

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

SAMSUNG ELECTRONICS CO., LTD.,
Petitioner

v.

RJ TECHNOLOGY LLC,
Patent Owner.

Case IPR2023-01183

Patent No. 7,749,641

DECLARATION OF DR. YANLIANG LIANG

I. Introduction

1. My name is Yanliang (Leonard) Liang. This declaration describes experiments I conducted based on U.S. Patent Application Publication No. 2002/0012830 (“Uemura”).

2. Unless otherwise noted, the statements made herein are based on my personal knowledge, and if called to testify about this declaration, I could and would do so competently and truthfully.

3. My CV is being submitted herewith as Ex. 2032 to this proceeding and is summarized in Section I, *infra*.

4. I have been retained by counsel for Patent Owner in these proceedings for the exclusive purpose of independently replicating certain testing and experiments described in Uemura at the direction of Dr. Yan Yao. I am being compensated at my customary rate for the time I spend on this matter. My compensation is not contingent on the content of my opinions nor the outcome of the *inter partes* review proceeding. I have no direct financial interest in any of the parties or this proceeding.

5. The opinions I set forth herein are my own and are based on my education, experience, training, and skill I have accumulated.

II. Qualifications

6. I am a co-founder and the CTO of LiBeyond LLC. LiBeyond LLC is a company based in Texas that focuses on research and development of new batteries, including testing of such batteries before they are sold on the market.

7. My primary research focuses on organic functional materials, solid-state batteries, multivalent metal batteries, aqueous batteries, and lithium and sodium batteries.

8. I received my undergraduate Bachelor of Sciences degree chemistry at Nankai University in Tianjin China, in 2007. While pursuing my undergraduate degree I was recognized as a Merit Student of Nankai University and received an Outstanding Student Scholarship (2004-2007).

9. In 2012, I completed a Ph.D. in materials physics and chemistry at Nankai University. During that time, I received various honors and awards including the Outstanding Graduate Student Scholarship (2008-2011), Excellent Paper Award at the 27th Chinese Chemical Society Congress in Xiamen, ChemComm Poster Prize at the 5th ChemComm International Symposium (2011), Yang Shi-Xian Scholarship (the highest honor a student can receive from the College of Chemistry) (2011), Sinopoly Battery Scholarship (2012), and the Excellent Graduates of NKU (awarded to the top 7% graduates) (2012).

10. After completing my graduate studies, I joined the Cullen College of Engineering in 2012 as a Postdoctoral Fellow working with Dr. Yan Yao. In this time, I have been the Principal Investigator (“PI”) or Co-Principal Investigator (“Co-PI”) of various research projects related to batteries that have received over \$2.5 million in funding. These include the projects: “Reliable Fabrication of All-Solid-State Lithium Batteries with High Cell-Level Specific Energy” funded by the U.S. Department of Energy (“DOE”), Office of Science, “Lithium and transition metal-free high-energy fast-charging batteries” funded by the U.S. DOE, ARPA-E, “Bioderived and biodegradable redox polymers for flexible wearable batteries” funded by Meta Platforms, Inc., and “Multidimensional diagnostics of the interface evolutions in solid-state lithium batteries” funded by the U. S. DOE, Vehicle Technology Office.

11. Based on my research on lithium-ion batteries, I have authored or contributed to 67 papers related to material sciences (many of which focus specifically on different types of batteries), been awarded 13 patents (5 U.S. Patents), provided 7 media reports, and delivered 10 invited talks, seminars, or guest lectures. In 2015 I was awarded the Nano Research Poster Award (Silver Award) at the 10th Sina-US Nano Symposium in Wuhan, China.

12. I am an inventor in the field of material science. I have been awarded 13 different patents, all related to electrochemistry and many deal specifically with

electrode materials for batteries. In 2021, I co-founded LiBeyond, LLC which strives to develop innovative battery solutions that meet or exceed the requirements for energy storage applications in the 21st century and beyond. More specifically, we pioneer novel battery chemistries and engineer solutions and then move these from initial discovery into larger formats demonstrating scalability with practical solutions for further development and evaluation with our vertical integration partners. We are currently focused on two technology platforms. The first is a Mg-Organic battery system that will compete with Li battery technologies with fast charging capability specifically targeted for electric vehicle applications. This business platform is supported with ARPA-E funding (OPEN 2021, subrecipient of UH) and includes Toyota North America as one of our vertical partners. The second, which is still in the ideation phase, is a variety of solid-state battery materials and manufacturing solutions also targeting electric vehicle applications.

13. I also actively participate in various professional organizations and outreach programs. I am a Senior Member of the Institute of Electrical and Electronics Engineers (IEEE). In 2019, I participated in the NASA High School Aerospace Scholars (HAS) lab tour with hands-on activity, Program for Mastery in Engineering Studies (PROMES) Engineering Summer Camp lab tour with hands-on activity, and NSF Research Experiences for Undergraduates lab tour. In 2014 I was

a judge at the International Sustainable World (Energy, Engineering & Environment) Project Olympiad (I-SWEEEP) International Science Fair.

III. Overview of experiments performed

14. I was asked to perform testing based on experimental procedures described in Uemura. Specifically, I was asked to (1) synthesize the cathode active materials in Examples 1-4 of Uemura, and if successful, assemble and test batteries using those cathode active materials under charge and discharge conditions described in Uemura; and (2) reproduce the battery as described as Example 2 in Uemura.

15. In this declaration, I provide details related to the experiments I conducted, including the synthesis of the cathode active materials, the characterization of the cathode active materials, cell assembly, and electrochemical testing.

IV. Synthesis of $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$ according to Uemura does not result in a layered material, as described in Uemura

16. I will first discuss the synthesis I performed. The cathode active materials in Examples 1-4 of Uemura follow the same chemical formula: $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$.¹ “x” is the amount of lithium in the cathode active material. The positive electrode (cathode) active material in Example 1 has 0.67 Li, the one in

¹ Ex. 1009, Fig. 3.

Example 2 has 0.83 Li, and Example 3 and Example 4 use the same cathode active material, which has 0.967 Li.²

17. Uemura states that “[t]he positive electrode materials of the rechargeable lithium batteries in examples 1 to 9 were prepared by the use of a coprecipitation method described” in paragraph 71.³

18. I tried to synthesize $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$ with the method described in paragraph 71 of Uemura. In particular, $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (9.19 g) and $\text{Mn}(\text{NO}_3)_2$ (50 wt.% aqueous solution, 11.30 g) were dissolved with H_2O into a 1 M solution, which was added dropwise into a stirred 10 wt.% solution of $\text{LiOH} \cdot \text{H}_2\text{O}$ (7.95 g) in H_2O over 30 min. The pH stayed higher than 9 throughout. The mixture was then filtered and washed with H_2O until the pH approximated 7. The resulting cake was vacuum-dried at 60 °C to yield the intermediate as deep gray powders.

19. The intermediate (1.5 g) was then finely ground with different amounts of $\text{LiOH} \cdot \text{H}_2\text{O}$ (0.464 g for $\text{Li}_{0.67}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$; 0.574 g for $\text{Li}_{0.83}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$, 0.669 g for $\text{Li}_{0.967}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$) with mortar and pestle. The mixtures were annealed in argon at 500 °C for 5 h then 900 °C for 24 h before left to cool to room temperature.

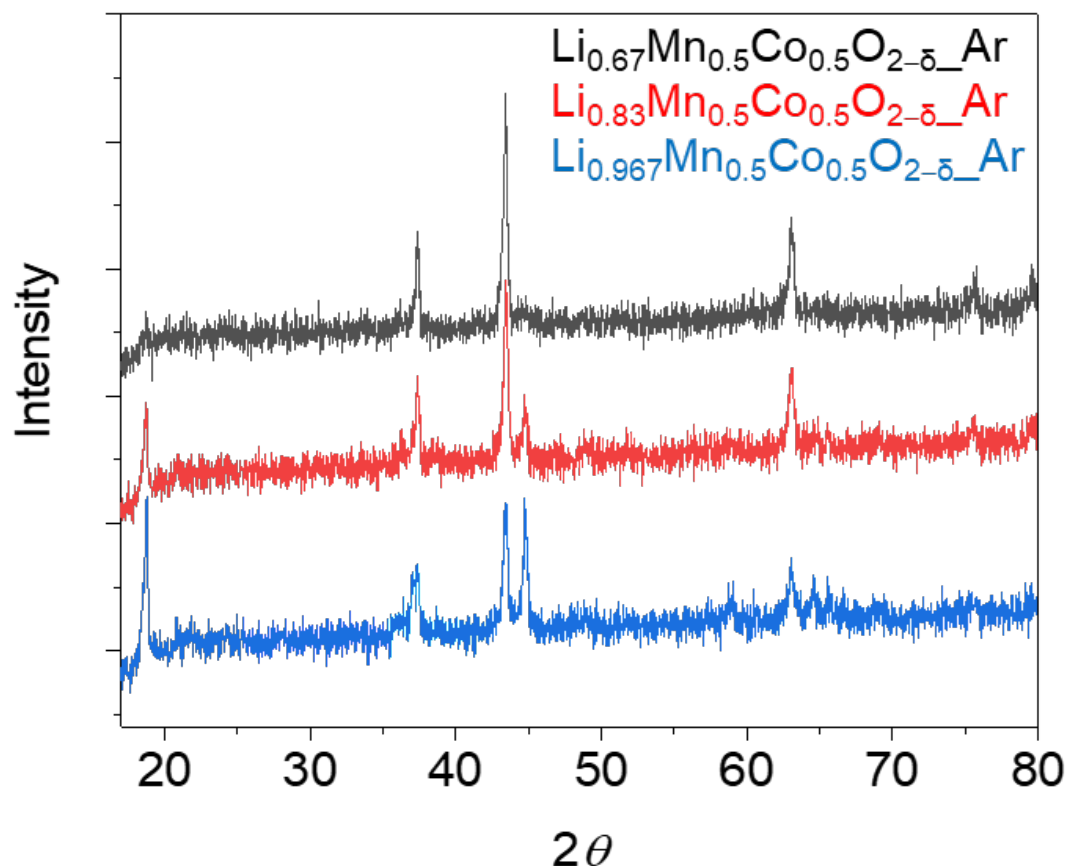
² *Id.*

³ *Id.*, ¶69.

20. The resulting products were deep gray powders. I then prepared each of the three samples (one for Example 1, one for Example 2, and one for Examples 3-4, which use identical cathode active material according to Uemura) for XRD testing.

21. XRD, or X-ray diffraction, is a widely used method to study the structural properties of a material. In XRD, a sample is irradiated with X-rays and the X-rays diffracted by the sample are recorded by a detector. XRD measures the way in which a crystalline structure diffracts X-rays. Based on the diffraction of the X-rays, the technique produces a pattern of peaks. The position, intensity, width, and shape of the peaks form the diffraction pattern. As a general matter, each crystalline form of a compound has its own characteristic or unique diffraction pattern. I used the XRD testing to ascertain whether the three samples had crystal structures that matched the ones described in Uemura. This was to ensure that my charge and discharge tests would be performed on the same types of materials as those in Uemura and that the materials in Uemura could in fact be produced with the method disclosed in Uemura.

22. The XRD patterns for the three samples are shown below:



23. According to the generated XRD patterns, the resulting powders likely have any one of the following structures: rock salt structure,⁴ a spinel structure,⁵ or a mixture of rock salt structure and spinel structure. The patterns also demonstrate

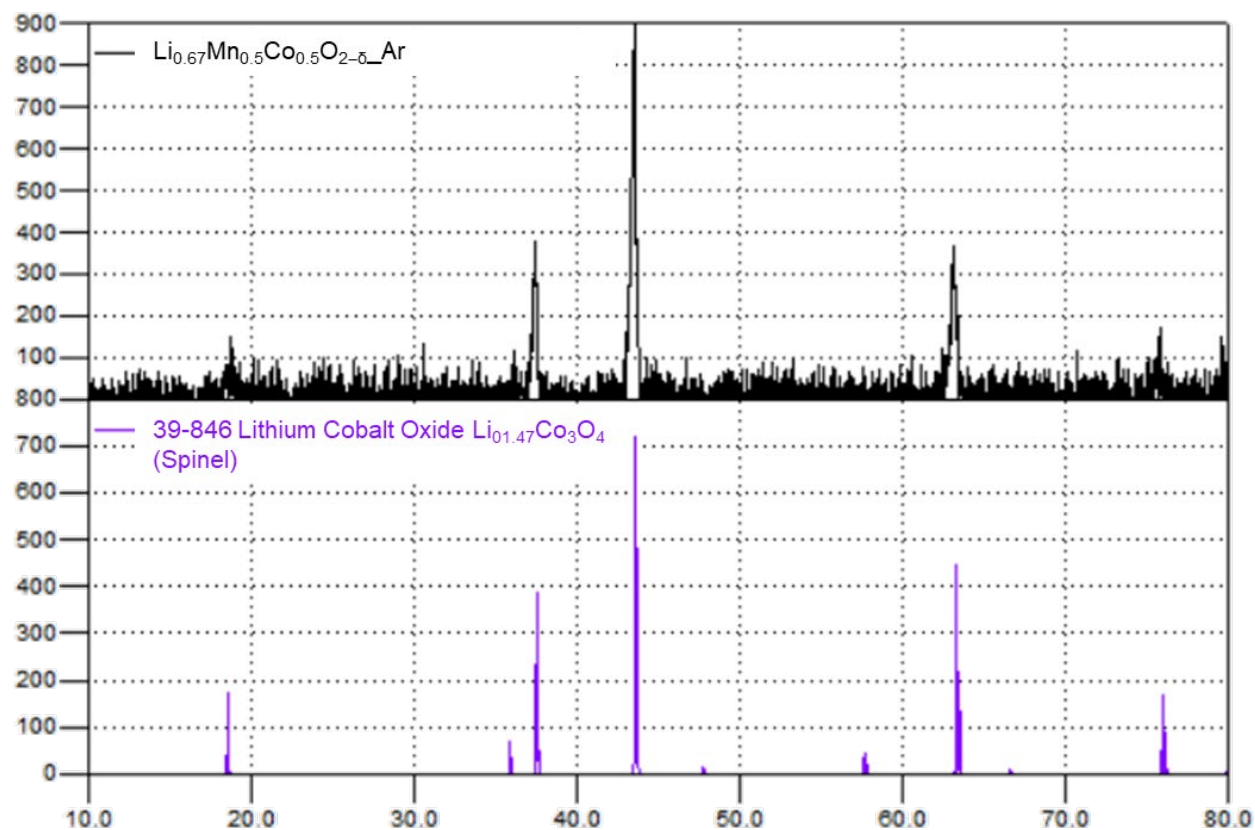
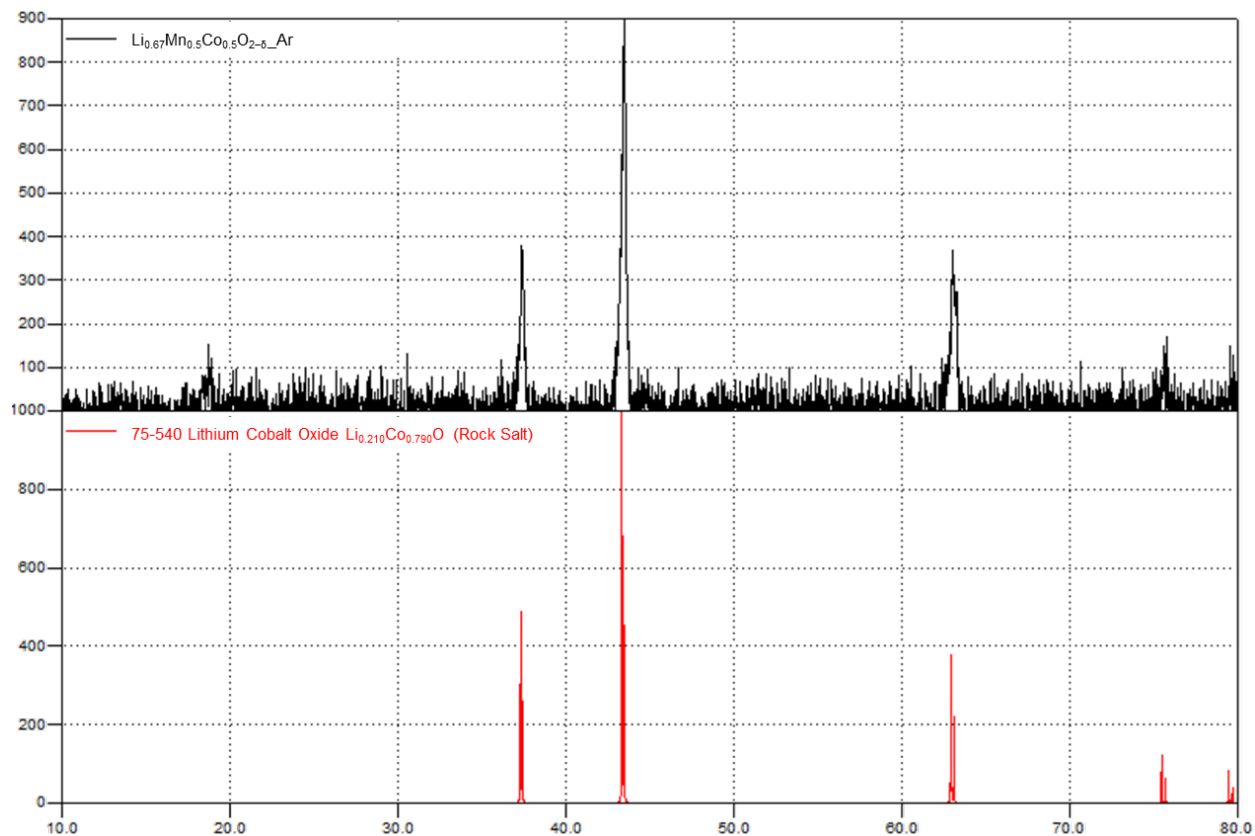
⁴ See Ex. 2034, W.D. Johnston et al., *The preparation, crystallography, and magnetic properties of the $\text{Li}_x\text{Co}_{(1-x)}\text{O}$ system*, J. Phys. Chem. Solids. Pergamon Press 1958, Vol. 7, 1-13.

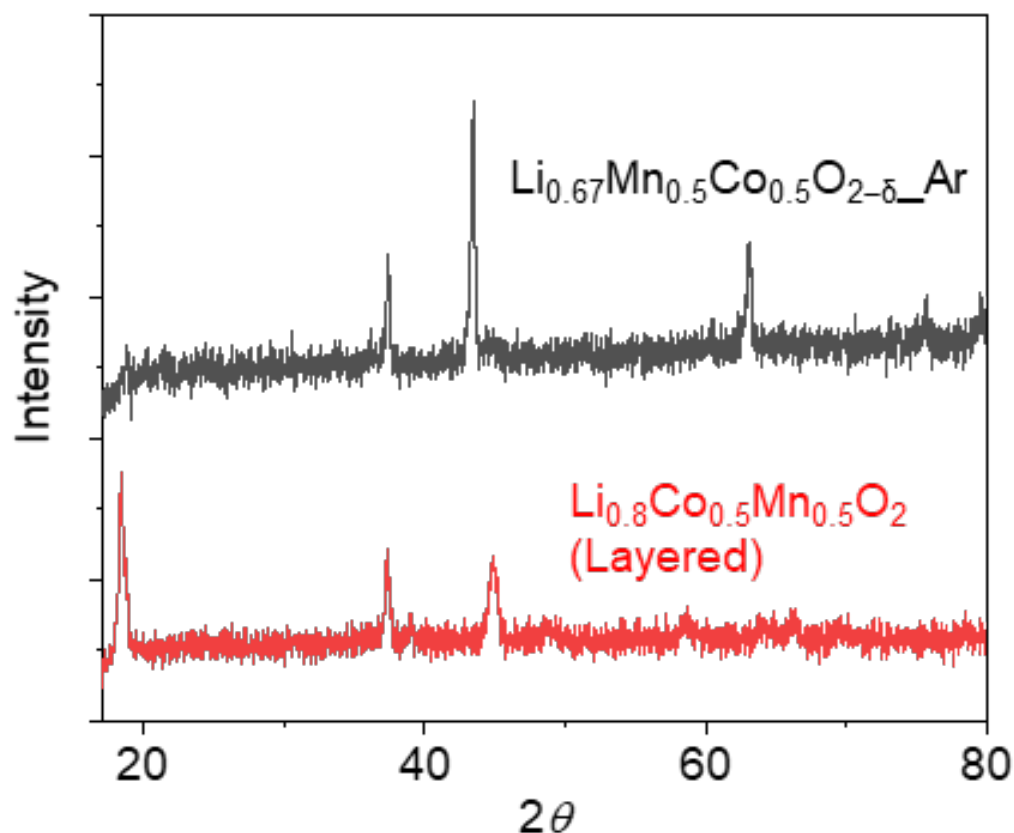
⁵ Ex. 2036, M.M. Thackeray et al., *Lithium insertion into Co_3O_4 : a preliminary investigation*, 17 Solid State Ionics 175 (1985).

to one of ordinary skill that the three samples do not possess a layered structure as described in Uemura.⁶

24. The samples do not have a layered structure because their peak patterns are more consistent with peak patterns of materials with a rock salt and/or spinel structure: if two compounds have similar XRD patterns, their crystal structures should be similar too. As shown below, the peak pattern of $\text{Li}_{0.67}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$ _Ar (i.e., the material I synthesized according to Uemura's Example 1) is consistent with the peak pattern for lithium cobalt oxides with rock salt and/or spinel structures. The peak pattern is also different from the XRD pattern of a doped layered LiMnO_2 . I thus conclude that the three samples I synthesized by following Uemura's experimental procedures do not have a layered structure.

⁶ Uemura, Fig. 3,





V. Testing of Uemura’s non-layered cathode material shows that Uemura’s synthesis procedure does not produce a working cathode

25. I then assembled three test cells (again, one for Example 1, one for Example 2, and one for Examples 3 and 4, as described above) with the materials prepared above (i.e., annealed in argon) and performed charge and discharge tests on each of them under charging and discharging conditions similar to Uemura’s.⁷ These tests were conducted in half-cell configurations, i.e., with lithium metal as the counter electrode, to determine the charging capacity of these synthesized materials.

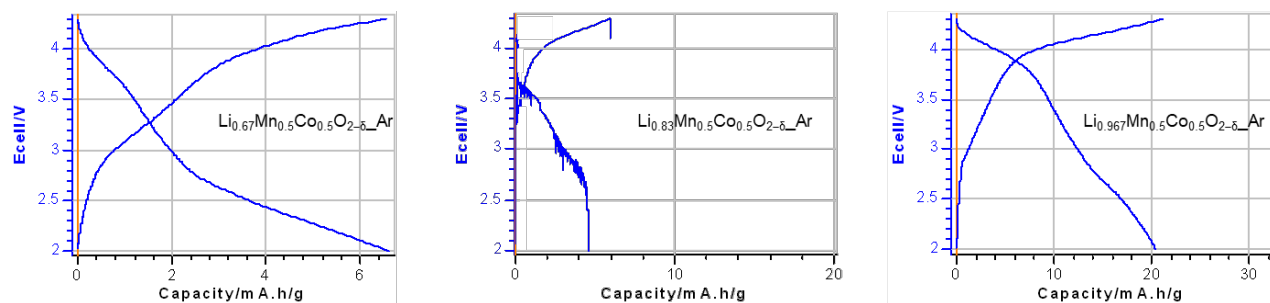
⁷ Uemura, ¶¶75, 81.

To perform these tests, I performed the following steps: (i) I mixed each of the synthesized materials with acetylene black and PTFE in the weight ratio of 80:16:4 to form a single self-standing electrode with a diameter of 7.14 mm and an areal loading of $\sim 13 \text{ mg/cm}^2$, (ii) I assembled the cathode into a Swagelok cell with a Mo current collector, a polyethylene separator, a lithium metal counter electrode (diameter: 7.94 mm), and a solution of LiPF_6 (1 M) in ethylene carbonate and dimethyl carbonate (1:1 w/w), and (iii) I galvanostatically cycled each cell at 0.47 mA/cm^2 at 60°C between 2.0 and 4.3V. I note that some of the parameters for this testing are not given in Uemura (e.g., the type of current collector). I therefore replicated Uemura's experiment drawing from my knowledge and experience.

26. The figures below show the first charge curve and the first discharge curve of the three test batteries assembled above. For the three samples of $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}\text{-Ar}$ prepared based on the experimental procedures described by Uemura, all three test batteries had a minimal amount of capacity under the charging conditions set forth above.

27. In particular, the first plotted graph on the left, which corresponds to the first battery cell, shows two curves in which capacity (on the x axis) is plotted against applied voltage (on the y axis). The curve trending upward corresponds to the first charge cycle; and the curve trending downward corresponds to the first discharge cycle. As can be seen in the upward trending curve, increasing the applied

voltage up to 4.3V produced a charge capacity of the cathode of approximately 6 mAh/g, which a person of ordinary skill would recognize is insufficient for a working secondary lithium-ion battery. The other two plotted graphs have similar charge capacity characteristics (i.e., the charge capacities were below 10mAh/g and around 20mAh/g, respectively). This would lead a person of ordinary skill to understand that in all three examples, attaining an acceptable cathode capacity would likely require the battery to be charged at voltages well over 4.3V, or the material synthesized simply could not be used as a working cathode material.



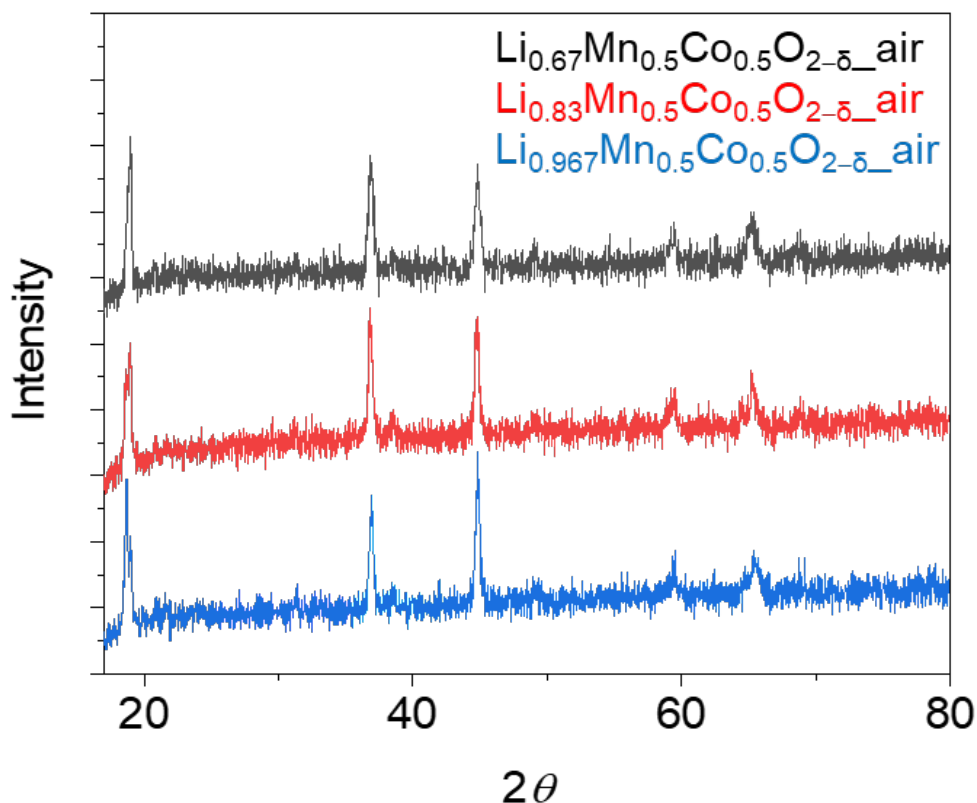
28. Thus, the cathode material produced following Uemura's synthesis procedure does not produce a working cathode.

VI. Synthesis of $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$ according to a process that improves Uemura's procedure likewise does not result in a layered material, as described in Uemura

29. Based on my experience, layered structure of lithium metal oxides is typically annealed in air. I thus tried to see if layered structure of $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$ can be obtained by annealing the sample in air. I revised the last step of the experimental procedure in Uemura to anneal the mixtures in air (rather than argon,

as described in Uemura) at 500 °C for 5 hours then 900 °C for 12 hours. The other steps remained the same.

30. I then prepared the samples for XRD analysis, and the XRD patterns are shown below.



31. Based on the XRD peak patterns, the products have mixed phases which appear to be spinel structures.⁸

⁸ See Ex. 2035, K. Banov et al., *High voltage cathode materials based on lithium cobaltate with nickel and manganese doping*, Bulgarian Chemical Communications, (continued...)

32. Layered and spinel structures synthesized at high temperatures such as 900 °C typically can be distinguished from their XRD patterns. One distinguishing characteristic is the peaks around 66°. If there are two distinct peaks, that sample may have a layered structure. In contrast, if the sample only has one peak around 66°, the most reasonable interpretation is that the sample has a spinel structure. Here, for all three samples, there is one peak around 66°, thus confirming that those samples had a spinel structure.

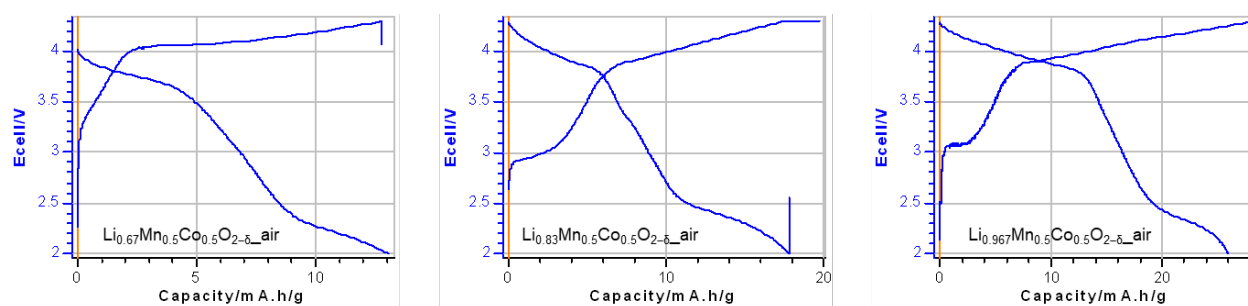
VII. Testing of the non-layered cathode material shows that the improved procedure also does not produce a working cathode

33. I assembled test cells in the same way as the test cell using argon-annealed synthesized materials (as discussed in section IV). Below are figures showing the first charge and the first discharge of each of those test cells assembled (annealed in air). The air annealed samples similarly had cathodes with a minimal amount of capacity.

34. In particular, using the first plotted graph on the left as an example (which corresponds to the first cell with cathode active material annealed in air), there are two curves. The curve trending upward corresponds to the first charge

Volume 50, Special Issue A, 171 (2018); Ex. 2021, C. Julien et al., *Combustion synthesis and characterization of substituted lithium cobalt oxides in lithium batteries*, 135 Solid State Ionics 241 (2000).

cycle, and the curve trending downward corresponds to the first discharge cycle. As can be seen in the upward trending curve, increasing the applied voltage to 4.3V produced a charge capacity of about 13 mAh/g, which a person of ordinary skill would recognize is insufficient for a working lithium-ion battery. The other two plotted graphs have similar charge characteristics (i.e., the charge capacity was around 18 mAh/g and 26 mAh/g, respectively).



35. Thus, the cathode material produced following the improved synthesis procedure does not produce a working cathode either.

VIII. Replication of Uemura Example 2 with cathode synthesis using the ion-exchange method

36. Example 2 of Uemura purports to use $\text{Li}_{0.83}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$ as the cathode active material and a hard carbon with approximately 13% of irreversible capacity.⁹

⁹ Uemura, Fig. 3.

37. Because the experimental procedures described in Uemura¹⁰ do not yield $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$ with a layered structure, to reproduce the battery described in Example 2 of Uemura, I conducted literature review to see how layered $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$ can be synthesized. In 2002, Akihisa Kajiyama and colleagues reported successful synthesis of layered $\text{Li}_x\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_2$ with the ion-exchange method.¹¹ Kajiyama states that by using their ion-exchange method, it was possible to successfully synthesize a Co-doped layered LiMnO_2 with up to 0.8 lithium (i.e., $\text{Li}_{0.8}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_2$).¹² Kajiyama states that at that time, efforts were underway to “obtain a stoichiometric compound $\text{LiMn}_{0.5}\text{Co}_{0.5}\text{O}_2$ ”—a cathode material with a full lithium atom.¹³

38. I note that the cathode materials described in Examples 3 and 4 of Uemura (for which I did not apply the ion-exchange method) contain an active material with the chemical composition of $\text{Li}_{0.967}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$. This material is very similar stoichiometrically to $\text{LiMn}_{0.5}\text{Co}_{0.5}\text{O}_2$, as to which Kajiyama reports synthesis

¹⁰ Uemura, ¶71.

¹¹ Ex. 2017, Akihisa Kajiyama et al., *Synthesis and electrochemical properties of $\text{Li}_x\text{Co}_{0.5}\text{Mn}_{0.5}\text{O}_2$* , Solid State Ionics 149 (2002) 39-45 (“Kajiyama”).

¹² Kajiyama at 42.

¹³ *Id.*

efforts had not been successful as of its publication in 2002. Accordingly, a person of ordinary skill would understand that as of Kajiyama's publication date in 2002, the cathode active material in Examples 3 and 4 had not been successfully synthesized with Kajiyama's method.

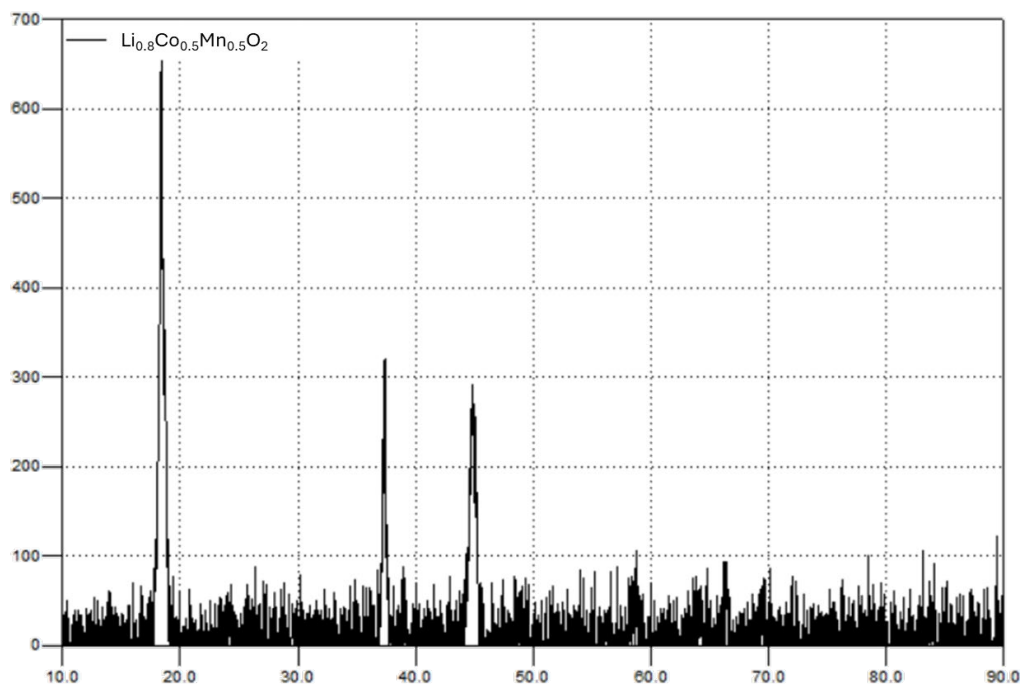
39. Following the procedure described in Kajiyama, I was able to synthesize $\text{Li}_{0.8}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_2$ with a layered structure, which is similar to the cathode material described in Example 2 of Uemura ($\text{Li}_{0.83}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_{2-\delta}$). The experimental procedures are laid out below:

An aqueous solution of $\text{CoAc}_2 \cdot 4\text{H}_2\text{O}$ (3.48 g), MnAc_2 (2.42 g), and $\text{NaAc} \cdot 3\text{H}_2\text{O}$ (3.04 g) was rotavaped to dryness at 80 °C. The mixture was annealed in air at 280 °C for 90 min then 700 °C for 20 h before quenching to room temperature. The resulting dark powders were mixed with LiNO_3 (21.89 g) and LiCl (1.84 g) and heated in air at 280 °C for 21 h. After cooling to room temperature, the mixture was dissolved with H_2O , filtered, washed with H_2O , and vacuum-dried at 60 °C to yield the product as dark powders.

40. After following the procedure set forth above, I tested the sample using XRD. The XRD pattern is shown below, which shows a well-defined layered structure, consistent with the XRD pattern reported in Fig. 4 of Kajiyama.¹⁴ This well-defined layered structure is consistent with the cathode active material Uemura

¹⁴ Ex. 2017 at Fig. 4.

purports to have used for Example 2.¹⁵ Accordingly, the cathode material I synthesized was suitable for replicating Example 2 of Uemura, which I describe below.



¹⁵ Uemura, Fig. 3.

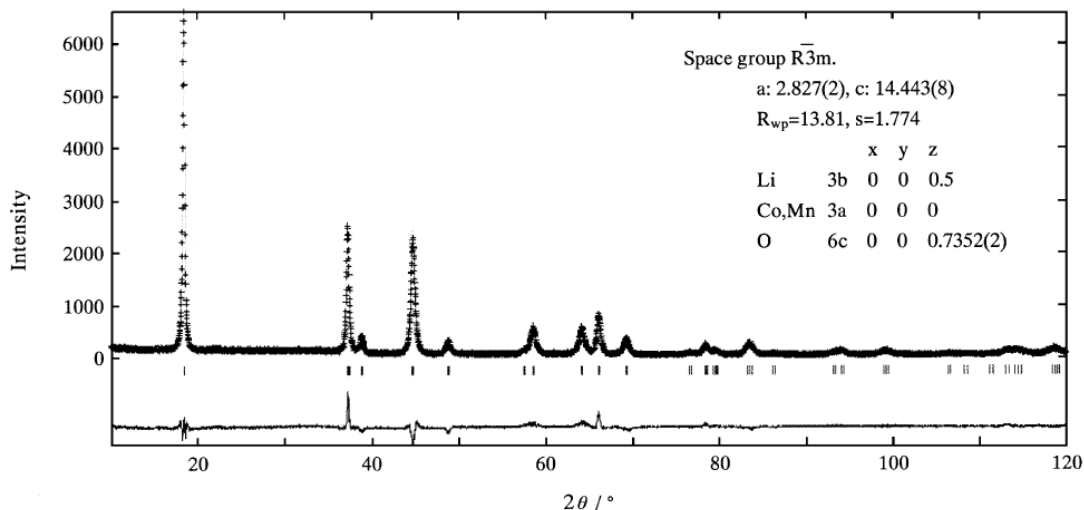


Fig. 4. XRD pattern of the lithium compound which was obtained by ion exchanging.

41. I mixed the synthesized layered $\text{Li}_{0.8}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_2$ with acetylene black and PTFE in the weight ratio of 80:16:4 to form a self-standing electrode with a diameter of 7.14 mm and an areal loading of $\sim 4 \text{ mg/cm}^2$. I then assembled the cathode into a Swagelok cell with a Mo current collector, a polyethylene separator, a counter electrode (lithium metal or hard carbon) (diameter: 7.94 mm; loading of hard carbon: $\sim 6 \text{ mg/cm}^2$), and a solution of LiPF_6 (1 M) in ethylene carbonate and dimethyl carbonate (1:1 w/w). I assembled two such cells with lithium metal as the counter electrode, and another cell with hard carbon as the counter electrode.

42. The two cells with lithium metal as the counter electrode were galvanostatically cycled at 0.14 mA/cm^2 at 60°C within 3.5–4.3V, 3.5–5.0V, respectively. For the test cell with hard carbon as the counter electrode, it was galvanostatically cycled at 0.14 mA/cm^2 at 60°C within 2.0–4.3V.

43. The hard carbon electrode had an areal loading of 6 mg/cm^2 , a discharge capacity of 390 mAh/g , and an irreversible capacity of 11%.

44. As shown below, when the two test cells with lithium metal as the counter electrode were charged between 3.5-4.3V and 3.5-5.0V, respectively, the data collected were comparable to the data reported in Kajiyama.¹⁶ As shown in Fig. 7 of Kajiyama, at 4.3V, the first charge capacity was close to 100 mAh/g , while the data I obtained had a first charge capacity of about 70 mAh/g . When the voltage increased to $\sim 5\text{V}$, Kajiyama reports that the charge capacity was almost 200 mAh/g , which is consistent with the data I obtained below: charge capacity surpassed 190 mAh/g at $\sim 4.8\text{V}$ before side reactions prevented charging from reaching higher voltages. This was likely due to the choice of the electrolytes solvent which were more akin to Uemura's (ethylene carbonate–dimethyl carbonate) than Kajiyama's (propylene carbonate). This further proves that a layered structure of $\text{Li}_{0.8}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_2$ was successfully synthesized. This material mirrors the cathode active material reportedly used in Example 2 of Uemura.

¹⁶ Kajiyama 2002 at 43-44.

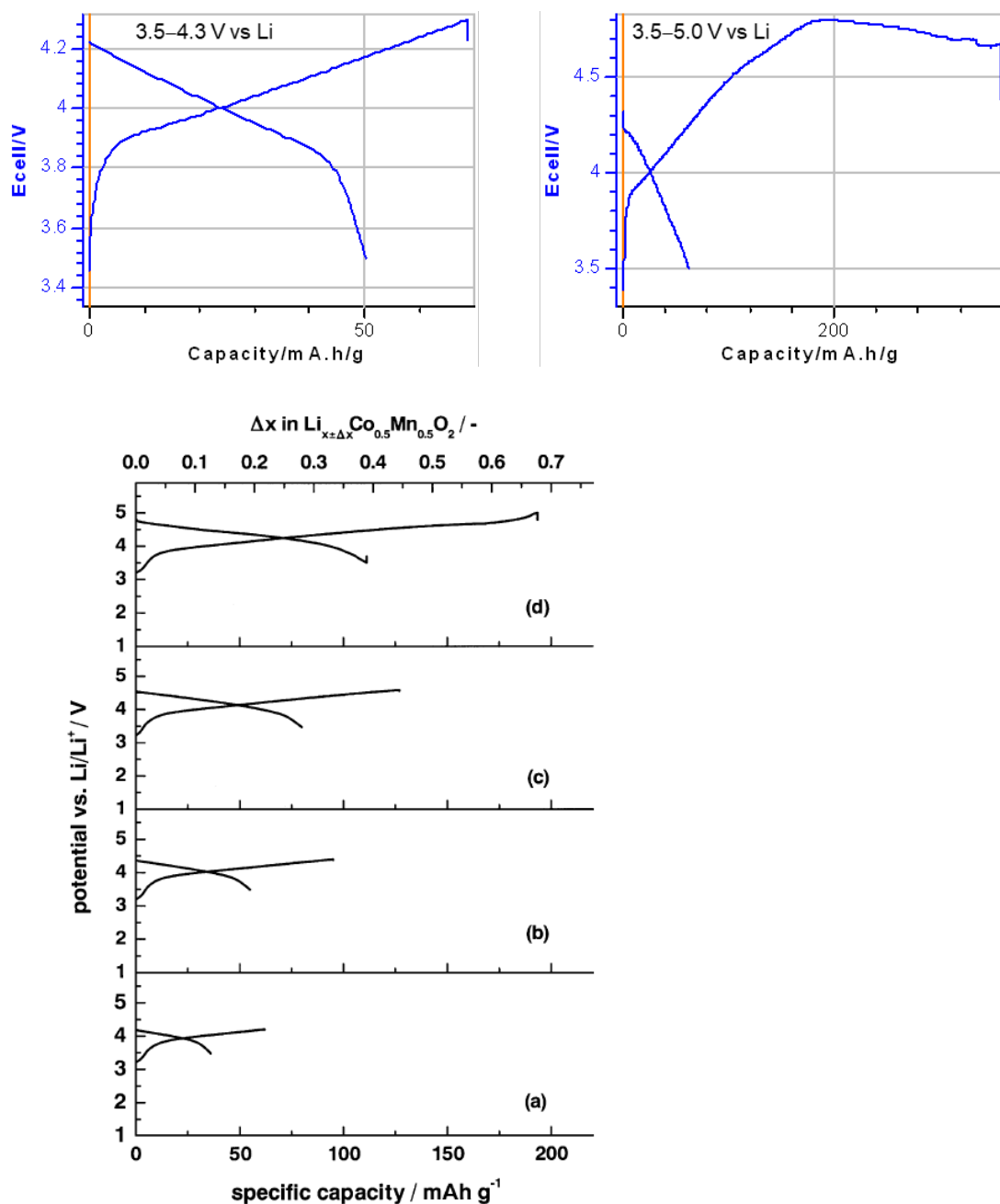
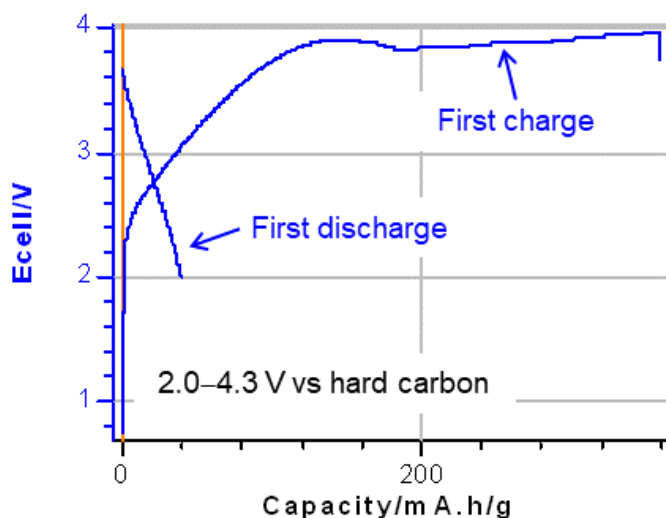


Fig. 7. Charge–discharge curves of the cell; $\text{Li}/\text{Li}_{0.7}\text{Co}_{0.5}\text{Mn}_{0.5}\text{O}_2$ during the first cycle with various charge depths. Charge depths correspond to changes in lithium composition of 0.22 (a), 0.33 (b), 0.44 (c) and 0.66 (d).

45. Below is the charge and discharge curve when the synthesized cathode active material ($\text{Li}_{0.8}\text{Mn}_{0.5}\text{Co}_{0.5}\text{O}_2$) was assembled into a full cell with hard carbon as the anode material. As shown in the figure below, before the voltage reaches 4V,

there is evidence of side reactions occurring and of the battery starting to degrade. Further, the downward trending curve (i.e., the first discharge cycle) shows a capacity of about 40 mAh/g, which is significantly lower than the first charge capacity (based on the upward trending curve, the first charge capacity was probably more than 100 mAh/g before the cell started to degrade).



46. A person of skill would conclude from these results that the resulting battery cells (i.e., those described in Uemura's Example 2 but with the cathode prepared using the ion exchange method), to the extent at all rechargeable, are not configured to be charged to a maximum voltage above 4V because of their performance degradation after a single charge cycle at or around that voltage.

IX. Declaration

47. I hereby declare that all statements made herein of my own knowledge are true and all statements made herein on information and belief were and are believed by me to be true, and that all statements herein were and are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that any such willful false statements may jeopardize the validity of the application or any patents issued thereon. If called to testify as to the truth of the matters stated herein, I could and would testify competently.

48. I declare under penalty of perjury that the foregoing is true and correct.

Respectfully submitted,

A handwritten signature in black ink, appearing to read 'Yanliang Liang', is written above a horizontal line.

Dr. Yanliang Liang

Dated: May 15, 2024