Contents lists available at ScienceDirect

Nano Energy

journal homepage: www.elsevier.com/locate/nanoen

Operando monitoring of the open circuit voltage during electrolyte filling ensures high performance of lithium-ion batteries

Hao Cui^a, Dongsheng Ren^a, Mengchao Yi^{b,c}, Sixuan Hou^{b,d}, Kai Yang^a, Hongmei Liang^a, Xuning Feng^b, Xuebing Han^b, Youzhi Song^{a,*}, Li Wang^{a,*}, Xiangming He^{a,*}

^a Institute of Nuclear and New Energy Technology, Tsinghua University, Beijing 100084, China

^b State Key Laboratory of Automotive Safety and Energy, Tsinghua University, Beijing 100084, China

^c R&D, Beijing WeLion New Energy Technology Co., LTD, Beijing 100084, China

^d College of Mechanical Engineering, University of Shanghai for Science and Technology, Shanghai 200082, China

ARTICLE INFO

Keywords: Open circuit voltage Wetting H-type electrolytic cell Equivalent circuit analysis Lithium-ion battery

ABSTRACT

The wetting process plays an important role in battery production efficiency and battery quality, including available energy density, cycling life, power, and battery consistency. A convenient and efficient method for characterizing electrolyte filling, which becomes more crucial for lithium-ion batteries (LIBs) with a large format or super energy density, is desperately needed in the battery industry. Herein, we propose the operando monitoring of the open circuit voltage (OCV) during the electrolyte filling process. It is found that battery OCV drops dramatically to -0.80 V at the beginning of filling (within 300 ms), and then characteristically recovers to 0.10 V with the wetting process, involving valuable information about the electrolyte filling process. Insights of the correlation between electrolyte wetting process and battery OCV are further stimulated using an equivalent circuit model. The recovery rate of OCV can be a critical indicator to quantify the electrolyte wetting process, verifying by batteries used separators with different wettability. This study provides a practical and effective tool to ensure the high-quality electrolyte infiltration process of LIBs.

1. Introduction

Due to the increasing demand for mobile electronics [1], electric vehicles [2,3], and large-scale energy storage stations [4], lithium-ion batteries (LIBs) with a large format and high density are in high demand [5], which necessitates significant increases in electrode size and compaction density. For example, blade batteries recently unveiled by BYD have an increased pole area (108 × 883 mm) of up to even 5 times that of conventional batteries [6]. The desired electrode porosity of high-energy LIBs is around 25% or even lower [7]. Thus, there are challenges to be overcome with regard to electrolyte infiltration and consequently high-throughput manufacturing [8,9].

Electrolyte infiltration, which occurs when the electrolyte wets the electrodes and separators after injection, is a time-consuming process during battery manufacturing [10–12] and plays an extremely important role in battery quality [13]. The electrode wetting state directly determines the in-plane reaction uniformity within an LIB and the consistency between different layers of cathode and anode [14], which further determines any potential decreases in energy utilization, cycling

life and power density [15]. Furthermore, homogeneous electrolyte immersion is necessary for uniform current distribution, and uneven current distribution will cause locally high current density and lithium plating [16,17], which not only accelerate the fading of active materials but also cause thermal failure concerns [18–20]. In battery production, different electrode wetting states can lead to cell-to-cell variations [21, 22]. Vacuum-pressure injection followed by long-term settling is generally used in the process of electrolyte filling for battery manufacturing, where a vacuum and pressure are simultaneously applied to the battery and electrolyte, respectively, to promote the electrolyte infiltration process [23]. With the continuous increase of electrode size and compaction density, homogeneous and efficient electrolyte filling is becoming challenging to ensure high performance of LIBs.

During the filling process, the liquid electrolyte first quickly (in seconds) enters the cavity of the cell and then slowly (in tens of hours) penetrates into the porous electrode and the separator [24,25]. As the injection process requires a critically dry and inert gas environment, the very time-consuming electrolyte infiltration process places a burden on

* Corresponding authors. *E-mail addresses:* syouzhi@126.com (Y. Song), wang-l@tsinghua.edu.cn (L. Wang), hexm@tsinghua.edu.cn (X. He).

https://doi.org/10.1016/j.nanoen.2022.107874

Received 21 August 2022; Received in revised form 30 September 2022; Accepted 4 October 2022 Available online 6 October 2022 2211-2855/© 2022 Elsevier Ltd. All rights reserved.







Fig. 1. Scheme of cell structure and OCV variation during electrolyte injection. (a) Illustration of a winding-type pouch battery. (b) Scheme of battery filling and OCV monitoring. (c) Profiles of the real-time OCV data during electrolyte filling.

the cost of manufacturing [26]. This process becomes severe for batteries with high energy density, whose electrodes are characterized by increased compaction density and large size. Moreover, the increased electrode size results in less redundant space inside the battery for electrolyte injection during a single filling operation. A larger and denser electrode necessitates a longer electrolyte infiltration path with higher diffusion resistance [27]. These factors unavoidably multiply the cost of the electrolyte injection process. In order to monitor the electrolyte wetting process, an in-plane imbibition method is proposed to probe the wetting speed and influence factors of electrodes and separators [28-30]. X-ray tomography [31], neutron radiography [23] and ultrasonic testing [32] have been used to monitor the electrolyte injection process of full batteries. X-ray testing is sensitive to the structural changes of electrode particles during cycling. Because of the sensitivity of neutrons to light elements, neutron-based characterization methods can be used to investigate electrolyte wetting [23], gas generation [33], and lithium-ion distribution [34] inside batteries. Nevertheless, the extremely high cost of equipment, such as synchrotron radiation sources. inconvenience and human bodily harm caused bv neutron-scattering methods limit the wide application of neutron tests for batteries. Due to its high sensitivity to gas, porosity, and electrode materials, ultrasound transmission is used to scan and test the internal status of the battery [35-38]. The characterization of electrolyte infiltration by ultrasonic testing has the characteristics of being nondestructive, inexpensive and harmless to humans and its detection efficiency is improving gradually [39].

Considering the importance of electrolyte wetting to battery performance, an efficient and easy online monitoring method needs to be further developed. The open circuit potential (OCV) of a battery, as an extremely important and easily monitored parameter, reflects important information about the battery [23,40]. The OCV is generated the moment the electrodes contact the electrolyte in a localized zone and is determined by the potential of active materials.

In this work, the electrolyte wetting behavior is probed by monitoring the OCV of a 1 Ah $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ (NCM622)/graphite (Gr) pouch cell. First, the OCV variation during the battery injection process is explored by an H-type electrolytic cell. Then, the OCV variation is correlated to the wetting states of different battery components with the help of equivalent circuit analysis and in situ ultrasonic imaging measurements. Third, separators with different wettability are employed to investigate their influence on the cell wettability rate. Finally, the mechanism of OCV generation and change during injection is valued as a reflection of the cell infiltration status, and an effective method for evaluating the cell infiltration state with electrolyte detection via OCV curves is proposed and confirmed.

2. Results and discussion

Fig. 1a presents an illustration of the winding-type pouch cell. The 1 Ah pouch cell consists of an NCM622 cathode and Gr anode. A commercial polyethylene (PE) microporous membrane is used as a separator. A summary of the cell design information is listed in Table S1. Digital images of packed and unpacked cells before filling are presented in Figure S1. A gas bag is set aside to collect and emit the emissions generated during cell formation. To activate the cell, 3 mL of liquid electrolyte (1 M LiPF₆ in a mixture of carbonate solvents) is injected into the battery through the air bag. Moreover, a high-precision digital recorder (1 ms resolution) is employed to capture the OCV profiles during filling (Figure S2). As presented in Fig. 1b, the liquid electrolyte flows from the two open sides into the separator and electrodes at the beginning of filling. Fig. 1c shows the profile of the real-time OCV data during electrolyte filling. Surprisingly, the OCV drops from zero to approximately - 0.80 V at the moment the electrolyte fills the cell. After settling at - 0.80 V for approximately 200 ms, the OCV surges to



Fig. 2. Investigating the mechanism of OCV variation during filling. (a) Scheme of the H-type electrolytic model cell. (b) OCV profiles of the Al and Cu current collectors when wetted by a liquid electrolyte. (c) Voltage profiles between different components of the cell. (d) Simulation circuit of the cell during filling. (e) Schematic representation of electrolyte infiltration at different stages after injection.

approximately - 0.20 V and then rises slowly. After 72 h, the OCV is stable at approximately 0.10 V, implying that the infiltration process is mainly completed. The above voltage variation can be divided into four periods. Before electrolyte injection, the OCV remains at zero (Stage I). Then, the OCV drops to approximately - 0.80 V once the electrolyte fills the cell (Stage II). Third, the OCV begins to gradually increase (Stage III) and approaches a final state with a stable value of approximately 0.10 V.

Subsequently, an H-type electrolytic cell is employed to thoroughly examine the voltage change mechanism during the injection process. Fig. 2a and Figure S3 present the structure of the H-type electrolytic cell. Each electrode has an affective area of 1 cm². The two electrodes are connected to each other by adding approximately 30 mL of liquid electrolyte. Fig. 2b presents the OCV profiles for the aluminum (Al)/ lithium metal (Li) and copper (Cu)/Li cells during filling, where the current collector (Al or Cu) foil and Li are used as the working electrode and reference electrode, respectively. For Al/Li, the OCV increases immediately to approximately 2.30 V as the aluminum foil makes contact with the electrolyte and is stabilized for approximately 300 ms. Then, the OCV drops to approximately 2.09 V and stably remains there. In the case of the Cu/Li cell, the OCV increases immediately to approximately 3.10 V as the Cu foil makes contact with the electrolyte and is stabilized for approximately 300 ms. Then, the voltage drops to a final value of 2.16 V within 500 ms. Furthermore, the difference between Al/Li and Cu/Li cells (marked as Δ (Al/Cu)) is calculated and presented in Fig. 2b. Remarkably, the OCV value of Δ (Al/Cu) decreases

to approximately - 0.80 V, which is exactly the same as the minimum value of the 1 Ah pouch cell within 300 ms. Then, the OCV value of Δ (Al/Cu) returns to approximately - 0.10 V.

These results indicate that the initial quick decrease in the OCV reflects the electrolyte wetting process of the Al and Cu current collectors. To further confirm this, the electrodes of the 1 Ah NCM622/Gr pouch cell are replaced by bare Al and Cu foils (denoted as the Al/Cu pouch cell). As expected, the Al/Cu pouch cell exhibits similar OCV profiles during filling with a minimum value of -0.80 V and a final value of -0.10 V (Figure S4). This result indicates that the OCV behavior at the beginning of injection is a result of the potential difference between the Al and Cu current collectors. Fig. 2c displays the OCV profiles of different electrode couples measured by the H-type cell. The OCVs of the pristine NCM622 cathode and Gr anode vs. Li electrode are 3.16 V and 3.04 V, respectively, with Δ (Ca(NCM622)/An(Gr)) values of 0.12 V. Therefore, the final OCV (approximately 0.10 V) of the 1 Ah pouch cell when fully soaked is a result of the potential difference between the NCM622 cathode and Gr anode.

Considering that the infiltration of the electrolyte into the main components of the cell, such as the cathode, anode and Al/Cu current collectors, is a time-consuming process, the OCV varies at different wetting states. Therefore, the circuit inside the cell is further investigated during different stages of the injection process. Fig. 2d shows a proposed simulation circuit. The area of the exposed current collector part is denoted as S_1 , the internal resistance is recorded as R_1 , and the



Fig. 3. Characterization of different components in the cell. (a) SEM images of the Gr anode, NCM622 cathode, PE and Al_2O_3 @PE separators. (b, c) Time dependence of electrolyte contact angles of the electrodes and separators. (d) Electrolyte uptake of the electrodes and separators.

electric potential of the formed circuit is denoted as E_1 . Similarly, the area of the electrode that has been immersed is denoted as S_2 , the electric potential of the formed circuit is denoted as E_2 , and the internal resistance is recorded as R_2 . Based on the internal structure of the cell, E_1 and E_2 are connected in parallel.

Therefore, the OCV can be expressed as:

$$OCV = \frac{E_1 R_2 + E_2 R_1}{R_1 + R_2}$$
(1)

The simplification is as follows:

$$OCV = E_2 - \frac{E_2 - E_1}{R_1 / R_2 + 1}$$
(2)

The variation profiles of the OCV are divided into four stages based on the electrolyte infiltration processes (Fig. 2e). Before injection, the battery is in the state of disconnection, and the OCV remains at zero in Stage I. The moment the electrolyte is injected in Stage II, the electrolyte rapidly fills the voids in the cell, and then the exposed current collectors near the tabs are immersed in the electrolyte and generate E_1 . This process is so quick that S_1 and R_1 appear unchanged over time. When the electrolyte gradually infiltrates into the separator and electrodes (Stage III), a voltage between the electrodes (E_2) is generated. As the infiltration process proceeds, the wetted areas of the electrodes ($S_2 = S_2(t)$) keep increasing with time. Because the internal resistance of the cell is inversely proportional to the wetted area of the electrodes when all other conditions are equal, the following equation can be obtained:

$$R_2 \propto 1/S_2 \tag{3}$$

Thus,

$$\frac{R_1}{R_2} \propto S_2(t) \tag{4}$$

Based on the above analysis, it can be seen that the OCV rises with time and eventually converges to E_2 . When the cell is fully immersed, the cell voltage is almost equal to the potential difference between the cathode and anode because the electrode area is much larger than the exposed collector area. As presented in Figs. 2b and 2c, the OCV drops from zero (Stage I) to approximately - 0.80 V once the electrolyte fills the cell (Stage II) and then begins to gradually increase and approaches a stable value of 0.10 V (Stage III). Clearly, the OCV variation obtained from the proposed simulation circuit matches perfectly with the experimental results.

To further understand the immersion behavior, the surface chemistry of the current collectors, electrode materials, and separators are carefully analyzed. Scanning electron microscopy (SEM) was first applied to observe the surface morphologies of the materials. As presented in Figure S5, the calendared Al foil exhibits a smooth surface, while the electrolytic Cu foil shows a relatively rough morphology. Fig. 3a displays the surface morphologies of the electrodes and separators. The Gr anode is composed of layered particles with a particle size of 5–20 μ m and a calendaring density of 1.65 g cm⁻³. The NCM622 cathode is made up of single crystal particles with a diameter of 2–6 μ m and a calendaring density of 3.4 g cm⁻³. The bare PE separator exhibits a porous surface with a pore size of approximately tens of nanometers. In addition, an alumina (Al₂O₃) ceramic-coated separator (Al₂O₃ @PE) with



Fig. 4. OCV profile and wettability of the 1 Ah pouch cell using different separators. (a, b) OCV profiles of the cell using PE and Al₂O₃ @PE separators. (c) Ultrasonic transmission images of the cells after electrolyte injection. (d) OCV profiles of the Al₂O₃ @PE Cell with different amounts of electrolyte injection.

improved wettability is employed for further comparative experiments. Further details about the commercial separators are shown in supporting information and Table S3. Moreover, dynamic electrolyte contact angles (CAs) for the current collectors, electrodes and separators are measured to investigate their wettability (Figure S5b, 5c and Figs. 3b, 3c). The CAs for the dense Al and Cu foils are 42.4° and 55.6°, respectively, at the beginning of the test (Figure S5b). Little change is observed as the time increases (Figure S5c). However, the CA of the porous Gr anode decreases dramatically from 20.5° to only 5.1° within 10 s. Similarly, the positive electrode also exhibits good electrolyte wettability. Nevertheless, the CA of the porous PE separator remains at approximately 36.9°, indicating its poor electrolyte compatibility. For the Al₂O₃@PE separator, the CA drops from 20.2° to 10.3° during the test, demonstrating its superior wettability. In addition, attenuated total reflection Fourier transform infrared spectroscopy (ATR-FTIR) and energy dispersive spectroscopy (EDS) are utilized to study the surface chemistry of the separator. The enhanced wettability of the Al₂O₃ @PE sample is attributable to the polar functional group-rich coating layer (Figure S6,

Figure S7). The electrolyte uptake (EU) of the electrodes and separators is measured to characterize their adsorption capacity for the electrolyte. As presented in Fig. 3d, the EU values are 19.2 wt%, 31.8 wt%, 78.4 wt % and 110.9 wt% for the NCM622 cathode, Gr anode, PE and Al_2O_3 @PE separators, respectively. The results indicate that the cathode and anode used here have better wettability than the PE separator, and the Al_2O_3 @PE separator with improved wettability has great potential for accelerating the cell wetting process.

The electrolyte infiltration processes of the cells assembled with different separators were visualized through in situ ultrasonic scanning (Figure S8) and OCV monitoring. The OCV profiles of the cell using the PE and Al₂O₃ @PE separators are presented in Figs. 4a, 4b. At the beginning of the test, their OCVs both drop from zero to approximately – 0.80 V. As time passes, the OCV of the PE Cell increases obviously later and slower than that of the Al₂O₃ @PE Cell. Fig. 4c shows the corresponding in situ ultrasonic scanning images. The blue color represents the noninfiltrated area, and the red color indicates the well-saturated area.

As the time increased, the ultrasonic imaging showed that the blue area decreased and finally disappeared (Figure S9). Moreover, the OCV values of the PE and Al₂O₃ @PE Cell rise from negative to 0.10 V (Table S2), but the voltage of the PE Cell arises obviously later and slower than that of the Al₂O₃ @PE Cell. The consistency of PE and Al₂O₃ @PE Cell are confirmed by parallel tests. The results are shown as OCV curves with error area (Figure S10). However, by comparing their OCV values and ultrasonic imaging results, it can be seen that the Al₂O₃ @PE Cell has a superior saturation speed than the PE Cell. As time increases, the blue area decreases for both cells. After 36 h, the Al₂O₃ @PE Cell is completely wetted, while the PE Cell still has 41.2% of its area in a semi soaked state. Moreover, the OCV values of the PE and Al₂O₃ @PE Cells increase to 0.04 V and 0.10 V, respectively. Based on the equivalent circuit analysis in Eq. 2 and experimental above, the relationship of OCV and percentage of soaked area (marked as *x*) can be expressed as:

$$OCV = 0.1 - \frac{0.2}{\left(\frac{1.7x}{1-x}\right) + 1}$$
(5)

Figure S11 shows the comparison of the simulation and test data. Clearly, the OCV variation obtained from the proposed simulation circuit matches well with the experimental results. Video S1 provides a visual comparison between the two cells within 24 h, which indicates that the soaking rate of the cell after filling can be significantly accelerated by improving the wettability of the separator. The impedance of PE and Al₂O₃ @PE Cell are tested at different time during wetting process (Figure S12). As time increasing, the impedance of both PE and Al₂O₃ @PE Cell decrease, which indicates the improvement of wetting process. However, the Al₂O₃ @PE Cells has lower impedance than PE Cell due to the better wettability. Figure S13 shows the cycling performance of the 1 Ah NCM622/Gr cells using different separators. The cells are cycled at 1 C after being filled with electrolyte for 12 h and the formation process at 0.1 C for one cycle. The OCV curves during filling process indicates that Al2O3 @PE Cell is nearly completely wetted compared with PE Cell. The discharge capacities are 904 mAh and 952 mAh for the cells using the PE and Al₂O₃ @PE separators, respectively. As the cycle number increases, the reversible capacity of the PE cell simultaneously increases. The Al₂O₃ @PE cell exhibits superior cycling stability. The result indicates that the performance of the PE cell is not immediately and fully maximized due to the lack of sufficient wetting. Fig. 4d shows the influence of the amount of electrolyte on the OCV of the Al₂O₃ @PE Cell. When the electrolyte volume is less than 3 mL, the OCV rises slower and cannot reach 0.10 V because the electrodes cannot be totally wetted. When the injected electrolyte is more than 3 mL, the difference between their OCV curves decreases, and all of their OCV values can reach 0.10 V because the cells are fully wetted. These results further justify that the OCV effectively reflects the status and quality of battery saturation.

Supplementary material related to this article can be found online at doi:10.1016/j.nanoen.2022.107874.

3. Conclusion

In summary, we propose the operando monitoring of the OCV for a 1 Ah NCM622/Gr pouch cell during electrolyte filling. It is observed that the OCV drops dramatically to -0.80 V at the beginning of filling and then changes characteristically with the wetting process. In addition, the wetting rate is significantly boosted by improving the wettability of the separators, and an equivalent circuit is proposed to enhance our understanding of the mechanism of this wetting process.

CRediT authorship contribution statement

Hao Cui: Conceptualization, Investigation, Methodology, Formal analysis, Validation, Data curation, Writing - original draft. **Dongsheng Ren:** Investigation, Methodology, Formal analysis. **Mengchao Yi**: Formal analysis. Sixuan Hou: Formal analysis. Kai Yang: Formal analysis. Hongmei Liang: Formal analysis. Xuning Feng: Methodology. Xuebing Han: Methodology. Youzhi Song: Conceptualization, Validation, Writing - review & editing. Li Wang: Conceptualization, Methodology, Investigation, Writing-review & editing. Xiangming He: Investigation, Formal analysis, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

We are grateful to the National Natural Science Foundation of China (No. U21A20170 (X. He), 52007099 (D. Ren), 52206263 (Y. Song)), the Ministry of Science and Technology of China (No. 2021YFB2501900 (D. Ren), No. 2019YFE0100200 (X. He) and 2019YFA0705703 (L. Wang)).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.nanoen.2022.107874.

References

- [1] Van Noorden, R., A BETTER BATTERY. Nature 2014, 507 (7490), 26-28.
- [2] G. Harper, R. Sommerville, E. Kendrick, L. Driscoll, P. Slater, R. Stolkin, A. Walton, P. Christensen, O. Heidrich, S. Lambert, A. Abbott, K.S. Ryder, L. Gaines, P. Anderson, Recycling lithium-ion batteries from electric vehicles, Nature 575 (7781) (2019) 75–86.
- [3] N. Nitta, F. Wu, J.T. Lee, G. Yushin, Li-ion battery materials: present and future, Materials Today 18 (5) (2015) 252–264.
- [4] K. Liu, Y.Y. Liu, D.C. Lin, A. Pei, Y. Cui, Materials for lithium-ion battery safety, Sci. Adv. 4 (6) (2018) eaas9820.
- [5] E. Pomerantseva, F. Bonaccorso, X.L. Feng, Y. Cui, Y. Gogotsi, Energy storage: the future enabled by nanomaterials, Science 366 (6468) (2019) eaan8285.
- [6] X.-G. Yang, T. Liu, C.-Y. Wang, Thermally modulated lithium iron phosphate batteries for mass-market electric vehicles, Nat. Energy 6 (2) (2021) 176–185.
- [7] J.M. Tarascon, M. Armand, Issues and challenges facing rechargeable lithium batteries, Nature 414 (6861) (2001), 359-67.
- [8] J.B. Goodenough, Y. Kim, Challenges for rechargeable Li batteries, Chem. Mat. 22 (3) (2010) 587–603.
 [9] D.L. Wood, J. Li, C. Daniel, Prospects for reducing the processing cost of lithium ion
- [9] D.L. Wood, S. Li, C. Danier, Property for Fedding the processing cost of infinition for batteries, J. Power Sources 275 (2015) 234–242.
 [10] Handbook of Battery Materials, 2nd Edition. Wiley-V C H Verlag Gmbh: Weinheim,
- 2011; p 1–989.
- [11] Y.-Z. Song, Y. Zhang, J.-J. Yuan, C.-E. Lin, X. Yin, C.-C. Sun, B. Zhu, L.-P. Zhu, Fast assemble of polyphenol derived coatings on polypropylene separator for high performance lithium-ion batteries, J. Electroanal. Chem. 808 (2018) 252–258.
- [12] Y.-Z. Song, J.-J. Yuan, X. Yin, Y. Zhang, C.-E. Lin, C.-c. Sun, L.-F. Fang, B. Zhu, L.-P. Zhu, Effect of polyphenol-polyamine treated polyethylene separator on the ionic conduction and interface properties for lithium-metal anode batteries, J. Electroanal. Chem. 816 (2018) 68–74.
- [13] D.L. Wood, J.L. Li, C. Daniel, Prospects for reducing the processing cost of lithium ion batteries, J. Power Sources 275 (2015) 234–242.
- [14] S.J. An, J. Li, C. Daniel, H.M. Meyer, S.E. Trask, B.J. Polzin, D.L. Wood, Electrolyte volume effects on electrochemical performance and solid electrolyte interphase in Si-graphite/NMC lithium-ion pouch cells, ACS Appl. Mater. Interfaces 9 (22) (2017) 18799–18808.
- [15] D.H. Jeon, Wettability in electrodes and its impact on the performance of lithiumion batteries, Energy Storage Mater. 18 (2019) 139–147.
- [16] Y. Xie, H.L. Zou, H.F. Xiang, R. Xia, D.D. Liang, P.C. Shi, S. Dai, H.H. Wang, Enhancement on the wettability of lithium battery separator toward nonaqueous electrolytes, J. Membr. Sci. 503 (2016) 25–30.
- [17] M.S. Wu, T.L. Liao, Y.Y. Wang, C.C. Wan, Assessment of the wettability of porous electrodes for lithium-ion batteries, J. Appl. Electrochem. 34 (8) (2004) 797–805.
- [18] X.N. Feng, S.Q. Zheng, D.S. Ren, X.M. He, L. Wang, H. Cui, X. Liu, C.Y. Jin, F. S. Zhang, C.S. Xu, H.J. Hsu, S. Gao, T.Y. Chen, Y.L. Li, T.Z. Wang, H. Wang, M.G. Li,

H. Cui et al.

M.G. Ouyang, Investigating the thermal runaway mechanisms of lithium-ion batteries based on thermal analysis database, Appl. Energy 246 (2019) 53–64.

- [19] G.F. Hu, P.F. Huang, Z.H. Bai, Q.S. Wang, K.X. Qi, Comprehensively analysis the failure evolution and safety evaluation of automotive lithium ion battery, Etransportation (2021) 10.
- [20] Y. Song, L. Wang, H. Cui, H. Liang, Q. Hu, D. Ren, Y. Yang, H. Zhang, H. Xu, X. He, Boosting battery safety by mitigating thermal-induced crosstalk with a Bicontinuous separator, Adv. Energy Mater. (2022) 2201964, https://doi.org/ 10.1002/aenm.202201964. In press.
- [21] M. Schindler, J. Sturm, S. Ludwig, J. Schmitt, A. Jossen, Evolution of initial cell-tocell variations during a three-year production cycle, Etransportation 8 (2021) 100102.
- [22] L. Wildfeuer, M. Lienkamp, Quantifiability of inherent cell-to-cell variations of commercial lithium-ion batteries, Etransportation 9 (2021) 100129.
- [23] W.J. Weydanz, H. Reisenweber, A. Gottschalk, M. Schulz, T. Knoche, G. Reinhart, M. Masuch, J. Franke, R. Gilles, Visualization of electrolyte filling process and influence of vacuum during filling for hard case prismatic lithium ion cells by neutron imaging to optimize the production process, J. Power Sources 380 (2018) 126–134.
- [24] M. Armand, J.M. Tarascon, Building better batteries, Nature 451 (2008) 652–657, 652-7.
- [25] A.S. Arico, P. Bruce, B. Scrosati, J.M. Tarascon, W. Van Schalkwijk, Nanostructured materials for advanced energy conversion and storage devices, Nat. Mater. 4 (5) (2005) 366–377.
- [26] S.G. Lee, D.H. Jeon, Effect of electrode compression on the wettability of lithiumion batteries, J. Power Sources 265 (2014) 363–369.
- [27] B. Dunn, H. Kamath, J.-M. Tarascon, Electrical energy storage for the grid: a battery of choices, Science 334 (6058) (2011) 928–935.
- [28] A. Davoodabadi, J. Li, H. Zhou, D.L. Wood, T.J. Singler, C. Jin, Effect of calendering and temperature on electrolyte wetting in lithium-ion battery electrodes, J. Energy Storage 26 (2019) 101034.
- [29] A. Davoodabadi, J. Li, Y. Liang, D.L. Wood III, T.J. Singler, C. Jin, Analysis of electrolyte imbibition through lithium-ion battery electrodes, J. Power Sources 424 (2019) 193–203.
- [30] A. Davoodabadi, C. Jin, D.L. Wood, T.J. Singler, J. Li, On electrolyte wetting through lithium-ion battery separators, Extrem. Mech. Lett. 40 (2020) 100960.
- [31] Y.Z. Song, X. Liu, D.S. Ren, H.M. Liang, L. Wang, Q. Hu, H. Cui, H. Xu, J.L. Wang, C. Zhao, X.B. Zuo, G.L. Xu, H.L. Amine, X.M. He, Simultaneously blocking chemical crosstalk and internal short circuit via gel-stretching derived nanoporous nonshrinkage separator for safe lithium-ion batteries, Adv. Mater. 34 (2) (2022) 2106335.
- [32] M.C. Yi, F.C. Jiang, L.G. Lu, S.X. Hou, J.Q. Ren, X.B. Han, L.L. Huang, Ultrasonic tomography study of metal defect detection in lithium-ion battery, Front. Energy Res. 9 (2021) 806929.
- [33] B. Michalak, H. Sommer, D. Mannes, A. Kaestner, T. Brezesinski, J. Janek, Gas evolution in operating lithium-ion batteries studied in situ by neutron imaging, Sci. Rep. 5 (2015) 15627.
- [34] J.B. Siegel, X.F. Lin, A.G. Stefanopoulou, D.S. Hussey, D.L. Jacobson, D. Gorsich, Neutron imaging of lithium concentration in LFP pouch cell battery, J. Electrochem. Soc. 158 (5) (2011) A523–A529.
- [35] A.G. Hsieh, S. Bhadra, B.J. Hertzberg, P.J. Gjeltema, A. Goy, J.W. Fleischer, D. A. Steingart, Electrochemical-acoustic time of flight: in operando correlation of physical dynamics with battery charge and health, Energy Environ. Sci. 8 (5) (2015) 1569–1577.
- [36] Ladpli, P.; Kopsaftopoulos, F.; Nardari, R.; Chang, F.K. In Battery charge and health state monitoring via ultrasonic guided-wave-based methods using built-in piezoelectric transducers, Conference on Smart Materials and Nondestructive Evaluation for Energy Systems, Portland, OR, Mar 27–28; Spie-Int Soc Optical Engineering: Portland, OR, 2017.
- [37] L. Gold, T. Bach, W. Virsik, A. Schmitt, J. Muller, T.E.M. Staab, G. Sextl, Probing lithium-ion batteries' state-of-charge using ultrasonic transmission - Concept and laboratory testing, J. Power Sources 343 (2017) 536–544.
- [38] R.D. Schmidt, J. Sakamoto, In-situ, non-destructive acoustic characterization of solid state electrolyte cells, J. Power Sources 324 (2016) 126–133.
- [39] Z. Deng, Z.Y. Huang, Y. Shen, Y.H. Huang, H. Ding, A. Luscombe, M. Johnson, J. E. Harlow, R. Gauthier, J.R. Dahn, Ultrasonic scanning to observe wetting and "unwetting" in Li-ion pouch, Cells Joule 4 (9) (2020) 2017–2029.
- [40] H. Wang, Z. Tao, Q. Ma, Y. Fu, H. Bai, Y. Zhu, H. Xiao, H. Bai, Impact of initial open-circuited potential on the consistency of lithium ion battery, IOP Conf. Ser.: Earth Environ. Sci. 153 (2) (2018), 022023.



Hao Cui is currently a candidate for Ph.D. of chemical engineering and technology at Institute of Nuclear and New Energy Technology (INET), Tsinghua University. He has got a bachelor's degree of polymer science and engineering at Department of Chemical Engineering in Tsinghua University in 2018. His research focuses on electrochemical mechanism, separators and electrolytes of lithium-ion batteries.

Dr. Dongsheng Ren is now a postdoctoral researcher in

Institute of Nuclear and New Energy Technology (INET),

Tsinghua University. He received his B. E. (2014) and Ph. D.

(2020) degree in School of Vehicle and Mobility, Tsinghua

University. His research focuses on electrochemical/thermal

modeling of lithium-ion battery.



Dr. Mengchao Yi is the senior engineer in department of solid state battery, R&D, Beijing WeLion New Energy Technology Co., LTD, focusing on battery failure analysis and advanced characterization techniques. Proposed the multi-probe ultrasonic detection technology and battery failure quantification method based on ultrasonic signal.



Sixuan Hou is studying for a master's degree at University of Shanghai for Science and Technology at present. His research direction is ultrasonic nondestructive testing of lithium-ion battery.



Kai Yang received his Bachelor degree in Chemical Engineering and Technology from Anhui Polytechnic University (China) in 2019. He is currently pursuing his MS degree under the supervision of Prof. Hong Xu at Tsinghua University (China). His research interests focus on exploiting Metal-Organic Frameworks separator to tailor Li-ion solvation and accelerate Li-ion transportation for high-energy density and long-life Li-ion batteries and Li-metal batteries.

Nano Energy 104 (2022) 107874

H. Cui et al.

Nano Energy 104 (2022) 107874



Hongmei Liang received her bachelor's and master's degrees from Central South University in 2015 and 2018, respectively. She is currently a PhD candidate with Prof. Xiangming He at the Institute of Nuclear and New Energy Technology, Tsinghua University, China. Her research mainly focuses on lithium metal anode and liquid electrolyte for lithium-ion batteries.



Dr. Youzhi Song received his Ph.D. degree under Prof. Baoku Zhu from the Department of Polymer Science and Engineering at Zhejiang University in 2018. Currently, he is a postdoctoral in Prof. Xiangming He's group in the Institute of Nuclear and New Energy Technology, Tsinghua University. His current research interests are focused on the rational design of polymeric membrane separators for safe and endurable lithiumbased batteries.



Dr. Xuning Feng received the B.E. Degree and the Ph.D. degrees in the Department of Automotive Engineering from Tsinghua University, Beijing, China, in 2011 and 2017, respectively. He is currently an associate Professor of School of Vehicle and Mobility of Tsinghua University. His research interests include battery management system and battery safety.



Dr. Li Wang is an associate professor at the Institute of Nuclear and New Energy Technology, Tsinghua University, China. She received his B.S. in 1999 and Ph.D. in Analytical Chemistry in 2004 from the Tsinghua University. Her research expertise includes new electrode material preparation technology, failure mechanism of lithium-ion battery, safety power-battery technology and fundamental understanding of related electrochemical processes.



Dr. Xuebing Han is an assistant professor of the state key laboratory of automotive safety and energy in School of Vehicle and Mobility from Tsinghua University. His research is focused on the lithium ion batteries, aiming to the smart battery design, intelligent battery management and the advanced energy system.



Dr. Xiangming He is a professor and the group leader of the Lithium-ion battery Laboratory in the Institute of Nuclear and New Energy Technology, Tsinghua University. He received his bachelor's and master's degrees from the School of Chemical Engineering and Ph.D. from Institute of Nuclear and New Energy Technology, Tsinghua University. His research focuses on the design and application of functional materials for energy storage and conversion and fundamental understanding of related electrochemical processes.