Lithium-CO₂ Batteries



A Long-Cycle-Life Lithium–CO₂ Battery with Carbon Neutrality

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Lithium–CO₂ batteries are attractive energy-storage systems for fulfilling the demand of future large-scale applications such as electric vehicles due to their high specific energy density. However, a major challenge with Li-CO₂ batteries is to attain reversible formation and decomposition of the Li₂CO₃ and carbon discharge products. A fully reversible Li-CO₂ battery is developed with overall carbon neutrality using MoS₂ nanoflakes as a cathode catalyst combined with an ionic liquid/dimethyl sulfoxide electrolyte. This combination of materials produces a multicomponent composite (Li_2CO_3/C) product. The battery shows a superior long cycle life of 500 for a fixed 500 mAh g⁻¹ capacity per cycle, far exceeding the best cycling stability reported in Li-CO₂ batteries. The long cycle life demonstrates that chemical transformations, making and breaking covalent C-O bonds can be used in energy-storage systems. Theoretical calculations are used to deduce a mechanism for the reversible discharge/charge processes and explain how the carbon interface with Li₂CO₃ provides the electronic conduction needed for the oxidation of Li₂CO₃ and carbon to generate the CO₂ on charge. This achievement paves the way for the use of CO₂ in advanced energy-storage systems.

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Lithium-ion batteries are widely used as electrochemical energy storage systems for consumer electronics;^[1] however, technologies with higher specific energy are needed for electrified transportation applications.^[2] Therefore, beyond Li-ion battery chemistries such as rechargeable Li-O₂ batteries have recently garnered much attention due to their higher theoretical energy density.^[3,4] Li-O₂ batteries generally have limited cyclability, though several studies have reported new concepts that have achieved long cycle life.^[5,6] Although far less studied, the Li-CO₂ battery is another beyond Li-ion technology with a theoretical energy density of 1876 Wh kg⁻¹,^[7,8] far exceeding that of Li-ion batteries (≈ 265 Wh kg⁻¹). This type of battery involves CO2 reduction and evolution reactions during discharge and charge, respectively, on the surface of a porous cathode with an electrolyte based on lithium salts.

Despite the potentially high-energy den-

sity of Li-CO₂ batteries, the electrochemical reactions remain poorly understood. Generally, it is believed that the discharge products in a Li-CO2 battery should involve formation of Li₂CO₃ and carbon to balance the reaction stoichiometry, while charge results in regeneration of CO₂, although other possible products, such as O₂, have been observed.^[9] Previous Li-CO₂ battery studies have shown evidence for Li₂CO₃ upon discharge and for its disappearance upon charge with cycle life generally less than 100 cycles.^[10–19] However, none of these studies have achieved full reversibility during cycling where carbon can react with Li₂CO₃ to regenerate CO₂ in the charging process. Recently, two studies have provided evidence for carbon reversibility in a Li–CO₂ battery using Raman spectroscopy,^[20,21] but not for more than one cycle. This is important since during the discharge process, other side reactions can occur that can degrade the electrolyte. Moreover, accumulation of carbon and other side products on the catalyst surface can lead to clogging of active sites, which results in the battery failure. Thus, it is crucial to operate Li-CO₂ batteries under a carbon neutral condition in order to reversibly balance the electrochemical reactions during discharge and charge processes.

Our recent findings on the superior electrocatalytic activity of nanostructured transition metal dichalcogenides for CO₂ reduction^[22–25] and O₂ reduction in a Li–O₂ battery,^[5,26,27] have led us to investigate whether this type of catalyst would enable carbon and Li₂CO₃ reversibility in a Li–CO₂ battery. Using a MoS₂ electrocatalyst with an ionic liquid/dimethyl sulfoxide (DMSO) electrolyte, we have been able to achieve a long-cycle-life Li–CO₂ battery with evidence from various in situ and ex situ techniques for reversibility with carbon neutrality.

We assembled a custom-made Swagelok type Li-CO₂ cell using lithium as the anode and MoS₂ nanoflakes (NFs) coated on a gas diffusion layer as the cathode with the electrolyte composed of an 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIM-BF₄)/DMSO (25%/75%) solvent with 0.1 M LiTFSI as a lithium salt. The MoS₂ NFs synthesis and characterization were performed and confirmed based on our previously established methods.^[24,26,27] The assembled cell was first purged with pure CO₂ and then connected to a battery analyzer for cycling experiments. This Li-CO₂ battery was found to cycle up to 500 consecutive discharge and charge cycles with a capacity of 500 mAh g^{-1} per cycle at a current density of 500 mA g⁻¹ as shown in Figure 1a. Comparing the discharge voltage of the first (2.92 V) and 500th (2.75 V) cycles at the capacity of 500 mAh g⁻¹ indicates only a 5% decrease in the discharge potential. The charge potential also shows about a 12% increase during 500 cycles. These results confirm the stable and sustainable performance of the cell during 500 continuous cycles.

We also tested the rate capability of the Li–CO₂ battery at different current densities of 100, 500, and 1000 mA g⁻¹. Figure 1b–d illustrates the polarization gap of the cell as a function of the number of cycles up to 200 cycles. The results indicate that at the current density of 100 mA g⁻¹, the battery shows the lowest polarization gap (0.7 V) at the first cycle and then this potential gap increases to ~1.45 V after 200 cycles (Figure 1b). Polarization gaps of 1.7 and 2.45 V were also obtained for higher current densities of 500 and 1000 mA g⁻¹ after 200 cycles, respectively (Figure 1c,d). These are 0.25 and 1 V larger than the polarization gap obtained at the current density of 100 mA g⁻¹.

To identify the composition of discharge/charge products during cycling especially the fate of carbon that should be formed during discharge, we used X-ray photoelectron spectroscopy (XPS) and Raman spectroscopy. The XPS was performed on the surface of cycled cathodes of MoS_2 NFs coated on Al mesh. **Figure 2**a,b depicts the XPS spectra of the cathode at the first discharge and charge cycles. All spectra were calibrated based on C–C bonding energy at 284.8 eV. The O–C–O and C–C peaks in the XPS spectra are representatives of the so called adventitious carbon contamination which has been discussed in the literature extensively.^[28–31] The C 1s and Li 1s spectra of the product show peaks at 289.5 and 55.2 eV, respectively, corresponding to formation of Li₂CO₃.^[5,17,20] This is consistent with our X-ray diffraction pattern (XRD) results (Section S4, Supporting Information).

Comparing the XPS spectra of C 1s and Li 1s for the discharge and charge cycles indicates that Li₂CO₃ is completely decomposed in the charge process, confirming the reversible formation and decomposition of Li₂CO₃. The XPS results also did not show any evidence of other products such as Li₂O, Li₂O₂, or LiOH. Ex situ Raman spectroscopy was also performed on the cathodes at the first charge and discharge cycles and results are shown in Figure 2e. The Raman spectra in discharge consist of three distinct Raman peaks at around 1580 and 1350 cm⁻¹ corresponding to carbon and 1088 cm⁻¹ corresponding to Li₂CO₃. Both the carbon and Li₂CO₃ Raman peaks completely disappeared on the charge, providing further evidence for the reversibility of the Li–CO₂ cell, i.e., Li₂CO₃ and carbon formation and decomposition during cycling.

In situ differential electrochemical mass spectroscopy (DEMS) was carried out to determine the gases produced during first charge and discharge cycles in the Li–CO₂ cell. Figure 2f shows DEMS profile to measure the e^-/CO_2 ratio during first cycle of the discharge process. The experiments were performed by discharging the battery up to 1600 mAh g⁻¹ capacity and comparing the moles of CO₂ in the headspace of the cell before and after first cycle of the discharge process. It was observed that moles of CO₂ noticeably decreased compared to that of the cell before the discharge process, suggesting the consumption of the CO₂ during this process. The measurements indicate an



Figure 1. The electrochemical performance of Li–CO₂ battery using MoS₂ NFs as cathode materials and a hybrid electrolyte (IL/DMSO) with 0.1 M LiTFSI. a) Discharge and charge voltages profile up to 500 cycles with the capacity of 500 mAh g⁻¹ per cycle. b–d) Polarization gap (V) for 500 mAh g⁻¹ capacity as a function of number of cycles at different current densities.









Figure 2. Spectroscopic investigation of MoS_2 NFs cathode coated on Al mesh. a,b) The XPS spectra of C 1s and Li 1s after the first discharge, and c,d) after the first charge cycle. e) Raman spectra showing the reversible formation/decomposition of Li_2CO_3 and carbon during the first cycle (D and G stand for the disordered and graphitic peaks of carbon, respectively). f) CO_2 moles during the first discharging process after the cell was discharged up to 1600 mAh g⁻¹. g) CO_2 moles detected before and after the charge process in first cycle.

e⁻/CO₂ ratio of 4.05 for the discharge process. Figure 2g presents the DEMS results during the first charge cycle, which was pre-discharged up to 2000 mAh g⁻¹. The results indicate an immediate rise only in the CO₂ partial pressure confirming that CO₂ is the only gas evolved during the charge process. The e^{-}/CO_{2} ratio of 4.07 during the first cycle of the charge process was obtained by comparing the evolved moles of CO₂ with calibration data (Figure S3, Supporting Information). This indicates a four-electron transfer process during the evolution of CO2 gas. No evidence of other evolved gases (e.g., O2, CO, and H₂) was observed during the charge process (Figure S6, Supporting Information). The DEMS results for the charge and discharge processes indicate that they both involve a four-electron reaction confirming the full reversibility of the battery at the first cycle. In order to quantify the discharge product, we used the Coulometry approach (Equation (1)) that was previously used by McCloskey et al.^[32] to determine the cathode weight gain;

$$\frac{3.6}{F \cdot b} \times M_i = \Delta w_i \left[\text{mg mAh}^{-1} \right]$$
(1)

where Δw_i is the cathode weight increase by formation of *i* species, F is the Faraday constant, *b* is the number of transferred electrons, and M_i is the molecular weight of *i* species. Considering $b = 4.05 \text{ e}^-/\text{CO}_2$ obtained from DEMS and molecular weight of 78.89 for Li₂CO₃, we obtained $\Delta w_i = 0.726 \text{ mg mAh}^{-1}$, which is 98.8% of the theoretical value of 0.735 mg mAh⁻¹ for a four-electron transfer process. Knowing the current of 500 mA over 1 h time period in our system, the amount of Li₂CO₃ was found to be 0.03 µg during the discharge process.

We also performed additional characterizations at higher cycles to examine the reversibility of product formation and decomposition. Figure 3a,b shows scanning electron microscopy (SEM) images of the cathode after 25 cycles of discharge and charge processes. The SEM images confirmed the presence of discharge products in the form of nanoparticles with an average size of 100-200 nm. The deposited nanoparticles on the surface of cathode completely disappeared after the charge experiment (Figure 3b) verifying the XPS and Raman data. A typical cathode area after 25 discharge cycles is depicted in the transmission electron microscopy (TEM) image shown in Figure 3c. Our electron energy-loss spectroscopy (EELS) and TEM results show the presence of four major components: i) crystalline Li₂CO₃, ii) amorphous carbon, iii) crystalline carbon, and iv) the MoS2 NF catalyst. The elemental assignments shown in Figure 3c) for the MoS₂ catalyst, the Li₂CO₃ and amorphous carbon, is based on EELS, high-resolution imaging, and electron diffraction pattern analysis, similar to the previously reported results by Asadi et al.^[5]

For example, crystalline Li_2CO_3 is found using highresolution phase contrast (HRTEM) imaging and electron diffraction (Figure 3d–f), while HRTEM images of the surface of the cathode after 25 discharge cycles is used to identify crystalline MoS_2 NFs (Figure 3f). This was further confirmed by measuring the intensity spacing across the blue line (6.2 nm), which is consistent with MoS_2 interlayer spacing (Figure 3g). We also performed EELS of the C–K edge to distinguish between crystalline and amorphous forms of carbon (Section S5, Supporting Information).

Raman spectroscopy was also performed on the cycled cathodes up to 100 charge/discharge cycles and results further







Figure 3. SEM and TEM images of cathode sample after 25 cycles working in the Li– CO_2 battery. a,b) SEM image of cathode surface after discharge (a) and charge (b) processes. c) TEM image showing the three components present in the sample: crystalline Li₂CO₃, amorphous carbon and MoS₂ catalyst. d) Li₂CO₃ crystals oriented toward [103] zone axis. e) Experimental diffraction pattern. f) HRTEM image of an MoS₂ NFs. g) The intensity across the blue line. The spacing is consistent with MoS₂ interlayer spacing. h) Raman spectra showing the reversible formation/decomposition of Li₂CO₃ and carbon up to 100 cycles.

confirm the reversibility at higher cycles (Figure 3h). Electrochemical impedance spectroscopy (Section S6, Supporting Information) and XPS results (Figure S1, Supporting Information) carried out at 10th cycle of charge and discharge also provide a strong evidence for the reversible formation and decomposition of products.

To examine the stability of the electrolyte, we performed ¹H NMR and ¹³C NMR experiments on the fresh and used electrolyte (after 100 discharge/charge cycles). Results shown in Figure S8 in the Supporting Information indicate that there is no evidence of electrolyte degradation for the used electrolyte.

Moreover, we studied the deep discharge of this system for possible application in primary batteries where no reversibility is needed. The experiments were performed at the same current densities used for the cycling experiments (Section 6, Supporting Information). Results shown in Figure S1 in the Supporting Information illustrate a discharge/charge capacity of about 60 000 mAh g⁻¹ (600 h charge and discharge) at a current density of 100 mA g⁻¹. Capacities of 35 000 mAh g⁻¹ (70 h) and 30 000 mAh g⁻¹ (30 h) were also obtained at current densities of 500 and 1000 mA g⁻¹, respectively.

The experimental investigations have provided evidence for carbon neutrality during long-term cycling of this Li–CO₂ battery, i.e., both the presence of carbon in the discharge product and its reincorporation into CO₂ gas with Li₂CO₃ decomposition. Despite these observations, many of the relevant mechanistic details of these reactions still remain unclear. In turn we carried out computational studies to provide further insight into the charge and discharge mechanisms during cycling experiments at a capacity of 500 mAh g⁻¹. This involved initially determining energies for some possible reactions using highly accurate quantum chemical wave-function-based calculations, followed by more detailed density functional theory (DFT) studies of possible reaction steps. The mechanisms are likely to be very complex since the discharge products are a mixture of crystalline Li₂CO₃ and amorphous carbon based on the TEM

studies (Figure 3c) and our studies are meant to provide insight into feasible mechanisms.

Energies for four possible discharge reactions have been determined with the very accurate G4MP2 method^[33] with the following assumptions: i) the effects of solvation have been added to gas phase G4MP2 energies in our calculations using a continuum model (SMD^[34]), assuming DMSO as the solvent, although these effects on the reaction energies do not change any conclusions (see Table S3, Supporting Information), ii) in the discharge reactions, the carbon and lithium are assumed to be in atomic form and the rest of the species CO, CO₂, Li₂CO₃, and Li₂O, are in their molecular forms. The results of these energies, when combined with experimental results, provide evidence for the overall reaction on discharge.

The first reaction examined was a two-electron reduction with CO gas being one of the products along with Li₂CO₃:

$$2\text{Li} + 2\text{CO}_2 \rightarrow \text{Li}_2\text{CO}_3 + \text{CO} \quad \Delta G = -4.82 \text{ eV}$$
 (2)

This two-electron reduction reaction is very thermodynamically favorable, but since no CO is observed during discharge this is not the likely reaction mechanism for formation of Li_2CO_3 . This reaction may be unfavorable due to kinetic factors (energy barriers) for the reaction mechanism or because other reactions involving solid-state products are more favorable. The second reaction examined involves formation of Li_2O and CO:

$$2\text{Li} + \text{CO}_2 \rightarrow \text{Li}_2\text{O} + \text{CO} \quad \Delta G = -2.22 \text{ eV}$$
 (3)

This reaction is also exothermic according to the calculations. However, Li_2O , as well as CO, are not observed experimentally, so this can also be ruled out as a discharge reaction. The third reaction is similar to reaction (3) but involves carbon formation:

$$4\text{Li} + \text{CO}_2 \rightarrow 2\text{Li}_2\text{O} + \text{C} \quad \Delta G = 1.91 \text{ eV}$$
⁽⁴⁾







Figure 4. Theoretical calculations of Li–CO₂ battery charge and discharge mechanisms. a) Schematic showing discharge and charge processes of a model for an Li_2CO_3/C composite on an MOS_2 cathode. b) $MO_{15}S_{21}$ cluster model used for CDFT calculations of diabatic charge hopping from MOS_2 NF to CO₂. c) CDFT calculations of electronic couplings between $[MO_{15}S_{21}]^0[CO_2]^0$ and $[MO_{15}S_{21}]^+[CO_2]^-$ as a function of separation between the active site and CO₂. The fit gives $H_{ab} = 2712e^{-0.85r_{Mo-CO2}}$. d) Potential-dependent mechanistic analysis of CO_3^{-2-} ion formation on MOS_2/IL co-catalyst that can lead to Li_2CO_3/C discharge product growth. Potential dependence is calculated through a computational hydrogen electrode approach and all voltages are with reference to the Li⁺/Li electrode. e) Cluster model $C_{55}H_{18}/(Li_2CO_3)_2$ used as model of amorphous carbon and lithium carbonate to calculate the charge potential for decomposition of Li_2CO_3/C . f) A reaction sequence that was investigated for oxidation, Li⁺ elimination and CO₂ elimination from cluster in (e) of an Li₂CO₃ dimer bonded to cluster (hydrogens are not shown in the diagram).

Since reaction (4) is endothermic, it is not likely, and also Li_2O is not observed. The fourth reaction we examined involves the formation of carbon and Li_2CO_3 :

$$2\text{Li} + 3/2\text{CO}_2 \rightarrow \text{Li}_2\text{CO}_3 + 1/2\text{C} \quad \Delta G = -1.33 \text{ eV}$$
(5)

Reaction (5) is thermodynamically favorable and is the most likely reaction based on the lack of experimental evidence for reactions (2)–(4). The TEM and Raman studies also show the presence of carbon and DEMS shows a four-electron reduction, which are consistent with reaction (5).

In addition to above results, the observed discharge potential of ≈ 2.9 V is consistent with the calculated thermodynamic potential for reaction (5) of ≈ 2.90 V versus Li/Li^{+, [35]} The calculated thermodynamic potential for reaction (4), which involves formation of Li₂O is 1.89 V, much lower than observed in Figure 1, further confirming that this reaction is unlikely to occur. Thus, we conclude that the discharge process involves reduction of CO₂ followed by reaction with (Li⁺ + e⁻) pairs to form a mixture of discharge products including crystalline Li₂CO₃ and some form of carbon.

The schematic in **Figure 4**a illustrates the formation of the discharge product where reduction of CO_2 is considered the first

step in the eventual formation of Li₂CO₃ and C. However, based on DFT calculations the reduction of CO_2 ($CO_2 + e^- \rightarrow CO_2^-$) in solution occurs at ≈0.6 V versus Li/Li⁺, much lower than the discharge potential. This would indicate that the MoS₂ interface with adsorbed CO₂ greatly affects the electron transfer. To investigate the electron transfer mechanism, constrained DFT (CDFT) calculations^[36-38] were used to investigate the role of the MoS₂/IL co-catalyst system for CO₂ reduction. Using an Mo₁₅S₂₁ cluster model with adsorbed (EMIM $^+$ + e^-) pairs to expose an isolated Mo atom on the catalyst edge, we consider the process of non-adiabatic electron hopping from the catalyst to a CO₂ molecule as shown in Figure 4b. Figure 4c shows the electronic couplings (H_{ab}) between the $[Mo_{15}S_{21}]^{0}[CO_{2}]^{0}$ and $[Mo_{15}S_{21}]^{+}[CO_{2}]^{-}$ charge states calculated as a function of distance between the active site and CO2. The CDFT-calculated Hab values decrease exponentially as expected, due to exponential decay of the diabatic wave function overlap as a function of donor-acceptor separation. However, we found that the coupling between the two diabats is strong even at large separation, suggesting that charge transfer is most likely adiabatic (i.e., CO₂ is spontaneously reduced due to the strong binding to the edge of the flake). Thus, this suggests that the discharge potential will correspond to the thermodynamic potential of the reaction, i.e., ≈2.9 V.

To further investigate the mechanism of the Li2CO3/C discharge product formation, a mechanistic study was performed on a periodic MoS₂ nanoribbon with the presence of EMIM cations, shown in Figure 4b. Potential dependence is calculated through a computational hydrogen electrode approach and all voltages are referenced to the Li/Li⁺ electrode^[39] and is shown in Figure 4d. Following the initial adsorption of CO₂ (downhill by 1.58 eV), we found that dissociative adsorption of a second CO_2 molecule is also favorable (by 0.60 eV), which leads to the formation of a co-adsorbed carbonate (CO₃*) and carbon monoxide (CO*). The CO* desorption is unfavorable by 2.23 eV, consistent with the experimental result wherein CO is not detected as a gaseous byproduct. Adsorbed CO₃* is assumed to react with Li⁺ ions in solution to form Li₂CO₃ where this electrochemical step is favorable below 2.31 V versus Li/Li⁺, which is approximately consistent with the experimental discharge potential. What remains unclear, however, is the process by which the amorphous carbon forms to satisfy the bulk reaction stoichiometry. Our DFT calculations suggest that a third CO₂ could, in principle, react with the edge containing adsorbed CO^* to form a second CO_3^* and a single carbon atom (C*), although we found that the thermodynamics for such a process are uphill by 0.94 eV. We speculate that this CO* conversion into CO3* and C* will be favorable in the presence of defect sites on the catalyst or carbon product leading to amorphous carbon growth.

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Based on the DEMS and Raman data, the charge process involves decomposition of crystalline Li_2CO_3 and amorphous carbon. The decomposition of Li_2CO_3 and carbon:

$$2\mathrm{Li}_{2}\mathrm{CO}_{3} + \mathrm{C} \rightarrow 3\mathrm{CO}_{2} + 4\mathrm{Li}^{+} + 4e^{-}$$
(6)

will occur at 2.9 V based on thermodynamics.^[35,40] However, the observed charge potential always occurs much higher around 3.8-4.5 V in our work and other experimental studies.^[20,41] The reason is probably because of an overpotential due to the oxidation potential of Li2CO3 required for the electrochemical reaction. From B3LYP density functional calculations, the oxidation potentials of Li₂CO₃ monomer and dimer are 4.2 and 4.4 V versus Li/Li⁺, respectively. The lower charge potential in Figure 1 of ≈4 V for the earlier cycles may be due to the mechanism by which decomposition occurs. The amorphous carbon may provide an electrically conducting network to enable the charge process that has to start with oxidation of the discharge product since Li₂CO₃ is not electronically conductive.^[42] From B3LYP density functional calculations, the oxidation potential of molecular Li₂CO₃ is lowered if it is bound to a carbon defect site in a model for amorphous carbon. Thus, the decomposition may occur at the interface between carbon and Li₂CO₃. This would require maintenance of an interface between Li₂CO₃ and carbon throughout decomposition to CO₂ and C. Our DFT calculations on a model system show that maintenance of such an interface is possible with loss of CO₂ and C (see Figure S12, Supporting Information).

We have also investigated the energy for reaction (6) by investigating in detail various reaction pathways for sequential oxidation and decomposition of a cluster model of $\text{Li}_2\text{CO}_3/\text{C}$ (see Section 11, Supporting Information for details). We used a C_{54} cluster model for amorphous carbon (with 18 hydrogens on the edges) and a single carbon adatom (representing a defect site) as shown in Figure 4. Sequential reactions involving oxidation, loss of Li⁺, and CO₂ elimination were found with an upper charge potential of \approx 4 V, consistent with experiment, with the highest energy step involving oxidation of the cluster. One of these reaction cycles is shown in Figure 4f. The exact role of the MoS₂ catalyst during charge other than providing a conductive network with the amorphous carbon is unclear based on this mechanism.

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In summary, we developed a rechargeable Li–CO₂ battery based on MoS_2 NFs that shows reversible cycling at 500 mAh g⁻¹ for 500 cycles, as well as a very high charge/discharge capacity of 60 000 mAh g⁻¹ for one cycle. Our experimental investigation confirmed the presence of crystalline Li₂CO₃ and amorphous C as the main discharge products, with evidence that they are reversibly decomposed in the subsequent charge cycle, i.e., it maintains carbon neutrality. Computational studies have provided new insight into feasible discharge and charge mechanisms involving formation and decomposition of the Li₂CO₃/C composite. The long cycle life demonstrates that complex C–O bond making and breaking chemical transformations can be used in energy storage systems, in addition to the widely studied alkali metal (Li, Na, K)–oxygen bond making and breaking transformations.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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and P.A. performed the DEMS and verified the data analysis. P.A. and L.M. performed XPS and verified the data analysis. L.M., Z.H., and S.R. carried out Raman experiments. S.R. performed NMR experiments. R.E.W., R.S.A., B.N., P.C.R., A.N., M.V., J.G., and L.A.C. performed the computational studies of the MoS_2/IL and reaction mechanisms during charge/discharge cycling. J.J. and R.K. performed the TEM and HRTEM experiments. A.S.K., L.A.C., A.C., M.A., R.E.W., and R.K. wrote the manuscript. All of the authors discussed the results and reviewed the manuscript.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

carbon neutrality, density functional theory (DFT), energy storage, Li anodes, Li–CO $_{\rm 2}\,{\rm batteries}$

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