In-situ measurements of mechanical and volume change of LiCoO₂ lithium-ion batteries during repeated charge–discharge cycling by using digital image correlation

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Abstract In-situ morphological evolution of displacement in pouch-type commercial lithium-ion batteries during multiple fifty-five electrochemical charging-discharging cycles was measured via digital image correlation technique. The maximum principal strain on the battery surface reached 0.35% during 55 cycles. The whole volume change analysis of LIBs shows that the maximum volume change rate arrives at 4.27% at the fully 52nd charging end, and the maximum residual volume change rate is about 2.89% at the 54th discharging end. The surface morphologies of cathodes and anodes before and after electrochemical cycling were observed by scanning electron microscopy. The elastic modulus of the copper foil in LIBs decrease from as-received 16.7 GPa to 10.6 GPa after 55 cycles by using tensile tests.

Keywords Lithium-ion battery; Digital image correlation; Volume change rate;

In-situ measurements

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1. Introduction

Lithium-ion batteries (LIB) are the preferred energy storage devices for electric and hybrid-vehicle applications. They are increasingly being used as power sources for portable electronics because of their many advantages, such as high power and energy densities, high power outputs, slow self-discharge, superior cycle performance, and long cycle life [1-3]. In practice, it is known that the chemical compositions of the active materials vary with charge-discharge cycles, which may result in strain and volume change of electrode, the increasing of its impedance, and the deteriorating of individual commercial LIB capacity. Finally, it would lead to the serious imbalance of the electrochemical properties (discharging capacity, open circuit voltage (OCV), and cell impedance) [4-9]. Furthermore, the accumulated strain and stress due to the electrode swelling upon cycling may induce crack nucleation and propagation, and other failure in batteries [10-12]. It is important to quantify the effect of strain on the mechanical-chemical properties of batteries to evaluate their reliability and life **in service** [13]. Quite a few in-situ characterization studies have been concentrated primarily at particle and electrode levels by using atomic force microscopy nanoindentation [14, 15], nanomechanical tensile tests [16], scanning probe microscopy [17], X-ray diffraction [9, 18, 19], online electrochemical mass spectrometry [20] and substrate curvature [21]. However, to the authors' knowledge, those conventional displacement methods including linear voltage displacement transducers and strain gauges are still used. It is difficult to obtain the full/local displacement or strain field of cathodes and anodes in LIBs. In recent years, digital

image correlation (DIC) technique has been increasing quickly as a powerful tool for non-contact and non-destructive displacement measurement method **under different scales and experimental temperatures** [22-24]. However, to the best of our knowledge, less effort has been devoted to use DIC techniques to in-situ study the evolution of full/local strain fields and volume change in batteries subjected to typical electrochemical operations. Chen *et al.* studied the roles of binders in the mechanical integrity of electrodes for LIBs by the coupled micro-scratch and two-dimensional DIC techniques [25]. Leung *et al.* applied three-dimensional (3D) DIC technique to real-time observe the evolution of displacement and strain of a pouch-type LIB under the first five cycles [26]. However, the measurement was limited to several charge-discharge cycles and it was difficult to draw implication for full lifetime of the batteries. Therefore, the effect of external mechanical forces on the overall performances of LIBs remains poorly understanding. It is difficult to predict how the overall expansion or contraction of LIBs arises from stress and strain within its constituent elements after many charge–discharge cycles.

The aim of the present study is to explore the strain variation and volume change of pouch-type commercial LIBs under multiple fifty-five cycles by using 3D DIC technique. The qualitative displacement and volume variation were provided as a function of state-of-charge (SOC) along the x -, y -, and z -directions, respectively. The surface morphologies of the electrodes were observed by scanning electronic microscopy (SEM), and the elastic modulus of copper foil in LIBs before and after cycling was measured by **tensile tests**. The results may improve the understanding of

the effects of the internal structure of an LIB on its mechanical integrities before and after many cycles.

2. Experimental

2.1 DIC tests

The commercial pouch-type lithium-ion polymer batteries used in this investigation were Lishen SP505456SA cells with a nominal capacity of 1920 mA h. Cobalt oxide and graphite were used as active materials for the cathode and anode, respectively. The interior structure of the aluminum plastic pouch was constructed by winding the ribbon-like electrode and separator to form a “jelly-roll” structure, as shown in Fig. 1. Cells with copper and aluminum foil were used current collectors for the negative and positive electrodes, respectively. The cell dimension is 4.9 mm × 54.5 mm × 56.5 mm. During 3D strain measurements, the galvanostatic charging–discharging was performed with 1 C rate at room temperature by using a battery testing system TC53 (CT-3008W-5V3A-S1, NEWARE TECHNOLOGY LIMITED, Shenzhen, China). As shown in Fig. 2, the battery was charged at a constant current of 1 C to 4.2 V for an overall charge cycle of 1 h, and then discharged at a rate of 1 C down to 3.0 V. The SOC of the battery was defined as the percentage of the charged capacity over its rated value (c.a. 1920 mA h).

As shown in Fig. 3, 3D DIC analysis was conducted by using a GOM Aramis system (ARAMIS, GOM Co., Germany). Before the tests, a random thin black speckle pattern was sprayed on the surface of the battery specimen. A resolution 1624 × 1236 pixels charge coupled device camera (GOM Co., Germany) equipped with a lens of 50 mm focal length was used to observe and record the dynamic movements

of the patterns. The frame rate of the camera can achieve up to 15 Hz, with reduced image resolution up to 30 Hz. The sampling rate of the camera was set as one image per second in this work. The detailed information was described in our previous works [23, 27-31]. Two illumination white lights (GOM Co., Germany) were used to compensate the light intensity and decrease the influence of high temperature thermal radiation. During the displacement analysis, the interesting region was divided into a lot of small square reference subsets of 15×13 pixels. The gap between two subsets was 7×7 pixels. The scale factor was 0.1 mm/pixels. The strain post-processing analysis was achieved with the available commercial DIC professional software (ARAMIS, Version 6.2, GOM, Germany).

After electrochemical cycling experiments, the fully discharged batteries were carefully disassembled in a glove box. The cathodes and anodes were immediately placed in a diethyl carbonate solution to remove lithium salts that had adsorbed on the electrodes. The treated electrodes were then dried in a drying box for SEM observations. To compare the electrode morphology change before and after cycling, an as-received battery specimen was disassembled and observed by SEM.

2.2 Tensile tests

The uniaxial tensile tests were performed to study the variation of elastic modulus of the copper foils before and after electrochemical cycling. The coating materials of the discharged and as-received battery specimens were carefully removed, and the treated copper foils were dried in the drying box. Copper foil specimens were subsequently cut along 0° to the rolling direction with a gauge length of 100 mm and a width of 15 mm. Tensile tests were performed at room temperature by using a

universal testing machine (Model: REGER 2000) with the speed rate of 1 mm/min.

3. Results and discussion

3.1. Analysis of displacement and strain

3.1.1. Displacement distribution during electrochemical cycling

During 1 C charging–discharging cycling, the displacements along the x -, y -, and z -directions of the LIB at the center of the battery surface were measured by using DIC system under different SOC. Fig. 4 illustrates the variation of displacement, u_{xx} , along the x -direction at 30%, 70%, and 100% SOC under different cycle numbers. As shown in Fig. 4(a), u_{xx} increases up to 9.0 μm at the fully charged state and then decreases down to 3.0 μm after the discharge process. These results are consistent well with the available data tested by Leung et al [26]. In our work, after the 25th cycle, the maximum value on the battery surface is about 18.0 μm at the fully charged state, and then it gradually decreases to 16.0 μm after the discharge process, as shown in Fig. 4(b). It is worth noting from Fig. 4(c) that after the 50th cycle, the u_{xx} distribution displays apparent compressive and tensile displacement state with an upper and lower axial symmetric feature. It is the first time to find these interesting results by using DIC measurements under multiple over 50 charging-discharging cycles. Similarly, the maximum value on the battery surface is about 23.0 μm at the fully charged state, and then it gradually decreases to 19.0 μm after the discharge process. It clearly indicates that the SOC and cycle number have an important influence on the deformation of LIBs in practice applications.

Fig. 5(a) illustrates that the y -displacements, u_{yy} , increases up to 24.0 μm at the

fully charged state and then decreases to 13.0 μm after the 1st discharge process. After the 25th and 50th cycles, the variation of u_{yy} became slightly less obvious. In figures 5(b) and (c), the maximum value is about 32.0 and 35.0 μm , respectively, at the fully charged state. They decrease to 21.0 and 25.0 μm after the discharge process, respectively. These results are in good agreement with that obtained by using LVDT [32]. In the initial state, the displacement mapping was relatively uniform over the battery surface. However, after many charging-discharging cycles, the compressive and tensile u_{yy} concentrates on the right and left sides of the battery surface.

It is found from Fig. 6 that the displacement value in the z -direction, u_{zz} , is relatively larger than the increments of the x - and y -displacements under the same experimental conditions. As shown in Fig. 6(a), during the 1st charge-discharge cycle, u_{zz} on the lower left-hand side of the battery increases steadily and reaches 113 μm at 100% SOC, which is about 12.6 and 4.7 times larger than those in x - and y -directions, respectively. And then it decreases to 32 μm after the 1st discharge process. As the cycle number increases, u_{zz} becomes more apparent. Figs. 6(b) and (c) demonstrate that the maximum value of u_{zz} increases up to 155 and 195 μm , respectively, at the fully charged state. They gradually decrease to 82 and 120 μm after the discharge process. During the 1st, 25th and 50th charge-discharge cycles, given that the initial thickness of the battery is 4.9 mm along the z -direction, the resulting strain values along the z -direction were as high as 2.31%, 3.16%, and 3.98%, respectively. These results are similar to 3.67% [26] and 1.5% [32] by using charge-discharge rates of 1 and 0.4 C, respectively. It is found that the u_{zz} variation is dependent on the thickness

of the jelly-roll structure. The evolutions of u_{xx} , u_{yy} and u_{zz} were attributed to the expansion and contraction of the distinct anode/separator/cathode layer due to the charge–discharge effect. The lattice swelling and shrinking processes of the active materials may induce the reversible trends of the x -, y -, and z -displacement. Generally, the graphite anode materials exhibit volume expansion during charging [33, 34], whereas the LiCoO_2 cathode materials undergo contraction. As the volume expansion of the negative electrode is usually far more significant than the volume contraction of the positive electrode, the whole expansion deformation can be observed. The volume expansion and contraction of active materials are highly reversible processes [32, 34]. It means that the active materials can contract back to their original size at the end of the discharge cycle. By contrast, the physical and electrochemical swelling caused by binder expansion and particle stacking are generally less reversible [34, 35]. These reasons may be attributed to the phenomenon of residual z -displacement observed after electrochemical cycling.

To further discuss the change of u_{zz} , the data was extracted and shown in Fig. 7. It can be seen the evolution of u_{zz} under different three phases, 1st–5th, 21st–25th and 51st–55th charge–discharge cycles at 1 C rate. In the first part of the curve, u_{zz} increases from 0 to about 113.4 μm during the first charging phase, and then gradually decreases up to about 34.7 μm during the first discharging end. The maximum displacement difference, $\Delta u_{zz,\text{max}}$, between the 1st and 5th cycles strong displays the effect of the electrochemical process on the mechanical deformation of battery. It changes repeatedly with this form during subsequent charging-discharging cycles. It

is interesting to observe that the maximum value is about 117.3 μm at the end of the second charging phase, which is larger than 113.4 μm above. Similarly, at the end of second discharging phase, the residual u_{zz} is about 45.2 μm larger than 34.7 μm last phase. It strong verifies that the accumulated residual displacement may damage the performances of LIBs and accelerate their failure. In the 21st charge-discharge cycle, u_{zz} increases from the residual value 85.6 μm to the peaking 155.3 μm . Eventually, it decreases slowly to about 85.5 μm . The effect of the accumulated displacement phenomena seems to weaken. At the end of the 24th discharging cycle, the residual u_{zz} is about 96.8 μm . For the 51st–55th charge–discharge cycles, it is found that the residual u_{zz} after each charge–discharge cycle increases from 122.7 μm to 140.6 μm . The peaking value of u_{zz} increases from 195.2 μm to 196.7 μm , as shown in the third part of the curve in Fig. 7. The variation of u_{zz} in our work clearly shows the significant accumulated displacement effect.

3.1.2. Major strain distribution during electrochemical cycling

Fig. 8 shows the evolution of the major strain of LIB in the x - y plane at 30%, 70%, and 100% SOC under different cycle numbers. As shown in Fig. 8(a), the major strain seems to a few change during the first cycle. With increasing cycle number, however, the change of the principal strain increases rapidly. It is found from fig. 8(b) that the principal strain on the upper-right side of the battery is larger than that on the lower-left side of the battery during the 25th charging process. This phenomenon is more apparent in Fig. 8(c), where the maximum principal strain on the right side of the battery can reach about 0.35%, and the principal strain on the upper-right side of

the battery tends to be larger than that in other areas of the battery. The observed principal strain distributions are tightly associated with the design of the jelly-roll battery structure.

3.2 The volume change of LIBs during electrochemical cycling

Fig. 9 shows the relative volume change, $\Delta V / V_0$, of lithium-ion battery under different charge–discharge cycling at 1 C rate, where $\Delta V = V_n - V_0$ is the volume change, V_n and V_0 are the n^{th} charging and original volume of the tested battery, respectively. In the first part of the curve, the relative volume expansion increases from 0 to about 2.37% during the first charging phase, and then the relative volumetric shrinkage gradually decreases up to about 0.73% during the first discharging end. It changes repeatedly with the similar form during the subsequent charging-discharging cycles. Here ΔV_{max} denotes the increment of the relative volume change at the peaking in each cycle, $\Delta V_{\text{max}} = (V_{n,\text{max}} - V_{k,\text{max}}) / V_0$, where $V_{n,\text{max}}$ and $V_{k,\text{max}}$ are the maximum volume of the n^{th} and k^{th} charging-discharging cycles ($n > k$), respectively. The detailed ΔV_{max} evolution is shown in Fig. 10(a). It is interesting to observe from Fig. 9 that the maximum $\Delta V / V_0$ is about 2.46% at the end of the 2nd charging phase, which is larger than 2.37% above. Similarly, at the end of the 2nd discharging phase, the volumetric shrinkage is about 0.92% larger than 0.73% last cycle. In the second part of the curve, the volume expansion rate increases from the residual value 1.78% to the peaking 3.25% during the 21st charge-discharge cycle. Finally, the volumetric shrinkage gradually decreases to about 1.79%. At the end of the 24th discharging cycle, the volumetric shrinkage is about 1.98%. For the

51st–55th charge–discharge cycles, it is found that the volume expansion rate after each charge–discharge cycle increases from 2.55% to 2.89%. The peaking value of $\Delta V / V_0$ increases from 4.05% to 4.27%, as shown in the third part of the curve. These results indicate that the residual volume expansion does play a significant influence on the performances of LIBs.

To further discuss the volume increment tendency of LIBs during repetitive charging-discharging processes, the evolution of $\Delta V_{\max}$ in our tests is extracted and shown in Fig. 10(a). In the first part of the curve, $\Delta V_{\max}$ increases from 0 to about 2.33% during the first full charging cycle, and then it gradually decreases up to about 0.09% during the second full charging cycle. It increases linearly with the increasing of 2st–5th charge–discharge cycles. In the second part of the curve, $\Delta V_{\max}$ is about 0.038% at the 22nd charging cycle. It is interesting to observe that $\Delta V_{\max}$ also displays the similar linear increasing trends, compared with that in the former 1st – 5th cycles. But the increment amplitude gradually becomes small. At the end of the 25th cycle, $\Delta V_{\max}$ is about 0.052%. During the 51st – 55th cycles, it is found that $\Delta V_{\max}$ varies within a range of 0.92% to –0.14%. The residual volume change rate of LIBs after each electrochemical cycle increases with a parabola function within 55 cycles in our work, as shown in Fig. 10(b). The accumulated volume may affect the performances and applications of LIBs.

3.3. Electrode microstructure observations

The tested battery was unpacked in a drying box after different charge-discharge cycles. The microstructures of the individual electrodes were examined by SEM. For

comparison, an as-received battery was also disassembled. Fig. 11 shows that there seems to be little apparent microstructure change of surface morphologies of as-received and cycled cathodes. The results are similar to the available results [36]. However, the surface morphologies of the corresponding anode active materials represent a few apparent change before and after electrochemical cycling, as shown in Fig. 12(b). It can be seen that a passive film exists on the electrode surface. It would be effectively hinder lithium ions from penetrating through to the active anode [37], thereby contributing to increase in the surface resistance. The passive film formation also causes the capacity to fade because of the consumption of the electrolyte components [36].

3.4. Elastic modulus measurements of copper foils

Fig. 13 shows the stress–strain curves of copper foil before and after different cycles at 1 C rate. During the linear elastic stage, the strain of the copper foil increases linearly with the increasing of external tensile loads at the *ab* and *de* curves, respectively. The calculated elastic modulus of the copper foil of the as-received sample is approximately 16.7 GPa. The elastic modulus of the copper foil after 55 electrochemical cycling is about 10.6 GPa under the same tensile rate of 1 mm/min. It is seen that the mechanical performances of copper foil gradually decreases with the increasing of electrochemical cycling.

4. Conclusions

The displacement and strain distributions of pouch-type LIBs were in-situ monitoring by using 3D DIC under over 50 multiple charging-discharging cycles. The

surface morphologies of electrodes in LIBs before and after charging-discharging cycles were observed by SEM. The main conclusions can be summarized as follows,

(1) The evolution of the x -, y -, and z -directional displacements were mainly determined as the functions of state-of-charge at room temperature. The increments of z -displacement were more apparent than those of other two direction displacements. The maximum principal strain of the battery reached 0.35% after 55 cycles.

(2) The volume change rate of LIBs under 1st–5th, 21st–25th and 51st–55th charge–discharge cycles at 1 C rate was successfully real-time measured by using DIC technique. The maximum volume change rate arrives at 4.27% at the fully 52nd charging end, and the maximum residual volume change rate is about 2.89% at the 54th discharging end. The electrochemical charge-discharge process plays a significant role in the shrinkage and swelling of the internal structure in LIBs.

(3) SEM observations show that the surface morphologies of the anodes have a few apparent change before and after 55 charge-discharge cycles. The elastic modulus of the copper foil before and after electrochemical cycling decreases from 16.7 GPa as-received to 10.6 GPa by tensions.

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Figure captions

Fig. 1 Schematic of the commercial pouch type lithium-ion polymer battery and the internal jelly-roll structure of LIBs.

Fig. 2 The charge–discharge voltage profile at 1 C rate in the present work.

Fig. 3 (a) Schematic of a typical lithium-ion battery test system, (b) Setup of the digital image correlation and battery test systems in our laboratory.

Fig. 4 The evolution of x -displacement distributions in LIBs at 30%, 70%, and 100% SOC after different cycle numbers, (a) the 1st cycle, (b) the 25th cycle, and (c) the 50th cycle.

Fig. 5 The distributions of y -displacement in LIBs at 30%, 70%, and 100% SOC with different cycle numbers, (a) the 1st cycle, (b) the 25th cycle, and (c) the 50th cycle.

Fig. 6 The distributions of z -displacement in LIBs at 30%, 70%, and 100% SOC with different cycle numbers, (a) the 1st cycle, (b) the 25th cycle, and (c) the 50th cycle.

Fig. 7 The evolution of the z -displacement of a representative lithium-ion battery under the 1st–5th, 21st–25th and 51st–55th charge-discharge cycles at 1 C rate.

Fig. 8 The change of major strain in the lithium-ion battery at 30%, 70%, and 100% SOC with different cycle numbers, (a) the 1st cycle, (b) the 25th cycle, and (c) the 50th cycle.

Fig. 9 The volume variation of LIBs under different continuous charge–discharge cycles at 1 C rate, only displaying the 1st–5th, 21st–25th and 51st–55th three charge-discharge phases.

Fig. 10 (a) The evolution of $\Delta V_{\max}$ during different cycles. (b) The relationship of residual $\Delta V / V_0$ after each cycle and cycle number.

Fig. 11 SEM images of the cathodes in fully discharged lithium-ion batteries after different cycle numbers at 1 C rate, (a) as-received, (b) after 55 cycles, (c) a magnified view of the spot marked with square in (a), (d) a magnified view of the spot marked with square in (b).

Fig. 12 SEM images of the anodes in fully discharged lithium-ion batteries with different cycle numbers at 1 C rate, (a) as-received, (b) after 55 cycles, (c) a magnified view of the spot marked with square in (a), (d) a magnified view of the spot marked with square in (b).

Fig. 13 Stress–strain curves of the copper foil before and after charge-discharge cycles at 1 C rate.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

(c)
Fig. 5

Fig. 6

Fig. 7

(a)

(b)

(c)

Fig. 8

Fig. 9

Fig. 10.

Fig. 11

Fig. 12

Fig. 13