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Review on electrode-level fracture in lithium-ion batteries*

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Fracture occurred in electrodes of the lithium-ion battery compromises the integrity of the electrode structure and would exert bad influence on the cell performance and cell safety. Mechanisms of the electrode-level fracture and how this fracture would affect the electrochemical performance of the battery are of great importance for comprehending and preventing its occurrence. Fracture occurring at the electrode level is complex, since it may involve fractures in or between different components of the electrode. In this review, three typical types of electrode-level fractures are discussed: the fracture of the active layer, the interfacial delamination, and the fracture of metallic foils (including the current collector and the lithium metal electrode). The crack in the active layer can serve as an effective indicator of degradation of the electrochemical performance. Interfacial delamination usually follows the fracture of the active layer and is detrimental to the cell capacity. Fracture of the current collector impacts cell safety directly. Experimental methods and modeling results of these three types of fractures are concluded. Reasonable explanations on how these electrode-level fractures affect the electrochemical performance are sorted out. Challenges and unsettled issues of investigating these fracture problems are brought up. It is noted that the state-of-the-art studies included in this review mainly focus on experimental observations and theoretical modeling of the typical mechanical damages. However, quantitative investigations on the relationship between the electrochemical performance and the electrode-level fracture are insufficient. To further understand fractures in a multi-scale and multi-physical way, advancing development of the cross discipline between mechanics and electrochemistry is badly needed.

Keywords: fracture, electrode, lithium-ion battery

PACS: 62.20.mm, 82.45.Fk ,82.47.Aa **DOI:** 10.1088/1674-1056/ab6841

1. Introduction

During the charge/discharge cycle process of a lithium-ion battery, lithium diffusion into/out of active materials drives expansion/contraction of the materials and this lithiation/de-lithiation induced deformation generally depends on the specific active materials. Heterogeneous Li composition or Li concentration gradient generates inhomogeneous deformation in the active materials and consequently brings about an internal stress.[1–9] This internal stress appearing and evolving in the repeated charge–discharge cycles is responsible for possible fracture initiation and propagation in the electrode materials.

It has been revealed that a close relation exists between fracture of active particles and aging of the battery. Quite a lot of research work has shown that fracture at the particle level would accelerate capacity fading and shorten the battery lifetime.[10–14] On one hand, fracture at the particle level can accelerate battery fading by increasing the impedance of electronic and ionic transport. Observations have revealed that fracture of active particles will block internal pathway for electric conduction which finally results in capacity fading.[14,15] Cathode materials such as $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ (NCA) generally exist as secondary particles formed by an agglomerate of smaller primary particles. Inter-granular fracture is prone to occur at the boundary between primary particles due to anisotropic changes on two sides and leads to a poor ionic and electric contact between primary particles with reduced electrochemical activity.[10] Except separation between primary particles within a secondary particle, debonding may also occur at the interface between active particles and binders, which leads to partial separation or even complete isolation of the active particles from the rest of the electrode. This is known to electrically disconnect active particles from conductive agent or current collector, resulting in capacity loss.[3,16–18] For active materials with large volume expansion such as silicon, pulverization and breakdown of active particles will completely lose its cycling ability.[3,19]

On the other hand, new surface created by fracture at the

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particle level may expose to the electrolyte. Under a certain voltage window, decomposition of the electrolyte would take place at the new fracture surface. This decomposition reaction will deplete lithium ions in the electrolyte as well as in the active particles, leading to irreversible capacity loss.[20,21] What is more, production of the decomposition reaction will deposit on the opening fracture face to form solid electrolyte interphase (SEI). SEI usually has low electric conductivity and its formation will definitely increase the electric resistance. This phenomenon is quite typical for electrode materials with large volume expansion ratios.

As reported above, a lot of research work has been conducted on revealing and comprehending the relation between fracture and electrochemical performance at particle level. Actually, the multi-scale response is a critical factor leading to the complexity of battery problem.[22–24] The renowned Newman's model delicately links electrochemical responses at particle level with those at electrode level, indicating that fracture at electrode level is at least equally important to that at particle level and its occurrence affects the whole cell performance as well.[25,26] In this case, fracture occurred at electrode level and how it affects the electrochemical performance are quite worthy of an elaborate discussion.

An electrode in a lithium-ion battery commonly includes a metallic current collector whose one side or both sides are connected with active layers, with the lithium metal electrode being the exception (see Subsection 4.2). An active layer in commercial electrodes is typically an aggregate of active particles, conductive additives (CA), binders, and pore-filling electrolyte.[27,28] Meanwhile, under laboratory conditions, an active layer is sometimes prepared preferably in the form of a thin film containing specific active material.[29,30] These two forms of active layers are of significant practical or scientific values. But it is noticed that fracture phenomena and mechanisms are not quite the same in composite electrodes and thin-film electrodes. Details will be demonstrated in the following sections.

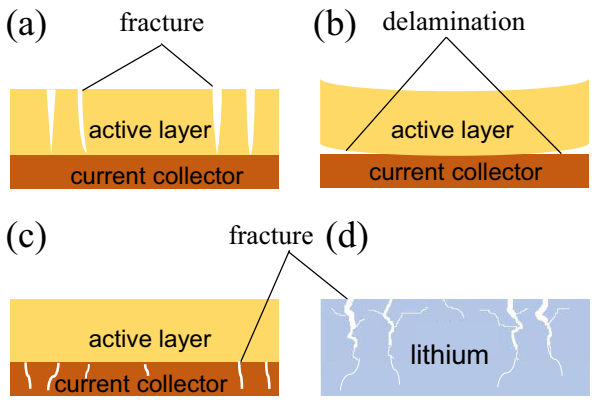


Fig. 1. Illustrations of (a) fracture of active layer, (b) interfacial delamination, (c) fracture of current collector, and (d) fracture of lithium metal electrode.

Due to complex composition of electrodes, the electrode-level fracture occurs in many different ways. This review will involve three typical types of electrode-level fractures, including the fracture of an active layer, the interfacial delamination, and the fracture of a metallic foil in electrodes (including current collectors and lithium metal electrodes), as illustrated in Fig. 1.

2. Fracture of active layer

In this section, the fracture occurred in the active layer is discussed. Sometimes fracture in the active layers can be observed before electrochemical cycling onset (for instance, silicon composite electrodes[31]), indicating fracture could be produced in the manufacturing process. However, in most cases fracture occurs during the electrochemical cycling process and is caused generally by the diffusion-induced stress within the electrode.[1,32] The thickness of an active layer is commonly much smaller than its length and width, indicating that the stress along the thickness direction is insignificant.[1,32] In other words, the in-plane crack induced by the in-plane stress (i.e., stress along the direction perpendicular to the thickness) is the major form of fracture occurred in the active layers.[28,33] Fracture in the active layer parallel to the surface was also reported[34,35] and will be discussed in Subsection 3.2.

2.1. Fracture of active layer in experimental observation

Cracks of active layers are frequently observed in electrodes and have been recognized to be responsible for the coupled mechanical-electrochemical degradation of electrodes in plenty of research work, regardless of the electrode type. For the active layer of a thin-film electrode, the fracture phenomenon is relatively simple to understand, since only fracture of the active content is involved. A number of experimental studies highlighted that the crack is a key factor of degradation in a thin-film electrode.[16,33,36–41]

Experimental studies also focused on the formation mechanism and the growing process of a crack in the active layer of thin-film electrodes. Beaulieu *et al.* reported that the crack of a thin-film Si–Sn electrode is caused by the shrinkage of the active layer upon delithiation and they also pointed out the crack pattern is similar to that found in dried mud,[36] as shown in Fig. 2. Li *et al.* found that the crack of active layer in thin-film silicon electrodes would become finer as the thickness of the film decreases.[33] Wang *et al.* observed that better electrochemical performance, which could even be superior to that of a Si film, would be obtained for Ti–Si films when cracks grow finer or denser.[37] Note that, phenomena in Refs. [33,37] seem to imply that the growing crack is not a relevant factor leading to the deteriorating electrochemical performances of an electrode. However, in those observations, the

layer thickness as well as the active material system was varying and consequently the observed result may be attributed to complex coupling mechanisms in the battery more than to the fracture itself. Chon *et al.* further addressed that the growth of a crack along the thickness direction of the active layer in thin-film silicon electrodes is actually complicated when there is a sharp crystalline-amorphous phase boundary[29] which commonly exists in the first cycle of silicon electrodes,[42–45] as shown in Fig. 3. A sequence of damage was revealed: upon delithiation, yielding of the amorphous Li_xSi layer first occurs, then the fracture of the amorphous layer, followed by the crack extension into the crystalline Si layer. By using the multibeam optical sensor (MOS) method, Pharr[46,47] and Choi[48] found that the fracture energies of Li_xSi and Li_xGe layers are functions of the lithium concentration. The indentation method has also been reported to measure the fracture toughness of the active layer in the thin-film Sn electrodes.[49] Although the fracture phenomenon in thin-film electrodes is much easier to understand, its strong relation to the electrode structure and different components makes it complicated.

Fracture in the active layer of composite electrodes is distinctly different from and even more complicated than that in thin film electrodes, owing to the complicated formula and mesostructure. The common morphology and cross-section view are shown in Fig. 4. According to the composition of the active layer, three possible types of fractures at the particle level, namely, fracture of active particles,[15,50–54] fracture of CA/binder composite,[55–58] and adhesive debonding between active particles and CA/binder composite,[59–61] would occur. The electrode-level fracture in composite electrodes may include multiple types of these particle-level fractures at the same time.[62,63] In other words, the binder has to be involved. Thus, the initiation and growing of the crack in the active layer of composite electrodes are sensitive to the type and content of the involved binder[27,31] and this fracture mechanism still calls for in-depth investigations via methodologies in composite material mechanics.

The crack in an active layer is considered as a key factor of degradation for composite electrodes as well in quite a lot of research work.[27,28,64–72] By adopting the methods of x-ray tomography and diffraction in Si and Si/C electrodes, Vanpeene *et al.* investigated the microstructure in composite electrodes and suggested that the crack in an active layer is critical.[71,72] The coupling of multiple mechanisms, including the gas evolution, was also referred to. A variety of experimental attempts can be made for controlling the crack in an active layer in complex composite electrodes. Material modification seems to provide one of the practical solutions. For instance, Vanpeene *et al.* proposed a maturation step, which consists of storing the electrode in a humid atmosphere for a few days before drying and cell assembly, to restrain the crack of the active layer in a silicon composite electrode.[73] The mechanism of suppressing cracks in the active layer through this manipulation is not very clearly explained but it is possibly related to the alteration of mesostructure and material property of the active layer during these maturation and drying processes.[74] Similarly, Du *et al.* also suggested that the drying process is critical for the crack initiation in the active layer of $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ electrodes.[70]

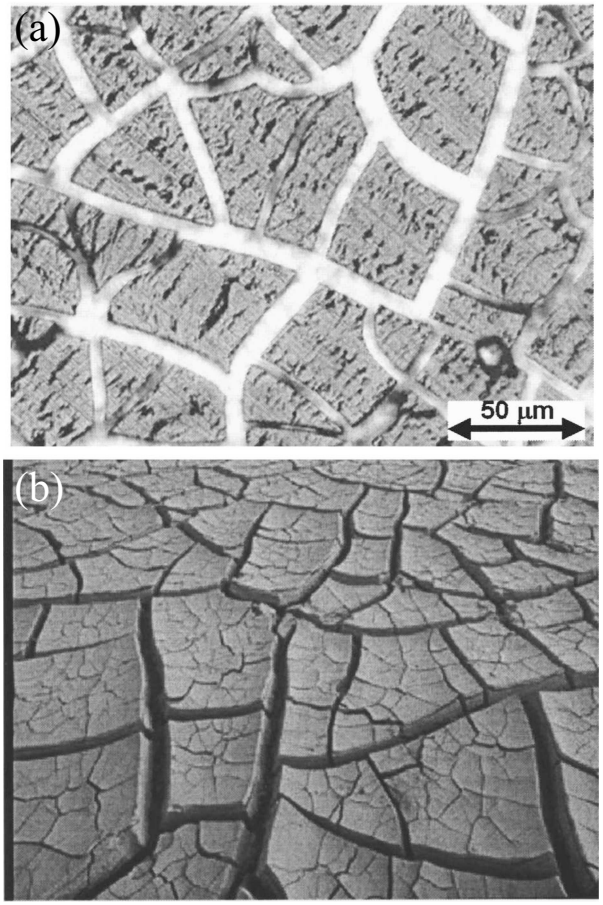


Fig. 2. Crack pattern: (a) cracks of a thin-film silicon electrode (b) dried mud. Reprinted with permission from Ref. [36]. Copyright 2001, The Electrochemical Society.

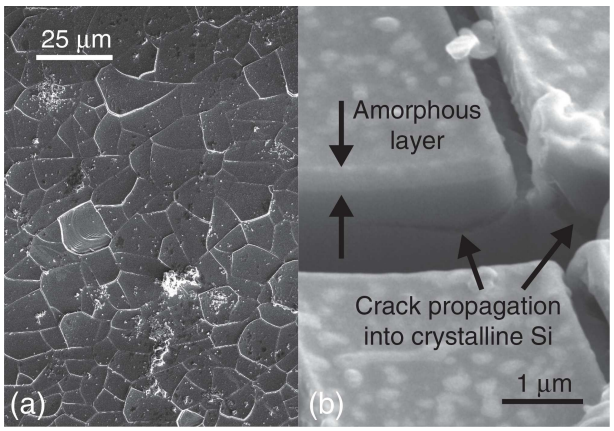


Fig. 3. Crack of active layer in thin-film silicon electrode: (a) surface morphology, (b) cross section view. Reprinted with permission from Ref. [29]. Copyright 2011, American Physical Society.

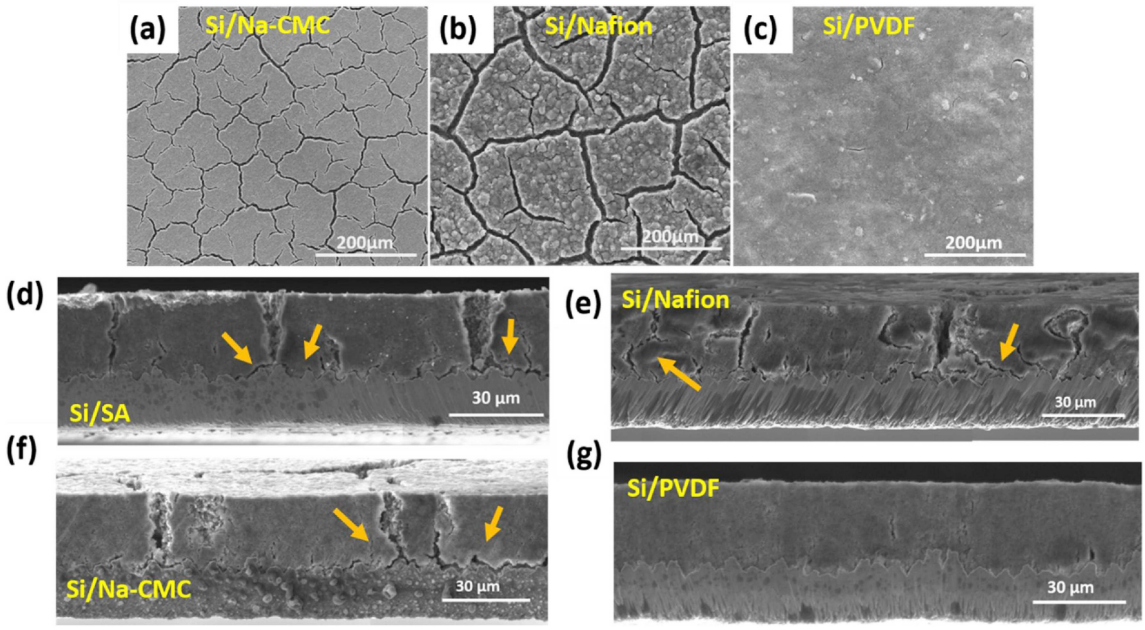


Fig. 4. Surface morphology of (a) Si/Na-CMC, (b) Si/nafion, and (c) Si/PVDF electrodes after the 1st delithiation. Cross-sections of (d) Si/SA, (e) Si/nafion, (f) Si/Na-CMC, and (g) Si/PVDF electrodes at the 2nd delithiation state prepared by focused ion beam. Reprinted with permission from Ref. [31]. Copyright 2019, Elsevier.

In this review, fracture at the electrode level in anodes attracts more attention, because the deformation of anode materials is commonly large, leading to severer damage. But it should not be interpreted as the unimportance of electrode-level fracture in cathodes. Numerical studies have shown the significance of the fracture at the electrode level in cathodes.[70,75,76] For instance, by using the nanoindentation on the thin-film electrode, the fracture properties of $Li_xMn_2O_4$[75] and $LiCoO_2$[76] were evaluated. The fracture toughness of the cathode material was found on the order of 0.1–10 MPa·$m^{1/2}$.

2.2. Impact of crack in active layer on electrochemical performance

As mentioned above, the fracture of the active layer, which can be easily observed and tested, serves as an indicator of the electrode degradation. But how this fracture affects the electrochemical performance of the battery in detail remains a major concern and an opening question. However, three widely recognized consequences brought about by electrode fracture can be generalized as follows: extra SEI formation caused by exposure of new surfaces to the electrolyte, loss of electrical conduction at particle level, and loss of electrical conduction caused by the interfacial delamination, as illustrated in Figs. 5(a)–5(c). Each one of these will be discussed individually in this review.

The extra SEI formation induced by fracture of the active layer or the active content in electrodes is similar to that of the active particle, as illustrated in Fig. 5(a), and is one of the major factors causing capacity fading in negative electrodes.[16,46,48,77–79] For a composite electrode, it is hard to evaluate quantitatively the impact of the extra SEI formation. This is because in the complex mesostructure of a composite electrode, it is unable to capture and characterize all newly created fracture surfaces which are exposed to the electrolyte. The unexpected additional exposure of the active material to the electrolyte in composite electrodes depends entirely on the particle-level fracture (including the fracture of the active particle and the adhesive debonding between the active particle and the CA/binder compound), which triggers another degradation mechanism, namely, the loss of electrical conduction at particle level.

Compared with the extra SEI formation, loss of electrical conduction induced by the particle-level fracture is an equally important degradation mechanism for composite electrodes,[31,64–67,71,80] as illustrated in Fig. 5(b). However, how this mechanism works still remains unclear. From a macro point of view, Li ions and electrons transport along the thickness direction, or in other words, they move parallel with cracks of the active layer.[1,32] In this way, the transport path seems not to be blocked by the crack in the active layer. It is also the case with thin-film electrodes.[68] Nevertheless, the degradation mechanism in terms of the electrical conduction loss may be explained via the mesoscopic characters of the crack in the active layer,[65–67,71–73,81] which still lacks comprehensive understanding.

It is noted that the crack in an active layer can be accompanied by other types of electrode-level fractures such as the interfacial delamination between the active layer and the current collector,[28,31,33,82] as illustrated in Fig. 5(c). The cracks separate the active layer into many islands.[31,33,36] Consequently, stress concentration occurs at the edge of these islands

and causes interfacial delamination.[31,36] The effect of interfacial delamination on electrode performance will be discussed in Section 3 in detail.

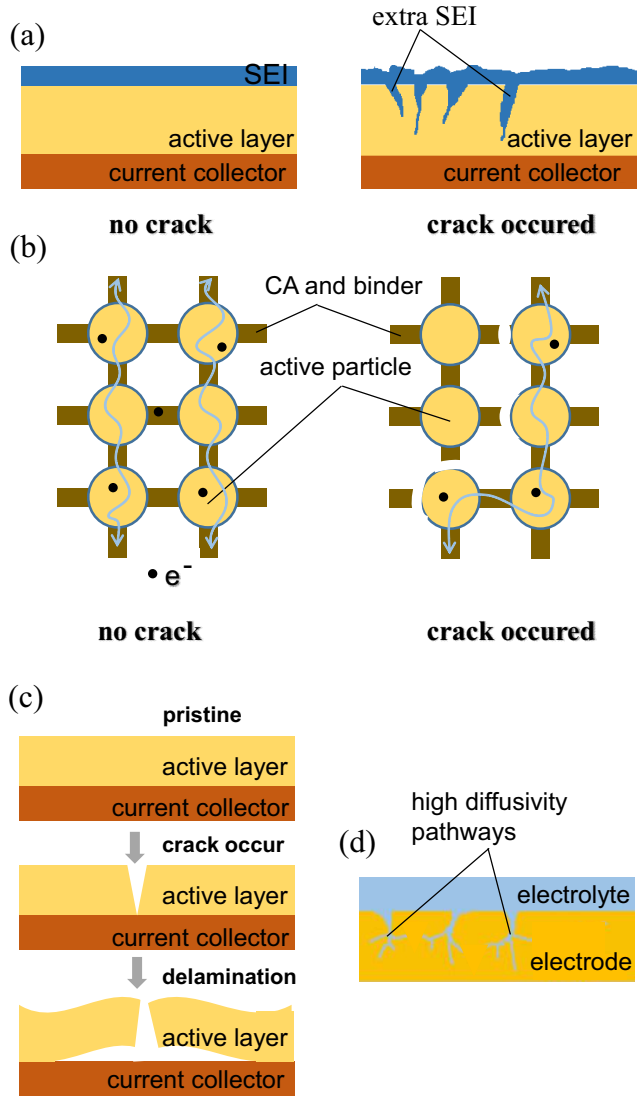


Fig. 5. Illustrations of (a) extra SEI formation caused by exposure of new surfaces, (b) loss of electrical conduction at particle level, (c) loss of electrical conduction caused by interfacial delamination, and (d) high diffusivity pathways.

Moreover, crack merging, especially the encountering of an in-plane crack and an interfacial one, would disengage blocks of active material from the electrode, leading to the loss of active material and deteriorating electrochemical performance.[33,46] In this mechanism, interfacial delamination is necessary and this typical fracture form will be discussed in Subsection 3.2.

In fact, the dominant degradation mechanism is highly related to the specific material system. For instance, Choi *et al.* suggested that fracture of the silicon particle would not directly lead to the fading of electrochemical performance.[80] By using a highly elastic binder (incorporation of 5 wt% polyrotaxane to polyacrylic acid binder), the crack propagation at the electrode level is resisted, and therefore the crack of the active layer barely appears, although the micron-scale silicon particles strongly pulverize. In this experiment, the silicon microparticle anode shows a remarkably stable cycle life. This indicates that preventing the crack of the active layer benefits the stability of the cyclic performance, or in other words, the crack of the active layer is an effective indicator of degradation. However, a recent study reported by Wang *et al.* addressed that the silicon composite electrode with the binder of polyvinylidene fluoride (PVDF) presented considerably fast capacity fading though very few cracks of the active layer were observed after electrochemical cycling,[31] as shown in Figs. 4(c) and 4(g). A possible explanation was provided that the main reason of capacity fading in Si/PVDF electrodes is the loss of conduction caused by adhesive debonding between Si particles and the CA/PVDF compound. But the adhesive debonding at the particle level does not necessarily lead to the fracture at the electrode level. In other words, at least for Si/PVDF electrodes, the crack of the active layer is neither a main cause nor an effective indicator of degradation. In fact, an experimental study reported by Maranchi *et al.* also suggested that the reversible capacity and cyclability of the thin-film silicon electrode are highly related to the adhesion between Si and Cu, even though the cracks of the active layer are triggered.[83] In this way, the importance of crack in active layers has been challenged. However, due to interaction of and competition among multiple mechanisms, it may be still too early to conclude the significance of crack in active layers for all electrode systems.

Interestingly, another mechanism possibly exists but has barely been mentioned in existing literatures. It has been reported that several types of defects (including electrode-level defects filled by electrolyte,[84] grain boundaries,[2] etc.) could improve the overall diffusivity. Since the liquid electrolyte commonly has a higher diffusion coefficient than that of the active material,[85] the crack filled by electrolyte, which is obviously sensible, can also provide high diffusivity pathways of lithium ions, as illustrated in Fig. 5(d). This may explain why the silicon composite electrodes with the binder of sodium-alginate (SA), carboxymethyl cellulose (Na-CMC), or nafion have considerable numbers of initial cracks[31] (as shown in Fig. 4) but still show better cyclic performances than that of Si/PVDF electrode.[86] The study of fracture in $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ cathodes reported by Rollag *et al.* also supported this hypothesis, although a strong and negative correlation between crack density and performance in terms of specific capacity was found.[87]

Moreover, Yang *et al.* recently observed the lithium redistribution around the crack tip in a thin-film active layer.[88] The high tensile stress at the crack tip lowers the electrochemical potential and drives lithium to gather at the crack tip. This mechanism also potentially affects the battery performance. The relationship between concentration localization and bat-

tery fading is unclear. Note that, the existence of this mechanism in composite electrode is still questionable, since the crack does not simply lie in the active material component.

In conclusion, mechanisms concerning cracks of active layers need to be investigated more sufficiently and evaluated specifically. The dominant degradation mechanism in terms of cracks in active layers can be totally distinct for different material systems.

2.3. Modeling of crack in active layer

Owing to the complexity of mechanical–electrochemical coupling in the battery system, it is quite complicated to isolate and focus on a single mechanism for intensive study via experimental approaches. In this case, theoretical approaches become important and irreplaceable. By using a modified spring-block model, the essential features of crack patterns in thin-film silicon electrodes were captured.[33] The phase field model can also be adopted to simulate the multiple mud-like crack problems.[89,90] However, due to randomness of cracks, possibly caused by inevitable defects in the battery,[2,91,92] directly modeling the exact pattern of the crack in the active layer is difficult in general.

For an active layer containing massive number of cracks, a sensible approach for characterization is introducing the crack density which describes the total crack length observed within the unit area of the surface of an active layer. The reciprocal of the crack density is defined as the average size of the crack pattern. Yang adopted the theory of energy principle and proposed an analytical method for describing the crack in an active layer.[38] The average damage size was calculated in his work. Xiao *et al.* proposed a very simple theoretical approach for predicting the average size of the crack pattern of active layer in a thin-film silicon electrode, with the assumption of uniformly distributed shear stress at the interface of the active layer and the current collector.[16] The formula of average size for the crack pattern was given by

$$L_{\mathrm{cr}} = \frac{2\sigma_{\mathrm{Y}}^{\mathrm{Si}}}{\tau_{\mathrm{cr}}^{\mathrm{int}}} h, \tag{1}$$

where L_{cr} is the critical size, $\sigma_{\mathrm{Y}}^{\mathrm{Si}}$ is the yield strength of Si, $\tau_{\mathrm{cr}}^{\mathrm{int}}$ is the interfacial shear strength between Si and Cu, and h is the film thickness. This predicted critical size is in excellent agreement with the experimentally observed crack spacing in thin-film Si electrodes.[16] A similar approach was employed to investigate the average damage size when the ratcheting effect was considered.[40] Chew *et al.* simulated the growth of a crack along the thickness direction in the active layer of thin-film silicon electrodes.[93] In their finite element analysis, it was found that the crack in the active layer would initiate at the lithiation step and subsequently form a pattern, as shown in Fig. 6. The average size of the crack pattern was also predicted in their modeling. It is noted that little research work can be found about theoretical modeling of cracks in active layers of composite electrodes, partially because of the complex mesostructure as well as the undetermined material properties of composite electrodes.

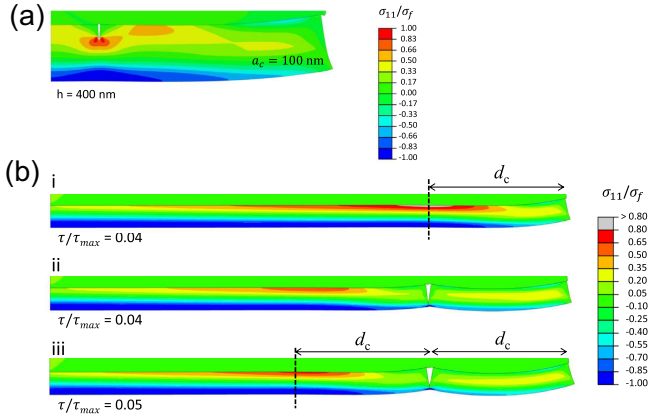


Fig. 6. Sequential cracking mechanism of the active layer in thin-film silicon electrode: (a) crack initiation, (b) formation of crack pattern. Reprinted with permission from Ref. [93]. Copyright 2014, Elsevier.

Researchers also pay close attention to how the crack of the active layer can be avoided. By using a simple one-dimensional model and calculating the stress intensity factor, Huggins and Nix estimated the critical thickness below which the fracture of the active layer would not occur.[39] Li *et al.*[33] also found the critical thickness by using a similar method proposed in Ref. [16]. The finite element analysis presented in Ref. [93] also provided a prediction of the critical film thickness. Most relevant studies agreed that the critical thickness of the active layer for no occurrence of fracture in a thin-film silicon electrode is $\sim$ 100–200 nm. However, the detailed mechanisms of the nucleation and progressive growth of the crack in thin-film silicon electrodes are quite complicated. Ding *et al.* found that as the lithium concentration increases, there exists a transition in fracture mechanism from intrinsic nanoscale cavitation to extensive shear banding ahead of the crack tip by using large-scale molecular dynamics (MD) simulations.[94] The atomistic simulations reported by Khosrownejad *et al.* further suggested that the mechanism of fracture in silicon is void nucleation and growth.[95]

Interestingly, most modeling studies focused on the first few electrochemical cycles. The evolution of the crack in the active layer along with electrochemical cycling process, which is a typical phenomenon observed in experiments,[27,28,31] still lacks sufficient understanding. Why the crack of the active layer occurs and how it would develop have attracted a lot of attention, but the mechanism on how it affects specifically the electrochemical performance is lack of understanding in these theoretical studies. Therefore, more progress needs to be made on the cross discipline between mechanics and electrochemistry.

3. Interfacial delamination

Interfacial debonding between the active layer and the current collector is another major type of mechanical failure occurred in electrodes and will be discussed in this section.

3.1. Delamination in experiment

Interfacial debonding in electrodes is mainly caused by deformation mismatch between the active layer and the current collector. The active layer swells or shrinks during the electrochemical cycling while the current collector resists its deformation. The deformation mismatch causes high interfacial stresses and finally results in the interfacial debonding.[96] Unlike cracks of the active layers which may propagate along an uncertain path, interfacial delamination develops on a fixed direction along the interface. However, interfacial delamination is hard to be observed *in situ* because this fracture conceals between the active layer and the current collector. Thus, most experimental studies of delamination in electrodes were qualitative or roughly quantitative investigations.

The most direct evidence of the occurrence of an interfacial delamination is the observation of an active layer peeling-off from the current collector. For example, He *et al.* found that some of the silicon islands which are formed after the fracture of the active layer would peel off from the current collector in a thin film silicon electrode,[97] as shown in Fig. 7(a). Xiao *et al.* suggested that there exists a critical size of island ($\sim$ 7–10 μm for thin-film silicon electrodes) beyond which the electrode islands cannot adhere to the substrate and consequently peel off.[16] A similar phenomenon of complete peeling off was also found in tin based ternary component alloy anodes.[98] However, the occurrence of delamination does not necessarily lead to the active layer's complete peeling-off, as shown in Fig. 7(b). As long as the active layer is still partially bonded to the current collector, the delamination cannot be observed from top view. In addition, the direct peeling off of the active layer is not very commonly seen in composite electrodes,[31] possibly owing to the existence of polymeric binder.

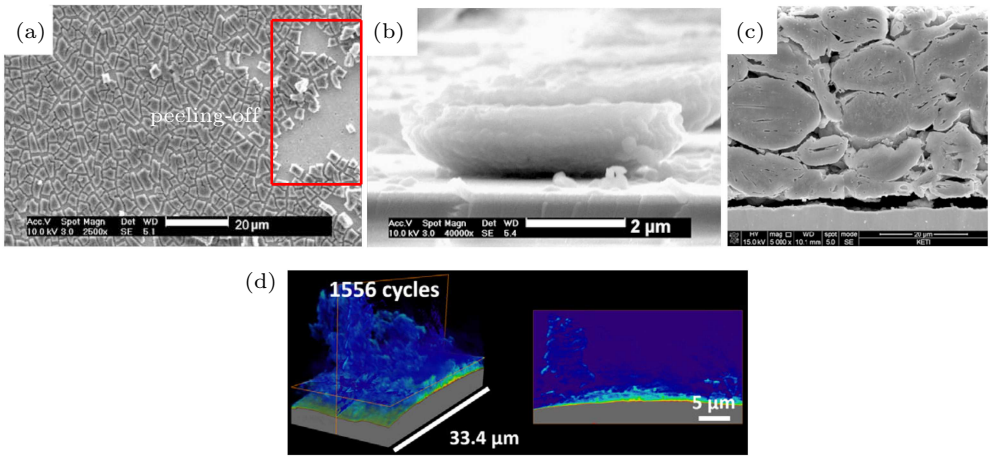


Fig. 7. Interfacial delamination (a) in thin-film electrode observed by top view (Si–Cu) (Reprinted with permission from Ref. [97]. Copyright 2012, The Electrochemical Society), (b) in thin-film island electrode observed by side view (Si–Cu) (Reprinted with permission from Ref. [99]. Copyright 2012, Elsevier), (c) in composite electrode observed by side view (graphite–Cu) (Reprinted with permission from Ref. [100]. Copyright 2013, Elsevier), and (d) in composite electrode observed by x-ray tomography method (Si–Cu) (Reprinted with permission from Ref. [35]. Copyright 2018, Elsevier).

Side view is the effective way to observe the interfacial delamination. For instance, by taking an observation of the side view of a silicon island electrode via SEM, He *et al.* captured the shape evolution of the island upon lithiation and delithiation.[99] The original silicon island was found to transform into the dome-like appearance after lithiation and change into bowl shape (Fig. 7(b)) after delithiation. The delamination of the silicon island with a width of 5 μm, which is below the critical size suggested in Ref. [16], was also observed, indicating that top view is a flawed way for confirmation of the interfacial delamination. In some other investigations, cross-sectioned electrodes were prepared to expose the concealed interfacial delamination between the active layer and the current collector. For instance, the delamination in the graphite composite electrode was observed in the cross-sectional SEM image,[100] as shown in Fig. 7(c). This method has been widely employed to characterize interfacial delamination in many electrode systems, including the $LiNi_{0.5}Mn_{1.5}O_4$ composite electrode,[101] the silicon composite electrode,[102] etc. It should be noted that the side-view observation is not perfect either, since only opening interfacial cracks (mode I) can be observed by side view while the sliding interfacial cracks (mode II) still cannot be achieved, although the mode II crack appears frequently in film/substrate systems.[96,103]

A superior approach to trace interfacial delamination is the x-ray tomography method. The three-dimensional morphological evolution can be visualized and quantified. For example, Tariq *et al.* presented this method to investigate lithia-

tion induced delamination of a composite silicon electrode,[34] as shown in Fig. 7(d). The delamination area was considerably large (44.1% of the initial interface) in this system. By using simultaneous voltage measurements, they also found that the increasing cell resistance was evidently related to the delamination process. Zhao *et al.* conducted synchrotron x-ray nanotomography studies and elucidated the failure mechanisms of the silicon electrodes from both the perspective of the mesoscopic scale and that of the macroscopic scale, which involve the adhesive debonding at the particle level and the gradual delamination at the electrode level.[35]

As mentioned in Subsection 2.2, the interfacial delamination sometimes accompanies the fracture of the active layer. This phenomenon has been widely reported,[28,31,33,36,82,83] but has not been fully dug out. For instance, how these two types of electrode-level fractures affect each other is still uncertain. Recently, Yang *et al.*[82] suggested an interaction mechanism between them in a thin-film silicon electrode, as shown in Fig. 8. They found that in the lithiation step, the surfaces of the crack in the active layer crushed each other, resulting in the interfacial delamination to release the energy. Numerical simulations were also conducted to prove this theory. Definitely, this is a sensible explanation of the interaction mechanism, but not necessarily the only one. To further clarify this issue, more effort should be put on designing experiments which allow observations of both the fracture of the active layer and the delamination.

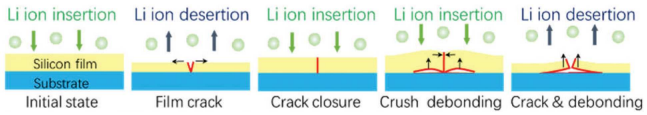


Fig. 8. Schematic illustration of interaction mechanism of crack and delamination in thin-film silicon electrodes. Reprinted with permission from Ref. [82]. Copyright 2018, Royal Society of Chemistry.

It is worth mentioning that there are two distinct micro mechanisms for comprehending the interfacial delamination in composite electrodes (for instance, silicon composite electrodes[104]): adhesive debonding and cohesive debonding. According to the manufacture procedure of composite electrodes, the active material is bonded to the current collector via the binder. If the bonding strength between the binder and the active particle is higher than that between the binder and the current collector, debonding takes place between the binder and the current collector, which is the adhesive debonding. Otherwise, the cohesive debonding is triggered between the binder and the active particle. Therefore, to suppress the onset and growth of the interfacial delamination in composite electrodes, one of the priorities is to determine the micro mechanism and figure out the weak interface to be improved.

Some other studies focused on the measurement of interfacial mechanical properties which are critically important for modeling the interfacial delamination as given in Subsection 3.3. Hu *et al.* employed 180° peel tests to determine the interfacial strengths of silicon composite electrodes.[104] The peel strength of Si/SA electrode is much higher than that of Si/PVDF electrode, which explains the better electrochemical performance of the former. Note that, the peel strength obtained by 180° peel tests also stands for the fracture toughness of mode I delamination. Recently, Guo *et al.* also used a similar method to determine the interfacial strength of graphite composite electrodes.[105] Surprisingly, the peel strength was found to increase with electrochemical cycles, indicating that the interfacial strength is not only determined by the binder, but also affected by the mesostructure change within the active layer.

Additionally, a similar phenomenon of interfacial delamination between the solid electrolyte and the electrode has also been noticed.[78,106–109] This delamination phenomenon is quite similar to that between the active layer and the current collector. They both occur in the layered structures of the battery, and are caused by the charge/discharge process. However, how their occurrences affect the electrochemical performance are distinct, which is not the focus of this review.

3.2. Impact of delamination on electrochemical performance

The interfacial delamination has been widely considered as a main reason of degradation. The mechanisms affecting electrochemical performances by delamination are much clearer than those for the fracture of the active layer. There are two primary mechanisms. One is the loss of the active material[16,35,97,98] and the other is the block of the electronic pathway at the debonding interface between the active layer and the current collector,[83,110] as shown in Figs. 9(a) and 9(b). Some other possible mechanisms, the extra SEI formation caused by the debonding areas for instance, may exist as well. But they are not predominant for delamination and are

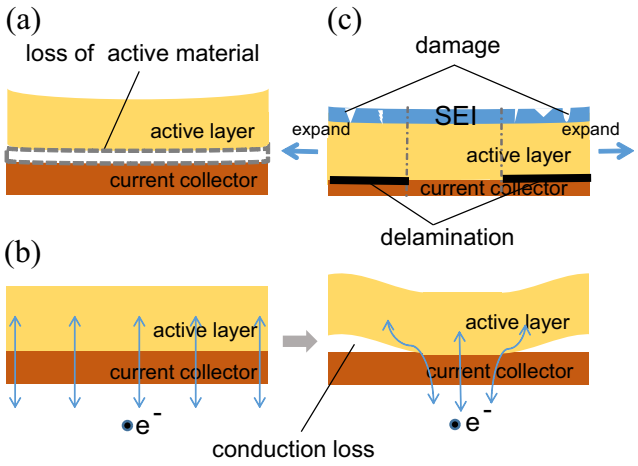


Fig. 9. Illustration of (a) loss of active material, (b) conduction loss, and (c) SEI damage induced by interfacial delamination.

not repeatedly discussed here. Another indirect mechanism induced by interfacial damage at the electrode level was recently reported by Zhang *et al.*[111] Because the damaged interface has very limited mechanical constraint on the deformation of the active layer, the freely deformed active layer would dramatically imperil the stability of the SEI in negative electrodes and consequently cause battery fading, as demonstrated in Fig. 9(c).

Note that, fracture in the active layer parallel to its surface or the interface was also reported in studies of silicon electrodes.[34,35] It is very likely to be trigged by the growth of interfacial delamination.[34] This type of electrode-level fracture is not discussed independently, as its impact on electrochemical performance is identical to that of the interfacial delamination.

However, in general, although the qualitative impact of the delamination on electrochemical performance is relatively clear, the quantitative characterizations, which include the quantitative relation between the delamination size and the battery fading (increasing resistance or decreasing capacity), are still insufficient. This is because most experimental means are unable to isolate efficiently the impact of delamination from that of the coupling failure mechanisms on the electrochemical performance.

3.3. Modeling of delamination

Owing to the difficulty in observing the delamination and explaining its mechanism via experimental approaches, theoretical approaches are indispensable. Note that, interfacial properties, including strength, fracture toughness, etc., are required for modeling the delamination. Besides the peel test (very likely only suitable for composite electrodes) mentioned in Subsection 3.1, atomistic simulations are able to provide information of these properties, especially for thin-film electrodes.[112,113]

There have been lots of modeling studies focusing on the interfacial delamination in lithium-ion battery electrodes. By using the theory of energy principle, Yang proposed an analytical method for describing the delamination in an electrode.[38] Their calculation results suggested that the onset of the crack in the active layer would be followed by the occurrence of interfacial debonding. Haftbaradaran *et al.* investigated the delamination of silicon island electrodes by employing the energy release rate in couple with the shear-lag model.[114] The critical sizes (width and radius) below which the delamination onset or the complete delamination can be avoided were provided. The critical size in Ref. [114] was expressed as follows:

$$l_{\mathrm{cr}} = \sqrt{\frac{2hE\Gamma}{(1-\upsilon^2)\tau_0^2}}, \tag{2}$$

where l_{cr} is the critical electrode size of avoiding the delamination initiation, E is the Young's modulus of the active layer, Γ is the interfacial fracture energy, υ is the Poisson's ratio of the active layer, and τ_0 is the interface shear strength. Interestingly, this formula has nothing to do with the lithium concentration. Haftbaradaran also investigated the stress intensity factor in couple with the solute segregation at the edge of film/substrate interface in thin-film electrodes.[115,116] The stress-induced solute segregation was found to have strong effects on the debonding onset. Guo *et al.* presented analytical solutions of the pop-up delamination between the active layer and the current collector in solid-state electrodes by calculating the energy release rate.[117] They suggested that the pop-up delamination can be suppressed by stiffening or thinning the solid electrolyte. Moreover, Lang *et al.* creatively proposed a theoretical model of spontaneous delamination induced by internal stress, which explained the formation of single Si nanosheets observed in experiments.[118] They also adopted MD to simulate the de-lithiation process of c-$Li_{13}Si_4$, and two key parameters (namely, mismatching stress and interfacial fracture energy) in the spontaneous debonding model were obtained. The critical thickness in the spontaneous delamination was written as

$$h_{\mathrm{cr}} = \frac{E\Gamma}{(1-\upsilon)\sigma_{\mathrm{m}}^2}, \tag{3}$$

where h_{cr} is the critical thickness and σ_{m} is the internal mismatch stress.

To comprehensively investigate the progressive growth of the interfacial delamination, the cohesive model (or cohesive zone method) has been commonly employed.[119–127] By using a numerical approach, Pal *et al.* evaluated the impacts of yield strength and modulus of the substrate on the progressive delamination in a thin-film silicon electrode.[119] Liu *et al.* further studied the effect of the elastoplastic behavior of the active layer on the delamination process in a thin-film silicon electrode by using the finite element analysis,[120,121] as demonstrated in Fig. 10. The shape of the active layer predicted in Ref. [120] (Fig. 10(b)) was quite similar to that observed in experiment[99] (Fig. 7(b)). Recently, Wen *et al.* also investigated numerically the effect of interfacial debonding and elastoplastic behavior on the macro deformation of the high-capacity negative electrode.[122] Additionally, both semi-analytical[96,123–125] and analytical approaches[126,127] have been developed in our previous work to evaluate the progressive delamination in electrodes. Key parameters were obtained in these studies and design suggestions to avoid delamination were given. In general, small sizes, soft materials, and low charge/discharge rates are beneficial for the integrity of an interface. Specifically, when the material system is fixed, two strategies are provided for suppressing delamination: lowering the size of the active layer to make it below the corresponding

critical value or partially charging/discharging the battery to given limits.[124] The expression of the critical size obtained in Refs. [124,126] is

$$l_{\rm cr} = \frac{6\Gamma}{\tau_0 \Omega c_{\max}}, \tag{4}$$

where $\Omega c_{\max}$ is the maximum volume change. By comparing Eq. (2) with Eq. (4), it can be found that formula (4) depends on the lithium concentration, but somehow is irrelevant to the modulus and the thickness of the active layer. Interestingly, the theoretical work of Ref. [124] successfully guided the experiment in Ref. [28] by following the idea of partial lithiation for suppressing the mechanical degradation and improving the long-term capacity of silicon composite electrodes.

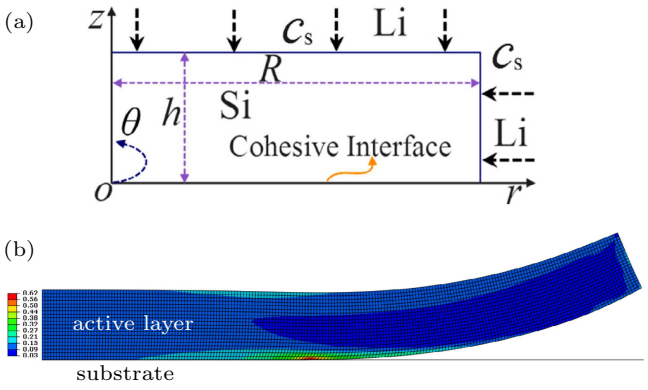


Fig. 10. Modeling of progressive delamination in silicon electrodes: (a) schematic diagram, and (b) illustration of modeling result. Reprinted with permission from Ref. [120]. Copyright 2015, Elsevier.

Although the cohesive model has been widely used to evaluate the progressive delamination in electrodes,[119–126] its reliability was challenged recently.[127] It was found that the cohesive law shape, commonly considered as an insignificant factor,[128–130] affects the modeling results when the interface is ductile. Especially for silicon island electrodes, different choices of cohesive laws may influence the theoretical predictions.

As a typical mechanical problem, delamination in electrodes can be modeled with well-developed theoretical methods. However, similar to the case of crack in active layers, more quantitative evaluations of the concrete influence of the delamination on the electrochemical performance are required.

4. Fracture of metallic foil in electrode

In this section, fracture of metallic foils existing in electrodes will be discussed. Two specific types, namely, the fracture of the current collector and that of the lithium metal electrode, will be involved. Since these two fracture phenomena both belong to the fracture of metallic foils under the electrochemical environment within batteries, discussions of them are included together in this section. The difference between them is that the current collector serves as an electrochemical inactive component while the lithium metal foil is electrochemical active.

4.1. Fracture of current collector

Metallic foils are commonly used as current collectors in composite electrodes. In a lithium-ion battery, aluminum and copper are typically chosen for the current collectors of cathode and anode, respectively.[131,132] In fact, corrosion and fracture of the aluminum and copper foils in the electrochemical environment within lithium-ion batteries have been noticed very early.[132] The pitting corrosion of aluminum by electrolytes as well as the environmentally assisted cracking of copper at or near the lithium potential was reported. The corrosion of the current collector has been widely investigated.[133–136] However, for quite a long time, researchers in both mechanical field and battery field did not pay enough attention to the fracture of the current collector.

Recently, due to the increasing deployment of electrical vehicles worldwide, mechanical abuse and subsequent failure of lithium-ion batteries have become a research focus.[137–141] In this case, the importance of the mechanical integrity of a current collector has been reevaluated. The attempt of decreasing the thickness of the current collector for an increase in the specific capacity of a battery brings about the problem of mechanical instability of the current collector.[142] When the battery is subject to an accidental impact loading, it is the metallic current collector that plays a major role in keeping the battery safe.[143] The composite active layers and the polymer separator show little structural resistance to mechanical compacts. If the current collectors fail mechanically, the structure within the battery would be compromised easily, leading to the direct contact of the conducting electrodes and consequently the onset of short-circuit followed by thermal runaway.

By observing 3D x-ray computed tomography of a spherically indented Li-ion battery, Wang *et al.* found the occurrence of brittle fracture in a large area of a copper current collector foil when the battery was mechanically abused.[144] As shown in Fig. 11, the mud-like cracks were observed within the copper layer. They also provided a sensible explanation why the fragmentation of the current collectors had been barely noticed and reported. The reason is the composite active layers containing polymeric binders can deform plastically and hold the materials together, which makes the fracture of the current collectors hide inside and not be seen under optical and electron microscopy.

Interestingly, Zhu *et al.* analyzed the failure mechanisms of 18650 battery cells under axial compression and found no fracture in the current collectors.[145] This indicates that the structural failure of the battery under mechanical abuse is sensitive to the type of the mechanical loading. The me-

chanical response of the electrode under an external force applied on the battery is not sufficiently understood. This actually brings about the issue of correlation between the response at the battery level and that at the electrode level, which may involve multiple phenomena including the anisotropic behavior of the current collectors[143] and the buckling of the electrode.[144,146]

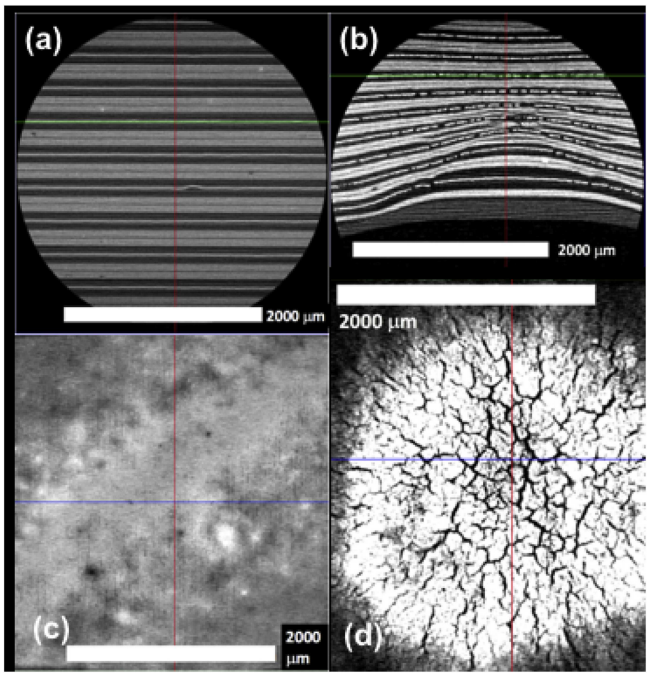


Fig. 11. The x-ray tomography images: cross-sections of (a) the virgin and (b) the indented cells; in-plane sections of (c) the virgin and (d) the indented cells. Reprinted with permission from Ref. [144]. Copyright 2017, Elsevier.

Nevertheless, an urgent issue for improving the battery safety is to find out when and how the fracture of the current collector occurs under mechanical abuses of the battery. Yet difficulties of settling this issue are manifest and need to be overcome. For instance, one of these difficulties is to expose the hidden fracture which is not easy to find. Another difficulty is dealing with the extreme conditions of the short circuit and the thermal runaway which usually evolves rapidly and can be quite dangerous.

4.2. Fracture of lithium metal electrode

Due to its high theoretical energy density and the lowest electrochemical potential, lithium metal is considered as a very promising anode material for high energy density batteries.[147–149] However, the application of lithium metal electrode in traditional rechargeable batteries is not successful, owing to the poor practical cycle life and safety issues caused by lithium dendrites.[140,148,150] Recently, due to increasing interest in Li–air and Li–sulfur batteries as well as the fast development of solid electrolytes, lithium metal electrodes were brought into sharp focus again.[151–155]

Investigations on lithium metal electrodes focused mostly on lithium dendrites,[156–159] yet interestingly, the intrinsic relation between fracture and dendrite has been barely discussed. When the battery is charged/discharged, the lithium metal electrode experiences electrochemical plating/stripping. Lithium ions form nuclei on the electrode surface and grow into dendrites during this process. Monroe and Newman found that the potential at the tip of the dendrite is different from that at its base due to the curvature of the surface.[160] Because of the difference in potential, the dendrite was found to grow preferentially along the protrusion direction.[152,161,162]

In fact, the existence of cracks in lithium metal electrodes has been reported by several research groups.[163,164] The fracture may initiate during the electrochemical cycling or during the manufacturing process before cycling. The edge of the crack provides an extremely high curvature of the surface and hence is believed to result in a convenient condition of dendrite growth.[164] The relationship between fracture and dendrite has been noticed, but is still lack of sufficient investigations.

The classical fracture problem of a metallic foil is not a latest mechanic subject.[165–167] It is the electrochemical environment within lithium-ion batteries that complicates the problem for which the deposition of lithium and the corrosion by electrolyte have to be considered. Further studies on fracture of metallic foils under multi-field environment are needed, whether they be experimental or theoretical.

5. Conclusion

In this review, fracture occurred at the electrode level in lithium-ion batteries has been focused on. Three typical types of electrode-level fractures, namely, the fracture of an active layer, the interfacial delamination, and the fracture of metallic foils in electrodes (including the current collector and the lithium metal electrode), have been discussed. Different fracture phenomena and their corresponding mechanisms in composite electrodes and thin-film electrodes have been revealed.

The crack of the active layer is considered as an indicator of mechanical-electrochemical degradation in plenty of investigations. But it is still unclear how the cracks of the active layer deteriorate the electrochemical performance of the battery. Four sensible mechanisms are concluded: the extra SEI formation caused by exposure of new surfaces, the loss of electrical conduction at the particle level, the loss of electrical conduction caused by interfacial delamination, and the high diffusivity pathways.

The mechanism influencing the electrochemical performance by delamination in electrodes is relatively manifest. Three mechanisms have been discussed: the loss of the active material, the conduction loss, and the SEI damage. The critical issue of investigating the interfacial delamination is how to make it easy to observe this phenomenon, since the crack conceals between the active layer and the current collector. Some

of the existing techniques used for observing the delamination have been mentioned. It is concluded that quantitative investigations on the relationship between the electrochemical performance and the mechanical damage are still rare.

Fracture of the metallic foil in electrodes was actually reported very early. But this fracture phenomenon and its consequences on the electrochemical performance in the lithium-ion battery did not receive enough attention, although it is of great importance for the cell safety. The fracture of metallic foils becomes complex under the electrochemical environment within a battery, which would involve the deposition of lithium and the corrosion by electrolyte. In this review, fracture problems in the current collector and the lithium metal electrode have been mentioned. The urgent issues in terms of extreme conditions for investigating metallic foil fracture in a battery need to be settled.

Fracture in electrodes of the lithium-ion battery is actually complex, since it may involve fractures in and between different components of the electrode and the electrochemical coupling needs to be included as well. Fracture damages the integrity of the electrode structure and compromises the whole cell performance. To further comprehend and avoid the electrode-level fractures with a multi-scale and multi-physical point of view, advancing the cross discipline between mechanics and electrochemistry is a priority.

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