cycles 10 (at C/3, dashed lines) and 50 (at 1C, solid lines).

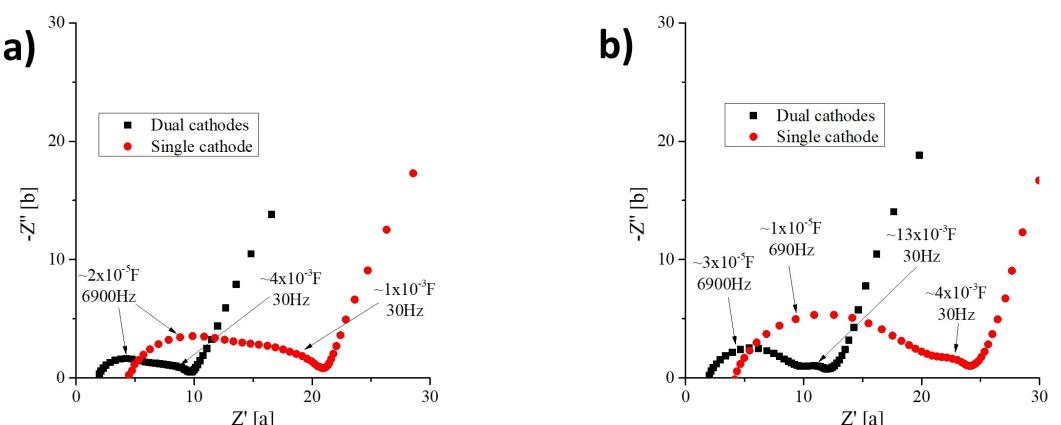


Figure 5. EIS spectra of the bilateral (C-A-C) and reference (C-A) cells after charge at a) cycle 20 and b) cycle 110.

can be used for studying other anode or cathode materials structured in bilateral electrodes. We believe that the simplicity of this architecture will allow other materials to be tested just as efficiently. This method allows developers of battery electrodes to test their products in a manner that preserves materials and equipment while maintaining performances similar to larger-scale jellyroll and pouch cells.

Conflict of Interest

The authors declare no conflict of interest.

Keywords: batteries • coin cells • electrochemistry • silicon • pouch cells

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