Enabling high energy density Li-ion batteries through Li₂O activation

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Abstract

Lithium oxide (Li₂O) is activated in the presence of a layered composite cathode material (HEM) significantly increasing the energy density of lithium-ion batteries. The degree of activation depends on the current rate, electrolyte salt, and anode type. In full-cell tests, the Li₂O was used as a lithium source to counter the first-cycle irreversibility of high-capacity composite alloy anodes. When Li₂O is mixed with HEM to serve as a cathode, the electrochemical performance was improved in a full cell having an SiO-SnCoC composite as an anode. The mechanism behind the Li₂O activation could also explain the first charge plateau and the abnormal high capacity associated with these high energy cathode materials.

Keywords

Li₂O activation; High energy composite cathode material; First cycle irreversibility.

Introduction

To extend the mileage range of current electric vehicles, it is essential to increase the energy density of the lithium-ion batteries (LIBs) used for energy storage. The increase of the energy density at the level of the materials will also be beneficial for reducing the cost associated with the inactive components in the battery, and hence would make the Li-ion technology affordable for the grid applications. In the last 20 years, mainly metal oxides and graphite have been used as the cathode and anode, respectively, in state-of-the-art LIBs. During these two decades, the LIB energy density has been improved by pursuing engineering processes that lead to a reduction in the amount of inactive components rather than new active materials. Theoretically, the capacity of the metal oxide cathode can be as high as 300 mAh/g due to the presence of Li, transition metal, and anions in the active material¹. Also, despite the existence of high-capacity anodes such as silicon, tin, and alloys, the use of these materials in commercial batteries is limited due to their large first-cycle irreversibility². In a full cell, a large excess of cathode material has been added to compensate for the irreversible capacity loss at the anode, but the energy density of the cell has only been increased slightly. One possibility is to use a small amount of lithium to partially pre-lithiate these anodes, but this causes a safety concern because of the potential fire hazard. Recently, a new stabilized lithium metal powder (SLMP) was introduced to overcome the first-cycle irreversibility in full cells³. This lithium powder, which is coated with lithium carbonate, can be mixed with anode materials such as silicon or graphite in a dry condition. The main drawback of SLMP is its incompatibility with the N-methylpyrrolidone or water solvent used to prepare electrodes, and the need of calendaring the electrode to fracture the Li_2CO_3 coating shell and thereby activate the lithium powder. Shanmukaraj et al. proposed the use of Li_3N as a source of lithium in the cathode; unfortunately, this material cannot be used for large battery applications due to its high reactivity⁴. Sun et al. report the use of nano-sized Li_2O embedded into Co matrix as additive to overcome the loss of silicon based anode using LiFePO_4 cathode⁵. The material was prepared by heat treatment of Co_3O_4 and

molten Li under Ar atmosphere at 200°C. When a nanomaterial of Li₂O was ball-milled with sub-micrometre-sized Co, a small capacity of less than 100 mAh/g was obtained.⁵

In this work, we demonstrate that micro-sized Li₂O can be electrochemically activated in LIBs when it is mixed with a high-capacity composite cathode material, such as Li₂MnO₃-LiMO₂ (M=Mn, Ni, Co), and can work as a lithium source to compensate for the first-cycle irreversibility of full cells having high capacity anodes such as silicon composites and alloys. We also present a possible explanation for the observations. This new, safe and practical approach can enable Si and Sn based high energy anodes.

Material and methods

The cathode material Li_{1.2}Ni_{0.15}Mn_{0.55}Co_{0.1}O₂ (0.5Li₂MnO₃• 0.5LiMn_{0.375}Ni_{0.375}Co_{0.25}O₂), named HEM, was obtained from Toda. The anode materials tested were graphite (from ConocoPhillips), Li₄Ti₅O₁₂ (LTO) (from Enerdel), and SiO-SnCoC (prepared by Spex ball milling). The electrolyte (Gen I) was prepared with 1M LiClO₄ in EC:EMC (3:7 ratio by weight). The electrolyte (Gen II) was 1.2 M LiPF₆ in EC/EMC (3:7 ratio by weight) from Tomiyama Chemical Industry (Japan). The HEM electrode was made with 92 wt% Li_{1.2}Ni_{0.15}Mn_{0.55}Co_{0.1}O₂, 4 wt% Timcal C-45, and 4 wt% Solvey 5130 PVDF binder and prepared by the Cell Fabrication Facility of Argonne National Laboratory. The HEM-Li₂O electrode was made of 50 wt% Li_{1.2}Ni_{0.15}Mn_{0.55}Co_{0.1}O₂, 20 wt% Li₂O, 20 wt% Super P, and 10 wt% PVDF. Electrochemical cycling experiments were conducted in 2032-type coin cells. Half-cells and full cells with graphite anodes were cycled between 2 and 4.7 V; and full cells with LTO anode were cycled between 0.5 V and 3.2 V. Full cells were made using cathode limited configuration (anode capacity/HEM-cathode capacity ratio was 3). Full cells with SnCoC-SiO anode were cycled between 2 V and 4.5 V. ICP-MS analysis was conducted on the harvested charged cathodes using a Perkin Elmer DRC II Inductively Coupled Plasma - Mass Spectrometer. X-ray powder diffraction data were collected at the 11-ID-C beam line in the Advanced Photon Source (APS) of Argonne National Laboratory. The X-ray wavelength used was pre-set to 0.1114Å.

Results

In this work we used a cathode in which Li_2O was mixed with a Li_2MnO_3 - LiMO_2 composite material, named thereafter HEM (“high energy material”). This HEM- Li_2O electrode was charged to 4.7 V with a lithium anode and the Gen I electrolyte. Interestingly, a large plateau at ~4.45 V was observed during the initial charge of the HEM- Li_2O electrode as shown in Figure 1. The first charge capacity of ~600 mAh/g of HEM is close to twice the charge capacity of the HEM cathode material by itself (300 mAh/g)⁶. Moreover, as discussed later, the resulting charge capacity depends highly on the current rate, nature of the salt in the electrolyte, and the anode used (Li metal, graphite, or $\text{Li}_4\text{Ti}_5\text{O}_{12}$). The large observed charging capacity is due to Li_2O being activated at high voltage as evidenced by the other experiments reported in this paper. The influence of the electrolyte and anode was then investigated. To understand the role of the electrolyte in the Li_2O activation, we conducted coin cell tests with electrolytes (Gen I) and (Gen II). The carbonate electrolytes were selected because of their relative stability at high voltage and the need to activate HEM at 4.7 V⁷. Similar capacities were obtained using either the Gen I or Gen II electrolytes in half-cell tests at the 10 mA/g rate as shown in Figure 1.

However, in the full-cell configuration with either a graphite or $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (LTO) anode at the 10 mA/g rate, the obtained plateau was shorter for cells using the Gen II electrolyte as shown in Figure 2. When the full HEM- Li_2O /graphite cell reaches 4.7V a charge capacity of 1083 mAh/g is obtained. These different behaviors suggest that the various anodes and electrolytes have an effect on the electrochemical mechanisms involving the activated Li_2O .

On the basis of inductive coupled plasma mass spectroscopy (ICP-MS) to measure Li loss from the charged cathode of the HEM- Li_2O /LTO full cell cycled at a 10 mA/g rate, the activation of Li_2O was determined to be ~28% for Gen I and 17% for Gen II electrolyte as shown in Table 1. The amount of activated Li_2O from ICP-MS analysis is below the calculated one based on the charge capacity when the full cell was cycled to 3.2 V, especially with the Gen I electrolyte (we should have observed only 440

mAh/g for the charge capacity instead of an observed capacity of 608 mAh/g (Figure 2)). The delivered charge capacities for HEM-Li₂O/LTO at 10 mA/g were 608 and 444 mAh/g in Gen I and Gen II electrolyte, respectively (Figure 2).

The activation process was also dependent on the charge current rate. A high ratio of Li₂O activation was achieved under a low current rate (3 mA/g). Indeed, 68% and 39% of Li₂O activation were obtained in Gen I and Gen II electrolytes, respectively (Table 1). High energy X-ray diffraction of the charged sample under the 3 mA/g current rate in the full cell with LTO anode confirmed that Li₂O is not totally activated/consumed as shown in Figure 3. This is consistent with the findings from the ICP-MS analysis that not all of Li₂O is activated. The Rietveld refinement of the initial phase composed of HEM/Li₂O led to the weight ratio of 0.653:0.347 and the composition of Li_{1.222}Mn_{0.778}O₂ (a=4.94545, b=8.55772, c=5.0197, β=108.9987) for the layered compound (Figure S1). This is in good agreement with experimental value of 71.4:28.6 and the prepared composition Li_{1.2}Co_{0.1}Mn_{0.55}Ni_{0.15}O₂. When this sample is charged to 4.7V, several new diffraction peaks appeared (Figure S2). Therefore, the powder pattern could be indexed successfully only using three phases [(Li_xMnO₂: C2/m, a=4.91276Å, b=8.51735Å, c=5.08128Å, β=109.1993), (Li₂O: Fm-3m, a= 4.6089Å), and (Li_yMnO₂: R-3m, a=2.84756Å, c=13.91076Å)]. The shoulder observed at 2θ= 1.374Å, corresponds to the (003) reflection of the rhombohedral phase whereas the highest intensity peak at 1.329Å corresponds to the (001) reflection of the monoclinic phase. Due to the difficulty in refining the lithium content using Powder XRD, we considered that the rhombohedral and monoclinic phases have the composition MnO₂. Consequently, we have been able to determine the amount of Li₂O remaining after the first charge (17.5 weight %). When this is compared to the initial amount of 34.7%, one can conclude that 50% of the Li₂O was activated. Even if we consider in the refinement two phases with Li_xMnO₂ compositions, similar Li₂O amounts were obtained.

To understand why the observed charge capacity is higher than the amount of activated Li_2O , we conducted the following experiment. First, we charged the HEM- Li_2O /LTO to determine the full capacity, then we harvested $\text{Li}_{4+x}\text{Ti}_5\text{O}_{12}$ from the cell and cycled it in the presence of a lithium counter electrode and determined the amount of lithium inserted in the LTO.

The results shown in Figure 4 indicate that the charge capacity obtained for the HEM- Li_2O /LTO cathode is not the same as the charge capacity of the harvested $\text{Li}_{4+x}\text{Ti}_5\text{O}_{12}$ electrodes (i.e. a difference of about $1000-608 = 450$ mAh/g with (Gen I) and $600-420 = 175$ mAh/g with (Gen II)). Based on the capacity of partially discharged $\text{Li}_{4+x}\text{Ti}_5\text{O}_{12}$, the amount of Li inserted in LTO in a full cell was less than the calculated amount based on the observed capacity during the voltage plateau at 3 V, but was close to the amount of Li loss in the charged HEM- Li_2O electrode as measured by ICP-MS (Table 1). Indeed, the amount of lithium inserted into LTO corresponds to a capacity of ~580 and ~450 mAh per gram of HEM for Gen I and Gen II electrolyte, respectively (Figure 4).

When the HEM electrode without Li_2O added was cycled with the same condition (i.e., with LTO anode until 3.2 V under 3 mA/g), only 250 mAh and 283 mAh per gram as capacities of HEM were recovered from the LTO with the Gen I and Gen II electrolytes, respectively (Figure 4). In the HEM- Li_2O /LTO full cell, the charge plateau was longer in Gen I than in Gen II electrolyte (Figure 4).

These results indicate that Li from activated/consumed Li_2O as well as from HEM is inserted into LTO. We also conducted half-cell and full-cell tests of the HEM- Li_2O electrode which showed no noticeable performance degradation (Figure S3 and S4).

Other evidence of the Li_2O activation/consumption effect came from the cycling of HEM- Li_2O in the half-cell configuration at the 3 mA/g rate, where the state of charge was limited to 350 mA/g (Figure 5).

Once the charge capacity reaches 350 mAh/g, the battery starts discharging. We repeat this sequence for many cycles (10 cycles). We found that for the first 6 cycles, the obtained discharge capacity of HEM- Li_2O is only ~ 170 mAh/ per gram of HEM. Normally, HEM alone delivers a charge capacity of 320

mAh/g and a discharge capacity of about ~ 270 mAh/g. This indicates that HEM is partially delithiated and some lithium sites in HEM are still full. Therefore, during discharge less lithium is re-intercalated into HEM and a smaller discharge capacity is obtained. One should mention also that the capacity coming from the activation of Li_2O is irreversible, which explains why less than 250 mAh/g was delivered during discharge.

Indeed, during the first discharge, the HEM- Li_2O material can insert an amount of lithium correspond to a discharge capacity of 166 mAh/g. This indicates that during the charge a partial delithiation of the HEM has occurred and that some of the capacity ($350 - 166 = 184$ mAh/g) is not due to the removal of lithium from HEM cathode. The cell was stable for more than 6 cycles, as indicated by the steady capacity. The sum of the capacity difference between the charge and the discharge for the 6 cycles was ~1200 mAh/g of HEM, which is beyond the theoretical capacity of Li_2O and HEM in the electrode (~990 mAh/g of HEM). HEM- Li_2O has the chemical formula of $\text{Li}_{3.575}\text{Mn}_{0.542}\text{Ni}_{0.144}\text{Co}_{0.094}\text{O}_{4.179}$. This material with a MW of 135.4 g/mol and 3.575Li has a theoretical capacity of 708 mAh/g of the total weight (HEM- Li_2O). This theoretical capacity is increased to ~990 mAh/g based on the HEM weight alone. This indicates that in addition to Li removal from HEM and Li activation/consumption from Li_2O , other electrochemical reactions not involving lithium mass transfer happened. As shown in Figure 5, the discharge capacity of the HEM- Li_2O /Li cell reached ~170 mAh/g in the first six cycles, even though the charge capacity was fixed to 350 mAh/g. The discharge capacity value is below the 250 mAh/g usually delivered by the HEM cathode alone. This finding indicates that the Li_2O activation/consumption started before the complete delithiation of HEM. More information can be elicited by simple cycling at various states of charge, so that the distinction between the delithiation of HEM and the activation/consumption of Li_2O can be traced (Figure S5 and S6). On the one hand, the delithiation of HEM is relatively reversible with first-cycle irreversibility of 10-25%, depending on the

state of charge. On the other hand, Li_2O activation is irreversible and can only be seen in the initial charge.

This behavior allows us to determine when the Li_2O activation is initiated. To that end, we determined the discharge capacities of an HEM- Li_2O /Li half-cell cycled under a low current rate of 3 mAh/g at various states of charge: 11% (130 mAh/g), 14% (160 mAh/g), 15% (180 mAh/g), 21% (250 mAh/g), 30% (350 mAh/g), 38% (450 mAh/g), 55% (650 mAh/g), and 100% (1175 mAh/g with cut-off voltage of 4.7 V). These results are summarized in Table 2 (and Figure S5 and S6) and show that Li_2O activation/consumption starts at a charge capacity of ~160 mAh/g, which corresponds to delithiation of the layered component (LiMO_2) of HEM. Despite charging the HEM- Li_2O to 38% state of charge (450 mAh/g) we only observed a limited removal of lithium from HEM. Indeed, based on the difference between the discharged capacity obtained between the state of charge 14% (160 mAh/g) and 38% (450 mAh/g), we concluded that only a small additional amount of lithium ($56 \text{ mAh/g} = 167 - 111$, see Table 2) was actually removed from HEM. Interestingly, the discharge capacity remains essentially unchanged between 30% and 55% state of charges. However, when the half-cell voltage reached 4.7 V (SOC = 100%), the discharge capacity increased to 273 mAh/g (Table 2), which now corresponds to the usual capacity of the HEM cathode as reported by others¹.

Li_2O in the HEM- Li_2O electrode is believed to be activated once lithium is removed from the layer component (LiMO_2) of HEM. A possible mechanism is that when HEM is partially delithiated, Li_2O undergoes oxidation reactions and loses electrons such as is shown in Figure 6.

The described process can cause the Li_2O activation through sequential electron loss and formation of various lithium-oxygen species. The resulting Li_xO_y species can potentially cause electrolyte decomposition and end up in SEI layers of either the anode or cathode. In the case of Gen I this may be responsible for the much larger extra capacity that was observed. From FTIR data it is clear that there are lithium alkyl carbonates formed on the cathode in the case of the Gen I electrolyte (Figure S7).

In the case of Gen II, the electrolyte degradation was not evident from the FTIR spectra and the additional capacity may result from electrochemical reactions that do not involve lithium consumption. One of the hypotheses is the presence of dissolved species having a redox shuttle type behavior (Figure 6).

The redox shuttle mechanism can also contribute to the charge capacity in the case of the Gen I electrolyte. When the cells using both electrolytes were opened no corrosion was observed on the aluminum current collector. The various possible mechanisms contributing to mass and charge transport including lithium consumption are illustrated in Figure S8.

Due to the electrolyte decomposition or redox shuttle, the cycling of the HEM material by itself in Gen II electrolyte at various states of charge shows the presence of internal currents (hundreds of μA) as self-discharge current. These currents are higher by at least four times those observed in a pure $\text{LiNi}_{0.75}\text{Co}_{0.18}\text{Mn}_{0.07}\text{O}_2$ layer structure (Figure S9). The Li_2O activation may explain the difference between charge capacity and the actual amount of lithium removed from the HEM materials as discussed next.

The fact that the Li_2O activation starts before the full delithiation of HEM suggests that the corresponding partial delithiated phase $\text{Li}_2\text{MnO}_3\text{-Li}_x\text{MO}_2$ has a role in catalyzing the activation of Li_2O . Indeed, in the second cycle no additional charge capacity (no Li_2O activation) is observed after reaching 4.7 V even though the x-ray shows that some Li_2O remains in the electrode. This result clearly points to the need of clarifying the role of the partial delithiated $\text{Li}_2\text{MnO}_3\text{-Li}_x\text{MO}_2$ phase as it could explain the difference between the charge capacity and the actual amount of lithium removed from these HEM-type materials. Earlier studies on the HEM cathode showed little change in the transition metal oxidation state after ~ 150 mAh/g state of charge during the first charge plateau⁸. The oxygen electroactivity with the formation of peroxide entities is possibly the origin of this capacity. Recent studies have shown that the high discharge capacity of the HEM cathode material can only be explained by the electrochemical

activity of oxygen⁹. Indeed, the high initial charge capacity cannot be simply attributed to Li₂O removal alone or to the transition metal oxidation. The removal of oxygen is observed only at the end of the charge, and no solid evidence has been shown for Mn oxidation activity (oxidation to 5+ or 7+ states). As shown in the insets of Figure 4 the HEM-type material displays a difference between the charge capacity and the real amount of lithium inserted into the anode. The charge capacity for the HEM/LTO full cell and that recovered from LTO, per gram of HEM, are 340 mAh/g - 257 mAh/g for Gen I electrolyte and 324 mAh/g - 283 mAh/g for Gen II electrolyte. Therefore, this HEM-Li₂O study could explain the first charge profile of the HEM material, where possible traces of Li₂O exist in the pristine material or during cycling, as recently reported¹⁰.

Finally, the activation of Li₂O can enable the use of a high energy density anode with high first-cycle irreversibility, such as silicon. As a practical proof of concept, we fabricated and tested HEM-Li₂O/SiO-SnCoC and HEM/SiO-SnCo full cells. The oxide-alloy anode provided good cycling performance but suffered from a high initial irreversible capacity of ~35%¹¹. The capacity obtained based on the HEM cathode weight is higher than 250 mAh/g with the presence of Li₂O, while it is less than 180 mAh/g without the Li₂O as shown in Figure 7. The high first charge capacity observed in the full cell is attributed to the activation of Li₂O which is an irreversible process. Part of the charge capacity is due to parasitic reactions such as electrolyte decomposition and redox shuttle activity.

Discussion

In this paper we have shown evidence that lithium oxide (Li₂O) is activated/consumed in the presence of a layered composite cathode material (HEM) and that this can significantly increase the energy density of lithium-ion batteries. The degree of activation depends on the current rate, electrolyte salt, and anode type. In full-cell tests, the Li₂O was used as a lithium source to counter the first-cycle irreversibility of high-capacity composite alloy anodes. The activation of Li₂O can enable the use of a high energy density anode with high first-cycle irreversibility, such as silicon. When Li₂O is mixed with the HEM to

serve as a cathode, the electrochemical performance was significantly improved in a full cell having an SiO-SnCoC composite as an anode. The ensemble of these results points to the likelihood of the role of Li₂O, present as traces in the pristine HEM type materials or formed during the charge process, in significantly extending the first charge plateau through electrolyte decomposition or redox shuttle reaction. It is true that in our example, 20% of Li₂O added in the electrode is a high amount because it is not yet optimized. Nevertheless, based on our ICP/X-ray measurements we found that the amount of activated Li₂O could reach as high as 68% of its 1675 mAh theoretical capacity (=1139 mAh/g). As example, in a cathode electrode 1.75 mg of Li₂O per cm² can generate a capacity of $1.75 \times 1675 \times 68\% = 2 \text{ mAh/cm}^2$. This capacity is needed to accommodate the 40% first cycle irreversibility of silicon based material. For example if we have a silicon based anode with a reversible capacity of 3 mAh/cm² (first cycle irreversibility of 40%) a cathode material with a capacity of $3/0.6 = 5 \text{ mAh/cm}^2$ is needed to produce a full cell of 3 mAh/cm². This capacity is obtained with the use of 25 mg/cm² of HEM (Li₂MnO₃-LiMO₂ (M=Mn, Ni, Co), with practical capacity of 200 mAh/g). However, 40% of this 25 mg/cm² HEM (10 mg/cm²) is consumed by providing lithium to form SEI on the silicon based material. As only Li from HEM is used, the other elements (nickel, cobalt, manganese and oxygen) are inactive elements and only adding dead weight to the cathode weight. We conclude that 1.75 g of Li₂O can possibly replace 10 mg of HEM. Instead of using 2/5 weight ratio of Li₂O on HEM (in our first attempt) a 1.75/25 weight ratio could solve the first irreversibility of silicon based material. This Li₂O/HEM weight ratio did not depend on the nature of the used anode material.

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Author contribution

AA, HW and KA proposed the concept. AA, YC and ZC conducted the laboratory experiments and acquired the data. HBY conducted the Rietveld refinement. RA and LAC performed the DFT calculation. AA, ZC, IB, LAC and KA discussed the results. AA, IB, LC and KA wrote the paper.

Competing financial interests

The authors declare no competing financial interests.

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List of Tables

Table 1. ICP-MS results for HEM/LTO and HEM-Li₂O/LTO full cells charged to 3.2 V at rate of 3 and 10 mA/g.

Electrode	Electrode composition (ICP)	Li ₂ O activated (%) (ICP)
HEM	$\text{Li}_{1.217}\text{Mn}_{0.542}\text{Ni}_{0.148}\text{Co}_{0.097}\text{O}_3$	Reference electrodes
HEM after charge in Gen I (3 mA/g)	$\text{Li}_{0.222}\text{Mn}_{0.542}\text{Ni}_{0.137}\text{Co}_{0.095}\text{O}_3$	
HEM after charge in Gen I (10 mA/g)	$\text{Li}_{0.376}\text{Mn}_{0.542}\text{Ni}_{0.132}\text{Co}_{0.091}\text{O}_3$	
HEM after charge in Gen II (3 mA/g)	$\text{Li}_{0.240}\text{Mn}_{0.542}\text{Ni}_{0.134}\text{Co}_{0.093}\text{O}_3$	
HEM after charge in Gen II (10 mA/g)	$\text{Li}_{0.323}\text{Mn}_{0.542}\text{Ni}_{0.143}\text{Co}_{0.099}\text{O}_3$	
HEM-Li ₂ O	$\text{Li}_{1.217}\text{Mn}_{0.542}\text{Ni}_{0.144}\text{Co}_{0.094}\text{O}_3; 1.179\text{Li}_2\text{O}$	
HEM-Li ₂ O after charge in Gen I (3 mA/g)	$\text{Li}_{0.222}\text{Mn}_{0.542}\text{Ni}_{0.153}\text{Co}_{0.105}\text{O}_3; 0.381\text{Li}_2\text{O}$	68%
HEM-Li ₂ O after charge in Gen I (10 mA/g)	$\text{Li}_{0.376}\text{Mn}_{0.542}\text{Ni}_{0.153}\text{Co}_{0.106}\text{O}_3; 0.843\text{Li}_2\text{O}$	28%
HEM-Li ₂ O after charge in Gen II (3 mA/g)	$\text{Li}_{0.240}\text{Mn}_{0.542}\text{Ni}_{0.155}\text{Co}_{0.107}\text{O}_3; 0.715\text{Li}_2\text{O}$	39%
HEM-Li ₂ O after charge in Gen II (10 mA/g)	$\text{Li}_{0.323}\text{Mn}_{0.542}\text{Ni}_{0.147}\text{Co}_{0.096}\text{O}_3; 0.977\text{Li}_2\text{O}$	17%

Table 2: Capacity of half-cell with HEM-Li₂O cathode at various states of charge at 3 mA/g rate.

State of Charge (mAh/g), (%)	Discharge capacity (mAh/g)
130 (11%)	105
160 (14%)	111
180 (15%)	129
250 (21%)	152
350 (30%)	166
450 (38%)	167
650 (55%)	168
1175 (100%)	273

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Figure 3. X-ray diffraction of charged HEM-Li₂O electrode in Gen I and Gen II electrolytes (I = 3 mA/g).

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Figure 5. Voltage profile versus cycling times of HEM-Li₂O/Li half-cell (lower curve) and capacity vs. cycle number (upper curve) at current rate of 3 mA/g and limited state of charge of 350 mAh/g (GEN I electrolyte).

Figure 6. Schematic of oxidative decomposition of Li₂O from density functional calculations. Charge overpotentials are not included.

Figure 7. First-cycle voltage profile versus capacity and cycling performance (inset) of HEM-Li₂O/SiO-SnCoC and HEM/SiO-SnCoC full cells (GEN I electrolyte).

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Figure 6. Schematic of oxidative decomposition of Li_2O from density functional calculations at the B3YP/6-31+G(d) level of theory. Oxidation potentials (V, Li/Li^+) are given. Charge overpotentials are not included. The calculations are based on molecular species. Solvation energies are computed using the SMD solvation model (using an acetone dielectric medium).

Figure 7. First-cycle voltage profile versus capacity and cycling performance (inset) of HEM–Li₂O/SiO-SnCoC and HEM/SiO-SnCoC full cells (GEN I electrolyte).

TOC Graphic:

Activation of Li_2O in in lithium ion battery with HEM- Li_2O