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REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

Kim et al. focused on the kosmotropic effect of anions and investigated how it affects the electrode structure and battery performance of NMC cathodes made via the coating of water-based slurries containing such anions. DFT, MD simulation, FTIR, and NMR were employed to discuss the difference in the bulk and interfacial solvation structures of anions with different kosmotropivities. The battery tests clarified the comparable cycle performance of the water-processed NMC811 containing kosmotropic anions with that made from the conventional NMP-based slurry under a high active-material loading of ca. 20 mg/cm² (i.e., the capacity retention of 81% vs. 82% after 400 cycles at 0.2C charging/0.5C discharging). In addition, the authors estimated how the transition from NMP-based slurries to water-based slurries with a high kosmotropic effect benefits cathode manufacturing, which expects a 46% reduction in energy consumption of the cathode production.

Overall, the results look great; ample computational and experimental results using various techniques will definitely be informative for researchers in the battery field to mitigate the reactivity of water with cathode active materials to enable stable cathode manufacturing using water as a slurry-processing solvent.

However, at this stage, before considering publication, the authors need to consider several questions and possibly additional experiments as provided below in order to further convince the readers of the benefits of the transition to cathode manufacturing using water-based slurries in terms of electrode properties and battery performance. Some of the comparisons currently provided do not give a completely fair picture.

The concerns are shown in detail as follows:

[a. Major Concerns]

(a-1) Do the results of battery tests really represent the positive effect of using the anions with a high kosmotropic effect?

The main issue of this manuscript is that the battery evaluation involves (at least) four different factors: (i) electrode structure (e.g., particle size distribution of NMC811), (ii) amount of leached Li⁺ ions from NMC811 (which affect the achievable capacity of the active material), (iii) state of Al current collector (whose corrosion would affect the overvoltage), and (iv) Li salt remained in the electrode layer (which would subsequently affect the CEI formation), and the evaluated cathodes are different in these aspects. This blurs the consequence of using the anions with a high kosmotropic effect, requiring some control experiments so that the results of battery tests truly represent the positive effect of using the anions with a high kosmotropic effect.

Firstly, I recommend performing a straightforward comparison between water-processed NMC811 and NMP-processed NMC811 by removing salts (used to mitigate the interfacial reaction of the active material in a water slurry) from the water processed electrodes before assembling the cells and undertaking the battery tests. This excludes the uninvestigated effect of (iv) and allows a fair comparison

between the consequences of using the anions with different kosmotropivities in a water-based slurry and that of an NMP-based slurry.

Secondly, making NMC811 cathodes using 1.0 M LiTFSI and 0.5 M Li₂SO₄ in NMP-based slurries will provide valuable insights into the structural modification of NMC811 particles by these salts. This is because, from Supplementary Figs. 10 and 19, readers might think that these salts themselves also have an impact on the breakage of the secondary particles of NMC811. (In contrast, the use of water without these salts provided relatively intact secondary particles.) Doing the cathode preparation in NMP using these salts will relieve this concern and enable the authors to say the breakage of the secondary particles is purely due to the interfacial reaction of water on the cathode surface and it subsequently affects the battery performance.

Thirdly, to state the effect of the studied anions on the CEI formation and its influence on the battery performance, the authors are recommended to perform battery tests using the NMC811 cathode (made via an NMP-based slurry) in the electrolyte solutions containing the Li salts of these anions additionally. This keeps (i)–(iii) in the same condition and allows the authors to discuss how these anions affect the battery performance (as well as the CEI composition).

(a–2) How about the rate and long-term cycle performances of NMC811 made via a water-based slurry with anions with a high kosmotropic effect?

Although the authors have tried to explain the significance of using the anions with a high kosmotropic effect in a water-based NMC811 by presenting the achieved capacity per the total cost of the cathode ingredients (Fig. 5c), the obvious difference in the battery performance between water-processed NMC811 and NMP-processed NMC811 has yet to be clarified. This should be clarified so that the authors can state the transition to the water-based slurry for cathode manufacturing is truly beneficial in addition to the estimated advantages of cost reduction and environmental friendliness.

Additional experiments that the authors could consider are:

Firstly, comparison of the charge/discharge rate performances between water-processed NMC811 and NMP-processed NMC811.

Secondly, the cycle test (Fig 5a) should be prolonged as much as possible (ideally, >1,000 cycles). This is because such a long-term cycle life has been expected for commercial electrodes (e.g., <https://www.energy.gov/eere/vehicles/articles/reducing-reliance-cobalt-lithium-ion-batteries>) and, in this respect, it is worthwhile to show how long water-processed NMC811 can keep up with NMP-processed NMC811. Furthermore, readers do not understand how their cells will fully degrade by only seeing Fig. 5a as both cathodes showed similar capacity decay up to 400 cycles. This necessitates a prolonged cycle test beyond 80% of the initial capacity.

The authors should address the aforementioned concerns and revise the manuscript to tell a clear story about the significance of their results in using the anions with a high kosmotropic effect for the cathode manufacturing of lithium-ion batteries.

In addition to the above considerations, the following point-by-point suggestions should be addressed in order to further improve the manuscript:

[b. Major comments]

(b-1) Line 85: Here, “material” should be changed to “CO₂ emission rate”. Revise the sentence accordingly.

(b-2) Lines 194–196: How was this surface observation of the Al current collectors performed? In particular, no information is available about how the dried electrode layers were removed to see the Al surface attached to the electrode slurries. Please provide the detailed experimental procedure in the manuscript or the Supplementary Information.

(b-3) Lines 200–204: To support the authors’ speculation, I recommend evaluating the particle size distribution of these NMC811 particles. This clarifies the breakage of secondary particles as a reduction in the particle size. For now, immersion of NMC811 in water appears to show almost no change in the particle size (Supplementary Fig. 10b vs. 10a).

(b-4) Lines 206–208: From Supplementary Fig. 11, I could not see the obvious difference in the I₀₀₃/I₁₀₄ ratio between the samples labelled “Kosmotropic”, “Chaotropic”, and “H₂O”, which contradicts Fig. 4c. Please double-check it.

(b-5) Line 219: From the caption of Supplementary Table 5, the NMC811 loading is 18.0 mg/cm². Which is true: 20 or 18.0 mg/cm²?

(b-6) Line 219: In relation to the above comment, I recommend specifying the NMC811 loading and the electrode density of all tested cathodes in the Supplementary Information. As a difference in both variables affects the battery performance, the authors need to show these values for all tested cathodes so that the observed difference in the battery performance is not due to the difference in NMC811 loading and/or electrode density.

(b-7) Lines 238–239: This is hardly believable based on the provided information (Supplementary Fig. 15). How about spectra in the N1s, P2p, and S2p regions?

In addition, how the cycled NMC811 was prepared for XPS (i.e., what was the washing process of the surface)?

I think it is too early to say there is no effect of a CEI on the battery performance.

(b-8) Lines 243–244: What are the possible reasons for a slightly poorer cycle performance of the higher Li₂SO₄-concentration samples than that prepared using 0.1 M Li₂SO₄?

(b-9) Lines 265–267 (and 275–276): I cannot fully agree with this sentence because this would also

relate to the reduced particle size of NMC811 as speculated in Supplementary Fig. 10c. In addition, reduced particle sizes will increase the tortuosity of ion conduction, hence increasing R_{ion} . As stated in (b-3), the measurement of the particle size distribution of NMC811 after immersing the aqueous solutions with different anions is necessary.

(b-10) Lines 276–277 and 376: What is the theory behind the E/C ratio of 2 or 3 g Ah⁻¹? Does this ratio always allow the electrolyte solution to fill all the pores in the electrode layers and separator?

The amount of the electrolyte solution should be decided based on the total pore volume of the porous cell components. Can you please calculate the pore volumes of the NMC811 cathode and separator to see if the volume of the electrolyte solution surpasses the total pore volume of the porous cell components?

(b-11) Lines 277–279: At least for Supplementary Fig. 20b, I do not think R_{ion} is stable from 0 to 12 h, which might disagree with this sentence. Please plot R_{ion} against the aging time to discuss this precisely.

(b-12) Fig. 5d: Are the charge/discharge curves in Fig. 5d measured at the first cycle? Please specify it.

(b-13) Fig. 5d: Gradual increase in the specific capacity with an increase in the areal capacity is surprising. How was it possible to realize? Were the experimental conditions for different areal capacities (e.g., electrode formulation, electrode density, applied currents during charging and discharging) the same as each other?

(b-14) Fig. 5f and Line 293: What was the AM composition (%) in This work precisely? I thought it was 95.5% from Line 361 (and probably from the caption of Supplementary Table 5), but based on this figure, it looks like 92% and Supplementary Table 6 says another value: 93.1%. Please use the same value consistently.

(b-15) Line 359 and thereafter: Please specify how the aqueous solutions containing different Li salts were used for slurry preparation. At this stage, readers cannot understand when and how the Li salts were added.

(b-16) Supplementary Table 3: Please specify how the cost information about each material was obtained. Any reference?

(b-17) Supplementary Table 4: Please specify how the cost information about each electrode component was obtained. Any reference?

(b-18) Supplementary Table 4: In addition, I think the Al current collector should be included here as well as in the cost estimation.

(b-19) Supplementary Table 4b: Why does the cost information about the solution differ from that in Supplementary Table 3b? (For instance, the cost of NMP in Supplementary Table 4b is 10.74 times higher than that in Supplementary Table 3b, but the cost of 1.0 m LiTFSI in H₂O in Supplementary Table 4b is

12.87 times higher than that in Supplementary Table 3b, i.e., no proportional increase)

[c. Minor comments]

(c-1) Lines 32 and 87: Please revise “approximately 45%” to “46%” to be exact.

(c-2) Line 73: Please add the reference(s) to describe some examples of using the kosmotropic effect in biochemistry.

(c-3) Line 76: Please add “, right” after “Fig. 1a”.

(c-4) Line 100: I guess “charge-delocalized” needs to be “charge-localized”. Please double-check it.

(c-5) Fig. 2a: For TFSI⁻, the ESP value for O atoms would need to be provided as these are the sites for hydrogen bonding. (The other three anions have provided the ESP values for O atoms, but TFSI⁻ has not provided it.)

(c-6) Line 219: Here, “tap densities” should be “electrode density” and “areal-mass-loading” can be “areal mass loading” (or “areal NMC811 loading”).

(c-7) Line 231: Please specify at which cycle number the specific capacity of 205 mAh/g was observed. (I presume it was observed at the first cycle.)

(c-8) Fig. 6a and Supplementary Fig. 22: I think “Blower 1” and “Blower 2” should be swapped in one of the figures to match the numbering.

(c-9) Lines 378–379: Was EIS conducted from the lowest frequency to the highest frequency? Please specify the direction of the measurement.

Finally, the authors should consider the recently published papers by Mecerreyes et al using aqueous binders which contain anions, which could be considered kosmotropic. Could the authors comment on this with respect to their analysis presented in the manuscript?

Eg. : Fluorine-Free Poly (ionic Liquid) s Binders for the Aqueous Processing of High-Voltage NMC811 Cathodes; AC Rolandi, C Pozo-Gonzalo, I de Meatza, N Casado, D Mecerreyes, ...
Advanced Energy and Sustainability Research 4 (12), 2300149; Carrageenans as Sustainable Water-Processable Binders for High-Voltage NMC811 Cathodes AC Rolandi, C Pozo-Gonzalo, I de Meatza, N Casado, M Forsyth, ...ACS Applied Energy Materials 6 (16), 8616-8625

Reviewer #2 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate

recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4 (Remarks to the Author):

Summary: The authors report the use of lithium salts with varying kosmotropivities (Li₂SO₄, LiOAc, LiOTf, and LiTFSI) enable the aqueous fabrication of LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ (NCM523) cathodes. The authors present the argument that the kosmotropic salt interacts with the water to prevent the water-induced lithium leaching and degradation of NCM811. Li₂SO₄, LiOAc, LiOTf, and LiTFSI were selected to have varying degrees of interaction with and structuring of water or kosmotropic (very strong interaction) to chaotropic (little interaction) variation. The authors begin by studying the structure of water in the aqueous lithium salt solutions using spectroscopic methods and simulations. Additionally, the performance of NCM batteries for cathodes cast with aqueous solutions of the two extreme salts LiTFSI (chaotropic) and Li₂SO₄ (kosmotropic) were evaluated. Tracking slurry pH and Li⁺ content were considered to check for leaching and material degradation and microscopy was used to identify changes in morphology of electrodes. The authors also examine electrodes with different mass loadings and NCM compositions. Finally, the authors provide an extensive cost/safety analysis of Li₂SO₄-based aqueous processing and propose alterations to existing NMP-based commercial processing system to transition to Li₂SO₄-based aqueous processing.

Overall, the authors present thorough analysis and discussion of their results, however, I believe the authors should add more discussion/comparisons of their results to prior work that report similar studies (see general comments below for papers) and explain how this work differs. Additionally, the authors should add details related to the experimental protocols to enhance reproducibility (please see general and specific comments for details) and to allow reviewers to fully evaluate the presented work. Currently, the experimental protocol for fabricating electrodes does not mention LiTFSI or Li₂SO₄ (or how they were integrated to the electrode fabrication process) making it difficult to fully evaluate the data presented. Additionally, the purity and source of all the chemicals utilized are not disclosed, making it impossible to evaluate the cost analysis presented by the authors. The findings of the manuscript could be of interest to the broader battery community; however, major revisions are necessary before determining the suitability of the manuscript for publication in Nature Communications. I have included my specific comments below.

General Comments:

- The authors should also add discussion to prior work and provide comparisons to the obtained results for the reader to clearly understand the significance and new findings.

References:

1. M. Heidebüchel, T. Schultz, T. Placke, M. Winter, N. Koch, R. Schmuck, & A. Gomez-Martin, ChemSusChem, e202202161 (2023).
2. H. Oh, S.-J. Shin, E. Choi, H. Yamagishi, T. Ohta, N. Yabuuchi, H.-G. Jung, H. Kim, and H. R. Byon, JACS Au, 1392-1402 (2023).
3. I. Dienwiebel, M. Diehl, B. Heidrich, X. Yang, M. Winter, M. Börner, Adv. Energy Sustainability Res., 2100075 (2021).

- The grade of lithium salts should be specified (for reproducibility and analysis of the presented cost

arguments).

- Do the salts used need to be lithium-based (since the anion seems critical to the water network)? Could alternative organic salts (ammonium, tetrabutylammonium, tetramethylammonium, etc.) also be used in this method? If other salts could be used, the authors should comment on the hypothesized effect of the cation's kosmotropicity on the observed stabilization for processing. For example, what would a chaotropic cation-based sulfate salt (such as tetramethylammonium sulfate) do? This is not a suggestion of additional experiments.
- EDS mapping of salt specific elements (perhaps sulfur) or thermal gravimetric analysis could provide more direct evidence/quantification of the precipitated salts present in the processed electrodes.

Specific Comments:

1. Abstract – The specific electrode material being studied should be mentioned in the abstract.
2. Figure 2 c – The line connecting the points should be removed, unless it represents a known fit between H-bond number and anion.
3. Figure 2 e – The data for the whole recovery time should be presented in the SI.
4. Page 9 – The method for getting the aluminum foil after casting should be specified. Was the electrode removed using solvent?
5. Figure 4 d-f – An arrow or label could be added to help the reader more readily identify the intergranular cracks.
6. Figure 5 a – Is this charge or discharge capacity? Please specify. The c-rate (two were provide in methods) should be specified.
7. Figure 5 d – The processing solution for the GCD data should be explicitly stated.
8. Methods Section – A comprehensive list of the materials/chemicals used, their purity, and supplying company should be provided.
9. Methods - Fabrication of electrodes– The preparation of the aqueous lithium salt solutions and how they were utilized in electrode fabrication should specified. The amount of liquid was used to formulate the slurry should be specified. The changes to slurry preparation for NCM622, NCM811, NCM9½½, and differing mass loading cathodes should be specified.
10. Methods – electrochemical Testing – The authors should specify any rest periods between battery assembly and electrochemical testing. Additionally, any “conditioning” cycles used before
11. Methods – EIS Testing – The voltage that EIS was centered at should be reported (i.e. 3 V with a 10 mV amplitude).
12. Figure S13 – Please specify the cycle shown and c-rate. Adding another panel with a later cycle (cycle 200) would provide the reader with a nice visual comparison of the battery performance vs. processing solvent.
13. Figure S18 – The figure caption should be clarified. It specifies a constant current density but the plot has different current densities. The number of cycles at each current density should be specified in the caption.
14. Figure S19 – EDS images for elements specifically found in TFSI and SO4 (sulfur and fluorine) should be added to this figure.
15. Supplementary Table 6 – Processing solution should be added as a column to allow the reader to easily compare the results. Additionally, sorting the whole table by specific capacity might help the reader see the trends more readily.

Manuscript Title: Kosmotropic aqueous solution for green lithium battery cathode manufacturing

Journal Name: Nature Communications

Manuscript Number: NCOMMS-24-14271-T

Reviewer's comments:

In submitted manuscript, authors have explored that kosmotropic anions in aqueous solution could be used for manufacturing of cathode for green lithium batteries. Ions in hydration shells are restructured to form a highly ordered state of anion–water clusters and regulate local hydration structure adjacent to cathode materials. Further it was also revealed that, scalable slurry-cast fabrication of various cathodes achieves a reduction in energy consumption. This is an applied nature; significant work.

This manuscript covers a detailed study which covered all details could be needed for reader. Data is supported with figures. All details have been provided in supplementary data.

Submitted manuscript could be accepted for publication after following minor points:

1. In summary, add some highlighted findings of submitted work in terms of different variables.
2. Mention objectives and novelty of submitted work in introduction section.
3. Eexplain about different softwares used to carry out computational analysis. Give brief understanding for readers.
4. In experimental section provide details about instrumentation were used for electrochemical evaluation.
5. In results and discussion section, on page number 6, discuss the order of strength of H-bond acceptor $\text{TFSI}^- < \text{OTf}^- < \text{OAc}^- < \text{SO}_4^{2-}$ with respect to different parameters such as electronegativity difference etc.
6. Literature citation in results and discussion section is totally missing. Provide relevant literature to support findings from computational analysis.
7. What are the future perspectives of submitted work in conclusions section?
8. There are so many grammatical mistakes throughout the manuscript.

Response to reviewer comments

Manuscript ID: NCOMMS-24-14271-A

Title: Kosmotropic aqueous processing solution for green lithium battery cathode manufacturing

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Reviewer #1

Kim et al. focused on the kosmotropic effect of anions and investigated how it affects the electrode structure and battery performance of NMC cathodes made via the coating of water-based slurries containing such anions. DFT, MD simulation, FTIR, and NMR were employed to discuss the difference in the bulk and interfacial solvation structures of anions with different kosmotropivities. The battery tests clarified the comparable cycle performance of the water-processed NMC811 containing kosmotropic anions with that made from the conventional NMP-based slurry under a high active-material loading of ca. 20 mg/cm² (i.e., the capacity retention of 81% vs. 82% after 400 cycles at 0.2C charging/0.5C discharging). In addition, the authors estimated how the transition from NMP-based slurries to water-based slurries with a high kosmotropic effect benefits cathode manufacturing, which expects a 46% reduction in energy consumption of the cathode production.

Overall, the results look great; ample computational and experimental results using various techniques will definitely be informative for researchers in the battery field to mitigate the reactivity of water with cathode active materials to enable stable cathode manufacturing using water as a slurry-processing solvent.

However, at this stage, before considering publication, the authors need to consider several questions and possibly additional experiments as provided below in order to further convince the readers of the benefits of the transition to cathode manufacturing using water-based slurries in terms of electrode properties and battery performance. Some of the comparisons currently provided do not give a completely fair picture.

→ **Response:** Many thanks for the reviewer's valuable comments. We've prepared a revision that can address the reviewer's comments well. Below are the details of the responses.

a-1. Do the results of battery tests really represent the positive effect of using the anions with a high kosmotropic effect?

The main issue of this manuscript is that the battery evaluation involves (at least) four different factors: (i) electrode structure (e.g., particle size distribution of NMC811), (ii) amount of leached Li^+ ions from NMC811 (which affect the achievable capacity of the active material), (iii) state of Al current collector (whose corrosion would affect the overvoltage), and (iv) Li salt remained in the electrode layer (which would subsequently affect the CEI formation), and the evaluated cathodes are different in these aspects. This blurs the consequence of using the anions with a high kosmotropic effect, requiring some control experiments so that the results of battery tests truly represent the positive effect of using the anions with a high kosmotropic effect.

Firstly, I recommend performing a straightforward comparison between water-processed NMC811 and NMP-processed NMC811 by removing salts (used to mitigate the interfacial reaction of the active material in a water slurry) from the water processed electrodes before assembling the cells and undertaking the battery tests. This excludes the uninvestigated effect of (iv) and allows a fair comparison between the consequences of using the anions with different kosmotropivities in a water-based slurry and that of an NMP-based slurry.

Secondly, making NMC811 cathodes using 1.0 M LiTFSI and 0.5 M Li_2SO_4 in NMP-based slurries will provide valuable insights into the structural modification of NMC811 particles by these salts. This is because, from Supplementary Figs. 10 and 19, readers might think that these salts themselves also have an impact on the breakage of the secondary particles of NMC811. (In contrast, the use of water without these salts provided relatively intact secondary particles.) Doing the cathode preparation in NMP using these salts will relieve this concern and enable the authors to say the breakage of the secondary particles is purely due to the interfacial reaction of water on the cathode surface and it subsequently affects the battery performance.

Thirdly, to state the effect of the studied anions on the CEI formation and its influence on the battery performance, the authors are recommended to perform battery tests using the NMC811 cathode (made via an NMP-based slurry) in the electrolyte solutions containing the Li salts of these anions additionally. This keeps (i)–(iii) in the same condition and allows

the authors to discuss how these anions affect the battery performance (as well as the CEI composition).

→ **Response:**

1. Comparison between water-processed NCM811 and NMP-processed NMC811 electrodes by removing salts from the water-processed electrodes

: In response to the reviewer's comment, in an effort to remove salts from water-processed electrodes, we immersed the electrodes in the same processing solution and then applied a sonication treatment. Unfortunately, this procedure caused the dissolution of the binder in the electrodes, leading to structural disruption, as illustrated in **Fig. R1**. Consequently, it was difficult to obtain reliable electrochemical data from the water-processed electrodes from which the salts had been removed. As an alternative approach, we incorporated salts into the NMP-based slurry to investigate their effect on the cycling performance of the cathodes, depicted in **Fig. R2**. The addition of salts into the NMP-based slurry had no beneficial effect on the cycling performance of the resulting cathodes. Further details are provided in the response #3.

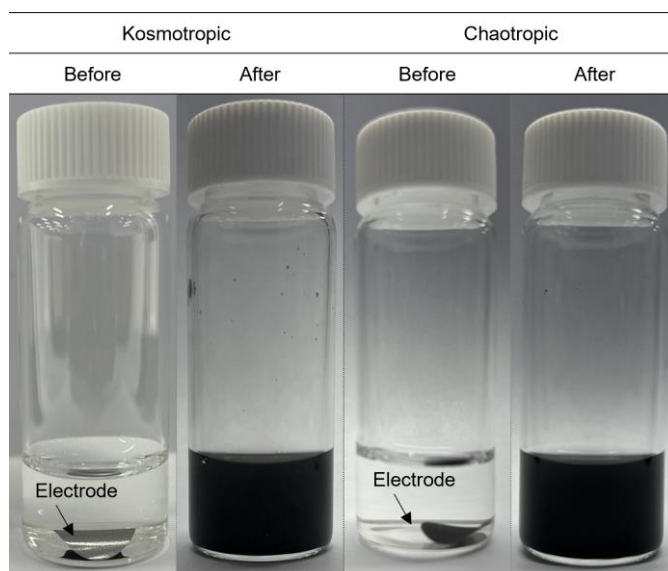


Fig. R1 | Photograph of the aqueous-processed electrodes immersed in water before and after the sonication treatment used to facilitate the removal of the salts: kosmotropic solution-processed electrode (left), and chaotropic solution-processed electrode (right).

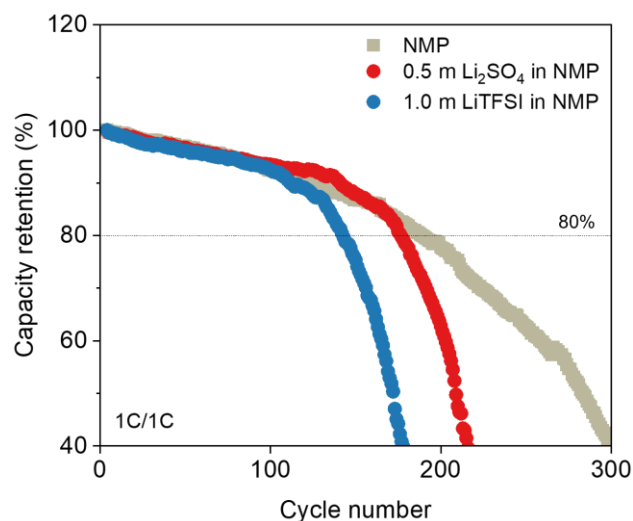


Fig. R2 | Cycling performance of the NCM811 cathodes prepared using NMP solutions with different salts at a current rate of 1 C/1 C ($= 3.7 \text{ mA cm}^{-2}$).

2. Making NMC811 cathodes using 1.0 M LiTFSI and 0.5 M Li₂SO₄ in NMP-based slurries to investigate an impact on the breakage of the secondary particles of NMC811.

: In response to the reviewer's comment, we prepared the NCM811 cathodes using 1.0 M LiTFSI and 0.5 M Li₂SO₄ in NMP-based slurries and then investigated the effect of the added salts on the change in the particle size of the NCM811. The particle sizes remained almost unchanged regardless of the inclusion of salt additives in the NMP-based slurries, indicating that no side reactions occurred between the salt and NCM811 cathode materials, as shown in **Fig. R3**. Additionally, the particle size of NCM811 in water-processed cathodes was analyzed, shown in **Fig. R4**. The particle size tends to decrease in the order of NMP > 0.5 M Li₂SO₄ in H₂O > 1.0 M LiTFSI in H₂O > H₂O. This finding demonstrates that the microstructural degradation of secondary NCM811 particles is ascribed to the undesired interfacial side reactions with water along the crystal grain boundaries, as previously discussed in the submitted manuscript and corroborated by the previous paper (ref.: ACS Appl. Mater. Interfaces 12, 52, 57963 (2020)). In response to the reviewer's comment, details regarding the particle size distribution have been included in the revised manuscript.

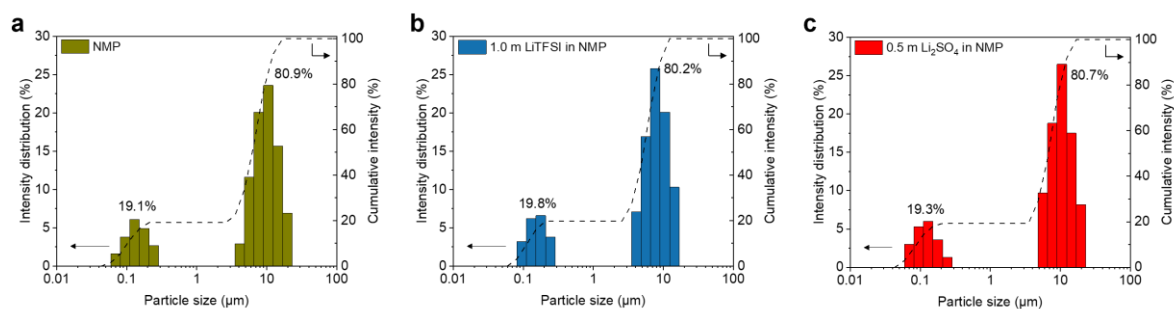


Fig. R3 | a-c, Particle size distribution of the NCM811 particles after immersion in various NMP-based solutions. **a-c,** NMP (**a**), 1.0 m LiTFSI in NMP (**b**), and 0.5 m Li₂SO₄ in NMP (**c**). The percentages indicate the proportion of primary and secondary particles present in each solution.

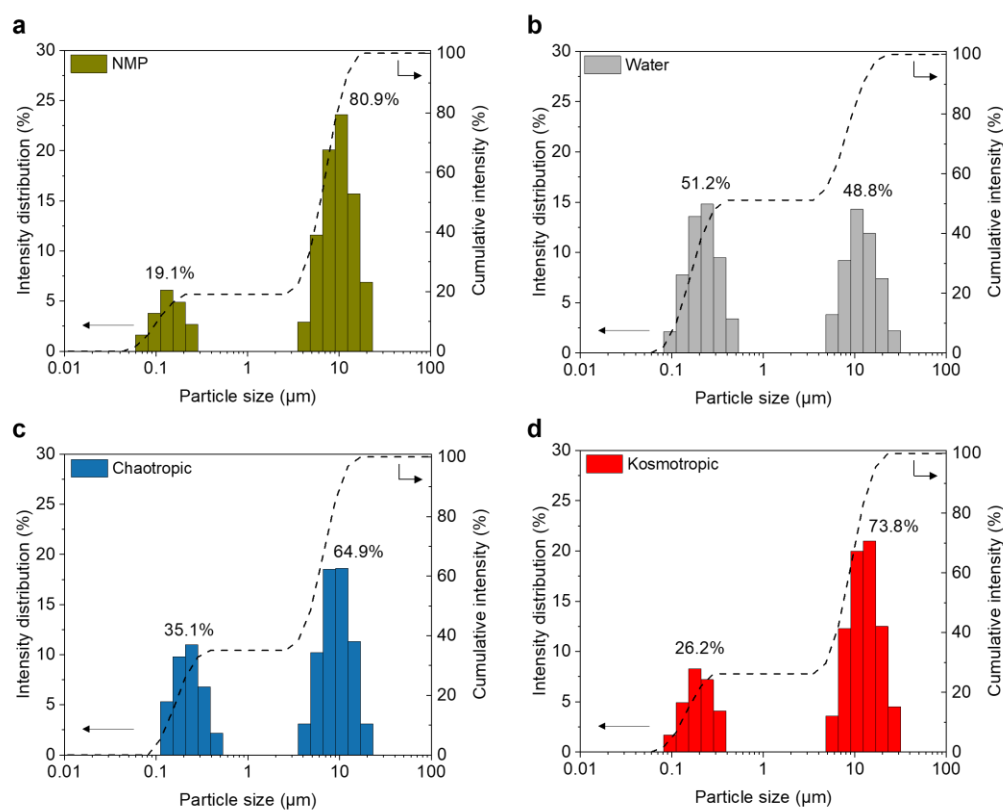


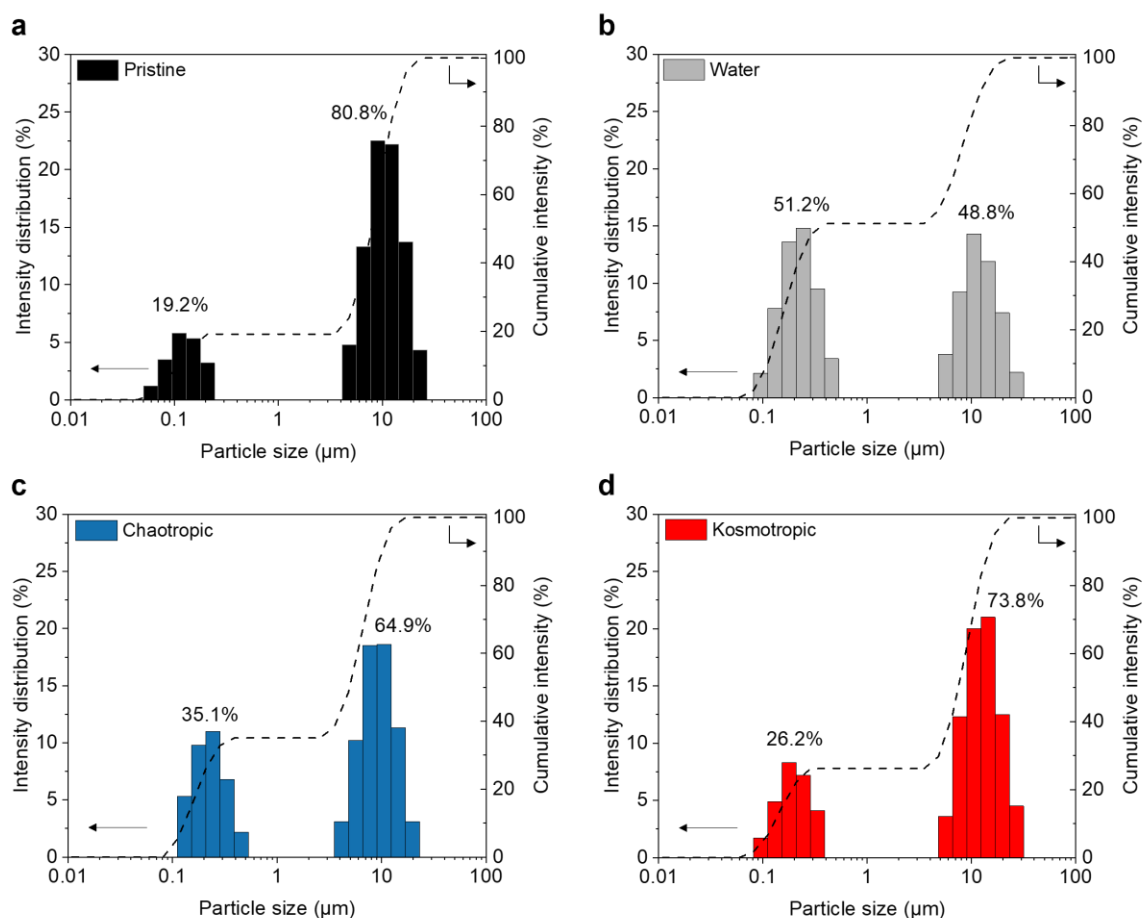
Fig. R4 | a-d, Particle size distribution of the NCM811 particles after immersion in various processing solutions. NMP (**a**), water (**b**), chaotropic solution (**c**), and kosmotropic solution (**d**). The percentages indicate the proportion of primary and secondary particles present in each processing solution.

[Revised Manuscript]

"After immersion in the water or chaotropic solutions, the secondary NCM811 particles disintegrated into primary particles. In contrast, particles immersed in the kosmotropic solution remained relatively intact. This observation was quantitatively confirmed by analyzing particle size distribution using Zetasizer (Supplementary Fig. 11). The particle size tends to decrease in the order of NMP > kosmotropic solution > chaotropic solution > water. This finding demonstrates that the microstructural degradation of secondary NCM811 particles is ascribed to the undesired interfacial side reactions with water along the crystal grain boundaries³⁴."

"Structural and physicochemical characterization

The surface and cross-sectional morphologies of the electrodes were investigated using field emission secondary electron microscopy (FE-SEM, S-4800, Hitachi), energy-dispersive X-ray spectroscopy (EDS, JSM 6400, JEOL), and field emission electron probe microanalyzer (JXA-8530F, JEOL). The particle size distribution of the NCM811 powders soaked in different processing solutions was analyzed using a Zetasizer (ELSZneoZS, Otsuka), with ethanol serving as the solvent. To observe the surface of Al current collectors after the electrode coating, the electrode active layers were gently removed from the electrode using a cotton swab soaked in dimethyl carbonate (DMC) solvent. The area of Al corrosion was quantified from binary images generated by applying 34% of the threshold program (Image J) to the SEM image of the Al current collectors. Cross-sectioned samples were thinned using focused ion beam (FIB, Helios Nano Lab, FEI) to analyze the microstructure of NCM811 particles."



Supplementary Fig. 11 | a-d, Particle size distribution of the pristine NCM811 particles (a), and NCM811 particles after immersion in various processing solutions (b-d): water (b), chaotropic solution (c), and kosmotropic solution (d). The percentages indicate the proportion of primary and secondary particles present in each processing solution.

3. Electrochemical performance of the NMP-based slurries containing Li salts

: In response to the reviewer's comment, we added the salts into the NMP-based slurry and then investigated their effect on the cycling performance of the resulting cathodes. The obtained NCM811 cathodes were cycled at a current rate of 1 C/1 C ($= 3.7 \text{ mA cm}^{-2}$). The cycling performance of the NCM811 cathodes tended to decrease in the following order: NMP > 0.5

m Li₂SO₄ in NMP > 1.0 m LiTFSI in NMP. This result reveals no beneficial effect of the incorporated salts on the cycling performance of the NMP-processed NCM811 cathodes. Such result was further verified by similar atomic percentile of the XPS analysis of the cycled NCM811 cathodes (**Fig. R5**). The inferior cycling performance of the salt-containing NCM811 cathodes is attributed to the precipitated salts-induced pore blockage.

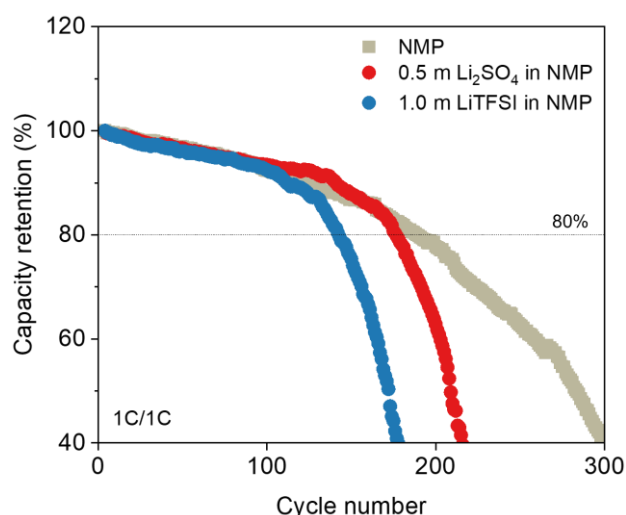


Fig. R2 | Cycling performance of the NCM811 cathodes prepared using NMP solutions with different salts at a current rate of 1 C/1 C (= 3.7 mA cm⁻²).

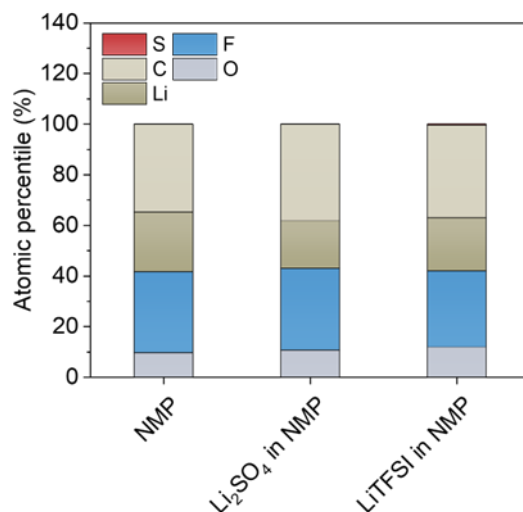


Fig. R5 | XPS elemental analysis of the surface of the cycled NCM811 cathodes (cycled at 1 C/1 C) processed with NMP, 0.5 m Li₂SO₄ in NMP, 1.0 m LiTFSI in NMP, respectively.

a-2. How about the rate and long-term cycle performances of NMC811 made via a water-based slurry with anions with a high kosmotropic effect?

Although the authors have tried to explain the significance of using the anions with a high kosmotropic effect in a water-based NMC811 by presenting the achieved capacity per the total cost of the cathode ingredients (Fig. 5c), the obvious difference in the battery performance between water-processed NMC811 and NMP-processed NMC811 has yet to be clarified. This should be clarified so that the authors can state the transition to the water-based slurry for cathode manufacturing is truly beneficial in addition to the estimated advantages of cost reduction and environmental friendliness.

Additional experiments that the authors could consider are:

Firstly, comparison of the charge/discharge rate performances between water-processed NMC811 and NMP-processed NMC811.

Secondly, the cycle test (Fig 5a) should be prolonged as much as possible (ideally, >1,000 cycles). This is because such a long-term cycle life has been expected for commercial electrodes (e.g., <https://www.energy.gov/eere/vehicles/articles/reducing-reliance-cobalt-lithium-ion-batteries>) and, in this respect, it is worthwhile to show how long water-processed NMC811 can keep up with NMP-processed NMC811. Furthermore, readers do not understand how their cells will fully degrade by only seeing Fig. 5a as both cathodes showed similar capacity decay up to 400 cycles. This necessitates a prolonged cycle test beyond 80% of the initial capacity.

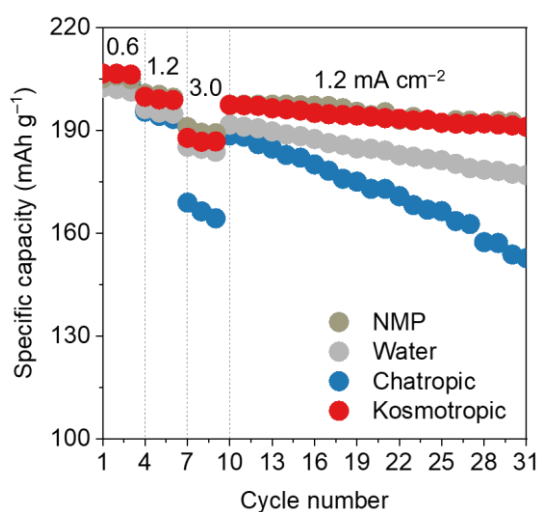
The authors should address the aforementioned concerns and revise the manuscript to tell a clear story about the significance of their results in using the anions with a high kosmotropic effect for the cathode manufacturing of lithium-ion batteries.

In addition to the above considerations, the following point-by-point suggestions should be addressed in order to further improve the manuscript:

→ **Response:** Thank you so much for the reviewer's valuable comments. Our response to the reviewer's comment is provided below.

Comparison of rate performance:

The rate performance result of the water-processed electrodes and NMP-processed electrode was already shown in the original manuscript (**Supplementary Fig. 19**). This result indicates that even at a high current density of 3.0 mA cm^{-2} , the kosmotropic aqueous solution-processed electrode exhibited the discharge capacities comparable to those of the NMP-processed electrodes.



Supplementary Fig. 19 | Discharge rate capability (discharge current rate = 0.6 mA cm^{-2} (for 3 cycles), 1.2 mA cm^{-2} (for 3 cycles), 3.0 mA cm^{-2} (for 3 cycles), and 1.2 mA cm^{-2} of the cathodes at an areal capacity of 6 mAh cm^{-2} under a fixed charge current density of 3.0 mA cm^{-2} .

Cycling performance

It is noted that the cycle life (> 1000 cycles) mentioned by the reviewer pertains to state-of-the-art Li-ion batteries (LIBs) with graphite anodes. On the other hand, we investigated the electrochemical performance of the aqueous solution-processed cathodes using a Li-metal cell configuration (Li metal was used as an anode), to focus exclusively on the cathode performance. In addition, since cell testing for practical energy-dense batteries requires high-areal-capacity electrodes and low electrolyte-to-capacity (E/C) ratios, we evaluated the Li-metal full cells with an areal capacity of 3.7 mAh cm^{-2} under an E/C ratio of 5 g Ah^{-1} . Such constrained conditions pose inherent challenges for long-term cycling stability, as described in the references from *Nat. Energy* **4**, 551 (2019), *Nat. Energy* **6**, 723 (2021), and *Angew. Chem. Int.*

Ed. **59**, 14935 (2020). To verify this discussion, we have included a comparison of areal capacities and cycle numbers between the cells with the kosmotropic aqueous solution-processed cathode (this study) and the previously reported Li-metal cells with aqueous solution-processed cathodes in **Fig. R6**, demonstrating the superior cycling performance of our study. Meanwhile, we have revised **Fig. 5a** to clarify the number of cycles needed to reach 80% of the initial capacity.

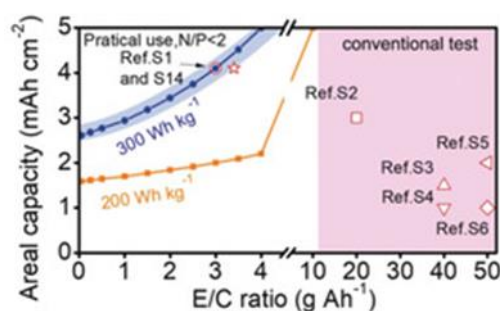


Fig. | A captured image from a research article published in *Angew. Chem. Int. Ed.* **59**, 14935 (2020)

what are practically required to achieve high energy. High cell-level energy density cannot be achieved unless realistic conditions are used^{17–20}, including high cathode loading, low electrolyte amount to cathode capacity (E/C) ratio and negative to positive areal capacity (N/P) ratio. Therefore, many ‘promising’ results obtained under unconstrained conditions may not be realized or repeated in realistic high-energy batteries.

Fig. | A captured image from a research article published in *Nat. Energy* **6**, 723 (2021)

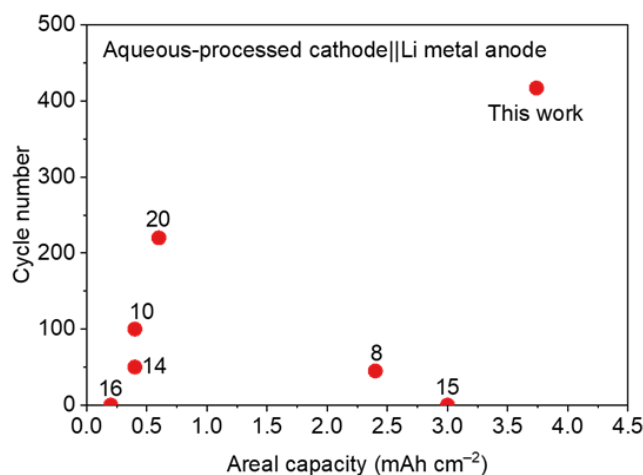


Fig. R6 | Comparison of the kosmotropic aqueous solution-processed cathode (this work) and the previously reported aqueous solution-processed cathodes, in terms of areal capacity of cathode and cycle number of Li-metal batteries.

[Revised manuscript]

“The capacity of the water- and chaotropic solution-processed cathodes degraded significantly after 200 cycles because of the active Li^+/H^+ exchange^{5,39} during the aqueous cathode fabrication (Supplementary Fig. 15 and Fig. 5a). In contrast, the kosmotropic solution-processed cathode delivered a high initial specific capacity of $205 \text{ mAh g}_{\text{NCM811}}^{-1}$ with a stable cycle retention (over 80% after 400 cycles), which is comparable to that of the NMP-processed cathode.”

“Furthermore, the kosmotropic solution-processed cathode demonstrated superior discharge rate capability, delivering up to 188 mAh g^{-1} under a fixed charge current density of 3.0 mA cm^{-2} , comparable to the NMP-processed cathode (Supplementary Fig. 19). In contrast, the chaotropic solution-processed cathodes exhibited inferior performance, which can be ascribed to the pore blockage as shown in the cross-sectional SEM images (Supplementary Fig. 20) and the reduced particle size of NCM811 that could increase the tortuosity of ion conduction (Supplementary Fig. 11).”

“For the electrochemical tests, CR2032 coin-type cells (Ni-rich cathode||polyethylene (PE)||Li-metal anode (thickness = $100 \text{ }\mu\text{m}$)) were assembled in an Ar-filled glove box. Thereafter, 1 M LiPF_6 in EC/DEC(= 1/1 (v/v)) with 10 wt.% of FEC and 2 wt.% VC was used as a liquid electrolyte. The electrolyte was injected into the cell with an electrolyte mass/electrode capacity (E/C) ratio of 5 g Ah^{-1} explore the cell performance for practical energy-dense Li-metal cells⁴⁰, if not specified. After the cell assembly, a rest period of 12 h was allowed to ensure complete wetting of the electrodes. The cells were then subjected to formation cycles (1 charge/discharge cycle at a current rate of 0.1C).”

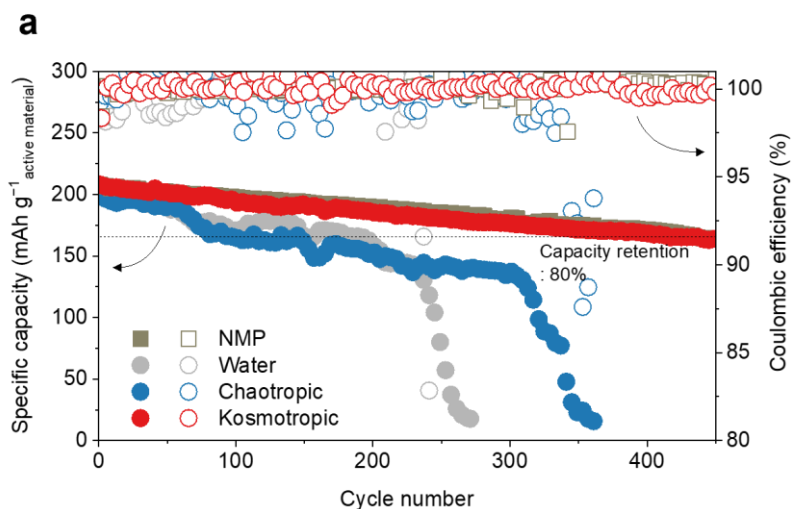
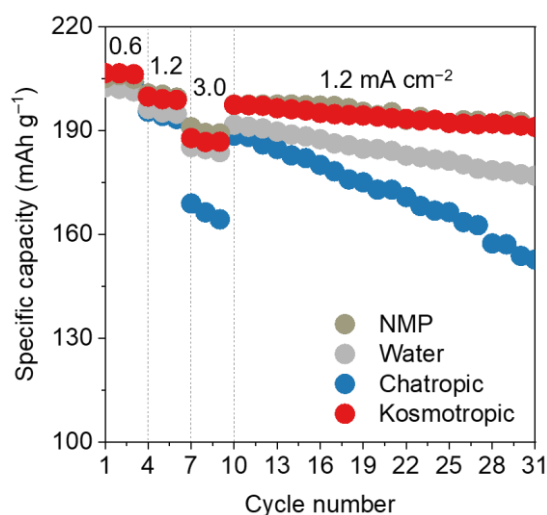


Fig. 5 | a, Cycling retention (expressed as **discharge** specific capacities of cathode materials) of the cathodes **at charge/discharge current rate of 0.2 C/0.5 C (= 1.9 mA cm⁻²)**.



Supplementary Fig. 19 | Discharge rate capability (**discharge current rate = 0.6 mA cm⁻² (for 3 cycles), 1.2 mA cm⁻² (for 3 cycles), 3.0 mA cm⁻² (for 3 cycles), and 1.2 mA cm⁻²** of the cathodes at an areal capacity of 6 mAh cm⁻² under a fixed charge current density of 3.0 mA cm⁻²).

b–1. Line 85: Here, “material” should be changed to “CO₂ emission rate”. Revise the sentence accordingly.

→ **Response:** In response to the reviewer's valuable comment, the sentence was changed to accurately deliver the message.

[Revised manuscript]

“In addition to the electrochemical benefits, the material cost of the kosmotropic solution is 96% lower than that of the NMP processing solvent. From a comprehensive techno-economic analysis, we propose that the use of the kosmotropic aqueous processing solution can reduce the operational (~ 23%), and capital (~ 95%) costs associated with cathode manufacturing by eliminating the conventional solvent recovery process. Furthermore, this approach substantially reduced energy consumption in electrode manufacturing by 46%, demonstrating its viability as a platform technology for environmentally friendly and cost-competitive electrode manufacturing applicable to various high-capacity electrode materials.”

b–2. Lines 194–196: How was this surface observation of the Al current collectors performed? In particular, no information is available about how the dried electrode layers were removed to see the Al surface attached to the electrode slurries. Please provide the detailed experimental procedure in the manuscript or the Supplementary Information.

→ **Response:** In response to the reviewer's valuable comment, the detailed experimental procedure used to obtain the Al foil for the corrosion analysis was added in the revised manuscript.

[Revised manuscript]

“Structural and physicochemical characterization

To observe the surface of Al current collectors after the electrode coating, the electrode active layers were gently removed from the electrode using a cotton swab soaked in dimethyl carbonate (DMC) solvent. The area of Al corrosion was quantified from binary images generated by applying 34% of the threshold program (Image J) to the SEM image of the Al current collectors. Cross-sectioned samples were thinned using focused ion beam (FIB, Helios Nano Lab, FEI) to analyze the microstructure of NCM811 particles.”

b–3. Lines 200–204: To support the authors’ speculation, I recommend evaluating the particle size distribution of these NMC811 particles. This clarifies the breakage of secondary particles as a reduction in the particle size. For now, immersion of NMC811 in water appears to show almost no change in the particle size (Supplementary Fig. 10b vs. 10a).

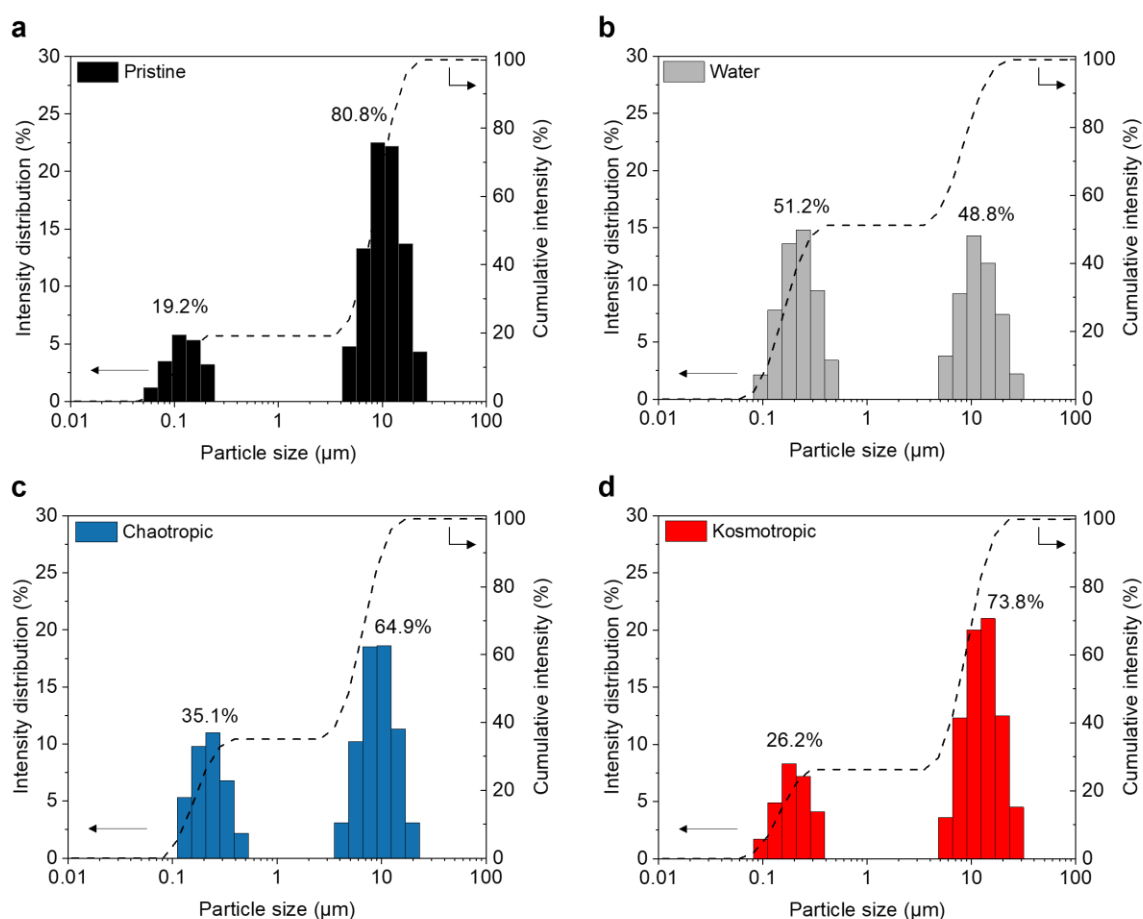
→ **Response:** Thank you for your insightful comment. In response to the reviewer’s comment, we quantitatively analyzed the particle size distribution of the NCM811 soaked in different processing solutions using a Zetasizer. The particle size of the resulting NCM811 tended to decrease in the following order: Pristine NCM811 \approx kosmotropic solution > chaotropic solution > water. This disintegration of the secondary NCM81 particles into the primary ones may be ascribed to the undesired interfacial side reactions with water along the crystal grain boundaries (ref.: Azhari, L. *et al.* Effects of Extended Aqueous Processing on Structure, Chemistry, and Performance of Polycrystalline $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ Cathode Powders. *ACS Appl. Mater. Interfaces* **12**, 57963-57974 (2020)). In comparison, the negligible change in the particle size of NCM811 soaked in the kosmotropic solution demonstrates the viability of the solution in retaining the structural stability of the NCM811 particles. This result aligns well with the SEM images and XRD patterns (shown in **Fig. 4c**, and **Supplementary Fig. 12**) of the submitted manuscript, exhibiting the significant effect of the processing solutions on the particle size of NMC811. In the response to the reviewer’s comment, the result of the particle size distribution was added (**Supplementary Fig. 11**) in the revised manuscript.

[Revised manuscript]

“The microstructural degradation of NCM811 was investigated using SEM (**Supplementary Fig. 10**). After immersion in the water **or chaotropic solutions**, the secondary NCM811 particles disintegrated into primary particles. **In contrast**, particles immersed in the kosmotropic solution remained **relatively** intact. **This observation was quantitatively confirmed by analyzing particle size distribution using Zetasizer (Supplementary Fig. 11).** The particle size tends to **decrease in the order of NMP > kosmotropic solution > chaotropic solution > water.** **This finding demonstrates that the microstructural degradation of secondary NCM811 particles is ascribed to the undesired interfacial side reactions with water along the crystal grain boundaries³⁴.**”

“Structural and physicochemical characterization

The total experimental time was approximately 3.7 h. All NMR experiments were performed at room temperature. The surface and cross-sectional morphologies of the electrodes were investigated using field emission secondary electron microscopy (FE-SEM, S-4800, Hitachi), energy-dispersive X-ray spectroscopy (EDS, JSM 6400, JEOL), and field emission electron probe microanalyzer (JXA-8530F, JEOL). The particle size distribution of the NCM811 powders soaked in different processing solutions was analyzed using a Zetasizer (ELSZneoZS, Otsuka), with ethanol serving as the solvent. To observe the surface of Al current collectors after the electrode coating, the electrode active layers were gently removed from the electrode using a cotton swab soaked in dimethyl carbonate (DMC) solvent.”

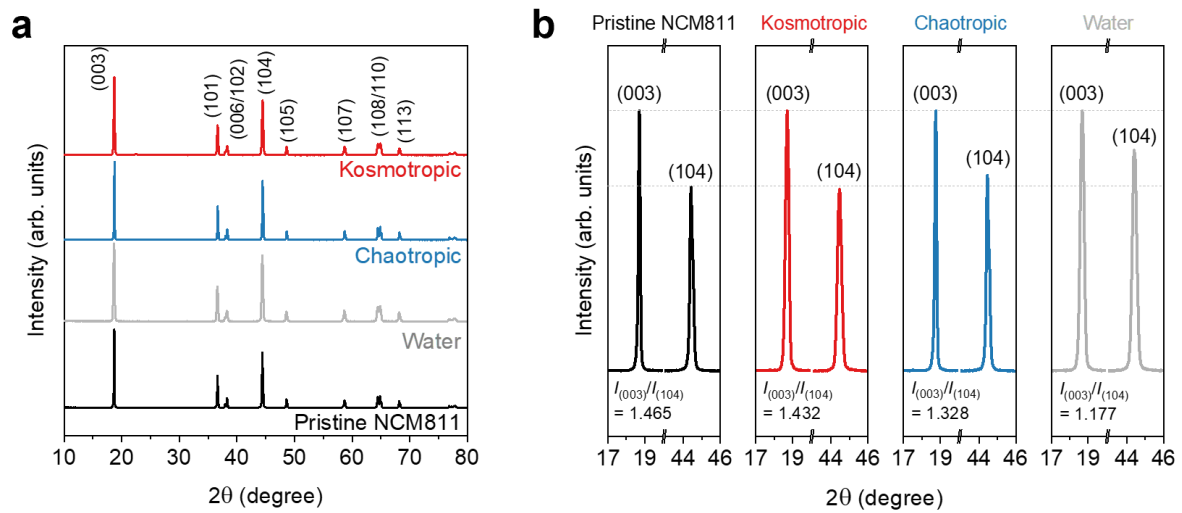


Supplementary Fig. 11 | a-d, Particle size distribution of the pristine NCM811 particles (a), and NCM811 particles after immersion in various processing solutions (b-d): water (b), chaotropic solution (c), and kosmotropic solution (d). The percentages indicate the proportion of primary and secondary particles present in each processing solution.

b–4. Lines 206–208: From Supplementary Fig. 11, I could not see the obvious difference in the I_{003}/I_{104} ratio between the samples labelled “Kosmotropic”, “Chaotropic”, and “H₂O”, which contradicts Fig. 4c. Please double-check it.

→ **Response:** Thank you so much for the reviewer’s valuable comment. In response to the reviewer’s comment, we added the high-magnification view for each sample in the revised **Supplementary Fig. 12**. This analysis provided the obvious difference in the I_{003}/I_{104} ratio between the different samples. The reviewer’s comment is highly appreciated again.

[Revised manuscript]



Supplementary Fig. 12 | a, X-ray diffraction (XRD) patterns of NCM811 powders after immersion in water, chaotropic solution, and kosmotropic solution for 6 h. **b**, Magnified XRD patterns, which is normalized by the intensity of (003) peak, highlighting the I_{003}/I_{104} ratio for the NCM811 powders.

b–5. Line 219: From the caption of Supplementary Table 5, the NMC811 loading is 18.0 mg/cm². Which is true: 20 or 18.0 mg/cm²?

→ **Response:** Thank you for the reviewer’s kind comment. The NMC811 loading was 18 mg cm⁻² for the cycle life comparison between the different processing solutions. In response to the reviewer’s comment, we revised the error in the revised manuscript.

[Revised manuscript]

“Electrochemical performance of the cathodes manufactured using the aqueous solutions

To test the aqueous processed cathodes, we prepared cells comprising of Li metal anodes and NMC811 cathodes with areal mass loading and electrode density densities of 18 mg_{NMC811} cm⁻² and 3.0 g cc⁻¹, respectively (See Methods and **Supplementary Table 2** for details). The aqueous cathode slurries exhibited dispersion stability similar to those formulated in NMP, suggesting that the incorporation of kosmotropic or chaotropic salts had a minimal effect on the dispersion of electrode slurries (Supplementary Fig. 13).”

b–6. Line 219: In relation to the above comment, I recommend specifying the NMC811 loading and the electrode density of all tested cathodes in the Supplementary Information. As a difference in both variables affects the battery performance, the authors need to show these values for all tested cathodes so that the observed difference in the battery performance is not due to the difference in MNC811 loading and/or electrode density.

→ **Response:** The reviewer’s valuable comments are highly appreciated. In response to the reviewer’s comment, we specified the mass loading and electrode density for all tested cathodes in the Supplementary Information and Methods.

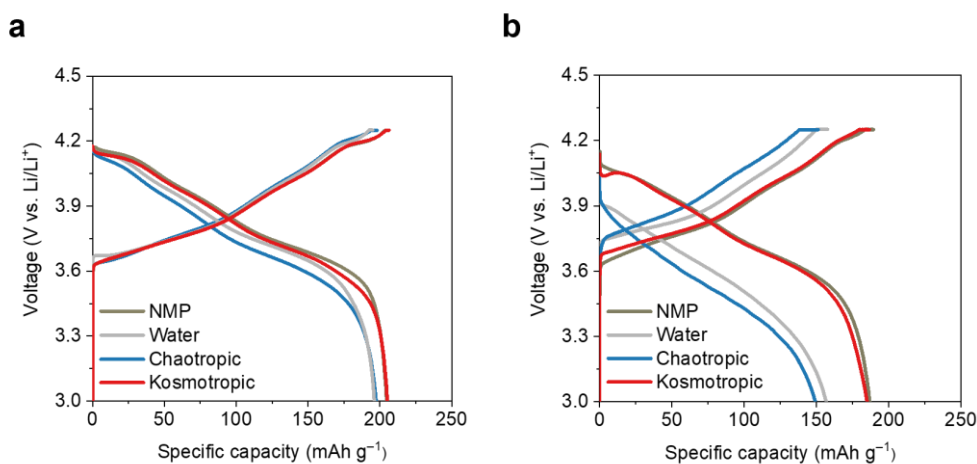
[Revised manuscript]

“To test the aqueous processed cathodes, we prepared cells comprising of Li metal anodes and NMC811 cathodes with areal mass loading and electrode density densities of 18 mg_{NMC811} cm⁻² and 3.0 g cc⁻¹, respectively (See Methods and **Supplementary Table 2** for details). The aqueous cathode slurries exhibited dispersion stability similar to those formulated in NMP, suggesting that the incorporation of kosmotropic or chaotropic salts had a minimal effect on the dispersion of electrode slurries (**Supplementary Fig. 13**). After calendaring, the structures of the electrodes were analyzed using cross-sectional SEM and corresponding EPMA images (**Supplementary Fig. 14**). Intergranular cracks were observed on the NCM811 particles in the water- and chaotropic solution-processed cathodes (red arrows, **Fig. 4d, e**). In contrast, the particles in the kosmotropic solution-processed cathodes remained comparatively unimpaired (**Fig. 4f**). These observations imply that external pressures can amplify the microstructural degradation of cathode materials after the aqueous processing.”

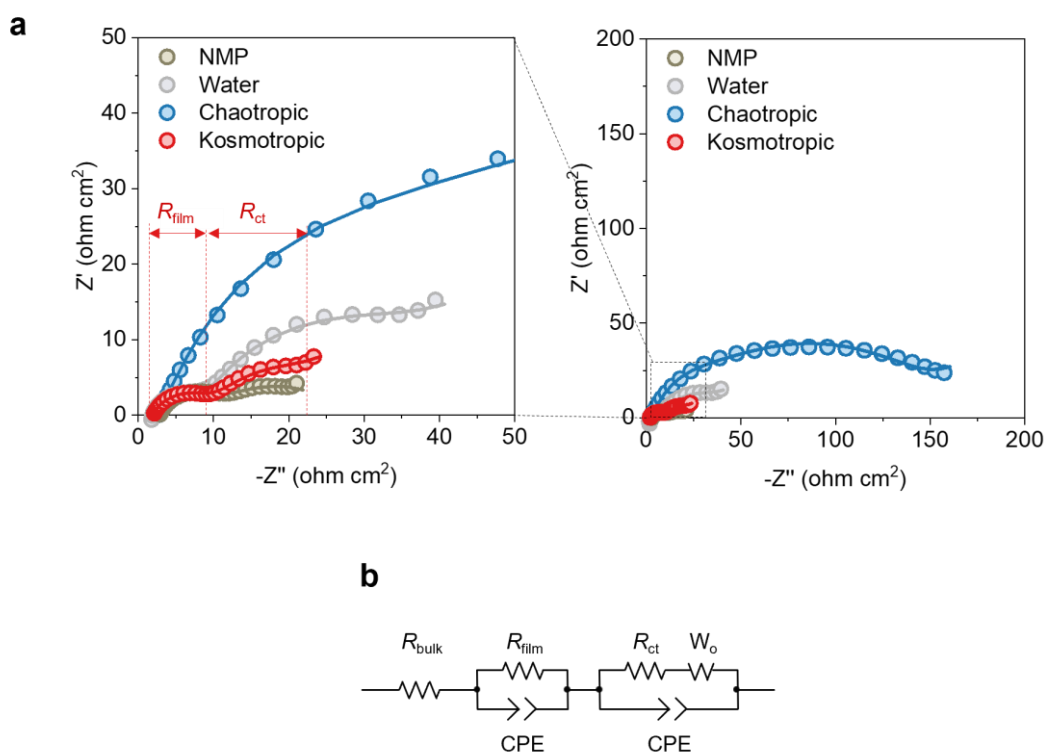
“Fabrication of electrodes, cell, and electrochemical characterizations

The electrode slurries were prepared with a composition of LiNi_{0.8}Co_{0.1}Mn_{0.1}O₂(NMC811)/carbon black/binder = 95.5/2.25/2.25 (w/w/w). Details of the electrode composition including residual salts are provided in **Supplementary Table 2**. For aqueous solution-processed electrodes, CMC and PAA were used as binders at a composition

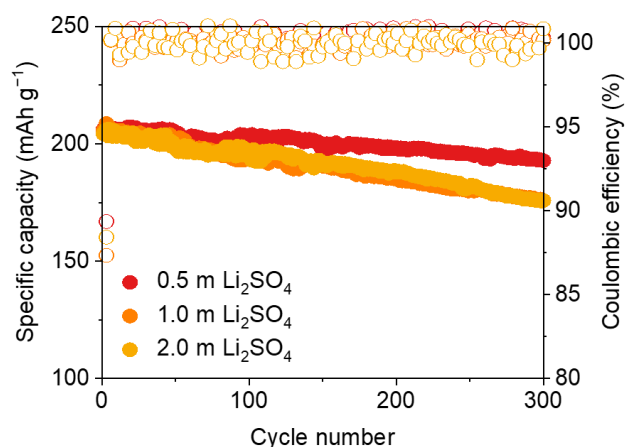
ratio of 1/1 (w/w). The salt additives were dissolved in water for 2 h and then the CMC and PAA binders were dissolved before the formulation of electrode slurries. The binder contents in the processing solutions are 3 wt.% and 10 wt.% for CMC and PAA, respectively. No additional water is added during the electrode fabrication, leaving 47 wt.% of water in the kosmotropic electrode slurry. The electrode slurries were cast onto Al current collectors and dried at 60°C for 6 h, followed by roll-pressing at 120°C. The electrode fabrication process was identical regardless of the mass loading and type of cathode materials. For the NMP-processed electrodes, PVDF was used as the binder, and dissolved in NMP. The concentration of the binder solution was set at 10 wt.%. The electrode slurries were cast onto Al current collectors and dried at 120°C for 6 h and then roll-pressed at 120°C. The density of the fabricated electrodes was set to 3.0 g cc⁻¹, and the electrodes were vacuum dried at 120°C for 12 h before the cell assembly.”



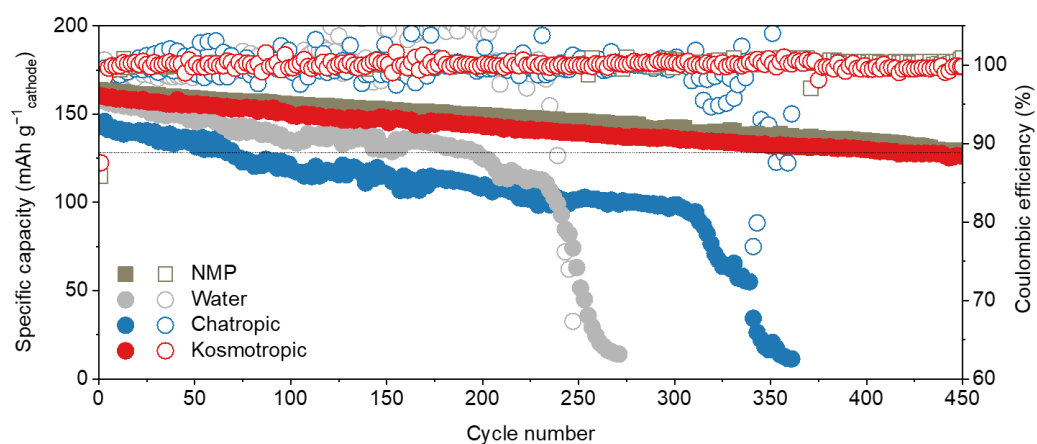
Supplementary Fig. 15 | a,b, Galvanostatic charge/discharge profiles of the cathodes at a mass loading of 18 mg cm⁻² at a charge/discharge current rate of 0.2 C/0.5 C (= 1.9 mA cm⁻²): 1st cycle (a) and 200th cycle (b).



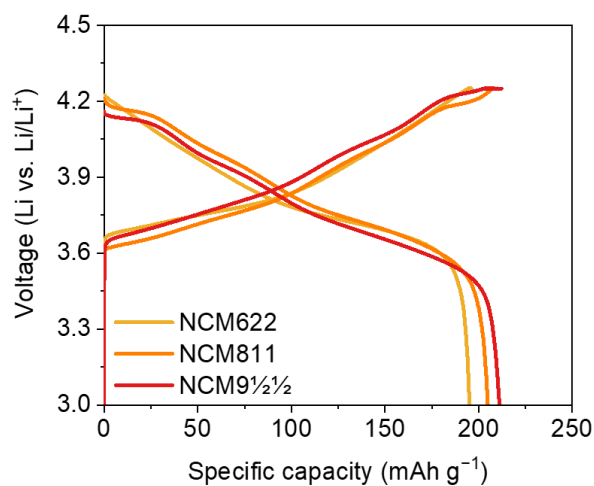
Supplementary Fig. 16 | **a**, Nyquist plots of the cycled cathodes at a mass loading of **18 mg cm⁻²** after the 400th cycle. **b**, The symbols and solid lines represent experimental data and fitted curves, respectively. **b**, Equivalent circuit used to fit the curves.



Supplementary Fig. 17 | Cycle retention of the kosmotropic solution-processed cathodes at a mass loading of **18 mg cm⁻²** as a function of the Li₂SO₄ concentration of the kosmotropic aqueous **processing** solution **at a current rate of 0.2 C/0.5 C (= 1.9 mA cm⁻²)**. The slightly poor cycle retention at 1.0 and 2.0 m Li₂SO₄ may be attributed to the pore blockage of the precipitated salt due to the excessive incorporation.



Supplementary Fig. 18 | Cycling retention (expressed as **discharge** specific capacities of cathodes based on their total mass including that of the salt additives) of the cathodes **at a current rate of 0.2 C/0.5 C ($= 1.9 \text{ mA cm}^{-2}$)**.



Supplementary Fig. 22 | Galvanostatic charge/discharge profiles of the kosmotropic solution-processed NCM622, NCM811, and NCM9½½ cathodes **at a current rate of 0.1 C/0.2 C ($= 0.8 \text{ mA cm}^{-2}$)**.

b-7. Lines 238–239: This is hardly believable based on the provided information (Supplementary Fig. 15). How about spectra in the N1s, P2p, and S2p regions?

In addition, how the cycled NMC811 was prepared for XPS (i.e., what was the washing process of the surface)?

I think it is too early to say there is no effect of a CEI on the battery performance.

→ **Response:** Many thanks to the reviewer's valuable comment. For the CEI analysis, we rinsed the cycled NCM811 using dimethyl carbonate (DMC) for 10 min to remove any residual salts and non-volatile solvents, which is a standard post-treatment procedure used for the SEI/CEI analysis (ref.: Review—Post-Mortem Analysis of Aged Lithium-Ion Batteries: Disassembly Methodology and Physico-Chemical Analysis Techniques *J. Electrochem. Soc.* **163**, A2149 (2016)). In the atomic percentile analysis of the XPS spectra, the phosphorus (P) and nitrogen (N) levels were hardly detected, while the sulfur (S) was present at a very low atomic percentile (0.12% to 0.13%, Please see **Fig. R7** below). In addition, more detailed analysis of the S 2p was difficult due to its very low content (**Fig. R8**).

In response to the reviewer's comment, we deleted the CEI results and related discussion in the revised manuscript to avoid any unwanted misunderstanding. As suggested by the reviewer, the CEI will be further investigated in future studies. The reviewer's valuable comment is highly appreciated.

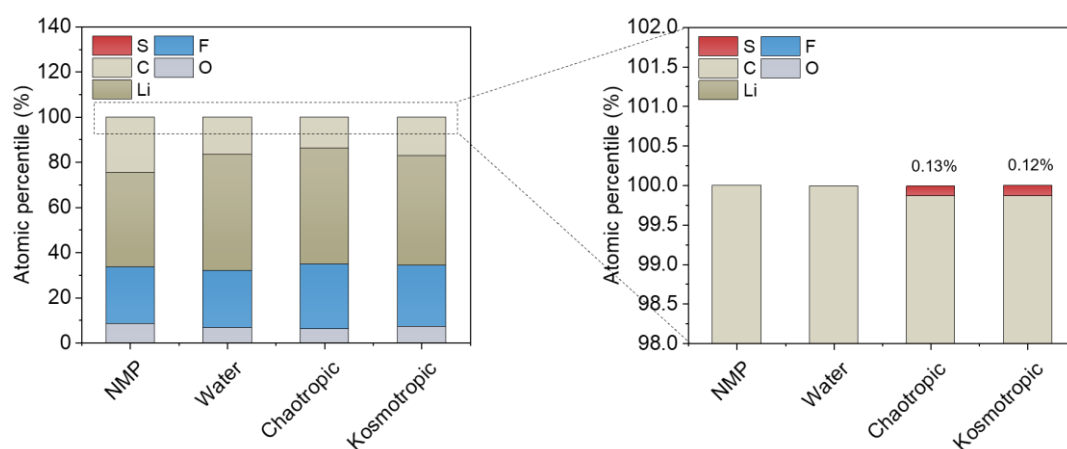


Fig. R7 | XPS elemental analysis of the surface of the cycled NCM811 cathodes (cycled at 0.2 C/0.5 C) processed with NMP, Water, chaotropic, and kosmotropic solution, respectively.

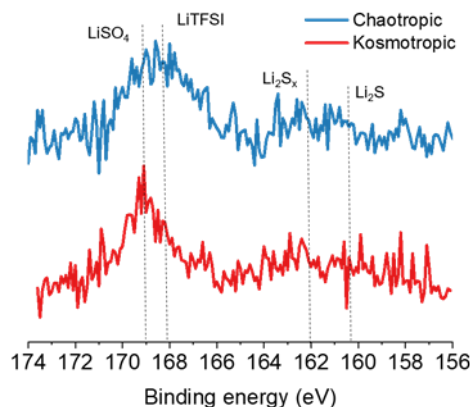


Fig. R8 | X-ray photoelectron spectroscopy (XPS) profiles of S2p of the cycled NCM811 cathodes.

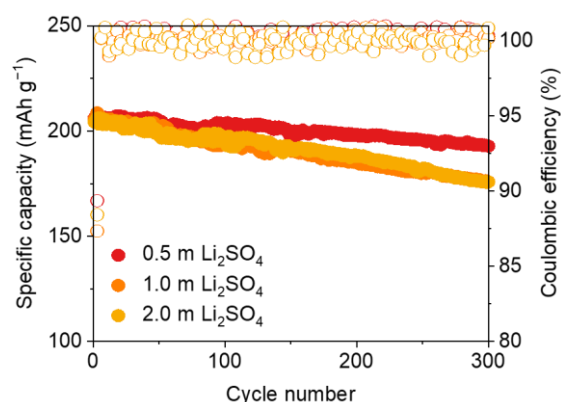
[Revised manuscript]

"The capacity of the water- and chaotropic solution-processed cathodes degraded significantly after 200 cycles because of the active Li^+/H^+ exchange^{5,39} during the aqueous cathode fabrication (Supplementary Fig. 15 and Fig. 5a). In contrast, the kosmotropic solution-processed cathode delivered a high initial specific capacity of $205 \text{ mAh g}_{\text{NCM811}}^{-1}$ with a stable cycle retention (over 80% after 400 cycles), which is comparable to that of the NMP-processed cathode. To elucidate the underlying electrochemical mechanisms responsible for this enhanced cycling stability, we analyzed the cathodes after 400 cycles using electrochemical impedance spectroscopy (EIS, Supplementary Fig. 16). The charge transfer resistance (R_{ct}) and the film resistance (R_{film}) of the kosmotropic solution-processed cathodes were comparable to those of the NMP-processed cathodes. Surface chemistry of the cathode-electrolyte interface (CEI) formed on the cycled cathodes were analyzed using X-ray photoelectron spectroscopy (XPS). No significant differences in the chemical compositions were observed (Supplementary Fig. 15). The observed congruence in the CEI chemical composition was attributed to the influence of the electrolyte during cycling. The vast disparities in the impedance, coupled with the consistent CEI composition, highlight the critical role of the chemical reactivity between water and cathode materials during cathode processing. Meanwhile, stable cycling performance (85% retention after 300 cycles) was also observed at higher Li_2SO_4 concentrations of 1.0 and 2.0 m (Supplementary Fig. 17)."

b–8. Lines 243–244: What are the possible reasons for a slightly poorer cycle performance of the higher Li₂SO₄-concentration samples than that prepared using 0.1 M Li₂SO₄?

→ **Response:** Thank you so much for the reviewer’s valuable comment. The slightly poor cycle retention at 1.0 and 2.0 m Li₂SO₄ may be attributed to the pore blockage of the precipitated salt. The addition of Li₂SO₄ in water is effective in mitigating the interfacial chemical reactions with NCM811, however, the excessive amount of Li₂SO₄ could block the pores of the resulting electrodes, resulting in an increase in the cell resistance. In response to the reviewer’s comment, additional explanation on this behavior was provided in the revised manuscript.

[Revised manuscript]

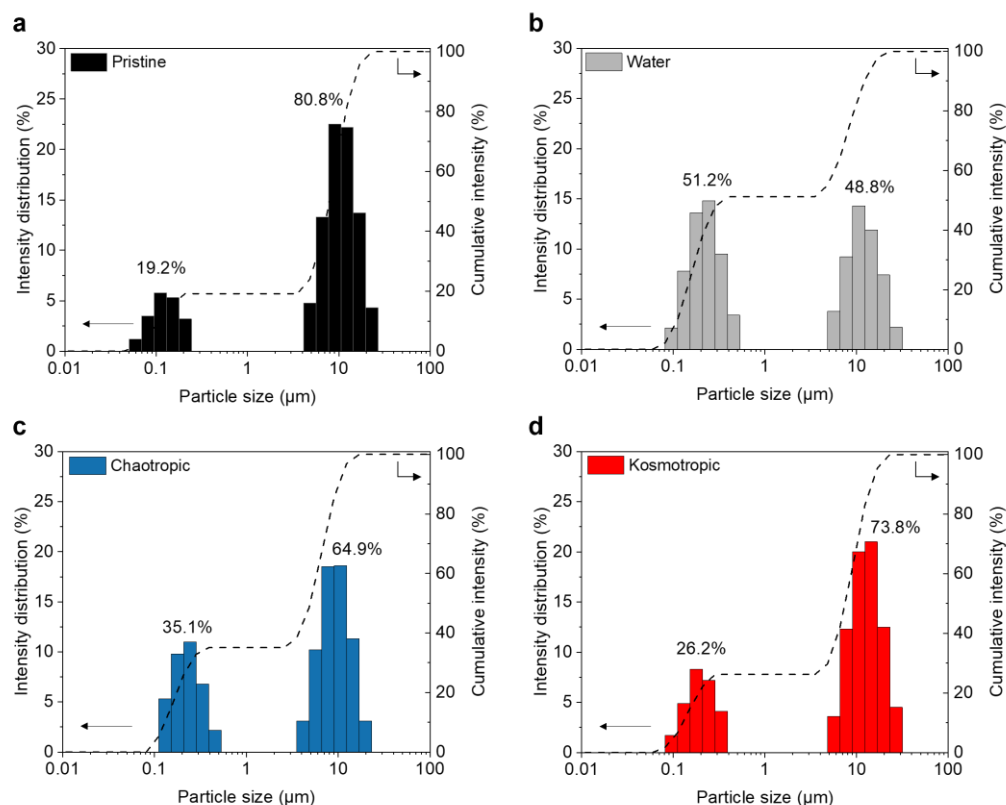


Supplementary Fig. 17 | Cycle retention of the kosmotropic solution-processed cathodes at a mass loading of **18 mg cm⁻²** as a function of the Li₂SO₄ concentration of the kosmotropic aqueous **processing** solution **at a current rate of 0.2 C/0.5 C (= 1.9 mA cm⁻²)**. The slightly poor cycle retention at 1.0 and 2.0 m Li₂SO₄ may be attributed to the pore blockage of the precipitated salt due to the excessive incorporation.

b–9. Lines 265–267 (and 275–276): I cannot fully agree with this sentence because this would also relate to the reduced particle size of NMC811 as speculated in Supplementary Fig. 10c. In addition, reduced particle sizes will increase the tortuosity of ion conduction, hence increasing R_{ion} . As stated in (b–3), the measurement of the particle size distribution of NMC811 after immersing the aqueous solutions with different anions is necessary.

→ **Response:** Many thanks for the reviewer’s valuable comment. In response to the reviewer’s comment, we quantitatively analyzed the particle size distribution of the NCM811 soaked in different processing solutions using a Zetasizer. The particle size of the resulting NCM811 tended to decrease in the following order: Pristine NCM811 \approx kosmotropic solution > chaotropic solution > water. This result was well consistent with the finding shown in **Supplementary Fig. 11** of the revised manuscript. We agree with the reviewer’s comment on the effect of the reduced particle size of NCM811. In response to the reviewer’s comment, we modified the corresponding sentence. Please see the revised manuscript. The reviewer’s valuable comment is highly appreciated, again.

[Revised manuscript]



Supplementary Fig. 11 | a-d, Particle size distribution of the pristine NCM811 particles (a),

and NCM811 particles after immersion in various processing solutions (b-d): water (b), chaotropic solution (c), and kosmotropic solution (d). The percentages indicate the proportion of primary and secondary particles present in each processing solution.

“The microstructural degradation of NCM811 was investigated using SEM (Supplementary Fig. 10). After immersion in the water or chaotropic solutions, the secondary NCM811 particles disintegrated into primary particles. In contrast, particles immersed in the kosmotropic solution remained relatively intact. This observation was quantitatively confirmed by analyzing particle size distribution using Zetasizer (Supplementary Fig. 11). The particle size tends to decrease in the order of NMP > kosmotropic solution > chaotropic solution > water. This finding demonstrates that the microstructural degradation of secondary NCM811 particles is ascribed to the undesired interfacial side reactions with water along the crystal grain boundaries³⁴.”

“Furthermore, the kosmotropic solution-processed cathode demonstrated superior discharge rate capability, delivering up to 188 mAh g⁻¹ under a fixed charge current density of 3.0 mA cm⁻², comparable to the NMP-processed cathode (Supplementary Fig. 19). In contrast, the chaotropic solution-processed cathodes exhibited inferior performance, which can be ascribed to the pore blockage as shown in the cross-sectional SEM images (Supplementary Fig. 20) and the reduced particle size of NCM811 that could increase the tortuosity of ion conduction (Supplementary Fig. 11).”

“The kosmotropic solution-processed cathode exhibited a low R_{ion} ($\sim 7.5 \Omega \text{ cm}^2$), closely resembling that of the water-processed cathode ($\sim 6.0 \Omega \text{ cm}^2$). In contrast, the chaotropic solution-processed cathode presented a significantly high R_{ion} at $\sim 80 \Omega \text{ cm}^2$, suggesting that the increased tortuosity stemming from the blocked pores and the reduced particle size of NCM811 further hinders ion transport.”

b–10. Lines 276–277 and 376: What is the theory behind the E/C ratio of 2 or 3 g Ah⁻¹? Does this ratio always allow the electrolyte solution to fill all the pores in the electrode layers and separator?

The amount of the electrolyte solution should be decided based on the total pore volume of the porous cell components. Can you please calculate the pore volumes of the NMC811 cathode and separator to see if the volume of the electrolyte solution surpasses the total pore volume of the porous cell components?

→ **Response:** Thank you so much for the reviewer’s insightful comment. In the submitted manuscript, the E/C ratio was determined by referring to previous papers (ref: *Nat. Nanotech.* **14**, 594 (2019), and *Nat. Energy* **6**, 495 (2021)), which suggested that an E/C ratio of 2–5 g Ah⁻¹ is recommended to closely mimic realistic cell conditions. In response to the reviewer’s comment, we calculated the pore volumes of the NMC811 cathode and separator. For the NMC811 electrode with a density of 3.0 g cc⁻¹ (area: 1.13 cm²), the theoretical pore volume is estimated to be 1.7 μL. For the PE separator with porosity of 47% (area: 2.54 cm²), the required pore volume is 1.9 μL. As a result, the total pore volume for the NMC811 cathodes and separator is approximately 5.3 μL. Meanwhile, in this study, an E/C ratio of 2 g Ah⁻¹ per electrode in symmetric cells corresponds to an electrolyte volume of approximately 16.7 μL, which is sufficient to fill the total pore volume of the cathode and separator. In response to the reviewer’s comment, the background on the E/C ratio and the pore volume data of the porous cell components were provided in the revised manuscript.

[Revised Manuscript]

Supplementary Table 6 | Calculation details on the pore volume of the electrode (a), and separator (b) used in this study.

a

Electrode	Material	Composition	Mass	True density	Volume	Volume
		[%]	[mg, 12 pi]	[g cc ⁻¹]	[cc, 12 pi]	[μL, 12 pi]
Active material	NCM811	95.5	20.61	4.3	0.0048	4.8

Conductive additive	Super P	2	0.43	1.6	0.00027	0.27
Binder	CMC	1.25	0.27	1.6	0.00017	0.17
	PAA	1.25	0.27	1.15	0.00023	0.23
Pore					0.0017	1.7
Sum			21.58		0.0071	
Electrode density			3.00			

b

Separator	Porosity [%]	thickness [μm]	Volume [cc, 18pi]	Volume [μL , 18 pi]
Polyethylene	47	16	0.0041	
Pore			0.0019	1.9

“Under the constrained amount ($\text{E/C ratio} = 2 \text{ g Ah}^{-1}$ per each electrode) of liquid electrolyte, which surpasses the total volume of the electrode and separator⁴⁰ (Supplementary Table 6). R_{ion} values appeared to be equilibrated after 6 h of aging, confirming complete wetting of the electrodes (Supplementary Fig. 21d). The kosmotropic solution-processed cathode exhibited a low R_{ion} ($\sim 7.5 \Omega \text{ cm}^2$), closely resembling that of the water-processed cathode ($\sim 6.0 \Omega \text{ cm}^2$).”

b–11. Lines 277–279: At least for Supplementary Fig. 20b, I do not think R_{ion} is stable from 0 to 12 h, which might disagree with this sentence. Please plot R_{ion} against the aging time to discuss this precisely.

→ **Response:** Many thanks for the reviewer’s valuable comments. In response to the reviewer’s comment, we have plotted R_{ion} against the aging time. This analysis reveals that R_{ion} appeared to be equilibrated after 6 h of aging (please see **Fig. R9** below). This result was added in the revised manuscript.

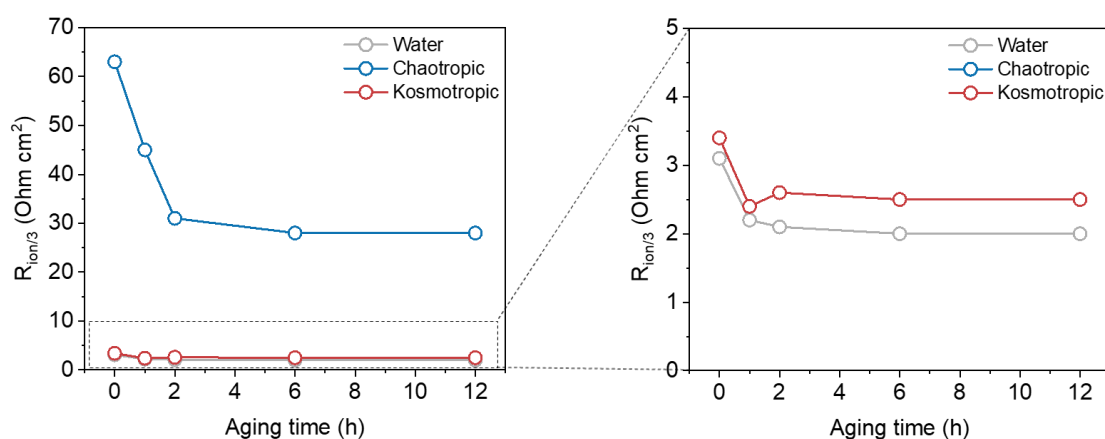
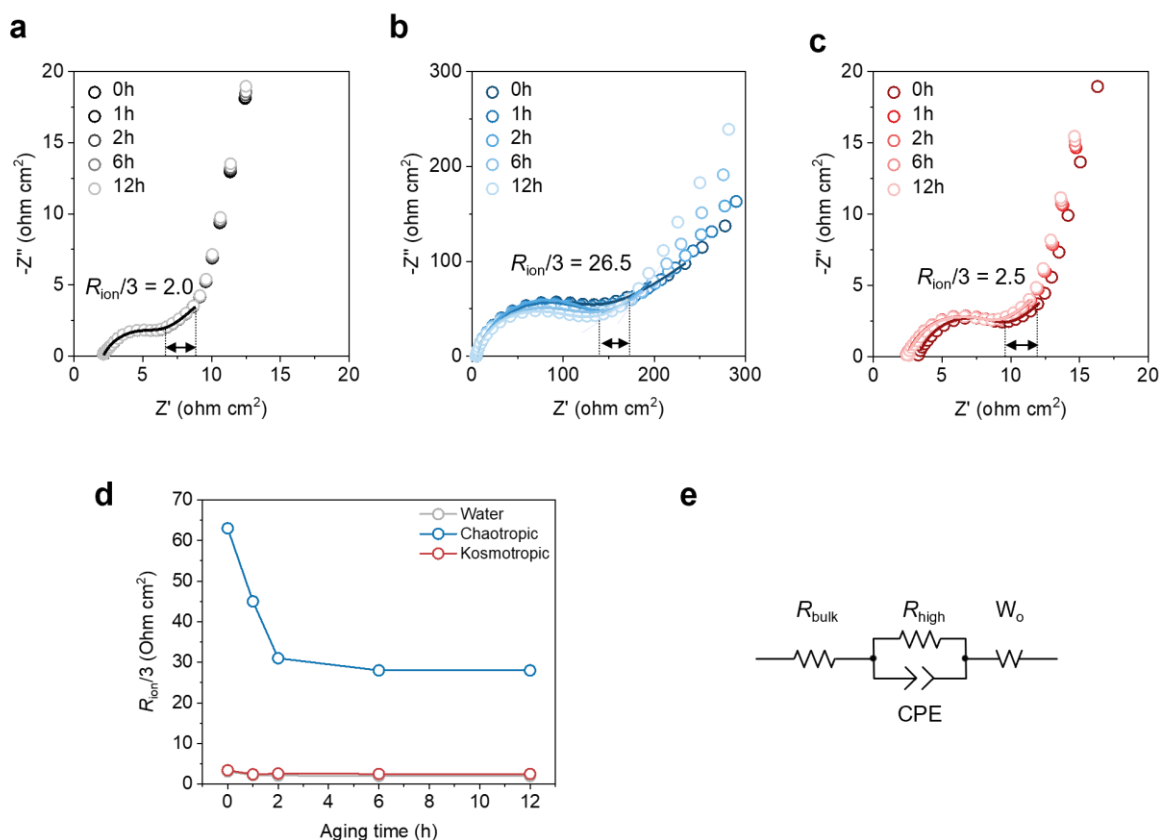


Fig. R9 | Ionic resistance inside of the electrodes as a function of the aging time.



Supplementary Fig. 21 | **a-c**, Nyquist plots of the water-processed (**a**), chaotropic solution-processed (**b**), and kosmotropic solution-processed (**c**) cathodes obtained using a symmetric cell configuration (electrode||electrode) as a function of the aging time under a limited amount of liquid electrolytes (electrolyte mass/electrode capacity (E/C) ratio of 2 g Ah⁻¹ **for each electrode**). The state of charge (SOC) of each electrode was set to 0% (fully-lithiated state). The symbol and solid lines represent experimental data and fitted curves based on a transmission line equivalent circuit model (TLM), respectively¹⁶. The projection of a slope to real axis (observed in the low-frequency region of Nyquist plot) reflects the ionic resistance inside the electrodes (R_{ion}) divided by 3. **d**, Change in the R_{ion} as a function of the aging time. **e**, Equivalent circuit model based on the generalized finite length Warburg element open circuit terminus (Z_w). W_0 was used to estimate R_{ion} ¹⁷.

“The ionic resistances of the electrodes (R_{ion}) were evaluated using EIS in a blocking-symmetric cell (electrode||electrode) configuration (**Supplementary Fig. 21**). Under the constrained amount (E/C ratio = 2 g Ah⁻¹ **per each electrode**) of liquid electrolyte, **which surpasses the total volume of the electrode and separator**⁴⁰ (**Supplementary Table 6**). R_{ion}

values appeared to be equilibrated after 6 h of aging, confirming complete wetting of the electrodes (**Supplementary Fig. 21d**).”

b–12. Fig. 5d: Are the charge/discharge curves in Fig. 5d measured at the first cycle? Please specify it.

→ **Response:** Thank you so much for the reviewer's valuable comment. The charge/discharge curves in **Fig. 5d** were measured at the first cycle after the formation. We have revised the figure caption to explicitly state this information.

[Revised Manuscript]

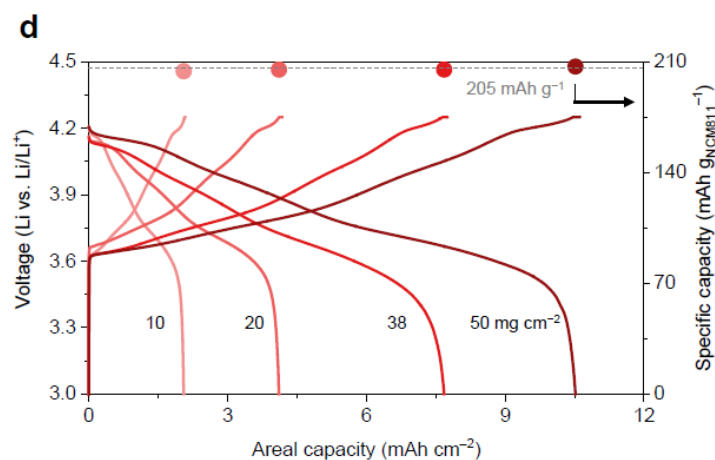


Fig. 5 | d, Galvanostatic **initial** charge/discharge profiles as a function of the mass loading of the **kosmotropic solution-processed** cathodes **at a current rate of 0.1C/0.1C**.

b–13. Fig. 5d: Gradual increase in the specific capacity with an increase in the areal capacity is surprising. How was it possible to realize? Were the experimental conditions for different areal capacities (e.g., electrode formulation, electrode density, applied currents during charging and discharging) the same as each other?

→ **Response:** Thank you so much for the reviewer’s detailed comments. In **Fig. 5d**, the specific capacity was not changed but stably maintained over the entire range of areal capacity. Please see the arrow on the upper right side. The all cathodes examined herein were prepared using the same fabrication process at the same composition ratio. In response to the reviewer’s comment, details on the cathode fabrication were added in the revised manuscript.

[Revised Manuscript]

“Fabrication of electrodes, cell, and electrochemical characterizations

The electrode slurries were prepared with a composition of $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2(\text{NCM811})/\text{carbon black}/\text{binder} = 95.5/2.25/2.25$ (w/w/w). Details of the electrode composition including residual salts are provided in **Supplementary Table 2**. For aqueous solution-processed electrodes, CMC and PAA were used as binders at a composition ratio of 1/1 (w/w). The salt additives were dissolved in water for 2 h and then the CMC and PAA binders were dissolved before the formulation of electrode slurries. The binder contents in the processing solutions are 3 wt.% and 10 wt.% for CMC and PAA, respectively. No additional water is added during the electrode fabrication, leaving 47 wt.% of water in the kosmotropic electrode slurry. The electrode slurries were cast onto Al current collectors and dried at 60°C for 6 h, followed by roll-pressing at 120°C. The electrode fabrication process was identical regardless of the mass loading and type of cathode materials. For the NMP-processed electrodes, PVDF was used as the binder, and dissolved in NMP. The concentration of the binder solution was set at 10 wt.%. The electrode slurries were cast onto Al current collectors and dried at 120°C for 6 h and then roll-pressed at 120°C. The density of the fabricated electrodes was set to 3.0 g cc⁻¹, and the electrodes were vacuum dried at 120°C for 12 h before the cell assembly.”

b–14. Fig. 5f and Line 293: What was the AM composition (%) in This work precisely? I thought it was 95.5% from Line 361 (and probably from the caption of Supplementary Table 5), but based on this figure, it looks like 92% and Supplementary Table 6 says another value: 93.1%. Please use the same value consistently.

→ **Response:** Many thanks for the reviewer’s valuable comment. The electrode composition ratio of this study is AM/carbon/binder (LiNi_{0.8}Co_{0.1}Mn_{0.1}O₂/carbon black/binder) = 95.5/2.0/2.5 (w/w/w) (**Supplementary Table 2a**). This composition ratio was provided without considering the salt additive to exclusively represent the AM content. Inclusion of the salt additive led to a change in the composition ratio to AM/carbon/binder/salt = 93.11/1.95/2.44/2.5 (w/w/w/w), as suggested in **Supplementary Table 2b** of the submitted manuscript. For a fair comparison with the previous works, the AM content of this study was suggested to be 93.1% (in **Fig. 5f** and **Supplementary Table 7**). In response to the reviewer’s comment, the X_{AM} (representing the AM content) was added along with the modified classification in **Fig. 5f**. The reviewer’s valuable comment is highly appreciated, again.

[Original manuscript]

Supplementary Table 2 | a, Composition of the electrode slurries containing 100 g of electrode components (cathode materials, binder, and conductive additive). **b**, Composition ratio of the aqueous-processed electrodes including that of the salt additives.

a

Processing solution	Cathode material [g]	Binder [g]	Conductive additive [g]	Salt additive [g]
H ₂ O	95.5	2.5	2.0	-
Chaotropic solution (1.0 m LiTFSI in H ₂ O)	95.5	2.5	2.0	11.5
Kosmotropic solution (0.5 m Li ₂ SO ₄ in H ₂ O)	95.5	2.5	2.0	2.6

b

Processing solution	Composition
---------------------	-------------

	[wt.%]		
	Cathode material	Binder & conductive additive	Salt additive
H ₂ O	95.5	4.5	-
Chaotropic solution (1.0 m LiTFSI in H ₂ O)	85.9	4.0	10.1
Kosmotropic solution (0.5 m Li ₂ SO ₄ in H ₂ O)	93.1	4.4	2.5

[Revised manuscript]

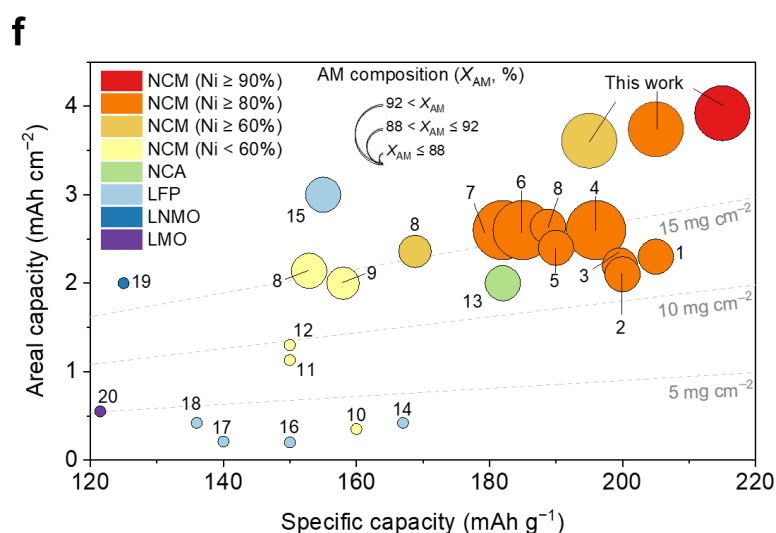


Fig. 5 | f, Comparison of kosmotropic solution-processed cathodes and previously reported aqueous solution-processed cathodes in terms of four parameters: specific capacity of cathode materials (x -axis), areal capacity (y -axis), types of cathode materials (heatmap), and cathode material composition (X_{AM}) of the electrode (diameter). The gray dashed lines represent mass loading of cathode materials. The number assigned to each circle corresponds to the serial number in **Supplementary Table 7** (ref.^{5,10,11,39,48-63}).

“Fabrication of electrodes, cell, and electrochemical characterizations

The electrode slurries were prepared with a composition of $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (NCM811)/carbon black/binder = 95.5/2.25/2.25 (w/w/w). **Details of the**

electrode composition including residual salts are provided in **Supplementary Table 2**. For aqueous solution-processed electrodes, CMC and PAA were used as binders at a composition ratio of 1/1 (w/w). The salt additives were dissolved in water for 2 h and then the CMC and PAA binders were dissolved before the formulation of electrode slurries. The binder contents in the processing solutions are 3 wt.% and 10 wt.% for CMC and PAA, respectively. No additional water is added during the electrode fabrication, leaving 47 wt.% of water in the kosmotropic electrode slurry. The electrode slurries were cast onto Al current collectors and dried at 60°C for 6 h, followed by roll-pressing at 120°C. The electrode fabrication process was identical regardless of the mass loading and type of cathode materials. For the NMP-processed electrodes, PVDF was used as the binder, and dissolved in NMP. The concentration of the binder solution was set at 10 wt.%. The electrode slurries were cast onto Al current collectors and dried at 120°C for 6 h and then roll-pressed at 120°C. The density of the fabricated electrodes was set to 3.0 g cc⁻¹, and the electrodes were vacuum dried at 120°C for 12 h before the cell assembly.”

b–15. Line 359 and thereafter: Please specify how the aqueous solutions containing different Li salts were used for slurry preparation. At this stage, readers cannot understand when and how the Li salts were added.

→ **Response:** Thank you so much for the reviewer’s valuable comment. For the preparation of the aqueous processing solution containing different Li salts, the salt additives were dissolved in water for 2 h and then the CMC and PAA binders were dissolved prior to the formulation of electrode slurries. The binder contents in the processing solutions are 3 wt.% and 10 wt.% for CMC and PAA, respectively. No additional water is added during the electrode fabrication, leaving 47 wt.% of water in the kosmotropic electrode slurry. In response to the reviewer’s comment, the detailed procedure for the aqueous processing solutions containing different Li salts was added in the revised manuscript.

[Revised manuscript]

“Fabrication of electrodes, cell, and electrochemical characterizations

The electrode slurries were prepared with a composition of $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2(\text{NCM811})/\text{carbon black}/\text{binder} = 95.5/2.25/2.25$ (w/w/w). Details of the electrode composition including residual salts are provided in Supplementary Table 2. For aqueous solution-processed electrodes, CMC and PAA were used as binders at a composition ratio of 1/1 (w/w). The salt additives were dissolved in water for 2 h and then the CMC and PAA binders were dissolved before the formulation of electrode slurries. The binder contents in the processing solutions are 3 wt.% and 10 wt.% for CMC and PAA, respectively. No additional water is added during the electrode fabrication, leaving 47 wt.% of water in the kosmotropic electrode slurry. The electrode slurries were cast onto Al current collectors and dried at 60°C for 6 h, followed by roll-pressing at 120°C. The electrode fabrication process was identical regardless of the mass loading and type of cathode materials. For the NMP-processed electrodes, PVDF was used as the binder, and dissolved in NMP. The concentration of the binder solution was set at 10 wt.%. The electrode slurries were cast onto Al current collectors and dried at 120°C for 6 h and then roll-pressed at 120°C. The density of the fabricated electrodes was set to 3.0 g cc⁻¹, and the electrodes were vacuum dried at 120°C for 12 h before the cell assembly.”

b–16. Supplementary Table 3: Please specify how the cost information about each material was obtained. Any reference?

→ **Response:** Many thanks for the reviewer’s insightful comment. The material costs were estimated based on commercially available batch-level prices provided by their suppliers. Detailed information on the materials, including their sources and prices, was included in the Methods section. The reviewer’s comment is highly appreciated, again.

[Revised manuscript]

“Material preparation

Lithium sulfate (Li_2SO_4 , 99.5%), Lithium acetate (LiOAc , 99.95%), Lithium trifluoromethanesulfonate (LiOTf , 96.0%), N-methyl-2-pyrrolidone (NMP, 99.5%), carboxymethyl cellulose (CMC, $M_w = 250,000$, degree of substitution = 0.9), and polyacrylic acid (PAA, $M_w = 45,000$, purity) were purchased by Sigma-Aldrich. Li bis(trifluoromethanesulfonyl)imide (LiTFSI , 99.0%) and polyvinylidene fluoride (PVDF) were bought from Samchun and Solvay, respectively. 1 M lithium hexafluorophosphate (LiPF_6) in ethylene carbonate (EC)/diethyl carbonate (DEC) (= 1/1 (v/v)), fluoroethylene carbonate (FEC, 99.9%), and vinyl carbonate (VC, 99.9%) were purchased by Enchem (Korea). A water purification system (Direct Q–5UV, Merck Millipore) was utilized to prepare deionized water for aqueous processing solutions. Al current collectors were purchased from MTI (China). The material cost information used in **Supplementary Tables 3–5** was obtained from the suppliers described above.”

Supplementary Table 3 | a,b, Cost information of materials (a) and processing solvents (or solutions) (b). Cost of the aqueous processing solutions was determined at an equimolar Li^+ concentration of 1.0 m in H_2O . **The material cost information was obtained from their suppliers.**

b–17. Supplementary Table 4: Please specify how the cost information about each electrode component was obtained. Any reference?

→ **Response:** Thank you so much for the reviewer's valuable comment. Our reply was the same as that provided in the comment b-16.

[Revised manuscript]

“**Material preparation**

Lithium sulfate (Li_2SO_4 , 99.5%), Lithium acetate (LiOAc , 99.95%), Lithium trifluoromethanesulfonate (LiOTf , 96.0%), N-methyl-2-pyrrolidone (NMP, 99.5%), carboxymethyl cellulose (CMC, $M_w = 250,000$, degree of substitution = 0.9), and polyacrylic acid (PAA, $M_w = 45,000$, purity) were purchased by Sigma-Aldrich. Li bis(trifluoromethanesulfonyl)imide (LiTFSI , 99.0%) and polyvinylidene fluoride (PVDF) were bought from Samchun and Solvay, respectively. 1 M lithium hexafluorophosphate (LiPF_6) in ethylene carbonate (EC)/diethyl carbonate (DEC) (= 1/1 (v/v)), fluoroethylene carbonate (FEC, 99.9%), and vinyl carbonate (VC, 99.9%) were purchased by Enchem (Korea). A water purification system (Direct Q–5UV, Merck Millipore) was utilized to prepare deionized water for aqueous processing solutions. Al current collectors were purchased from MTI (China). The material cost information used in **Supplementary Tables 3–5** was obtained from the suppliers described above.”

Supplementary Table 4 | **a**, Cost information of the **cathode** components. **Material specifications are described in Methods.** **b**, Calculation details of the specific capacity based on the material cost of cathodes **and processing solutions, which are normalized by electrode area of 1 cm².** The material cost information was obtained from their suppliers.

b–18. Supplementary Table 4: In addition, I think the Al current collector should be included here as well as in the cost estimation

→ **Response:** Thank you very much for the reviewer’s valuable comment. In response to the reviewer's comment, we have revised the **Supplementary Table 4** to include the cost of the Al current collector. Again, the reviewer's kind comment is greatly appreciated.

[Revised manuscript]

Supplementary Table 4 | **a**, Cost information of the **cathode** components. **Material specifications are described in Methods.** **b**, Calculation details of the specific capacity based on the material cost of cathodes **and processing solutions, which are normalized by electrode area of 1 cm².** The material cost information was obtained from their suppliers.

a

Component	Material	Weight [g]	Cost [USD]	Specific cost [USD g ⁻¹]
Cathode material	NCM811	500	345	0.69
Binder	PAA	250	407	1.63
	CMC	1,000	168	0.17
	PVdF	100	120	1.20
Conductive additive	SP-90	400	214	0.54

Component	Material	Area [m ²]	Cost [USD]	Areal cost [USD m ⁻²]
Current collector	Al	98	295	3.01

b

Cathodes (prepared using the following processing solution)	Areal capacity [mAh cm ⁻²]	Cathode cost [USD]	Solution cost [USD]	Capacity normalized by material cost [mAh USD ⁻¹]
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NMP	3.74	0.01367	0.00481	202
H ₂ O	3.61	0.01352	0.00005	266
Chaotropic solution (1.0 m LiTFSI in H ₂ O)	3.59	0.01352	0.01192	141
Kosmotropic solution (0.5 m Li ₂ SO ₄ in H ₂ O)	3.74	0.01352	0.00019	273

b–19. Supplementary Table 4b: Why does the cost information about the solution differ from that in Supplementary Table 3b? (For instance, the cost of NMP in Supplementary Table 4b is 10.74 times higher than that in Supplementary Table 3b, but the cost of 1.0 m LiTFSI in H₂O in Supplementary Table 4b is 12.87 times higher than that in Supplementary Table 3b, i.e., no proportional increase)

→ **Response:** We appreciate the reviewer’s valuable comment. I’m afraid that the reviewer’s comment may arise from our careless description in the Tables. In response to the reviewer’s comment, the description shown in **Supplementary Table 4b** was changed from “Processing solution” to “Cathodes (prepared with the following processing solution)”. In addition, the other descriptions were also modified. The reviewer’s valuable comment is highly appreciated, again.

[Revised manuscript]

Supplementary Table 4 | a, Cost information of the cathode components. **Material specifications are described in Methods.** **b,** Calculation details of the specific capacity based on the material cost of cathodes and processing solutions, which are normalized by electrode area of 1 cm². The material cost information was obtained from their suppliers.

a

Component	Material	Weight [g]	Cost [USD]	Specific cost [USD g ⁻¹]
Cathode material	NCM811	500	345	0.69
Binder	PAA	250	407	1.63
	CMC	1,000	168	0.17
	PVdF	100	120	1.20
Conductive additive	SP-90	400	214	0.54
Component	Material	Area [m ²]	Cost [USD]	Areal cost [USD m ⁻²]
Current collector	Al	98	295	3.01

b

Cathodes (prepared using the following processing solution)	Areal capacity [mAh cm ⁻²]	Cathode cost [USD]	Solution cost [USD]	Capacity normalized by material cost [mAh USD ⁻¹]
NMP	3.74	0.01367	0.00481	202
H ₂ O	3.61	0.01352	0.00005	266
Chaotropic solution (1.0 m LiTFSI in H ₂ O)	3.59	0.01352	0.01192	141
Kosmotropic solution (0.5 m Li ₂ SO ₄ in H ₂ O)	3.74	0.01352	0.00019	273

c-1. Lines 32 and 87: Please revise “approximately 45%” to “46%” to be exact.

→ **Response:** We appreciate the reviewer’s detailed comment. In response to the reviewer’s comment, the corresponding sentence was modified in the revised manuscript.

[Revised manuscript]

“Techno-economic analysis demonstrated that this kosmotropic solution approach reduced energy consumption in battery manufacturing by 46% compared to the NMP-based process, highlighting its practical and sustainable viability.”

“From a comprehensive techno-economic analysis, we propose that the use of the kosmotropic aqueous **processing** solution can reduce the operational (~ 23%), and capital (~ 95%) costs associated with cathode manufacturing **by eliminating the conventional solvent recovery process.** Furthermore, this approach substantially reduced energy consumption in electrode manufacturing by 46%, demonstrating its viability as a platform technology for environmentally friendly and cost-competitive electrode manufacturing applicable to various high-capacity electrode materials.”

c-2. Line 73: Please add the reference(s) to describe some examples of using the kosmotropic effect in biochemistry.

→ **Response:** Thank you so much for the reviewer's insightful comment. In response to the reviewer's comment, we added the relevant references that reported the kosmotropic effect in biochemistry.

[Revised manuscript]

“Herein, we introduce a class of aqueous **processing** solutions based on the kosmotropic effect, a phenomenon common in biochemistry^{12,13} but rarely applied to batteries.”

“Reference

12 Cacace, M., Landau, E. & Ramsden, J. The Hofmeister series: salt and solvent effects on interfacial phenomena. *Q. Rev. Biophys.* **30**, 241-277 (1997).

13 Jungwirth, P. & Cremer, P. S. Beyond hofmeister. *Nat. Chem.* **6**, 261-263 (2014).”

c-3. Line 76: Please add “, right” after “Fig. 1a”.
--

→ **Response:** Thank you so much for the reviewer’s detailed comment. In response to the reviewer’s comment, the corresponding sentence was modified in the revised manuscript.

[Revised manuscript]

“**Incorporating** the kosmotropic anions resulted in the restructuring of the ion hydration shell to form a highly ordered state of anion–water clusters and the regulation of the local hydration structure adjacent to cathode materials (**Fig. 1a, right**).”

c-4. Line 100: I guess “charge-delocalized” needs to be “charge-localized”. Please double-check it.

→ **Response:** Many thanks for the reviewer’s valuable comment. In response to the reviewer’s comment, we have carefully double-checked the term “charge-delocalized”. The term “charge-delocalized” is used to describe chaotropic ions in the Hofmeister series, as these ions are characterized by their charge-delocalized properties. Please refer to the figures and discussions in the following references for more detailed information. In response to the reviewer’s comment, we added the relevant references in the revised manuscript to clarify the term “charge-delocalized”.

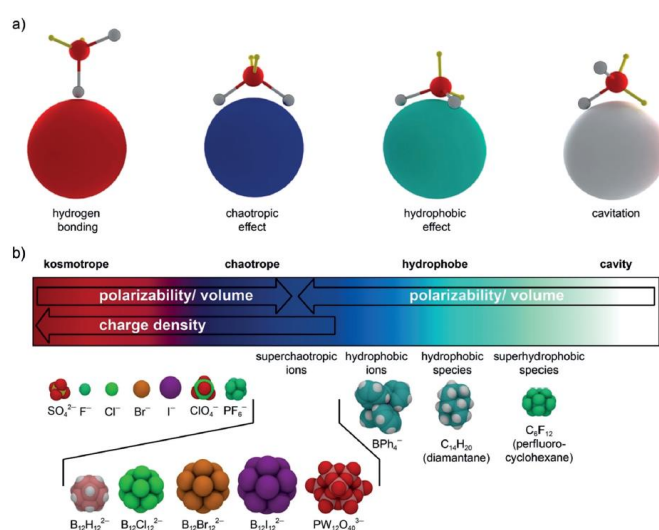


Figure 1. Principal aqueous solvation patterns for anions. a) Presumed favorable orientations of water molecules around a cavity and at the surface of different solutes: kosmotropic and chaotropic ions, hydrophobic molecules, and void space. Electron lone pairs are visualized in yellow. b) Extended Hofmeister scale with specification of the superchaotropic, hydrophobic ionic, and superhydrophobic regions; see Figure S1 in the Supporting Information for a two-dimensional illustration of the solvation pattern dependence on charge density and polarizability.

Chaotropic ions are typically large and charge-delocalized. The positioning of a chaotropic ion in the cavity causes less directional ion–dipole interactions to predominate, which results in a lateral orientation of the water molecules under alignment of their dipole moment (Figure 1 a, second from the left). The positioning of a hydrophobic solute in the cavity

Fig. | A captured image from a research article published in *Angew. Chem. Int. Ed.* 57, 13968–13981 (2018), “The Chaotropic Effect as an Assembly Motif in Chemistry”.

tural entropies.⁵⁶ The chaotropicity of anions increases generally with size and charge delocalization; large cluster anions have been referred to as being superchaotropic,^{7,8} because they are more chaotropic than SCN^- , ClO_4^- , and PF_6^- on the classical Hofmeister scale.^{59–62}

Figure. A captured image from a research article published in *J. Am. Chem. Soc.* 145, 13089–13098 (2023), “Metallacarborane Cluster Anions of the Cobalt Bisdicarbollide-Type as Chaotropic Carriers for Transmembrane and Intracellular Delivery of Cationic Peptides”.

[Revised manuscript]

“Ions, as categorized by the Hofmeister series, can influence the molecular structure and dynamics of water by altering the strength of its H-bond network¹⁶. The kosmotropic ion exhibits a high charge density and can strengthen the H-bond structure¹⁷. On the other side of the series, the chaotropic ion has a large and charge-delocalized molecular structure¹⁸, exhibiting weaker intermolecular interactions with water and disordering the H-bond networks.”

“Reference

- 18 Assaf, K. I. & Nau, W. M. The chaotropic effect as an assembly motif in chemistry. *Angew. Chem. Int. Ed.* **57**, 13968-13981 (2018).”

c-5. Fig. 2a: For TFSI⁻, the ESP value for O atoms would need to be provided as these are the sites for hydrogen bonding. (The other three anions have provided the ESP values for O atoms, but TFSI⁻ has not provided it.)

→ **Response:** Thank you so much for the reviewer's valuable comment. The ESP values shown in **Fig. 2a** indicate the regions of highest and lowest electron density of the respective anions. As the reviewer mentioned, the ESP value for O atoms in TFSI⁻ should be included to precisely examine hydrogen bonding capabilities. In response to the reviewer's comment, we included the ESP values (−0.157 Ha) for O atoms in TFSI⁻ in the revised manuscript. The reviewer's valuable comment is highly appreciated, again.

[Revised manuscript]

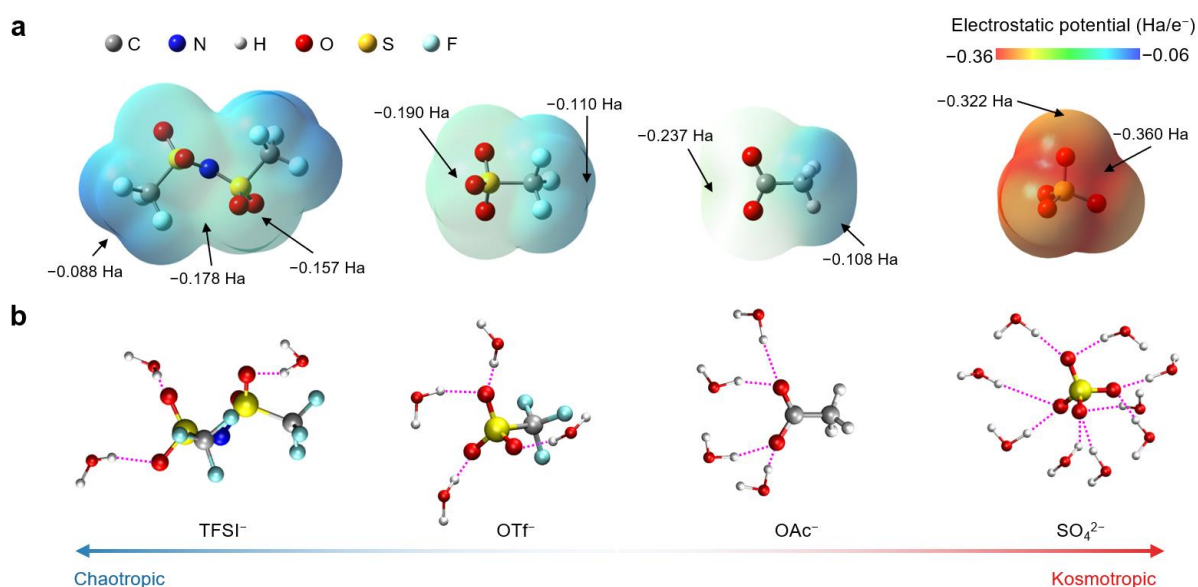


Fig. 2 | Reorganization of ion hydration shell by Hofmeister effect. a,b, Electrostatic potential maps (a) and hydration structures (b) of different anions (TFSI⁻, OTf⁻, OAc⁻, and SO₄²⁻) with varying chaotropicity and kosmotropicity. Gray, blue, white, red, yellow, and cyan spheres represent carbon, nitrogen, hydrogen, oxygen, sulfur, and fluorine, respectively.

c-6. Line 219: Here, “tap densities” should be “electrode density” and “areal-mass-loading” can be “areal mass loading” (or “areal NMC811 loading”).

→ **Response:** Thank you so much for the reviewer’s valuable comments. In response to the reviewer’s comment, the terms were modified in the revised manuscript.

[Revised manuscript]

“To test the aqueous processed cathodes, we prepared cells comprising of Li metal anodes and NMC811 cathodes with **areal mass loading** and **electrode density** densities of **18** mg_{NMC811} cm⁻² and 3.0 g cc⁻¹, respectively (See Methods and **Supplementary Table 2** for details).”

c-7. Line 231: Please specify at which cycle number the specific capacity of 205 mAh/g was observed. (I presume it was observed at the first cycle.)

→ **Response:** Thank you so much for the reviewer's valuable comments. The specific capacity of 205 mAh/g was observed at the first cycle after the formation. In response to the reviewer's comment, the cycle number for the specific capacity of 205 mAh/g was provided in the revised manuscript.

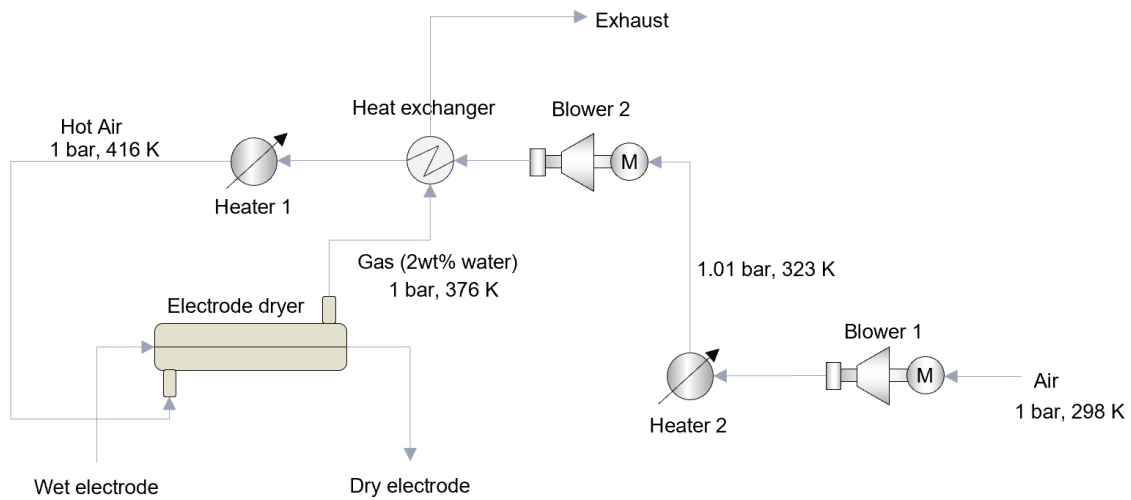
[Revised manuscript]

“In contrast, the kosmotropic solution-processed cathode delivered a high **initial** specific capacity of 205 mAh g_{NCM811}⁻¹ with a stable cycle retention **(over 80% after 400 cycles)**, which is comparable to that of the NMP-processed cathode.”

c–8. Fig. 6a and Supplementary Fig. 22: I think “Blower 1” and “Blower 2” should be swapped in one of the figures to match the numbering.

→ **Response:** We appreciate for the reviewer’s detailed comments In response to reviewer’s comment, the corresponding expression in **Supplementary Fig. 23** was modified in the revised manuscript.

[Revised manuscript]



Supplementary Fig. 23 | Process flow diagram of the electrode manufacturing based on kosmotropic aqueous **processing** solution.

c-9. Lines 378–379: Was EIS conducted from the lowest frequency to the highest frequency? Please specify the direction of the measurement.

→ **Response:** Thank you so much for the reviewer's detailed comment. In this study, the EIS analysis was conducted from the highest frequency (10^6 Hz) to the lowest frequency (10^{-2} Hz). In response to the reviewer's comment, the corresponding sentence was modified in the revised manuscript to clearly specify the measurement.

[Revised manuscript]

“The resistances of the electrodes were measured using electrochemical impedance spectroscopy (EIS) analysis in a frequency range that varies from 10^6 to 10^{-2} Hz and an applied amplitude of 10 mV using potentiostat/galvanostat (VSP-300, Bio-Logic).”

Additional comments. Finally, the authors should consider the recently published papers by Mecerreyes et al using aqueous binders which contain anions, which could be considered kosmotropic. Could the authors comment on this with respect to their analysis presented in the manuscript?

Eg. : Fluorine-Free Poly (ionic Liquid) s Binders for the Aqueous Processing of High-Voltage NMC811 Cathodes; AC Rolandi, C Pozo-Gonzalo, I de Meatza, N Casado, D Mecerreyes, ...

Advanced Energy and Sustainability Research 4 (12), 2300149; Carrageenans as Sustainable Water-Processable Binders for High-Voltage NMC811 Cathodes AC Rolandi, C Pozo-Gonzalo, I de Meatza, N Casado, M Forsyth, ...ACS Applied Energy Materials 6 (16), 8616-8625

→ **Response:** Thank you so much for the reviewer's valuable comments. The paper by Mecerreyes et al. investigated the use of aqueous binders for the aqueous processing of the NMC811 cathodes. They reported that the binders containing the anionic groups improved the electrochemical performance due to their enhanced ionic conductivity. For example, the improved cycling performance of the aqueous-processed cathodes with phosphate binders may indicate that the phosphate anion mitigates the side reaction between water and electrode active materials. Additionally, the SEM analysis supported this hypothesis by showing that the aqueous-processed electrode with the phosphate binder can inhibit the formation of bubbles on the electrode surface, which is typically a result of hydrogen gas production from the side reactions, as referenced in *ChemSusChem*. **11**, 5562 (2018).

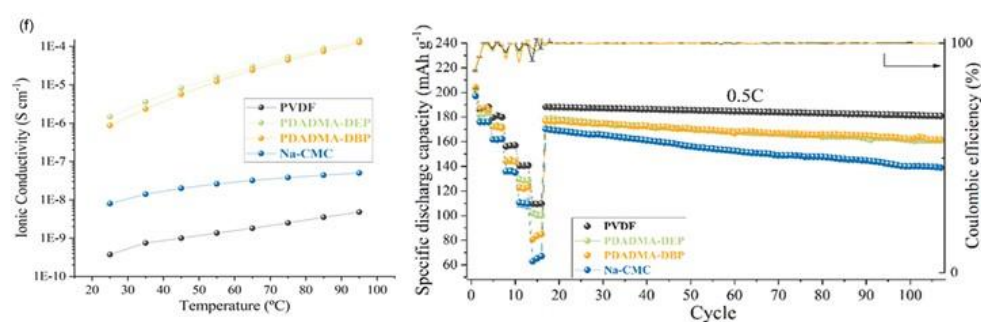


Fig. | A captured image from a research article published in *Adv. Energy Sustain. Res.* **4**, 2300149 (2023) “Fluorine-Free Poly (ionic Liquid) s Binders for the Aqueous Processing of

High-Voltage NMC811 Cathodes”. The ionic conductivity of various polymers (left), rate and cycle performance of cathodes containing respective polymer as binder.

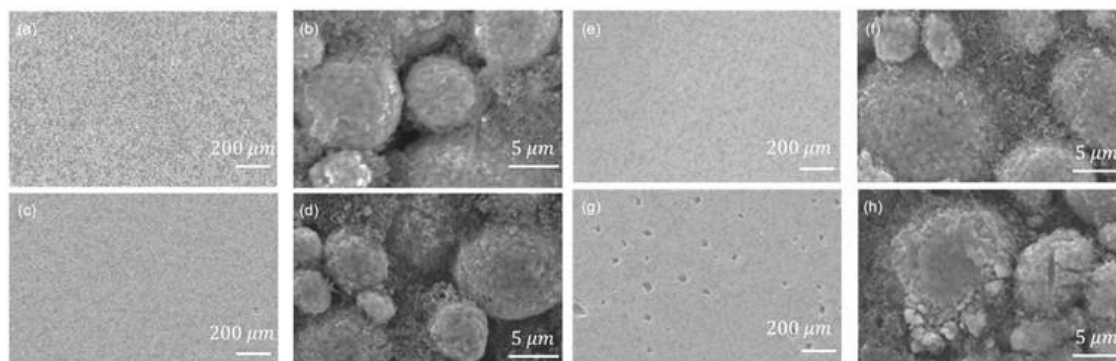


Fig. | A captured image from a research article published in *Adv. Energy Sustain. Res.* **4**, 2300149 (2023) “Fluorine-Free Poly(ionic Liquid)s Binders for the Aqueous Processing of High-Voltage NMC811 Cathodes”. SEM image of electrodes fabricated using PVDF (**a-b**), PDADMA-DEP (**c-d**), PDADMA-DBP (**e-f**), CMC (**g-h**) binder.

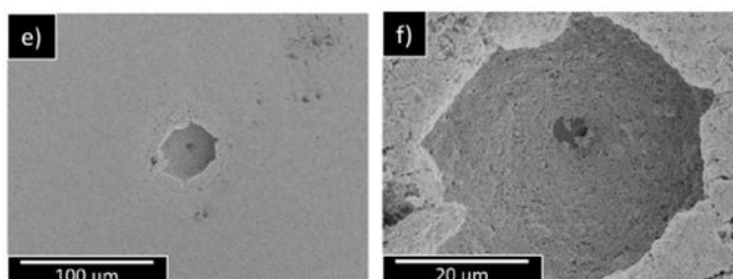


Fig. | A captured image from a research article published in *ChemSusChem.* **11**, 5562 (2018) “Complementary Strategies Toward the Aqueous Processing of High-Voltage $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ Lithium-Ion Cathodes”. SEM image of aqueous processed electrode surface

The other paper (ref.: *ACS Appl. Energy Mater.* **6**, 8616 (2023)) reported that the binders containing sulfonate groups exhibited the enhanced cycling performance with higher degree of sulfonate substitution. It can be suggested that the increased interactions between the sulfonate groups and water molecules may alleviate the reactivity of water, thereby improving the electrochemical stability of the resulting electrode.

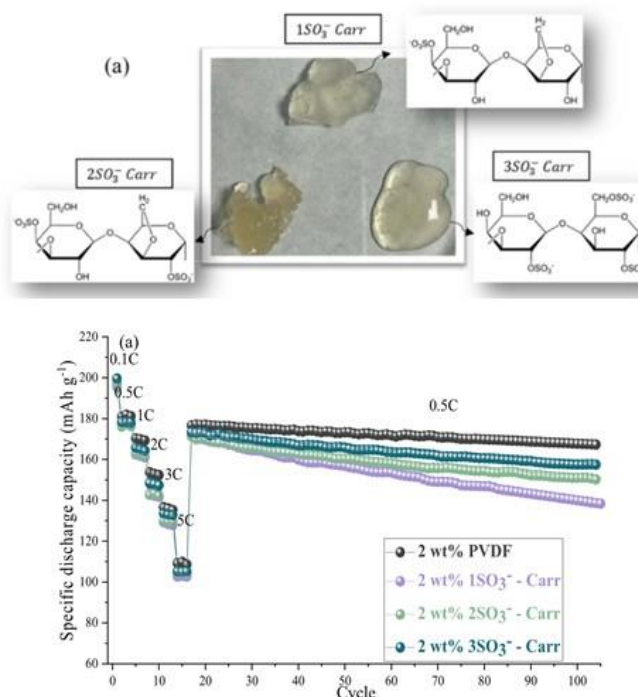


Fig. | A captured image from a research article published in *ACS Appl. Energy Mater.* **6**, 8616 (2023) “Carrageenans as Sustainable Water-Processable Binders for High-Voltage NMC811 Cathodes”. Photograph of polymer solution and their chemical structures, showing the different number of sulfonate groups (top). Rate and cycle performance of cathodes containing respective polymer as binder (bottom).

These previous papers mentioned above showed the potential of kosmotropic groups to stabilize water-based electrode fabrication processes through the use of functional binders. However, the binder content was of concern, as it was not sufficient to effectively suppress the interfacial side reactions between water and electrode active materials. In response to the reviewer’s comment, we have cited these previous papers in the revised manuscript and included a comparative analysis in **Fig. 5f** and **Supplementary Table 7**. Additionally, we have expanded our discussion on the theoretical implications of kosmotropic groups on other electrode components to highlight their broader potential impact.

[Revised manuscript]

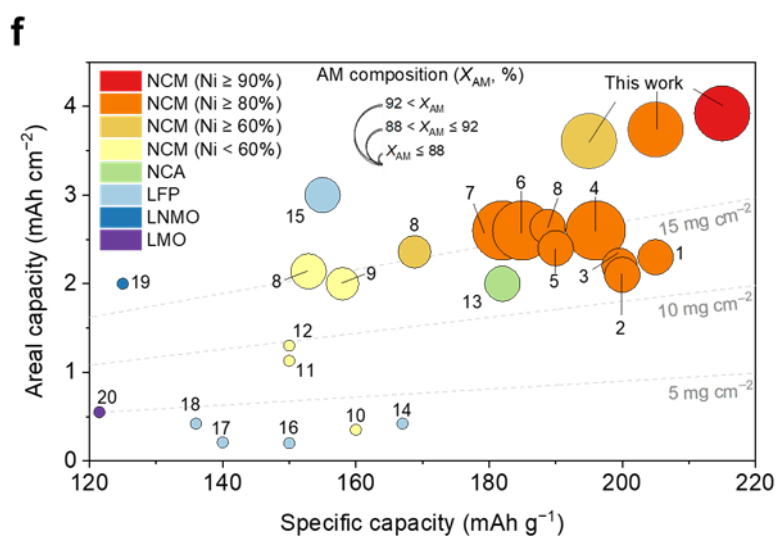


Fig. 5 | f, Comparison of kosmotropic solution-processed cathodes and previously reported aqueous solution-processed cathodes in terms of four parameters: specific capacity of cathode materials (x -axis), areal capacity (y -axis), types of cathode materials (heatmap), and cathode material composition (X_{AM}) of the electrode (diameter). The gray dashed lines represent mass loading of cathode materials. The number assigned to each circle corresponds to the serial number in **Supplementary Table 7** (ref.^{5,10,11,39,48-63}).

Supplementary Table 7 | Comparison of the kosmotropic solution-processed cathodes and previously reported aqueous solution-processed cathodes.

No.	Processing solution	Cathode material	Binder	Specific capacity [mAh g ⁻¹]	Areal capacity [mAh cm ⁻²]	AM composition [wt. %]	Ref.
This work	0.5 m Li ₂ O ₄ in H ₂ O	NCM9½½	CMC/PAA	215	3.9	93.1	This work
		NCM811	CMC/PAA	205	3.7, 7.8, 10.2		
		NCM622	CMC/PAA	195	3.6		
1	H ₂ O	NCM811	CMC/Acrylic emulsion	205	2.3	90	22
2	H ₂ O	NCM811	Carrageenan	200	2.2	90	23
3	H ₂ O	NCM811	PDADMA-DEP (or DBP)	200	2.1	90	24
4	H ₂ O	NCM811	SBR/CMC	196	2.6	94	25
5	Li ₂ SO ₄ in H ₂ O	NCM811	CMC	190	2.4	92	26
6	H ₂ O	NCM811	CMC/PAA	185	2.6	93	27
7	H ₂ O	NCM811	CMC/ICN	182	2.6	93	28
		NCM811		189	2.6	90	
		NCM622	CMC	169	2.4	90	
8	H ₂ O	NCM111		153	2.1	90	29
		NCM523	CMC/PAA/PEO	158	2	91	
9	LiOH in H ₂ O	NCM523	CMC/PAA/PEO	158	2	91	30
10	H ₂ O	NCM424	CMC	160	0.4	80	31

11	H ₂ O	NCM111	CMC	150	1.1	88	32
12	H ₃ PO ₄ in H ₂ O	NCM111	CMC	150	1.3	88	33
13	H ₂ O	NCA	CMC	182	2	92	34
14	H ₂ O	LFP	Xanthan gum	167	0.4	87	35
15	H ₂ O	LFP	NBR/CMC	155	3	90	36
16	H ₂ O	LFP	PAA	150	0.2	83	37
17	H ₂ O	LFP	CMC	140	0.2	85	38
18	H ₂ O	LFP	PEO	136	0.4	73	39
19	LiTFSI in H ₂ O	LNMO	CMC/SBR	125	2	71	14
20	H ₂ O	LMO	Alginate	122	0.6	75	40

“Conclusion

The widespread use of the hazardous chemical NMP in LIB cathode manufacturing has raised significant environmental concerns. To address this imperative issue, we designed a kosmotropic aqueous **processing** solution to mitigate the adverse interfacial chemical reactions between water and cathode materials by forming highly ordered ion hydration shells. The kosmotropic solution-processed cathodes exhibited a stable cycle life comparable to that of the NMP-processed cathode under practical conditions and achieved their theoretical specific capacity at high areal loadings. By conducting the techno-economic analysis, we demonstrated the superiority of the kosmotropic aqueous **processing** solution over NMP in electrode manufacturing, achieving significant reductions in energy consumption, CO₂ emissions, and cost savings for sustainable cathode production. **We anticipate that this kosmotropic solution-processed cathode approach could be extended to other electrode active materials and battery chemistries. In addition, incorporating this kosmotropic principle into other electrode components such as binders and conductive additives may be an effective way to mitigate water activity.** This strategy provides insight into environmentally sustainable aqueous processing methodologies and encourages a path toward green and carbon-neutral battery manufacturing.”

“Supplementary Reference

- 23 Radloff, S., Scurtu, R.-G., Hölzle, M. & Wohlfahrt-Mehrens, M. Applying established water-based binders to aqueous processing of LiNi_{0.83}Co_{0.12}Mn_{0.05}O₂ positive electrodes. *J. Electrochem. Soc.* **168**, 100506 (2021).
- 24 Radloff, S., Scurtu, R.-G., Hölzle, M. & Wohlfahrt-Mehrens, M. Water-based LiNi_{0.83}Co_{0.12}Mn_{0.05}O₂ electrodes with excellent cycling stability fabricated using unconventional binders. *J. Electrochem. Soc.* **169**, 040514 (2022).”

Reviewer #2

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #3

In submitted manuscript, authors have explored that kosmotropic anions in aqueous solution could be used for manufacturing of cathode for green lithium batteries. Ions in hydration shells are restructured to form a highly ordered state of anion–water clusters and regulate local hydration structure adjacent to cathode materials. Further it was also revealed that, scalable slurry-cast fabrication of various cathodes achieves a reduction in energy consumption. This is an applied nature; significant work.

This manuscript covers a detailed study which covered all details could be needed for reader.

Data is supported with figures. All details have been provided in supplementary data.

Submitted manuscript could be accepted for publication after following minor points:

3–1. In summary, add some highlighted findings of submitted work in terms of different variables.

→ **Response:** We highly appreciate the reviewer’s valuable comments. In response to the reviewer's suggestion, we have revised the Summary to include highlighted findings of our work, focusing on key variables such as kosmotropic anion effects, cycling performance, and manufacturing efficiency.

[Revised Manuscript]

“Summary

The global pursuit of carbon neutrality is driving efforts toward environmentally friendly aqueous electrode manufacturing. However, the inherent chemical reactivity of water with cathode materials remains a challenge to achieving this goal. Here, we design a class of aqueous processing solutions based on the kosmotropic effect. Ion hydration shells in the kosmotropic solutions are restructured to form an ordered state of anion–water clusters and to stabilize local hydration structure adjacent to cathode materials. Consequently, interfacial side reactions and structural degradation of Ni-rich cathode materials are mitigated. The kosmotropic solution-processed $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ cathodes achieved high gravimetric and areal capacities ($\geq 205 \text{ mAh g}^{-1}$ and $\geq 3.7 \text{ mAh cm}^{-2}$) together with stable cyclability, which appeared comparable to those of commercial *N*-methyl-2-pyrrolidone (NMP)-processed cathodes. Techno-economic analysis demonstrated that this kosmotropic solution approach reduced energy consumption in battery manufacturing by 46% compared to the NMP-based process, highlighting its practical and sustainable viability.”

3–2. Mention objectives and novelty of submitted work in introduction section.

→ **Response:** Many thanks for the reviewer’s valuable comments. In response to the reviewer's comment, we have revised the Introduction to clearly describe the objective and novel aspects of our research.

The primary objective of this study is to facilitate an urgent transition from the widespread use of the hazardous chemical NMP in lithium-ion battery cathode manufacturing to scalable cathode manufacturing with environmentally benign processing solvents, which can comply with the environmental restrictions and achieve sustainable battery production. In specific, to address the formidable challenge (inherent chemical reactivity of water molecules with Ni-rich cathode materials) of aqueous cathode manufacturing, herein, we propose a class of aqueous processing solutions based on the kosmotropic effect. Using the kosmotropic aqueous processing solution, we enabled the scalable slurry-cast fabrication of various Ni-rich cathodes with stable cyclability comparable to those of commercial toxic NMP-processed cathodes.

The novelty of this study lies in the restructuring of the ion hydration shells in the aqueous processing solution by introducing kosmotropic effect, a phenomenon common in biochemistry but rarely applied to batteries. The restructured ion hydration shells formed a highly ordered state of anion–water clusters that stabilize the local hydration environment around the cathode materials, which in turn mitigated the undesired side reactions at water–cathode material interfaces and prevented the structural degradation of the cathode materials during the cathode manufacturing. This aqueous processing solution strategy based on the kosmotropic effect provides insight into environmentally-sustainable and cost-competitive aqueous cathode manufacturing applicable to various high-capacity lithium electrode materials and encourages a path towards green and carbon-neutral battery manufacturing.

In response to the reviewer’s comment, the objective and novelty of the study were clearly provided in the revised manuscript.

[Revised Manuscript]

“Previous works implemented to resolve these problems have focused on modifying the surface of cathode materials⁹, adding mild organic acids for *in-situ* surface coating¹⁰, increasing Li⁺ concentration in water to reduce Li⁺ leaching (Le Chatelier’s principle)^{5,11}, and applying coatings on Al current collectors⁷. Despite these efforts, the underlying challenge of water’s inherent chemical reactivity with cathode materials remains unresolved. To address this

limitation and enable the use of water as a processing solvent for cathode manufacturing, it is crucial to understand and manipulate the local hydration environment around the cathode materials.

Herein, we introduce a class of aqueous **processing** solutions based on the kosmotropic effect, a phenomenon common in biochemistry^{12,13}, but rarely applied to batteries. **Incorporating** the kosmotropic anions resulted in the restructuring of the ion hydration shell to form a highly ordered state of anion–water clusters and the regulation of the local hydration structure adjacent to cathode materials (**Fig. 1a, right**). This reorganization of the ion hydration shell influences **water molecules' thermodynamics and physicochemical properties**, thus mitigating the undesired interfacial reactions at the cathode material surfaces **during cathode fabrication**. Owing to these advantageous effects, the kosmotropic aqueous **processing** solution enabled the scalable slurry-cast fabrication of various **Ni-rich** cathodes (**LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂ (NCM622)**, **LiNi_{0.8}Co_{0.1}Mn_{0.1}O₂ (NCM811)**, and **LiNi_{0.9}Co_{0.05}Mn_{0.05}O₂ (NCM9^{1/2}/2)**) with stable cycle life comparable to those of conventional NMP-processed cathodes (**Fig. 1b**). In addition, the kosmotropic solution-processed **Ni-rich** cathodes exhibited high gravimetric and areal capacities ($\geq 195 \text{ mAh g}^{-1}$ and **3.6** mAh cm^{-2} , respectively) compared to previously reported aqueous solution-processed cathodes. **In addition to the electrochemical benefits, the material cost of the kosmotropic solution is 96% lower than that of the NMP processing solvent.** From a comprehensive techno-economic analysis, we propose that the use of the kosmotropic aqueous **processing** solution can reduce the operational ($\sim 23\%$), and capital ($\sim 95\%$) costs associated with cathode manufacturing **by eliminating the conventional solvent recovery process.** **Furthermore, this approach substantially reduced energy consumption in electrode manufacturing by 46%, demonstrating its viability as a platform technology for environmentally friendly and cost-competitive electrode manufacturing applicable to various high-capacity electrode materials.**”

3–3. Explain about different softwares used to carry out computational analysis. Give brief understanding for readers.

→ **Response:** We greatly appreciate the reviewer's valuable comment. The computational techniques used in this study include Molecular Dynamics (MD) and Density Functional Theory (DFT). The software used for this computation are described below.

MD simulations predict the movement of molecules based on Newton's second law of motion, describing the classical behavior of particles to determine the structural and dynamic properties of systems. In this study, GROMACS software was used for the MD simulations. GROMACS is user-friendly, allowing intuitive setting of system parameters and topology. It can utilize both CPU and GPU to accelerate simulations, making it one of the most popular MD software packages. Driven by these advantages, many MD simulation studies have been conducted using GROMACS due to these advantages.

DFT, on the other hand, calculates the physical and chemical properties of a system based on quantum mechanics, specifically the Schrödinger equation, reformulated in terms of electron density rather than wavefunctions. We used Gaussian and VASP software for DFT calculations in this study. Gaussian simplifies the initial setup by allowing coordinates and parameters of molecules to be written in a single input script, and its output files can be easily visualized using GaussView software for Electrostatic Potential (ESP) visualization. VASP, suitable for calculating solid-state materials, was used to calculate the charges of NCM811 atoms. Its popularity and ease of post-processing computational results make VASP a widely used program in the field.

In response to the reviewer's comment, brief explanation about computational analysis were provided in the revised manuscript.

[Revised manuscript]

“Supplementary Note 1 | Computational analysis method

To investigate the effects of anions in aqueous processing solutions, we conducted MD and DFT simulations. MD simulations predict the movement of molecules to describe the behavior of particles, while DFT calculates the electron density of molecules based on quantum mechanics. Therefore, MD simulations can be used to understand the properties of the system composed of electrolytes and NCM811, while DFT can be used to calculate the ESP of molecules or ions.

All MD simulations were performed using the GROMACS² molecular dynamics simulation package using optimized potentials for liquid simulation-all-atom (OPLS-AA) force field parameters.”

3–4. In experimental section provide details about instrumentation were used for electrochemical evaluation.

→ **Response:** Thank you so much for the reviewer's valuable comment. In response to the reviewer's comment, we have revised the experimental section to include the detailed information about the instrumentation used for electrochemical evaluation.

[Revised manuscript]

“Fabrication of electrodes, cell, and electrochemical characterizations

For the electrochemical tests, CR2032 coin-type cells (Ni-rich cathode||polyethylene (PE)||Li-metal anode (thickness = 100 μm)) were assembled in an Ar-filled glove box. Thereafter, 1 M LiPF_6 in EC/DEC(= 1/1 (v/v)) with 10 wt.% of FEC and 2 wt.% VC was used as a liquid electrolyte. The electrolyte was injected into the cell with an electrolyte mass/electrode capacity (E/C) ratio of 5 g Ah^{-1} explore the cell performance for practical energy-dense Li-metal cells⁴⁰, if not specified. After the cell assembly, a rest period of 12 h was allowed to ensure complete wetting of the electrodes. The cells were then subjected to formation cycles (1 charge/discharge cycle at a current rate of 0.1C). The cells were cycled at a charge/discharge current rate of 0.2 C/0.5 C and a voltage range of 3.00–4.25 V at 25°C, if not specified. The resistances of the electrodes were measured using electrochemical impedance spectroscopy (EIS) analysis in a frequency range that varies from 10^6 to 10^{-2} Hz and an applied amplitude of 10 mV using potentiostat/galvanostat (VSP-300, Bio-Logic). The EIS analysis of the full cells was conducted after discharge at an open-circuit voltage (OCV) of 3.4 V. To analyze the ion resistance inside the electrodes (R_{ion}), a symmetric cell configuration (electrode||electrode) at a fully lithiated state was used. The charge/discharge performance of the cells was investigated using a cycle tester (PESC 05, PNE Solution).”

3–5. In results and discussion section, on page number 6, discuss the order of strength of H-bond acceptor $\text{TFSI}^- < \text{OTf}^- < \text{OAc}^- < \text{SO}_4^{2-}$ with respect to different parameters such as electronegativity difference etc.

→ **Response:** We appreciate the reviewer’s insightful comment. An important factor required for hydrogen bond acceptors is the presence of highly electronegative sites, identified as oxygen atoms of the anions by their electrostatic potential (ESP) values. The ESP values of the molecules are influenced by the inductive effect of their substituents. For example, the fluoroalkyl groups of TFSI^- and OTf^- anions exhibit a strong electron-withdrawing effect due to the high electronegativity of the fluorine atom. This effect significantly reduces the electron density around the oxygen atoms, making these anions weaker hydrogen bond acceptors. Conversely, the methyl group of OAc^- is a weak electron-donating group, resulting in a relatively higher electron density around the oxygen atoms. In SO_4^{2-} , the electronegativity difference between sulfur (2.58) and oxygen (3.44) causes the electron density to be pulled towards the oxygen atoms. This leads to a higher electron density around the oxygen atoms, making the sulfate ion the strongest hydrogen bond acceptor among the listed anions. Consequently, this trend in ESP values is consistent with the principles of the Hofmeister series: $\text{TFSI}^- < \text{OTf}^- < \text{OAc}^- < \text{SO}_4^{2-}$. This order is determined by the electron density around the oxygen atoms, which correlates with the anion’s ability to accept hydrogen bonds. In response to the reviewer’s comment, we have added a detailed description of the relationship between electron density around the oxygen atom and hydrogen bonding ability in the revised manuscript.

[Revised Manuscript]

“To gain insight into the hydration states of these anions, we calculated their electrostatic potential (ESP) values using density functional theory (DFT) (**Fig. 2a**). The dipole moment in water molecules causes the preferential coordination of H atoms (H-bond donors) with the highly electronegative sites of anions (e.g., O atoms as H-bond acceptors)^{20,21}. The DFT calculations revealed that the ESP values of the hydration shells gravitated towards more negative values with an increase in the kosmotropicity of anions. It is expected that the fluoroalkyl group of TFSI^- or OTf^- can act as an electron-withdrawing group, reducing the electron density of the O atoms. In contrast, the methyl group of OAc^- is a weak electron-donating group, resulting in a relatively higher electron density around the O atoms. In SO_4^{2-} ,

the electronegativity difference between sulfur (2.58) and oxygen (3.44) causes electron density towards the oxygen atoms, resulting in a higher electron density around the O atoms.

Therefore, the ability of H-bond acceptors is strengthened in the order: $\text{TFSI}^- < \text{OTf}^- < \text{OAc}^- < \text{SO}_4^{2-}$."

3–6. Literature citation in results and discussion section is totally missing. Provide relevant literature to support findings from computational analysis.

→ **Response:** We appreciate the reviewer’s insightful comment. In response to the reviewer’s comment, we have added the relevant references to support our computational analysis in the revised manuscript. The references focused on (1) the determination of the surface region using the number density of the solvent, (2) the higher presence of SO_4^{2-} on the anode surface compared to TFSI^- and its role in reducing reactive H^+ , and (3) the method of analyzing anion transport properties based on the distance from the electrode surface. The reviewer’s valuable comment is highly appreciated, again.

[Revised manuscript]

“The sulfur atom of SO_4^{2-} in the kosmotropic solution was notably concentrated near the surface ($r < 5.75 \text{ \AA}$), whereas the central nitrogen atom of TFSI^- in the chaotropic solution was located in the bulk region ($r > 5.75 \text{ \AA}$) (**Fig. 3a**, top)²⁷.”

“Higher concentrations of SO_4^{2-} near the cathode surface can interact with reactive H^+ at the NCM811–water interface, consistent with the previously reported result for LiCoO_2 in salt-in-water electrolytes³⁰.”

“To gain insight into the diffusion behavior of ion hydration clusters, the displacement of anions was monitored for 5 ps (**Fig. 3d**)³³.”

“Reference

27 Xing, L., Vatamanu, J., Borodin, O., Smith, G. D. & Bedrov, D. Electrode/electrolyte interface in sulfolane-based electrolytes for Li ion batteries: a molecular dynamics simulation study. *J. Phys. Chem. C* **116**, 23871-23881 (2012).

30 Oh, H. *et al.* Anion-Induced Interfacial Liquid Layers on LiCoO_2 in Salt-in-Water Lithium-Ion Batteries. *JACS Au* **3**, 1392-1402 (2023).

33 Kim, J. *et al.* Observation of H_2 evolution and electrolyte diffusion on MoS_2 monolayer by in situ liquid-phase transmission electron microscopy. *Adv. Mater.* **34**, 2206066 (2022).”

3–7. What are the future perspectives of submitted work in conclusions section?

→ **Response:** Thank you so much for the reviewer’s constructive comment. In response to the reviewer’s comment, a discussion of the future perspectives of this study was added in the conclusion of the revised manuscript. The reviewer’s valuable comments are highly appreciated, again.

[Revised Manuscript]

“Conclusion

The widespread use of the hazardous chemical NMP in LIB cathode manufacturing has raised significant environmental concerns. To address this imperative issue, we designed a kosmotropic aqueous **processing** solution to mitigate the adverse interfacial chemical reactions between water and cathode materials by forming highly ordered ion hydration shells. The kosmotropic solution-processed cathodes exhibited a stable cycle life comparable to that of the NMP-processed cathode under practical conditions and achieved their theoretical specific capacity at high areal loadings. By conducting the techno-economic analysis, we demonstrated the superiority of the kosmotropic aqueous **processing** solution over NMP in electrode manufacturing, achieving significant reductions in energy consumption, CO₂ emissions, and cost savings for sustainable cathode production. **We anticipate that this kosmotropic solution-processed cathode approach could be extended to other electrode active materials and battery chemistries. In addition, incorporating this kosmotropic principle into other electrode components such as binders and conductive additives may be an effective way to mitigate water activity.** This strategy provides insight into environmentally sustainable aqueous processing methodologies and encourages a path toward green and carbon-neutral battery manufacturing.”

3–8. There are so many grammatical mistakes throughout the manuscript.

→ **Response:** Thank you so much for the reviewer's valuable comment. In response to the reviewer's comment, this manuscript has been re-edited by a professional, native English-speaking editor. For the editor's reference, below is a copy of the manuscript that has been professionally proofread to ensure that all grammatical and stylistic errors have been corrected.



Fig. | A captured image of editing certificate.

[Revised Manuscript]

“Therefore, an urgent transition to scalable cathode manufacturing using environmentally benign processing solvents, which should **also be** compatible with existing electrode

fabrication processes, is essential to comply with environmental **regulations** and ensure sustainable battery production.”

“Water, comprised of two hydrogen atoms and one oxygen atom, is one of the most elementary molecules on Earth¹⁴. Despite its **simple molecular structure**, water **forms** intricate and dynamic configurations **via** extensive hydrogen bond (H-bond) networks, adopting an open tetrahedral arrangement, **unlike** other liquids with non-directional van der Waals interactions¹⁵.”

“The dynamics of water molecules in the surface region of NCM811 **were** investigated using the residence time correlation function ($C_i(t)$) from the MD trajectories (**Fig. 3c** and **Supplementary Note 3**).”

“The economic, environmental, and electrochemical advantages of the kosmotropic solution (0.5 m Li₂SO₄ in H₂O) over the NMP solvent **are** summarized in **Fig. 6e**. **The** kosmotropic solution exhibits reduced health risks, as **evidenced** by **its** favorable material hazard index of the solution (**Supplementary Table 11**). **Moreover**, the commitment to environmental compatibility is evident from the commendable green score in **Supplementary Table 12**. These benefits collectively contribute to a significant reduction in CO₂ emission associated with electrode manufacturing. **Utilizing** the fluorine-free binder is expected to further enhance the sustainable value chain of the batteries (**Supplementary Table 12b**).”

“We anticipate that this kosmotropic solution-processed cathode approach could be extended to other electrode active materials and battery chemistries. In addition, incorporating this kosmotropic principle into other electrode components such as binders and conductive additives may be an effective way to mitigate water activity. This strategy provides insight into environmentally sustainable aqueous processing methodologies and encourages a path toward green and carbon-neutral battery manufacturing.”

“Each bulk aqueous solution **comprised** 4476 water molecules and 80 LiX molecules (X: TFSI⁻, OAc⁻, and OTF⁻) to set 1 N Li⁺ concentration. In the Li₂SO₄ bulk solution system, 4476 water molecules and 40 Li₂SO₄ molecules were used.”

Reviewer #4

The authors report the use of lithium salts with varying kosmotropivities (Li_2SO_4 , LiOAc , LiOTf , and LiTFSI) enable the aqueous fabrication of $\text{LiNi}_{0.X}\text{Co}_{0.Y}\text{Mn}_{0.Y}\text{O}_2$ (NCMXY) cathodes. The authors present the argument that the kosmotropic salt interacts with the water to prevent the water-induced lithium leaching and degradation of NCM811. Li_2SO_4 , LiOAc , LiOTf , and LiTFSI were selected to have varying degrees of interaction with and structuring of water or kosmotropic (very strong interaction) to chaotropic (little interaction) variation. The authors begin by studying the structure of water in the aqueous lithium salt solutions using spectroscopic methods and simulations. Additionally, the performance of NCM batteries for cathodes cast with aqueous solutions of the two extreme salts LiTFSI (chaotropic) and Li_2SO_4 (kosmotropic) were evaluated. Tracking slurry pH and Li^+ content were considered to check for leaching and material degradation and microscopy was used to identify changes in morphology of electrodes. The authors also examine electrodes with different mass loadings and NCM compositions. Finally, the authors provide an extensive cost/safety analysis of Li_2SO_4 -based aqueous processing and propose alterations to existing NMP-based commercial processing system to transition to Li_2SO_4 -based aqueous processing.

Overall, the authors present thorough analysis and discussion of their results, however, I believe the authors should add more discussion/comparisons of their results to prior work that report similar studies (see general comments below for papers) and explain how this work differs. Additionally, the authors should add details related to the experimental protocols to enhance reproducibility (please see general and specific comments for details) and to allow reviewers to fully evaluate the presented work. Currently, the experimental protocol for fabricating electrodes does not mention LiTFSI or Li_2SO_4 (or how they were integrated to the electrode fabrication process) making it difficult to fully evaluate the data presented. Additionally, the purity and source of all the chemicals utilized are not disclosed, making it impossible to evaluate the cost analysis presented by the authors. The findings of the manuscript could be of interest to the broader battery community; however, major revisions are necessary before determining the suitability of the manuscript for publication in Nature Communications. I have included my specific comments below.

→ **Response:** Our deep appreciation is devoted to the reviewer's insightful comments, which helped us advance our research one step further. In response to the reviewer's comments, we

tried to enrich the details of the investigation in the revised manuscript. We carefully addressed the reviewer's comments by point-by-point response. Please see our replies below.

4-1. The authors should also add discussion to prior work and provide comparisons to the obtained results for the reader to clearly understand the significance and new findings.

References:

1. M. Heidbüchel, T. Schultz, T. Placke, M. Winter, N. Koch, R. Schmuch, & A. Gomez-Martin, *ChemSusChem*, e202202161 (2023).
2. H. Oh, S.-J. Shin, E. Choi, H. Yamagishi, T. Ohta, N. Yabuuchi, H.-G. Jung, H. Kim, and H. R. Byon, *JACS Au*, 1392-1402 (2023).
3. I. Dienwiebel, M. Diehl, B. Heidrich, X. Yang, M. Winter, M. Börner, *Adv. Energy Sustainability Res.*, 2100075 (2021).

→ **Response:** Many thanks for the reviewer's valuable comment. In response to the reviewer's comment, the comparisons with the previous works mentioned by the reviewer were provided in the revised manuscript.

The previous works by Heidbüchel et al. (ref.: *ChemSusChem* **16**, e202202161 (2023)) and Dienwiebel et al. (ref.: *Adv. Energy Sustainability Res.* **2**, 2100075 (2021)) showed that the addition of Li salts during the aqueous processing of cathodes contributed to the formation of protective layers on the cathode active materials, thereby stabilizing the electrodes. This approach prevented Li^+ leaching from the cathode active materials, resulting in the improved cycle life and stability.

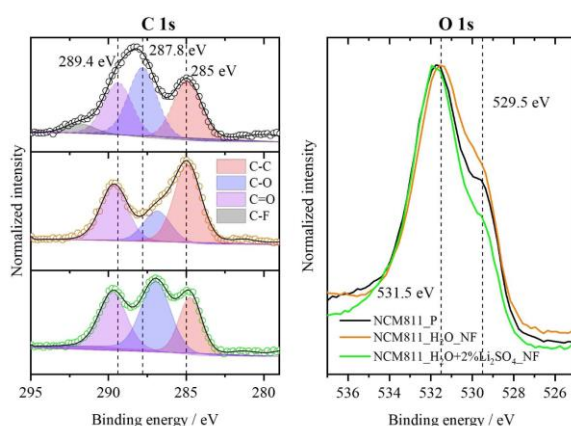


Fig. | A captured image from a research article published in *ChemSusChem* **16**, e202202161 (2023). XPS spectra of Ni-rich cathode materials in different processing methods.

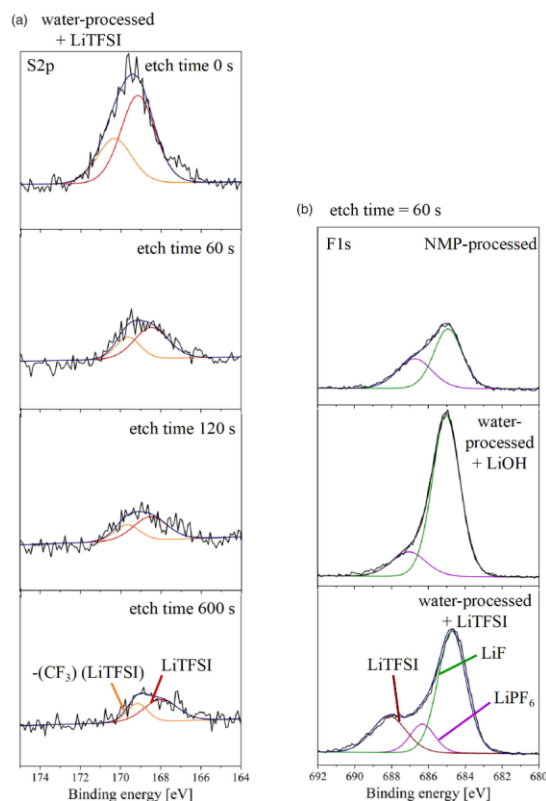


Fig. | A captured image from a research article published in *Adv. Energy Sustainability Res.* **2**, 2100075 (2021). XPS spectra of Ni-rich cathode materials in different processing methods.

The previous work by Oh et al. (ref.: *JACS Au* **3**, 1392 (2023)) reported a new aqueous liquid electrolyte (not an aqueous processing solvent for the cathode fabrication shown in our study) for aqueous LIBs. They found that the role of the liquid electrolytes in stabilizing $LiCoO_2$ electrode was correlated with the kosmotropic strength of anions (sulfate > nitrate > perchlorate > bistriflimide (TFSI)).

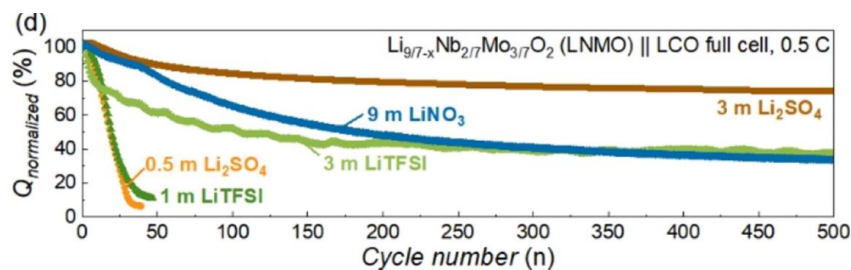


Fig. | A captured image from a research article published in *JACS Au* **3**, 1392 (2023). Cycle performance of aqueous LCO||LNMO full cells.

In comparison, the primary objective of this study is to facilitate an urgent transition from the widespread use of the hazardous chemical NMP in lithium-ion battery cathode manufacturing to scalable cathode manufacturing with environmentally benign processing solvents, which can comply with the environmental restrictions and achieve sustainable battery production. In specific, to address the formidable challenge (inherent chemical reactivity of water molecules with Ni-rich cathode materials) of aqueous cathode manufacturing, herein, we propose a class of aqueous processing solutions based on the kosmotropic effect. Using the kosmotropic aqueous processing solution, we enabled the scalable slurry-cast fabrication of various Ni-rich cathodes with stable cyclability comparable to those of commercial toxic NMP-processed cathodes. A novelty of this study lies in the restructuring of the ion hydration shells in the aqueous processing solution by introducing kosmotropic effect. The restructured ion hydration shells formed a highly ordered state of anion–water clusters that stabilize the local hydration environment around the cathode materials, which in turn mitigated the undesired side reactions at water–cathode material interfaces and prevented the structural degradation of the cathode materials during the cathode manufacturing. This aqueous processing solution strategy based on the kosmotropic effect provides insight into environmentally-sustainable and cost-competitive aqueous cathode manufacturing applicable to various high-capacity lithium electrode materials and encourages a path towards green and carbon-neutral battery manufacturing.

In response to the reviewer’s comment, we have included comparisons into the revised manuscript to provide a clearer context for the reader.

[Revised Manuscript]

“Previous works implemented to resolve these problems have focused on modifying the surface of cathode materials⁹, adding mild organic acids for *in-situ* surface coating¹⁰, increasing Li⁺ concentration in water to reduce Li⁺ leaching (Le Chatelier’s principle)^{5,11}, and applying coatings on Al current collectors⁷. Despite these efforts, the underlying challenge of water’s inherent chemical reactivity with cathode materials remains unresolved. To address this limitation and enable the use of water as a processing solvent for cathode manufacturing, it is crucial to understand and manipulate the local hydration environment around the cathode materials.

Herein, we introduce a class of aqueous processing solutions based on the kosmotropic effect, a phenomenon common in biochemistry^{12,13}, but rarely applied to batteries. Incorporating the

kosmotropic anions resulted in the restructuring of the ion hydration shell to form a highly ordered state of anion–water clusters and the regulation of the local hydration structure adjacent to cathode materials (Fig. 1a, right). This reorganization of the ion hydration shell influences water molecules' thermodynamics and physicochemical properties, thus mitigating the undesired interfacial reactions at the cathode material surfaces during cathode fabrication.

Owing to these advantageous effects, the kosmotropic aqueous processing solution enabled the scalable slurry-cast fabrication of various Ni-rich cathodes ($\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ (NCM622), $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (NCM811), and $\text{LiNi}_{0.9}\text{Co}_{0.05}\text{Mn}_{0.05}\text{O}_2$ (NCM9½½)) with stable cycle life comparable to those of conventional NMP-processed cathodes (Fig. 1b). In addition, the kosmotropic solution-processed Ni-rich cathodes exhibited high gravimetric and areal capacities ($\geq 195 \text{ mAh g}^{-1}$ and 3.6 mAh cm^{-2} , respectively) compared to previously reported aqueous solution-processed cathodes. In addition to the electrochemical benefits, the material cost of the kosmotropic solution is 96% lower than that of the NMP processing solvent. From a comprehensive techno-economic analysis, we propose that the use of the kosmotropic aqueous processing solution can reduce the operational ($\sim 23\%$), and capital ($\sim 95\%$) costs associated with cathode manufacturing by eliminating the conventional solvent recovery process. Furthermore, this approach substantially reduced energy consumption in electrode manufacturing by 46%, demonstrating its viability as a platform technology for environmentally friendly and cost-competitive electrode manufacturing applicable to various high-capacity electrode materials.”

“Higher concentrations of SO_4^{2-} near the cathode surface can interact with reactive H^+ at the NCM811–water interface, consistent with the previously reported result for LiCoO_2 in salt-in-water electrolytes³⁰.”

“Reference

- 5 Dienwiebel, I. *et al.* Enabling Aqueous Processing for $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ -Based Positive Electrodes in Lithium-Ion Batteries by Applying Lithium-Based Processing Additives. *Adv. Energy Sustain. Res.* **2**, 2100075 (2021).
- 11 Heidbüchel, M. *et al.* Enabling Aqueous Processing of Ni-Rich Layered Oxide Cathode Materials by Addition of Lithium Sulfate. *ChemSusChem* **16**, e202202161 (2023).
- 30 Oh, H. *et al.* Anion-Induced Interfacial Liquid Layers on LiCoO_2 in Salt-in-Water Lithium-Ion Batteries. *JACS Au* **3**, 1392-1402 (2023).”

Supplementary Table 7 | Comparison of the kosmotropic solution-processed cathodes and previously reported aqueous solution-processed cathodes.

No.	Processing solution	Cathode material	Binder	Specific capacity [mAh g ⁻¹]	Areal capacity [mAh cm ⁻²]	AM composition [wt. %]	Ref.
This work	0.5 m Li ₂ O ₄ in H ₂ O	NCM9½/½	CMC/PAA	215	3.9	93.1	This work
		NCM811	CMC/PAA	205	3.7, 7.8, 10.2		
		NCM622	CMC/PAA	195	3.6		
1	H ₂ O	NCM811	CMC/Acrylic emulsion	205	2.3	90	22
2	H ₂ O	NCM811	Carrageenan	200	2.2	90	23
3	H ₂ O	NCM811	PDADMA-DEP (or DBP)	200	2.1	90	24
4	H ₂ O	NCM811	SBR/CMC	196	2.6	94	25
5	Li ₂ SO ₄ in H ₂ O	NCM811	CMC	190	2.4	92	26
6	H ₂ O	NCM811	CMC/PAA	185	2.6	93	27
7	H ₂ O	NCM811	CMC/ICN	182	2.6	93	28
8	H ₂ O	NCM811		189	2.6	90	29
		NCM622	CMC	169	2.4	90	
		NCM111		153	2.1	90	

9	LiOH in H ₂ O	NCM523	CMC/PAA/PEO	158	2	91	30
10	H ₂ O	NCM424	CMC	160	0.4	80	31
11	H ₂ O	NCM111	CMC	150	1.1	88	32
12	H ₃ PO ₄ in H ₂ O	NCM111	CMC	150	1.3	88	33
13	H ₂ O	NCA	CMC	182	2	92	34
14	H ₂ O	LFP	Xanthan gum	167	0.4	87	35
15	H ₂ O	LFP	NBR/CMC	155	3	90	36
16	H ₂ O	LFP	PAA	150	0.2	83	37
17	H ₂ O	LFP	CMC	140	0.2	85	38
18	H ₂ O	LFP	PEO	136	0.4	73	39
19	LiTFSI in H ₂ O	LNMO	CMC/SBR	125	2	71	14
20	H ₂ O	LMO	Alginate	122	0.6	75	40

4-2. The grade of lithium salts should be specified (for reproducibility and analysis of the presented cost arguments).

→ **Response:** Thank you so much for the reviewer's valuable comment. We acknowledge that the specification of both the grade and the manufacturers of the lithium salts is necessary for the reproducibility and cost analysis. The information on the Li salts and other materials used in this study were added in the revised manuscript.

[Revised manuscript]

“Material preparation

Lithium sulfate (Li_2SO_4 , 99.5%), Lithium acetate (LiOAc , 99.95%), Lithium trifluoromethanesulfonate (LiOTf , 96.0%), N-methyl-2-pyrrolidone (NMP, 99.5%), carboxymethyl cellulose (CMC, $M_w = 250,000$, degree of substitution = 0.9), and polyacrylic acid (PAA, $M_w = 45,000$, purity) were purchased by Sigma-Aldrich. Li bis(trifluoromethanesulfonyl)imide (LiTFSI , 99.0%) and polyvinylidene fluoride (PVDF) were bought from Samchun and Solvay, respectively. 1 M lithium hexafluorophosphate (LiPF_6) in ethylene carbonate (EC)/diethyl carbonate (DEC) (= 1/1 (v/v)), fluoroethylene carbonate (FEC, 99.9%), and vinyl carbonate (VC, 99.9%) were purchased by Enchem (Korea). A water purification system (Direct Q-5UV, Merck Millipore) was utilized to prepare deionized water for aqueous processing solutions. Al current collectors were purchased from MTI (China). The material cost information used in **Supplementary Tables 3–5** was obtained from the suppliers described above.”

4–3. Do the salts used need to be lithium-based (since the anion seems critical to the water network)? Could alternative organic salts (ammonium, tetrabutylammonium, tetramethylammonium, etc.) also be used in this method? If other salts could be used, the authors should comment on the hypothesized effect of the cation's kosmotropicity on the observed stabilization for processing. For example, what would a chaotropic cation-based sulfate salt (such as tetramethylammonium sulfate) do? This is not a suggestion of additional experiments.

→ **Response:** Thank you so much for the reviewer's insightful comment. In this study, the design of salt additives was based on two main factors: (i) the kosmotropicity of ions, and (ii) the suppression of the $\text{Li}^+\text{-H}^+$ exchange reaction. Regarding the kosmotropicity of ions, as already described in the submitted manuscript, it is known that the hydrogen bonding network of water is more influenced by anions than by cations [ref: *Biophys. J.* **72**, 65-76 (1997)]. This is the main reason why we were focused on the kosmotropicity of anions, rather than cations. In response to the reviewer's comment, we investigated the effect of cations (e.g., Na^+ and Mg^{2+}) on the cycling performance of cells in the presence of the same kosmotropic sulfate.

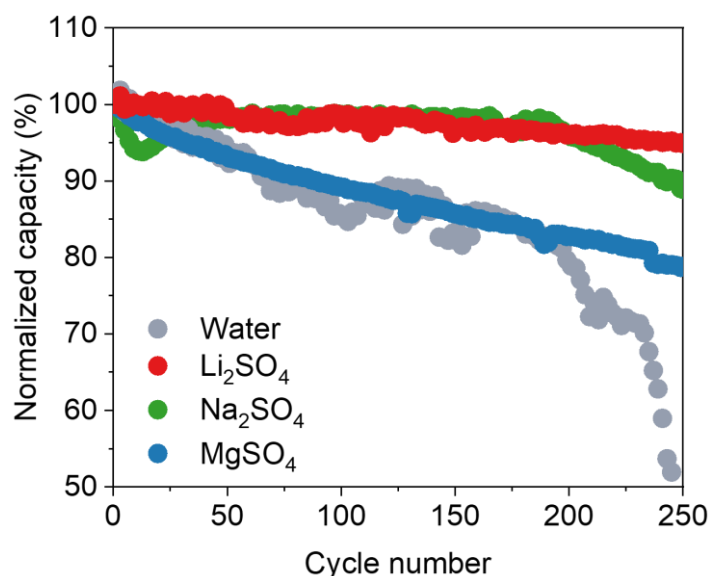


Fig. R10 | Cycle retention of the kosmotropic solution-processed cathodes as a function of the cations in the different salt additives (Na_2SO_4 vs. MgSO_4 vs. Li_2SO_4). The cells were cycled at a current rate of 0.2 C/0.5 C (= 1.9 mA cm^{-2}).

We observed that the addition of the salt additives improved the cycling performance, revealing the viable role of the kosmotropic SO_4^{2-} anion in suppressing the water activity. It is

known that the strength of cation's kosmotropicity decreases in the following order: $\text{Mg}^{2+} > \text{Li}^+ > \text{Na}^+$ [ref.: *Int. J. Mol. Sci.* **4**, 1932 (2021)]. However, the observed cycling performance tended to decrease in the following order: $\text{Li}^+ > \text{Na}^+ > \text{Mg}^{2+}$, which is not consistent with the strength of cation's kosmotropicity mentioned above.

The reason why the best cell performance was observed at Li^+ can be explained by considering the Li^+/H^+ exchange reaction. As already described in the submitted manuscript, according to the Le Chatelier's principle, increasing the Li^+ concentration on the right side of **Equation 1** thermodynamically suppresses the Li^+ leaching reaction from Li cathode materials.



These supplementary experimental results and theoretical considerations described above verify the effectiveness of Li^+ in the salt additives to mitigate the chemical activity of water molecules towards Li cathode materials.

Regarding the potential use of chaotropic cations suggested by the reviewer (*e.g.*, tetramethylammonium sulfate), it is known that chaotropic cations disrupt the hydrogen-bond network of water [ref.: *J. Am. Chem. Soc.* **124**, 12302 (2002)]. Therefore, the use of chaotropic cations in the design of salt additives for preparation of aqueous processing solutions is not appropriate.

In response to the reviewer's comment, the design concept of cations in the salt additives was added in the revised manuscript.

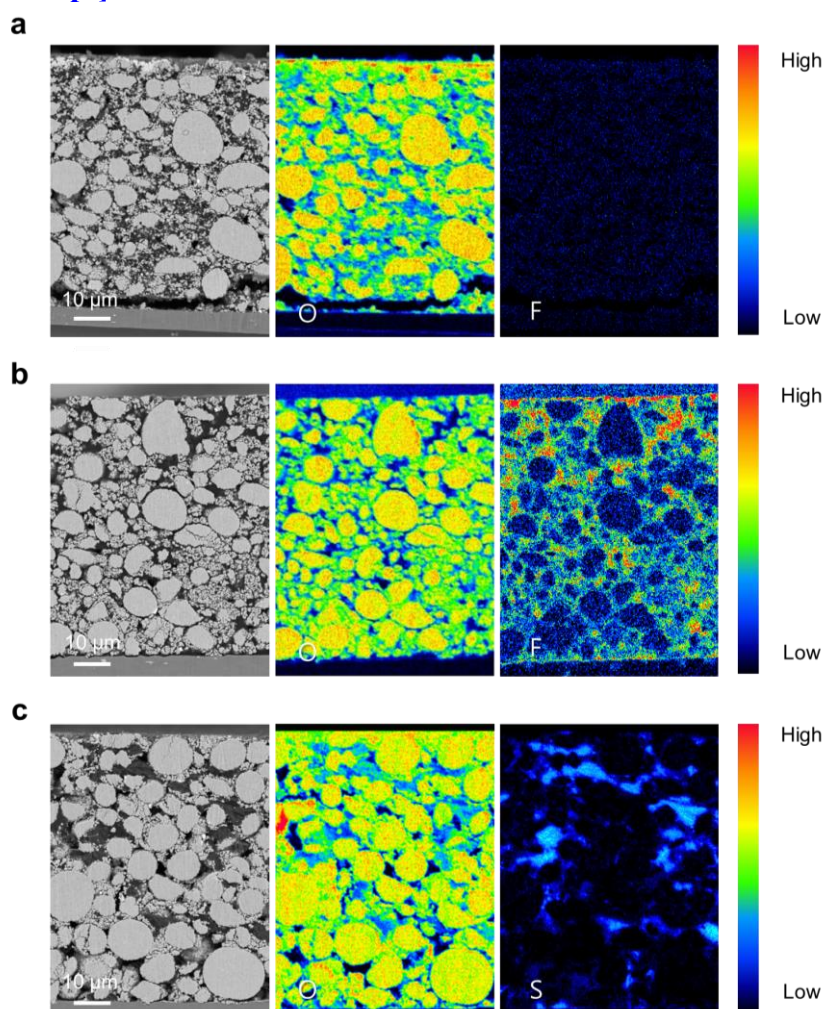
[Revised manuscript]

"Ions, as categorized by the Hofmeister series, can influence the molecular structure and dynamics of water by altering the strength of its H-bond network¹⁶. The kosmotropic ion exhibits a high charge density and can strengthen the H-bond structure¹⁷. On the other side of the series, the chaotropic ion has a large and charge-delocalized molecular structure¹⁸, exhibiting weaker intermolecular interactions with water and disordering the H-bond networks¹⁷. Owing to its molecular polarity, anions exert a more pronounced influence on the structure of water than cations¹⁹. Based on the kosmotropicity of anions, we investigated the effects of four representative Li salts: Li bis(trifluoromethanesulfonyl)imide (LiTFSI), Li trifluoromethanesulfonate (LiOTf), Li acetate (LiOAc), and Li sulfate (Li_2SO_4). Among various cation candidates, Li^+ was selected to effectively suppress the Li^+/H^+ exchange reaction in Equation (1) described above."

4–4. EDS mapping of salt specific elements (perhaps sulfur) or thermal gravimetric analysis could provide more direct evidence/quantification of the precipitated salts present in the processed electrodes.

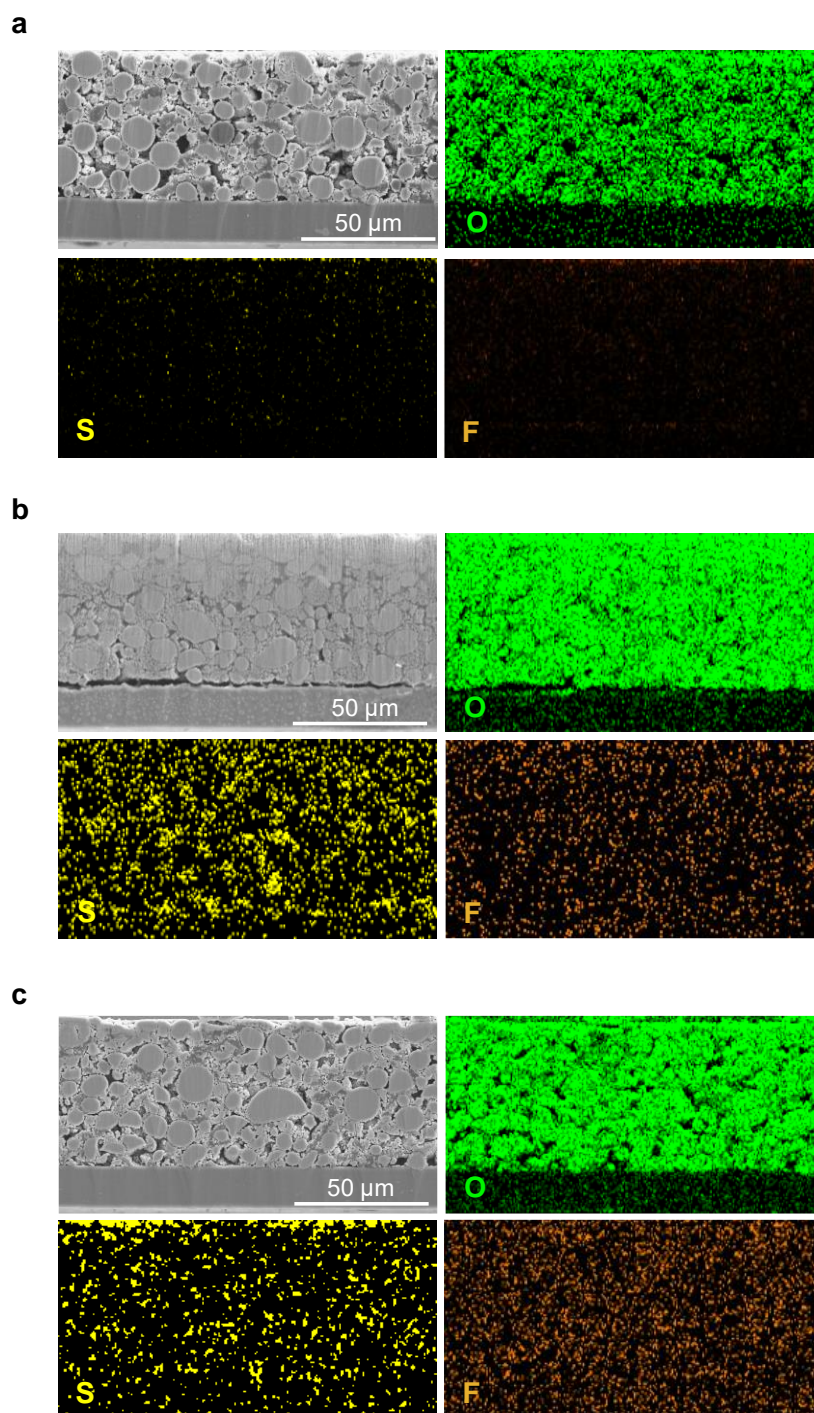
→ **Response:** Thank you so much for the reviewer’s insightful comment. In response to the reviewer’s comment, we have added the EDS images of the S and F elements in **Supplementary Fig. 20**. This result indicates the presence of the TFSI and SO₄ anions inside the fabricated electrodes. As such lightweight elements are difficult to detect accurately by the EDS analysis, we added the electron probe microanalysis (EPMA) elemental mapping images to identify the distribution of salt additives (**Supplementary Fig. 14**) in the revised manuscript. The reviewer’s valuable comment is highly appreciated, again.

[Revised Manuscript]



Supplementary Fig. 14 | a-c, Cross-sectional scanning electron microscopy (SEM) and electron probe microanalysis (EPMA) elemental mapping (element: O, F, and S) images of the

aqueous solution-processed cathodes: water (a), chaotropic solution (b), kosmotropic solution (c). The F and S elements indicate the presence and distribution of the salt additives (LiTFSI was used in the chaotropic solution and Li_2SO_4 was used in the kosmotropic solution).



Supplementary Fig. 20 | a-c, Cross-sectional SEM and corresponding EDS images (elements: O, S, and F) of the water-processed (a), chaotropic solution-processed (b), and kosmotropic solution-processed (c) NCM811 cathodes.

4–5. Abstract – The specific electrode material being studied should be mentioned in the abstract.

→ **Response:** Thank you so much for the reviewer's valuable comment. In response to the reviewer's comment, we have revised the abstract to mention the specific electrode material 'Ni-rich' cathode, as it includes NCM811 which is under studied.

[Revised Manuscript]

“Summary

The global pursuit of carbon neutrality is driving efforts towards environmentally friendly aqueous electrode manufacturing. However, the inherent chemical reactivity of water with cathode materials remains a challenge to achieving this goal. Here, we design a class of aqueous processing solutions based on the kosmotropic effect. Ion hydration shells in the kosmotropic solutions are restructured to form an ordered state of anion–water clusters and to stabilize local hydration structure adjacent to cathode materials. Consequently, interfacial side reactions and structural degradation of Ni-rich cathode materials are mitigated. The resulting kosmotropic solution-processed $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ cathodes achieved high gravimetric and areal capacities ($\geq 205 \text{ mAh g}^{-1}$ and $\geq 3.7 \text{ mAh cm}^{-2}$) together with stable cyclability, which appeared comparable to those of commercial *N*-methyl-2-pyrrolidone (NMP)-processed cathodes. Techno-economic analysis demonstrated that this kosmotropic solution approach reduced energy consumption in battery manufacturing by 46% compared to the NMP-based process, highlighting its practical and sustainable viability.”

4–6. Figure 2 c – The line connecting the points should be removed, unless it represents a known fit between H-bond number and anion.

→ **Response:** We greatly appreciate the reviewer's valuable comment. In response to the reviewer's comment, we have eliminated the line connecting the points in **Fig. 2c** of the revised manuscript.

[Revised manuscript]

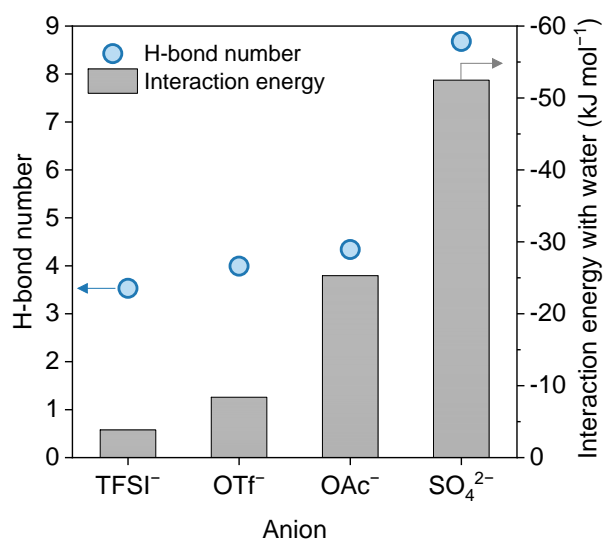


Fig. 2 | c, H-bond number of anion-water clusters and interaction energy between anion and water molecules calculated from the molecular dynamics (MD) trajectories.

4–7. Figure 2 e – The data for the whole recovery time should be presented in the SI.

→ **Response:** Thank you so much for the reviewer’s insightful comment. Please note that the data shown in **Fig. 2e** covered the whole recovery time for the T_1 measurements. It is known that a recovery time at least 5 times longer than the T_1 value, which allows for the recovery of the magnetization and the return to the equilibrium state, is required to obtain reliable results in the ^1H NMR measurement [ref.: *High-resolution NMR techniques in organic chemistry* 7 (Elsevier, 2016) and *Understanding NMR spectroscopy* (John Wiley & Sons, 2010)]. In this study, the T_1 values varied from 46.9 ms for the kosmotropic solution (*i.e.*, 0.5 m Li_2SO_4) to 74.3 ms for the chaotropic solution (*i.e.*, 1.0 m LiTFSI). Meanwhile, as shown in **Fig. 2e**, the T_1 measurement was performed over a time period of 6.4 s, which far exceeded the recommended minimum recovery time (at least 5 times longer than the T_1 value). Excessively long recovery times are known to cause experimental noise and potential instability in the magnetic field [ref.: *Principles of Nuclear Magnetic Resonance in One and Two Dimension*. (Oxford university Press, 1990) and *Spin Dynamics: Basics of Nuclear Magnetic Resonance* (Wiley, 2001)]. In response to the reviewer’s comment, we have added the relevant references describing this methodology in the **Methods** section of the revised manuscript. The reviewer’s valuable comment is highly appreciated, again.

[Revised manuscript]

“The longitudinal relaxation time (T_1) was calculated with Equation (1):

$$(1) \quad I = I_0(1 - \exp^{-t/T_1})$$

where I is the peak intensity at time t , I_0 is the saturation intensity, and T_1 is the longitudinal relaxation time. The total recovery time is 6.4 s, allowing the magnetization to recover sufficiently and return to its equilibrium state, thereby minimizing saturation effects^{42,43}.”

4–8. Page 9 – The method for getting the aluminum foil after casting should be specified. Was the electrode removed using solvent?

→ **Response:** Thank you so much for the reviewer's valuable comment. In response to the reviewer's comment, the method used to obtain the Al foil for the corrosion analysis has been specified in the revised manuscript.

[Revised manuscript]

“To observe the surface of Al current collectors after the electrode coating, the electrode active layers were gently removed from the electrode using a cotton swab soaked in dimethyl carbonate (DMC) solvent. The area of Al corrosion was quantified from binary images generated by applying 34% of the threshold program (Image J) to the SEM image of the Al current collectors.”

4–9. Figure 4 d-f – An arrow or label could be added to help the reader more readily identify the intergranular cracks.

→ **Response:** Thank you so much for the reviewer’s valuable comment. In response to the reviewer's comment, we have revised **Fig. 4d-f** to include the arrows in the revised manuscript.

[Revised manuscript]

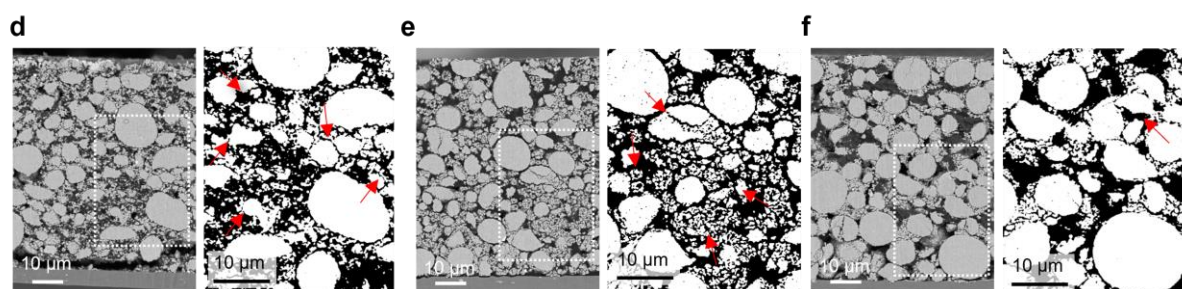


Fig. 4 | d-f, Cross-sectional scanning electron microscopy (SEM) images and corresponding binary images of aqueous solution-processed cathodes: water (**d**), chaotropic solution (**e**), kosmotropic solution (**f**). **Red arrows indicate intergranular cracks of NCM811 secondary particles.**

“Electrochemical performance of cathodes manufactured with aqueous processing solutions

To test the aqueous processed cathodes, we prepared cells comprising of Li metal anodes and NMC811 cathodes with **areal mass loading** and **electrode density** densities of **18** $\text{mg}_{\text{NCM811}} \text{cm}^{-2}$ and 3.0 g cc^{-1} , respectively (See Methods and **Supplementary Table 2** for details). The aqueous cathode slurries exhibited dispersion **stability** similar to those formulated in NMP, **suggesting that the incorporation of kosmotropic or chaotropic salts had a minimal effect on the dispersion of electrode slurries** (**Supplementary Fig. 13**). After calendaring, the structures of the electrodes were analyzed using cross-sectional SEM **and corresponding EPMA images** (**Supplementary Fig. 14**). Intergranular cracks were observed on the NCM811 particles in the water- and chaotropic solution-processed cathodes (**red arrows**, **Fig. 4d, e**). **In contrast**, the particles in the kosmotropic solution-processed cathodes remained comparatively unimpaired (**Fig. 4f**).”

4–10. Figure 5 a – Is this charge or discharge capacity? Please specify. The c-rate (two were provide in methods) should be specified.

→ **Response:** Many thanks for the reviewer’s insightful comment. In response to the reviewer’s comment, the experimental details of **Fig. 5a** were added in the revised manuscript.

[Revised manuscript]

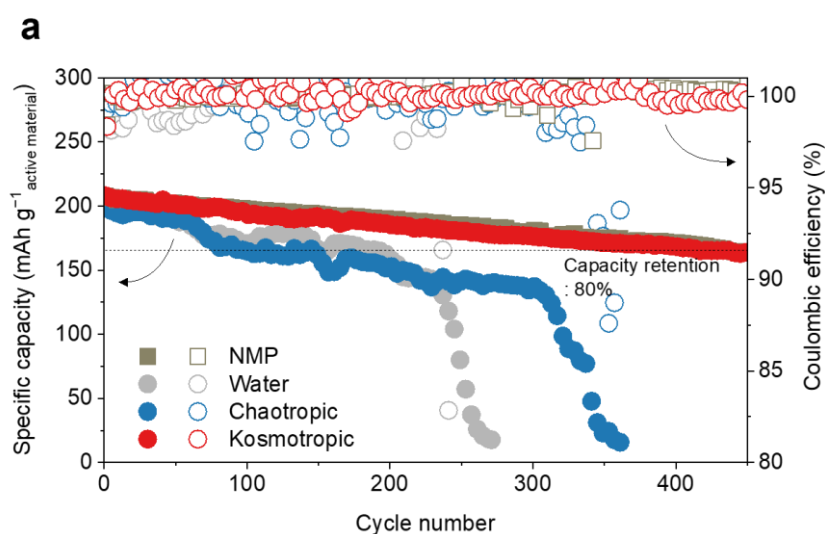


Fig. 5 | a, Cycling retention (expressed as **discharge** specific capacities of cathode materials) of the cathodes **at charge/discharge current rate of 0.2 C/0.5 C (= 1.9 mA cm⁻²).**

4–11. Figure 5 d – The processing solution for the GCD data should be explicitly stated.

→ **Response:** Thank you so much for the reviewer's valuable comment. In response to the reviewer's comment, we have revised the caption for **Fig. 5d** to state the processing solution used for the GCD data.

[Revised Manuscript]

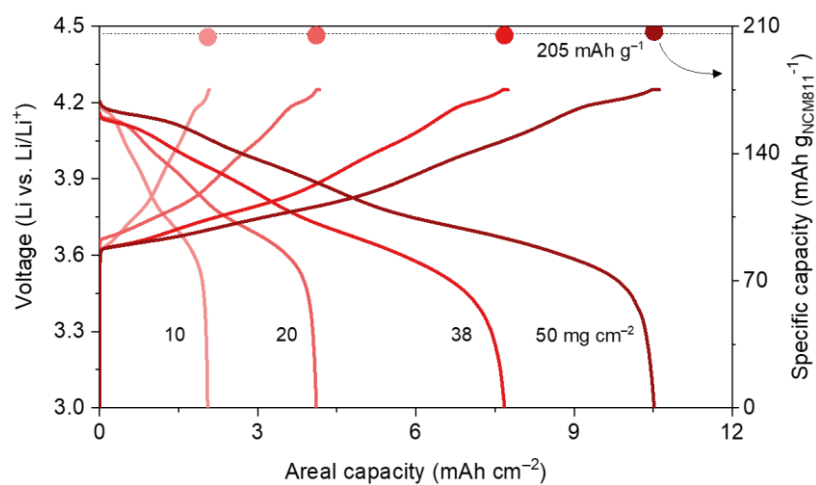


Fig. 5 | d, Galvanostatic **initial** charge/discharge profiles as a function of mass loading of **kosmotropic solution-processed** cathodes **at a current rate of 0.1C/0.1C**.

4–12. Methods Section – A comprehensive list of the materials/chemicals used, their purity, and supplying company should be provided.

→ **Response:** We appreciate the reviewer’s valuable comment. In response to the reviewer’s comment, we added the comprehensive list of the materials/chemicals, including their purity and supplying company. Please see Methods in the revised manuscript.

[Revised manuscript]

“Material preparation

Lithium sulfate (Li_2SO_4 , 99.5%), Lithium acetate (LiOAc , 99.95%), Lithium trifluoromethanesulfonate (LiOTf , 96.0%), N-methyl-2-pyrrolidone (NMP, 99.5%), carboxymethyl cellulose (CMC, $M_w = 250,000$, degree of substitution = 0.9), and polyacrylic acid (PAA, $M_w = 45,000$, purity) were purchased by Sigma-Aldrich. Li bis(trifluoromethanesulfonyl)imide (LiTFSI , 99.0%) and polyvinylidene fluoride (PVDF) were bought from Samchun and Solvay, respectively. 1 M lithium hexafluorophosphate (LiPF_6) in ethylene carbonate (EC)/diethyl carbonate (DEC) (= 1/1 (v/v)), fluoroethylene carbonate (FEC, 99.9%), and vinyl carbonate (VC, 99.9%) were purchased by Enchem (Korea). A water purification system (Direct Q–5UV, Merck Millipore) was utilized to prepare deionized water for aqueous processing solutions. Al current collectors were purchased from MTI (China). The material cost information used in **Supplementary Tables 3–5** was obtained from the suppliers described above.”

4–13. Methods - Fabrication of electrodes– The preparation of the aqueous lithium salt solutions and how they were utilized in electrode fabrication should be specified. The amount of liquid was used to formulate the slurry should be specified. The changes to slurry preparation for NCM622, NCM811, NCM9½½, and differing mass loading cathodes should be specified.

→ **Response:** Many thanks for the reviewer's detailed comments. In response to the reviewer's comment, we included the preparation details of the aqueous processing solutions and solid contents for electrode fabrication in the revised manuscript.

[Revised Manuscript]

“The electrode slurries were prepared with a composition of $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (NCM811)/carbon black/binder = 95.5/2.25/2.25 (w/w/w). Details of the electrode composition including residual salts are provided in Supplementary Table 2. For aqueous solution-processed electrodes, CMC and PAA were used as binders at a composition ratio of 1/1 (w/w). The salt additives were dissolved in water for 2 h and then the CMC and PAA binders were dissolved before the formulation of electrode slurries. The binder contents in the processing solutions are 3 wt.% and 10 wt.% for CMC and PAA, respectively. No additional water is added during the electrode fabrication, leaving 47 wt.% of water in the kosmotropic electrode slurry. The electrode slurries were cast onto Al current collectors and dried at 60°C for 6 h, followed by roll-pressing at 120°C. The electrode fabrication process was identical regardless of the mass loading and type of cathode materials. For the NMP-processed electrodes, PVDF was used as the binder, and dissolved in NMP. The concentration of the binder solution was set at 10 wt.%. The electrode slurries were cast onto Al current collectors and dried at 120°C for 6 h and then roll-pressed at 120°C. The density of the fabricated electrodes was set to 3.0 g cc⁻¹, and the electrodes were vacuum dried at 120°C for 12 h before the cell assembly.”

4–14. Methods – electrochemical Testing – The authors should specify any rest periods between battery assembly and electrochemical testing. Additionally, any “conditioning” cycles used before.

→ **Response:** Thank you so much for the reviewer’s valuable comment. In response to the reviewer’s comment, the details on the electrochemical testing were included in the revised manuscript.

[Revised Manuscript]

“Fabrication of electrodes, cell, and electrochemical characterizations

For the electrochemical tests, CR2032 coin-type cells (Ni-rich cathode||polyethylene (PE)||Li-metal anode (thickness = 100 μm)) were assembled in an Ar-filled glove box. Thereafter, 1 M LiPF_6 in EC/DEC(= 1/1 (v/v)) with 10 wt.% of FEC and 2 wt.% VC was used as a liquid electrolyte. The electrolyte was injected into the cell with an electrolyte mass/electrode capacity (E/C) ratio of 5 g Ah^{-1} explore the cell performance for practical energy-dense Li-metal cells⁴⁰, if not specified. After the cell assembly, a rest period of 12 h was allowed to ensure complete wetting of the electrodes. The cells were then subjected to formation cycles (1 charge/discharge cycle at a current rate of 0.1C). The cells were cycled at a charge/discharge current rate of 0.2 C/0.5 C and a voltage range of 3.00–4.25 V at 25°C, if not specified. The resistances of the electrodes were measured using electrochemical impedance spectroscopy (EIS) analysis in a frequency range that varies from 10^6 to 10^{-2} Hz and an applied amplitude of 10 mV using potentiostat/galvanostat (VSP-300, Bio-Logic). The EIS analysis of the full cells was conducted after discharge at an open-circuit voltage (OCV) of 3.4 V. To analyze the ion resistance inside the electrodes (R_{ion}), a symmetric cell configuration (electrode||electrode) at a fully lithiated state was used. The charge/discharge performance of the cells was investigated using a cycle tester (PESC 05, PNE Solution).”

4–15. Methods – EIS Testing – The voltage that EIS was centered at should be reported (i.e. 3 V with a 10 mV amplitude).

→ **Response:** Thank you so much for the reviewer's insightful comment. In response to the reviewer's comment, we have revised the Methods section.

[Revised Manuscript]

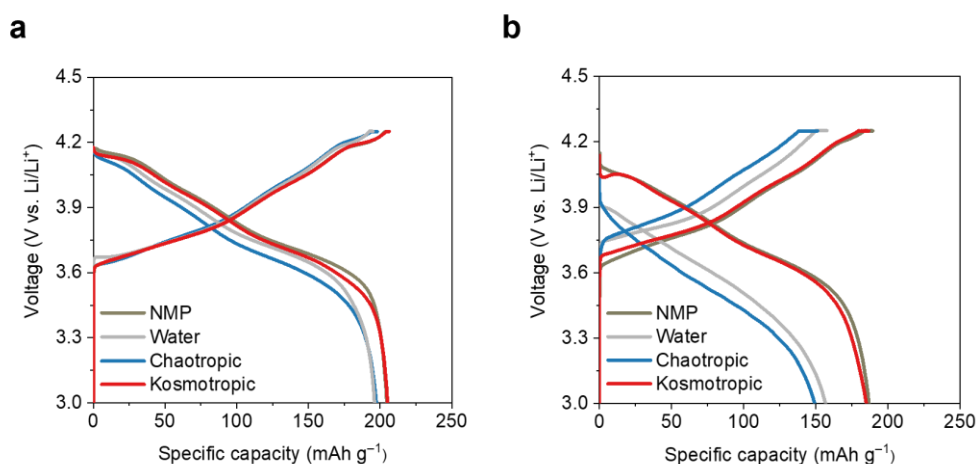
“Fabrication of electrodes, cell, and electrochemical characterizations

For the electrochemical tests, CR2032 coin-type cells (Ni-rich cathode||polyethylene (PE)||Li-metal anode (thickness = 100 μm)) were assembled in an Ar-filled glove box. Thereafter, 1 M LiPF_6 in EC/DEC(= 1/1 (v/v)) with 10 wt.% of FEC and 2 wt.% VC was used as a liquid electrolyte. The electrolyte was injected into the cell with an electrolyte mass/electrode capacity (E/C) ratio of 5 g Ah^{-1} explore the cell performance for practical energy-dense Li-metal cells⁴⁰, if not specified. After the cell assembly, a rest period of 12 h was allowed to ensure complete wetting of the electrodes. The cells were then subjected to formation cycles (1 charge/discharge cycle at a current rate of 0.1 C). The cells were cycled at a charge/discharge current rate of 0.2 C/0.5 C and a voltage range of 3.00–4.25 V at 25°C, if not specified. The resistances of the electrodes were measured using electrochemical impedance spectroscopy (EIS) analysis in a frequency range that varies from 10^6 to 10^{-2} Hz and an applied amplitude of 10 mV using potentiostat/galvanostat (VSP-300, Bio-Logic). The EIS analysis of the full cells was conducted after discharge at an open-circuit voltage (OCV) of 3.4 V. To analyze the ion resistance inside the electrodes (R_{ion}), a symmetric cell configuration (electrode||electrode) at a fully lithiated state was used. The charge/discharge performance of the cells was investigated using a cycle tester (PESC 05, PNE Solution).”

4–16. Figure S13 – Please specify the cycle shown and c-rate. Adding another panel with a later cycle (cycle 200) would provide the reader with a nice visual comparison of the battery performance vs. processing solvent.

→ **Response:** Thank you so much for the reviewer’s valuable comment. In response to the reviewer’s comment, we specified the cycle number and c-rate. In addition, another panel showing the charge/discharge curves after 200 cycles was added in the revised manuscript.

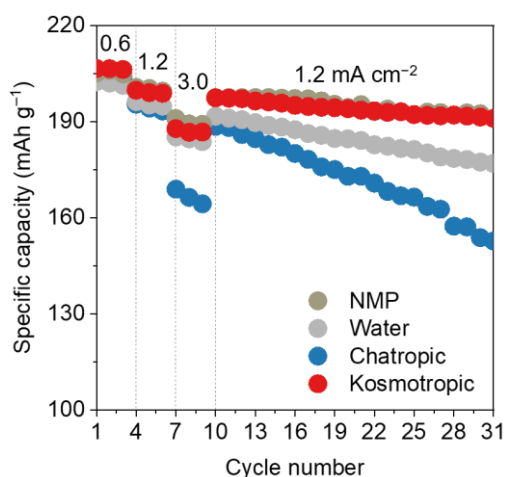
[Revised Manuscript]



Supplementary Fig. 15 | a,b, Galvanostatic charge/discharge profiles of the cathodes at a mass loading of 18 mg cm^{-2} at a charge/discharge current rate of $0.2 \text{ C}/0.5 \text{ C}$ ($= 1.9 \text{ mA cm}^{-2}$): 1st cycle (**a**) and 200th cycle (**b**).

4–17. Figure S18 – The figure caption should be clarified. It specifies a constant current density but the plot has different current densities. The number of cycles at each current density should be specified in the caption.

→ **Response:** Thank you so much for the reviewer's valuable comment. In response to the reviewer's comment, the figure caption were specified in the revised manuscript.



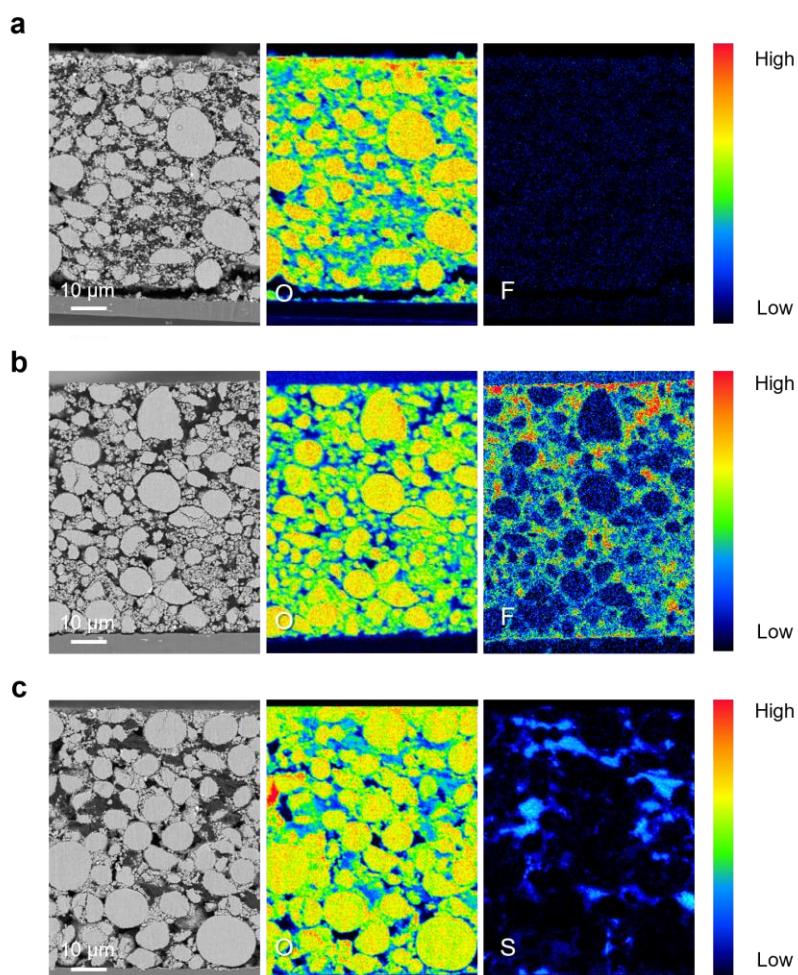
[Revised Manuscript]

Supplementary Fig. 19 | Discharge rate capability (discharge current rate = 0.6 mA cm⁻² (for 3 cycles), 1.2 mA cm⁻² (for 3 cycles), 3.0 mA cm⁻² (for 3 cycles), and 1.2 mA cm⁻² of the cathodes at an areal capacity of 6 mAh cm⁻² under a fixed charge current density of 3.0 mA cm⁻².

4–18. Figure S19 – EDS images for elements specifically found in TFSI and SO₄ (sulfur and fluorine) should be added to this figure.

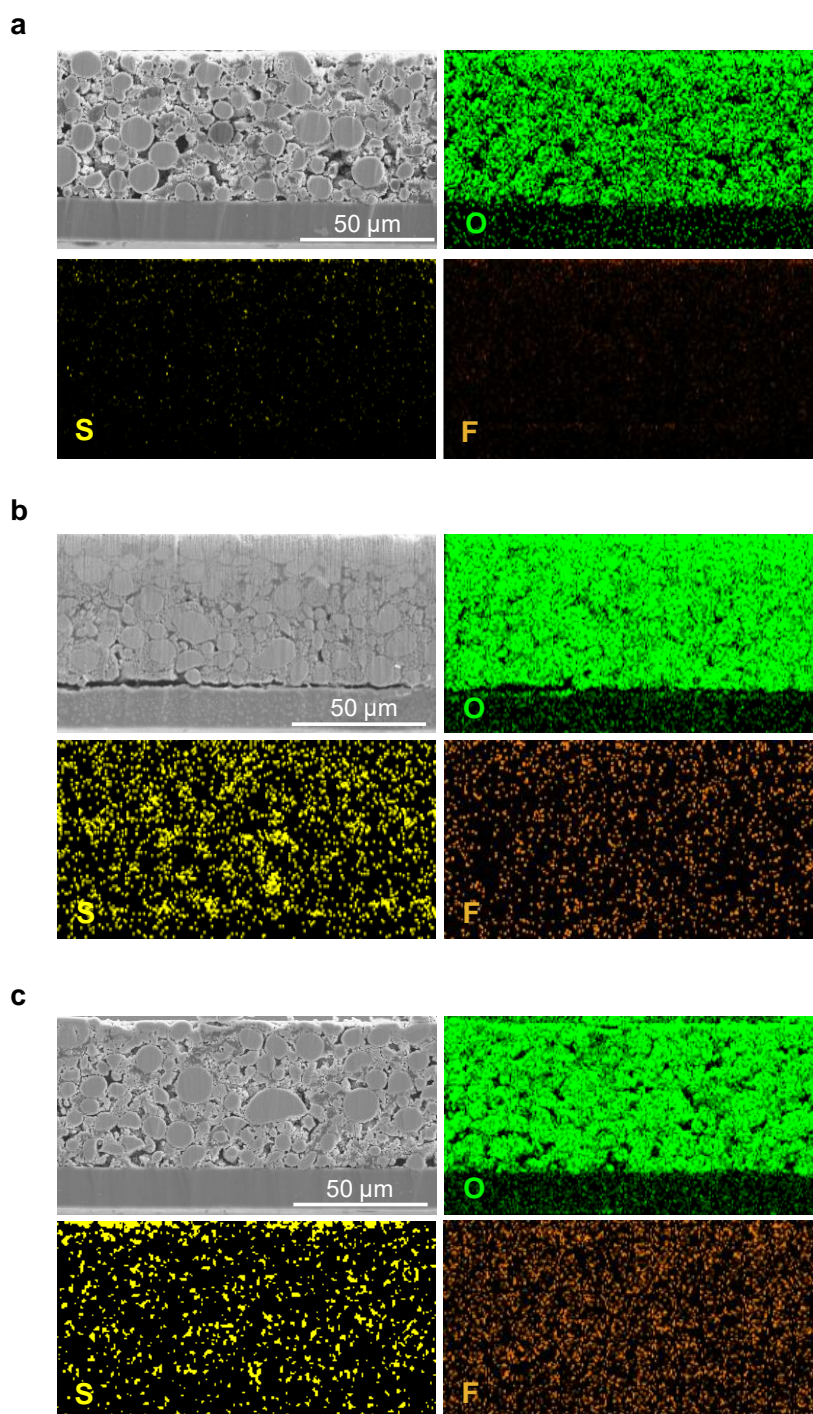
→ **Response:** Thank you so much for the reviewer’s insightful comment. In response to the reviewer’s comment, we have added the EDS images of S and F elements in the revised manuscript. This result indicates the presence of the TFSI and SO₄ anions inside the fabricated electrodes. As such lightweight elements are difficult to detect accurately by the EDS analysis, we added the electron probe microanalysis (EPMA) elemental mapping images to identify the distribution of salt additives (**Supplementary Fig. 14**) in the revised manuscript. The reviewer’s valuable comment is highly appreciated, again.

[Revised Manuscript]



Supplementary Fig. 14 | a-c, Cross-sectional scanning electron microscopy (SEM) and electron probe microanalysis (EPMA) elemental mapping (element: O, F, and S) images of the aqueous solution-processed cathodes: water (a), chaotropic solution (b), kosmotropic solution

(c). The F and S elements indicate the presence and distribution of the salt additives (LiTFSI was used in the chaotropic solution and Li_2SO_4 was used in the kosmotropic solution).



Supplementary Fig. 20 | **a-c**, Cross-sectional SEM and corresponding EDS images (elements: O, S, and F) of the water-processed (a), chaotropic solution-processed (b), and kosmotropic solution-processed (c) NCM811 cathodes.

4–19. Supplementary Table 6 – Processing solution should be added as a column to allow the reader to easily compare the results. Additionally, sorting the whole table by specific capacity might help the reader see the trends more readily.

→ **Response:** Thank you so much for the reviewer’s valuable suggestion. In response to the reviewer’s comment, the **Supplementary Table 7** was modified in the revised manuscript. The reviewer’s valuable comment is highly appreciated, again.

[Revised Manuscript]

Supplementary Table 7 | Comparison of the kosmotropic solution-processed cathodes and previously reported aqueous solution-processed cathodes.

No.	Processing solution	Cathode material	Binder	Specific capacity [mAh g ⁻¹]	Areal capacity [mAh cm ⁻²]	AM composition [wt. %]	Ref.
This work	0.5 m Li ₂ O ₄ in H ₂ O	NCM9½½	CMC/PAA	215	3.9	93.1	This work
		NCM811	CMC/PAA	205	3.7, 7.8, 10.2		
		NCM622	CMC/PAA	195	3.6		
1	H ₂ O	NCM811	CMC/Acrylic emulsion	205	2.3	90	22
2	H ₂ O	NCM811	Carrageenan	200	2.2	90	23
3	H ₂ O	NCM811	PDADMA-DEP (or DBP)	200	2.1	90	24
4	H ₂ O	NCM811	SBR/CMC	196	2.6	94	25

5	Li ₂ SO ₄ in H ₂ O	NCM811	CMC	190	2.4	92	26
6	H ₂ O	NCM811	CMC/PAA	185	2.6	93	27
7	H ₂ O	NCM811	CMC/ICN	182	2.6	93	28
		NCM811		189	2.6	90	
8	H ₂ O	NCM622	CMC	169	2.4	90	29
		NCM111		153	2.1	90	
9	LiOH in H ₂ O	NCM523	CMC/PAA/PEO	158	2	91	30
10	H ₂ O	NCM424	CMC	160	0.4	80	31
11	H ₂ O	NCM111	CMC	150	1.1	88	32
12	H ₃ PO ₄ in H ₂ O	NCM111	CMC	150	1.3	88	33
13	H ₂ O	NCA	CMC	182	2	92	34
14	H ₂ O	LFP	Xanthan gum	167	0.4	87	35
15	H ₂ O	LFP	NBR/CMC	155	3	90	36
16	H ₂ O	LFP	PAA	150	0.2	83	37
17	H ₂ O	LFP	CMC	140	0.2	85	38
18	H ₂ O	LFP	PEO	136	0.4	73	39
19	LiTFSI in H ₂ O	LNMO	CMC/SBR	125	2	71	14
20	H ₂ O	LMO	Alginate	122	0.6	75	40

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The manuscript has been revised according to the reviewers' comments, and I think it has addressed most of the concerns. The current version can tell outstanding information about the anion choice for processing water-based slurries of a high-Ni cathode active material (without losing its expected performance), which could stimulate our transition to eco-friendly battery manufacturing. This is a great work for a broad battery community and I think can be accepted for publication after the following comments are addressed.

1. One concern that needs to be addressed relates the original review (suggestion in comment (a-1). I believe Fig. 5a is still not a fair comparison as to the amount and type of salt remaining in the electrode layer, which therefore cannot fully support the sentence in Lines 244–246: “The capacity of the water- and chaotropic solution-processed cathodes degraded significantly after 200 cycles because of the active Li^+/H^+ exchange^{5,39} during the aqueous cathode fabrication (Supplementary Fig. 15 and Fig. 5a)”⁵; i.e., we cannot say the observed difference in cyclabilities between the four electrodes is purely due to interfacial side reactions between water and NCM811. This is because, as the authors have described in many parts (e.g., Fig R2, Supplementary Figs 17 and 20, their answer to my comment (b-8), etc.), the remaining salt seemingly contributes to pore blocking, which leads to a rapid decay in the capacity of an NCM811 cathode. Ideally, the comparison between the four electrodes after removing the salt residue is still required to clarify this point and should be considered in future work. Despite the authors mentioning that removing the salt residue is challenging, I believe, for example, this can be done using the same process as (or a similar process to) the authors' sample-preparation method for XPS (i.e., soaking an electrode in DMC for 10 min and drying, where the solvent can be replaced by the new one several times to ensure that the remaining salt is fully removed). Whilst I am not suggesting these experiments for the current manuscript, I believe it is important to be explicit that this could also be an issue that influences the capacity stability.

Additional comments to address below as well as some suggestions for the authors consideration would further improve the significance of the study:

2. As per my original suggestion of my comment (b-3). If the authors could briefly describe each sample-preparation step of NCM811 for particle-size analysis, that would be informative. I could not fully understand the procedure between (i) soaking NCM811 in different processing solutions and (ii) measuring particle size distribution of NCM811 using ethanol as the solvent (Lines 388–389). Does this mean the processing solutions were removed and dried NCM811 was re-dispersed in ethanol for analysis?
3. Supplementary Table 2: I think adding the information about the electrode using NMP as a processing solvent is helpful (although it would be the same as H_2O 's case.)
4. Line 399: Based on Supplementary Tables 2 and 6, “NCM811/carbon black/binder” might be “95.5/2.0/2.5 (w/w/w)”. Please double-check it.
5. Lines 412–413: I guess the information about the diameter of each electrode and separator is missing.

Please double-check it and include this information if missing. (I guess it is 12 mm for the cathode and 18 mm for the separator from Supplementary Table 6, but how about the anode diameter?)

6. Fig. 5d: This is related to my comment (b-12). Does “the first cycle after the formation” mean “the second cycle including the formation (=0.1C/0.1C for one cycle)”? If so, please specify this in the caption of Fig. 5d.

7. I think the definition of 1C in the authors’ study has yet to be provided. Please double-check this and provide the definition (e.g., 1C = 205 mA/g-NCM811) if they have not done so.

8. Line 299: Please note that 205 mAh/g-NCM811 is not the theoretical capacity of NCM811 (based on full Li⁺ intercalation/deintercalation; e.g., Batteries 2023, 9(4), 193; <https://doi.org/10.3390/batteries9040193>). The authors can use “practical” or just delete “theoretical”.

9. I think the information about carbon black (i.e., supplier, grade, purity, etc.) is missing. Please provide it in the “Materials preparation” section.

10. Supplementary Table 4b: It would be nice if the authors could explain how the solution cost was calculated. This is because I could obtain a value that is nearly twice as large as the authors’ estimation for “NMP” and “Water” based on the provided information. For instance, in the case of “NMP”, 3.74 mAh/cm² and 205 mAh/g-NCM811 gives 0.01824 g-NCM811 (per 1 cm²). If I assume 47 wt% of NMP is used for the slurry preparation, the amount of NMP is: $0.01824 \text{ g} / 0.955 \times 47 / (100 - 47) = 0.01694 \text{ g}$. Multiplying this by 0.5 USD g⁻¹ (from Supplementary Table 3b) gives 0.00847 USD (> 0.00481 USD in Supplementary Table 4b).

11. Supplementary Table 4b: I feel it would be a fair comparison if the authors could use the fixed areal capacity for all cases. (I do not think this would significantly affect the result though.)

Finally, and additional suggestion as per Lines 261–269: the authors would further strengthen the significance of water-based cathode manufacturing if they could consider the additional costs in NMP-based cathode manufacturing using a higher slurry drying temperature during the coating process than water-based cathode manufacturing and an NMP recovery (or collection) system attached to a coater. Even without a detailed cost estimation for this point, just mentioning this with some references would be fine.

Reviewer #2 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4 (Remarks to the Author):

The authors carefully considered, responded, and incorporated changes/updates to the manuscript/supplementary information for the all the reviewer comments. I feel the authors did an outstanding job fully addressing all of my comments in the revised manuscript and I am satisfied with the changes the authors have made. Below, I have included one very minor suggestion for the authors. I

believe the revised manuscript provides significant insight that will advance the aqueous processing of electrodes and I recommend accepting the manuscript for publication in Nature Communications.

Minor comment: The authors should consider adding Fig. R6 (from response to Reviewer #1's comment a-2) to the supplementary information, since it provides a very nice visual summary of Supplementary Table 7 for the reader.

Response to reviewer comments

Manuscript ID: NCOMMS-24-14271-B

Title: Kosmotropic aqueous processing solution for green lithium battery cathode manufacturing

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Reviewer #1

The manuscript has been revised according to the reviewers' comments, and I think it has addressed most of the concerns. The current version can tell outstanding information about the anion choice for processing water-based slurries of a high-Ni cathode active material (without losing its expected performance), which could stimulate our transition to eco-friendly battery manufacturing. This is a great work for a broad battery community and I think can be accepted for publication after the following comments are addressed.

1. One concern that needs to be addressed relates the original review (suggestion in comment (a-1)). I believe Fig. 5a is still not a fair comparison as to the amount and type of salt remaining in the electrode layer, which therefore cannot fully support the sentence in Lines 244–246: “The capacity of the water- and chaotropic solution-processed cathodes degraded significantly after 200 cycles because of the active Li^+/H^+ exchange^{5,39} during the aqueous cathode fabrication (Supplementary Fig. 15 and Fig. 5a)”; i.e., we cannot say the observed difference in cyclabilities between the four electrodes is purely due to interfacial side reactions between water and NCM811. This is because, as the authors have described in many parts (e.g., Fig R2, Supplementary Figs 17 and 20, their answer to my comment (b-8), etc.), the remaining salt seemingly contributes to pore blocking, which leads to a rapid decay in the capacity of an NCM811 cathode. Ideally, the comparison between the four electrodes after removing the salt residue is still required to clarify this point and should be considered in future work. Despite the authors mentioning that removing the salt residue is challenging, I believe, for example, this can be done using the same process as (or a similar process to) the authors' sample-preparation method for XPS (i.e., soaking an electrode in DMC for 10 min and drying, where the solvent can be replaced by the new one several times to ensure that the remaining salt is fully removed). Whilst I am not suggesting these experiments for the current manuscript, I believe it is important to be explicit that this could also be an issue that influences the capacity stability.

→ **Response:** We appreciate for the reviewer's valuable comment. As the reviewer pointed out, the kosmotropic/chaotropic solution-processed cathode contain residual salts, which could affect the cycling performance. Despite the presence of residual salts, the kosmotropic solution-processed cathode exhibited a longer cycle life than the water-processed cathode, indicating that the kosmotropic processing solution effectively mitigated interfacial side reactions and

structural degradation of cathode active materials during the aqueous electrode manufacturing. We acknowledge the reviewer's suggestion to compare the four electrodes after removing the salt residues, specifically by performing a salt removal process on the kosmotropic and chaotropic solution-processed cathodes (excluding the NMP- and water-processed cathodes). This additional analysis will be conducted in future work, as the reviewer suggested. To address the reviewer's comment, we have revised the discussion regarding the effect of the chaotropic processing solution on cycling performance to avoid any misunderstanding. The reviewer's valuable comment is highly appreciated, again.

[Revised Manuscript]

“The capacity of the water-~~and chaotropic~~-processed cathodes degraded significantly after 200 cycles because of the active Li^+/H^+ exchange^{5,39} during the aqueous cathode fabrication (Supplementary Fig. 15 and Fig. 5a). **The chaotropic solution-processed cathode showed improved cyclability than the water-processed cathode, however, failed to maintain the cycle retention after 300 cycles.** In contrast, the kosmotropic solution-processed cathode delivered a high initial specific capacity of 205 mAh g_{NCM811}⁻¹ with a stable cycle retention (over 80% after 400 cycles), which is comparable to that of the NMP-processed cathode. To elucidate the underlying electrochemical mechanisms responsible for this enhanced cycling stability, we analyzed the cathodes after 400 cycles using electrochemical impedance spectroscopy (EIS, Supplementary Fig. 16).”

2. As per my original suggestion of my comment (b–3). If the authors could briefly describe each sample-preparation step of NCM811 for particle-size analysis, that would be informative. I could not fully understand the procedure between (i) soaking NCM811 in different processing solutions and (ii) measuring particle size distribution of NCM811 using ethanol as the solvent (Lines 388–389). Does this mean the processing solutions were removed and dried NCM811 was re-dispersed in ethanol for analysis?

→ **Response:** We sincerely appreciate the reviewer’s valuable comment. As mentioned by the reviewer, the NCM811 particles soaked in different processing solutions were dried and then redispersed in ethanol for the particle size analysis. Ethanol was chosen as a solvent because of its chemical stability with NCM811 particles and its effectiveness in maintaining stable particle suspension (**ref.:** *Angew. Chem. Int. Ed.* **59**, 14935 (2020)).

In response to the reviewer’s comment, we have added the details in the **Methods** to clearly describe this procedure.

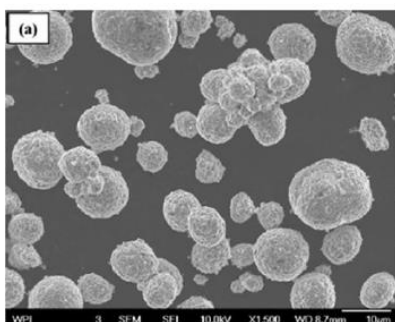


Figure 3. SEM images of NCM811 (a) dispersed for 24 h in ethanol with agitation of 500 rpm,

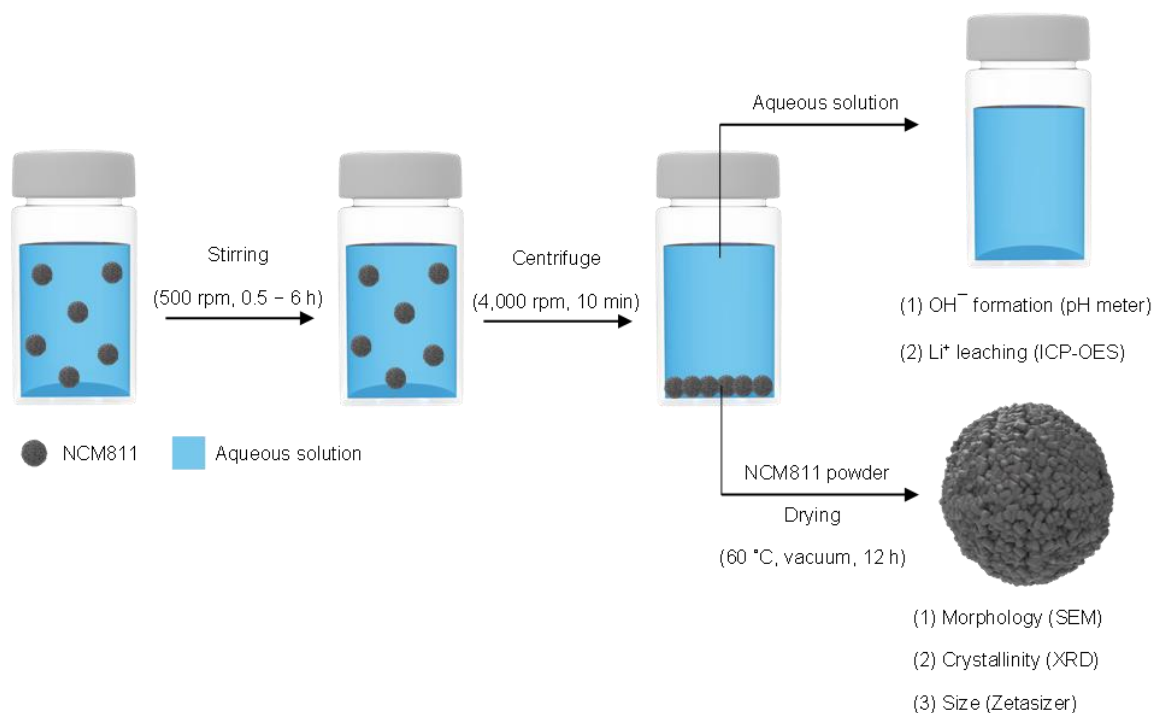
Fig. | A captured image from a research article published in *Angew. Chem. Int. Ed.* **59**, 14935 (2020).

[Revised manuscript]

“Structural and physicochemical characterization

To investigate chemical reactivity of the aqueous solutions with the cathode active materials, NCM811 particles were dispersed in different processing solutions and then subjected to centrifugation to collect the treated NCM811 particles and processing solutions, respectively. Details of the analysis procedure are provided in **Supplementary Fig. 8**. The change in pH value and Li^+ concentration of the obtained processing solutions were measured using a pH

meter (SevenExcellence S400, METTLER TOLEDO) and an inductively coupled plasma optical emission spectrometer (ICP-OES, 5110, Agilent), respectively. The collected particles were dried at 60°C for 12 h, and their X-ray diffraction (XRD) patterns were recorded using an X-ray diffractometer (SmartLab, Rigaku) at 45 kV and 200 mA using Cu K α radiation. For the particle size distribution analysis, the dried NCM811 particles were redispersed in ethanol and analyzed using a Zetasizer (ELSZneoZS, Otsuka).”



Supplementary Fig. 8 | Schematic depicting the study model used to investigate the chemical reactivity of the aqueous solutions with cathode materials for aqueous electrode processing.

3. Supplementary Table 2: I think adding the information about the electrode using NMP as a processing solvent is helpful (although it would be the same as H₂O's case.)

→ **Response:** Thank you so much for the reviewer's valuable suggestion. We have updated **Supplementary Table 2** to include information on the electrode fabricated using NMP as a processing solvent.

[Revised Manuscript]

Supplementary Table 2 | **a**, Composition of the electrode slurries containing 100 g of electrode components (cathode materials, binder, and conductive additive). **b**, Composition ratio of the aqueous-processed electrodes including that of the salt additives.

a

Processing solution	Cathode material [g]	Binder [g]	Conductive additive [g]	Salt additive [g]
NMP	95.5	2.5	2.0	-
H ₂ O	95.5	2.5	2.0	-
Chaotropic solution (1.0 m LiTFSI in H ₂ O)	95.5	2.5	2.0	11.5
Kosmotropic solution (0.5 m Li ₂ SO ₄ in H ₂ O)	95.5	2.5	2.0	2.6

b

Processing solution	Composition [wt.%]		
	Cathode material	Binder & conductive additive	Salt additive
NMP	95.5	4.5	-
H ₂ O	95.5	4.5	-
Chaotropic solution (1.0 m LiTFSI in H ₂ O)	85.9	4.0	10.1
Kosmotropic solution (0.5 m Li ₂ SO ₄ in H ₂ O)	93.1	4.4	2.5

4. Line 399: Based on Supplementary Tables 2 and 6, “NCM811/carbon black/binder” might be “95.5/2.0/2.5 (w/w/w)”. Please double-check it.

→ **Response:** Many thanks for the reviewer’s valuable comment. In response to the reviewer’s comment, we have revised the electrode composition ratio in **Methods**.

[Revised Manuscript]

“The electrode slurries were prepared with a composition of $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2(\text{NCM811})/\text{carbon black/binder} = 95.5/2.0/2.5$ (w/w/w).”

5. Lines 412–413: I guess the information about the diameter of each electrode and separator is missing. Please double-check it and include this information if missing. (I guess it is 12 mm for the cathode and 18 mm for the separator from Supplementary Table 6, but how about the anode diameter?)

→ **Response:** We appreciate the reviewer’s valuable comment. In response to the comment, we have added the diameters of the cell components in **Methods**.

[Revised Manuscript]

“For the electrochemical tests, CR2032 coin-type cells (Ni-rich cathode (diameter = 12 mm)||polyethylene (PE) (thickness = 16 μ m and diameter = 18 mm)||Li-metal anode (thickness = 100 μ m and diameter = 16 mm)) were assembled in an Ar-filled glove box.”

6. Fig. 5d: This is related to my comment (b-12). Does “the first cycle after the formation” mean “the second cycle including the formation (=0.1C/0.1C for one cycle)”?. If so, please specify this in the caption of Fig. 5d.

→ **Response:** Thank you for the reviewer’s valuable comment. As the reviewer mentioned, “the first cycle after the formation” refers to the second cycle, followed by the one formation cycle. To clarify this, we have revised the caption of **Fig. 5d**.

[Revised Manuscript]

Fig. 5 | Electrochemical performance of NCM811 electrodes prepared by aqueous processing solutions. d, Galvanostatic charge/discharge profiles as a function of mass loading of kosmotropic solution-processed cathodes at a current rate of 0.1C/0.1C. **These profiles were obtained after one formation cycle.**

7. I think the definition of 1C in the authors' study has yet to be provided. Please double-check this and provide the definition (e.g., 1C = 205 mA/g-NCM811) if they have not done so.

→ **Response:** Many thanks for the reviewer's detailed comment. In response to the reviewer's comment, we have added the definition of 1C in our study, specifying it as $1\text{ C} = x\text{ mA g}^{-1}\text{ active material}$ ($x = 195, 205, \text{ and } 215$ for NCM622, NCM811, and NCM 9½½ active material, respectively). This definition has been included in the **Methods**. The reviewer's comment is highly appreciated, again.

[Revised Manuscript]

““After the cell assembly, a rest period of 12 h was allowed to ensure complete wetting of the electrodes. The cells were then subjected to one formation cycle at a current rate of 0.1C (**1C = $x\text{ mA g}^{-1}\text{ active material}$ ($x = 195, 205, \text{ and } 215$ for NCM622, NCM811, and NCM 9½½ active material, respectively)**). The cells were cycled at a charge/discharge current rate of 0.2 C/0.5 C and a voltage range of 3.00–4.25 V at 25°C, if not specified.”

8. Line 299: Please note that 205 mAh/g-NCM811 is not the theoretical capacity of NCM811 (based on full Li⁺ intercalation/deintercalation; e.g., Batteries 2023, 9(4), 193; <https://doi.org/10.3390/batteries9040193>). The authors can use “practical” or just delete “theoretical”.

→ **Response:** Thank you for the reviewer’s valuable comment. In response to the reviewer’s comment, we have revised the sentence to “practical capacity”.

[Revised Manuscript]

“Electrochemical analysis was extended to high areal-loading NCM811 cathodes (**Fig. 5d**). The kosmotropic solution-processed cathodes stably maintained the specific capacity of NCM811 ($\sim 205 \text{ mAh g}_{\text{NCM811}}^{-1}$) up to an areal mass loading of 50 mg cm^{-2} ($\sim 10 \text{ mAh cm}^{-2}$). In addition, NCM622, NCM811, and NCM9 $\frac{1}{2}$ $\frac{1}{2}$ processed with the kosmotropic solution achieved capacities of 195, 205, and 215 mAh g⁻¹, respectively, aligning closely with their **practical** capacities (**Supplementary Fig. 22**).”

9. I think the information about carbon black (i.e., supplier, grade, purity, etc.) is missing. Please provide it in the “Materials preparation” section.

→ **Response:** Many thanks for the reviewer’s valuable comment. We have included this detailed information on the carbon black in the “Materials preparation” section. Specifically, carbon black (Super P) powders used as a conductive additive were supplied by TIMCAL.

“Material preparation

Lithium sulfate (Li_2SO_4 , 99.5%), Lithium acetate (LiOAc , 99.95%), Lithium trifluoromethanesulfonate (LiOTf , 96.0%), N-methyl-2-pyrrolidone (NMP, 99.5%), carboxymethyl cellulose (CMC, $M_w = 250,000$, degree of substitution = 0.9), and polyacrylic acid (PAA, $M_w = 45,000$, purity) were purchased by Sigma-Aldrich. Li bis(trifluoromethanesulfonyl)imide (LiTFSI , 99.0%) and polyvinylidene fluoride (PVDF) were bought from Samchun and Solvay, respectively. Carbon black (Super P) powders used as a conductive additive were supplied by TIMCAL.”

10. Supplementary Table 4b: It would be nice if the authors could explain how the solution cost was calculated. This is because I could obtain a value that is nearly twice as large as the authors' estimation for "NMP" and "Water" based on the provided information. For instance, in the case of "NMP", 3.74 mAh/cm² and 205 mAh/g-NCM811 gives 0.01824 g-NCM811 (per 1 cm²). If I assume 47 wt% of NMP is used for the slurry preparation, the amount of NMP is: $0.01824 \text{ g} / 0.955 \times 47 / (100 - 47) = 0.01694 \text{ g}$. Multiplying this by 0.5 USD g⁻¹ (from Supplementary Table 3b) gives 0.00847 USD (> 0.00481 USD in Supplementary Table 4b).

→ **Response:** Thank you so much for the reviewer's detailed analysis and for pointing out the discrepancy in the cost calculations. In response to the reviewer's comment, we have updated **Supplementary Table 4b** to reflect the corrected cost estimates. In addition, we have included the solid content of the electrode slurry to help readers better understand the cost estimation.

[Revised Manuscript]

Supplementary Table 4 | **a**, Cost information of the cathode components. Material specifications are described in Methods. **b**, Calculation details of the specific capacity based on the material cost of cathodes and processing solutions, which are normalized by electrode area of 1 cm². **The solid content of the electrode slurries was 53 wt.%. The material cost information was obtained from their suppliers.**

a

Component	Material	Weight [g]	Cost [USD]	Specific cost [USD g ⁻¹]
Cathode material	NCM811	500	345	0.69
Binder	PAA	250	407	1.63
	CMC	1,000	168	0.17

	PVdF	100	120	1.20
Conductive additive	Carbon black	400	214	0.54
Component	Material	Area [m ²]	Cost [USD]	Areal cost [USD m ⁻²]
Current collector	Al	98	295	3.01

b

Electrodes (prepared using the following solution)	Areal capacity [mAh cm ⁻²]	Cathode cost [USD]	Solution cost [USD]	Capacity normalized by material cost [mAh USD ⁻¹]
NMP	3.74	0.01367	0.00847	169
H ₂ O	3.61	0.01352	0.00008	265
Chaotropic solution (1.0 m LiTFSI in H ₂ O)	3.59	0.01352	0.02101	104
Kosmotropic solution (0.5 m Li ₂ SO ₄ in H ₂ O)	3.74	0.01352	0.00034	270

11. Supplementary Table 4b: I feel it would be a fair comparison if the authors could use the fixed areal capacity for all cases. (I do not think this would significantly affect the result though.)

→ **Response:** Thank you so much for the reviewer's valuable comment. In **Supplementary Table 4b**, the mass loading of the electrode active material is identical for all electrodes. Therefore, the variation in the areal capacity results from the difference in the discharge capacity observed for each electrode (shown in **Fig. 5a**). This indicates that the difference in the areal capacity reflects the performance characteristics of each electrode. We appreciate the reviewer's insightful comment, again.

12. Lines 261–269: the authors would further strengthen the significance of water-based cathode manufacturing if they could consider the additional costs in NMP-based cathode manufacturing using a higher slurry drying temperature during the coating process than water-based cathode manufacturing and an NMP recovery (or collection) system attached to a coater. Even without a detailed cost estimation for this point, just mentioning this with some references would be fine.

→ **Response:** Many thanks for the reviewer’s insightful comment. We agree that including the additional operating costs associated with the NMP-based cathode manufacturing, such as the higher slurry drying temperature and the use of an NMP recovery system, would provide a more comprehensive comparison. Please note that this information has been provided in **Fig. 6**, which describes the energy and cost analysis of water-based and NMP-based cathode manufacturing. The reviewer’s valuable comment is highly appreciated, again.

[Original Manuscript]

“Economic and environmental analysis of kosmotropic aqueous cathode manufacturing process

To explore the practical implications of these scientific findings for industrial application, we designed process flow diagrams (PFDs) for the manufacturing electrode with NMP solvent and kosmotropic aqueous processing solutions (**Fig. 6a**, **Supplementary Fig. 24**, **Supplementary Table 8**, and **Supplementary Note 4**). The NMP-processed cathodes were dried at high temperatures, followed by a recovery step of NMP vapor via condensation (emphasized within the dashed outline in **Fig. 6a**). The recycling step is imperative given the high cost of NMP and its environmental impacts upon atmospheric release, and accounts for 45% of the total energy consumption in electrode processing (including slurry mixing, coating, drying, solvent recovery) (**Supplementary Tables 9, 10**, and **Supplementary Note 5**).

In contrast, the kosmotropic solution-based process eliminates the need for a solvent recovery step. Furthermore, the drying process of the aqueous electrode consumes less energy because of its lower vapor pressure compared to NMP. Overall, the kosmotropic aqueous cathode processing results in a 97.4% decrease in the energy consumption and a 97.1% reduction in CO₂ emissions compared to the NMP-based process (**Fig. 6b** and **Supplementary Fig. 25**). Considering that the electrode processing accounts for 47% of energy consumption in battery manufacturing⁴¹, it implies that the kosmotropic aqueous electrode processing can

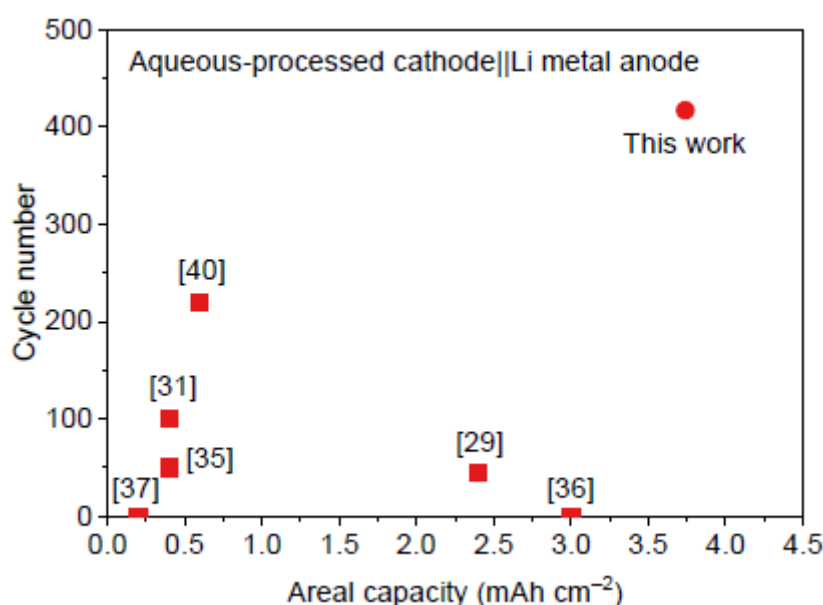
reduce 46% of the energy consumption, demonstrating its sustainable manufacturing. Moreover, the intrinsic simplicity of kosmotropic aqueous cathode processing translates to a 95% reduction in the initial capital expenditure (**Fig. 6c**) and a 23% decrease in the operating costs (**Fig. 6d**).”

Reviewer #4

1. The authors should consider adding Fig. R6 (from response to Reviewer #1's comment a–2) to the supplementary information, since it provides a very nice visual summary of Supplementary Table 7 for the reader.

→ **Response:** Thank you so much for the reviewer's valuable comment. In response to the reviewer's comment, we have included **Fig. R6** as **Supplementary Fig. 23** in the revised manuscript. Again, we highly appreciate the reviewer's comment.

[Revised Manuscript]



Supplementary Fig. 23 | Comparison of the kosmotropic aqueous solution-processed cathode (this work) and the previously reported aqueous solution-processed cathodes, in terms of areal capacity and cycle number of the cathode containing Li-metal batteries. The inset numbers correspond to the reference listed in **Supplementary Table 7**.

“To highlight the improved cell performance, the kosmotropic solution-processed cathodes and previously reported aqueous solution-processed cathodes are compared in **Fig. 5f**, **Supplementary Fig. 23**, and **Supplementary Table 7**.”

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

Review Comments for NCOMMS-24-14271B

The manuscript has been properly revised, and I can recommend its publication.

Just one minor comment related to my comment (10) in the previous round:

The authors would want to double-check the values in Supplementary Table 4b because I could not reproduce some values based on the provided information. Please collect my calculation sheet (Excel file) for double-checking (especially for the values colored in red) in attachment.

I believe no additional review is required. I look forward to seeing the published version shortly and its positive impact on the battery community in the foreseeable future.

Reviewer #2 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Response to reviewer comments

Manuscript ID: NCOMMS-24-14271-C

Title: Kosmotropic aqueous processing solution for green lithium battery cathode manufacturing

Reviewer #1

The manuscript has been properly revised, and I can recommend its publication.

Just one minor comment related to my comment (10) in the previous round:

The authors would want to double-check the values in Supplementary Table 4b because I could not reproduce some values based on the provided information. Please collect my calculation sheet (Excel file) for double-checking (especially for the values colored in red) in attachment.

I believe no additional review is required. I look forward to seeing the published version shortly and its positive impact on the battery community in the foreseeable future.

→ **Response:** We appreciate for the reviewer's valuable comment. When we double-checked the reviewer's calculations, we found two key errors:

1. The mass loading of active materials for all electrodes should be 18.2 mg/cm². Please note that the difference in the areal capacity is due to "under-utilization of specific capacity" in chaotropic and water-processed electrodes, rather than variations in mass loading.
2. The cost of super P should be included in the calculation for both chaotropic and kosmotropic-processed cathodes

Please see table below.

We hope these clarifications address the reviewer's concerns, and we are pleased to provide further details if needed. Once again, many thanks for the reviewer's constructive feedback.

Reviewer's calculation										
			g/cm2				USD/cm2			
Material		[USD g-1]	NMP	Water	Chatropic	Kosmotrop	NMP	Water	Chatropic	Kosmotropic
Electrode	NCM811	0.69	0.01824	0.01761	0.01751	0.01824	0.012588	0.0121509	0.0120819	0.0125856
Binder	PAA	1.628		0.00024	0.00024	0.00024		0.0003888	0.0003888	0.0003888
	CMC	0.168		0.00024	0.00024	0.00024		4.012E-05	4.012E-05	4.012E-05
	PVdF	1.2	0.00048				0.000573			
	Carbon black	0.535	0.00038	0.00038			0.000204	0.0002044	0	0
Solvent	NMP	0.5	0.01694				0.00847			
	Water	0.005		0.01635				8.175E-05		
	Chao	1.24			0.01626				0.0201624	
	Kos	0.02				0.01694				0.0003388
Current collector							0.0003	0.0003	0.0003	0.0003
Sum							0.022136	0.0131659	0.0329732	0.0136533
Cost (USD/cm2)							3.74	3.61	3.59	3.74
Specific capacity (mAh/USD)							168.957	274.19249	108.87638	273.92695
Our calculation										
			g/cm2				USD/cm2			
Material		[USD g-1]	NMP	Water	Chatropic	Kosmotrop	NMP	Water	Chatropic	Kosmotropic
Electrode	NCM811	0.69	0.01824	0.01824	0.01824	0.01824	0.012588	0.0125883	0.0125883	0.0125883
Binder	PAA	1.628		0.00024	0.00024	0.00024		0.0003888	0.0003888	0.0003888
	CMC	0.168		0.00024	0.00024	0.00024		4.012E-05	4.012E-05	4.012E-05
	PVdF	1.2	0.00048				0.000573			
	Carbon black	0.535	0.00038	0.00038	0.00038	0.00038	0.000204	0.0002044	0.0002044	0.0002044
Solvent	NMP	0.5	0.01694				0.00847			
	Water	0.005		0.01694				0.0000847		
	Chao	1.24			0.01694				0.0210056	
	Kos	0.02				0.01694				0.0003388
Current collector							0.0003	0.0003	0.0003	0.0003
Sum							0.022136	0.0136063	0.0345272	0.0138604
Cost (USD/cm2)							3.74	3.61	3.59	3.74
Specific capacity (mAh/USD)							168.957	265.31874	103.97607	269.83395