

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

AMPEREX TECHNOLOGY LTD.
Petitioner,

v.

SEMICONDUCTOR ENERGY LABORATORY CO., LTD.
Patent Owner

U.S. Patent No. 10,741,828

Issued: August 11, 2020

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Inventor: Teruaki Ochiai et al.

Title: Positive Electrode Active Material Including Lithium Cobaltate Coated with
Lithium Titanate and Magnesium Oxide

Case IPR2023-TBD

**DECLARATION OF TROY A. HAYES, PH.D., P.E.
IN SUPPORT OF PETITION FOR
INTER PARTES REVIEW OF U.S. PATENT NO. 10,741,828**

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LIST OF EXHIBITS

Exhibit	Description
1001	U.S. Patent No. 10,741,828 (“828 Patent”)
1002	File History of U.S. Patent No. 10,741,828 (“828 FH”)
1003	Declaration of Dr. Troy Hayes in Support of <i>Inter Partes</i> Review of U.S. Patent No. 10,741,828
1004	<i>Curriculum Vitae</i> of Dr. Troy Hayes
1005	Shim et al., <i>Synergistic Effects of Coating and Doping for Lithium Ion Battery Cathode Materials: Synthesis and Characterization of Lithium Titanate-Coated LiCoO₂ with Mg Doping</i> , 186 <i>Electrochimica Acta</i> , 201–08 (2015)
1006	Lee & Park, <i>Interface Characterization of MgF₂-Coated LiCoO₂ Thin Films</i> , 230 <i>Solid State Ionics</i> , 86–91 (2013)
1007	Yu et al., <i>The Investigation of Ti-Modified LiCoO₂ Materials for Lithium Ion Battery</i> , 262 <i>Journal of Power Sources</i> , 136–139 (2014)
1008	U.S. Patent No. 9,105,926 (“Fujiki”)
1009	Van Schalkwijk & Scrosati, <i>Advances in Lithium-Ion Batteries</i> (2002)
1010	Wang et al., <i>TEM Study of Electrochemical Cycling-Induced Damage and Disorder in LiCoO₂ Cathodes for Rechargeable Lithium Batteries</i> , 146 <i>Journal of the Electrochemical Society</i> , 473–80 (1999)
1011	Myung et al., <i>Effects of Al Doping on the Microstructure of LiCoO₂ Cathode Materials</i> , 130 <i>Solid State Ionics</i> , 47–56 (2001)

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1012	Tukamoto et al., <i>Electronic Conductivity of LiCoO₂ and Its Enhancement by Magnesium Doping</i> , 144 J. Electrochem. Soc., 3164–68 (1997)
1013	Shi et al., <i>Effect of Mg-doping on the Structural and Electronic Properties of LiCoO₂: A First-Principles Investigation</i> , 171 Journal of Power Sources, 908–12 (2007)
1014	Jang et al., <i>Electrochemical and Electron Microscopic Characterization of Thin-Film LiCoO₂ Cathodes Under High-Voltage Cycling Conditions</i> , 119–121 Journal of Power Sources 295–99 (2003)
1015	Bhattacharya et al., <i>A Transmission Electron Microscopy Study of Crack Formation and Propagation in Electrochemically Cycled Graphite Electrode in Lithium-Ion Cells</i> , 196 Journal of Power Sources, 8719–27 (2011)
1016	Fey et al., <i>MgAl₂O₄ Spinel-Coated LiCoO₂ as Long-Cycling Cathode Materials</i> , 146 Journal of Power Sources, 245–49 (2005)
1017	Int’l Patent No. WO 2013/048048 (“Song”)
1018	U.S. Patent No. 9,048,495 B2 (“Sun”)
1019	CN 102610806 (“Liu”) (translated)
1020	U.S. Patent Application 2007/0224506 A1 (“Ooyama”)
1021	Dedryvère et al., <i>Surface Film Formation on Electrodes in a LiCoO₂/Graphite Cell: A Step by Step XPS Study</i> , 174 Journal of Power Sources, 462–68 (2007)
1022	Solano, <i>Development of Artificial Surface Layers for Thin Film Cathode Materials</i> , Material Chemistry, Université de Bordeaux (2015)

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1023	Aurbach et al., <i>On the Capacity Fading of LiCoO₂ Intercalation Electrodes: The Effect of Cycling, Storage, Temperature, and Surface Film Forming Additives</i> , 47 <i>Electrochimica Acta</i> , 4291–306 (2002)
1024	Aykol et al., <i>Thermodynamic Aspects of Cathode Coatings for Lithium-Ion Batteries</i> , <i>Advanced Energy Materials</i> , 1400690 (2014)
1025	Kitagawa et al., <i>Application of Fluorine-containing Solvents in High Voltage Operation</i> , 78 <i>Electrochemistry</i> , 345–48 (2010)
1026	Muto et al., <i>Capacity-Fading Mechanisms of LiNiO₂-Based Lithium-Ion Batteries</i> , 156 <i>Journal of the Electrochemical Society</i> , A371–A377 (2009)
1027	Sun et al., <i>Role of AlF₃ Coating on LiCoO₂ Particles During Cycling to Cutoff Voltage Above 4.5 V</i> , 156 <i>Journal of the Electrochemical Society</i> , A1005–A1010 (2009)
1028	Park et al., <i>Amorphous Metal Fluoride Passivation Coatings Prepared by Layer Deposition on LiCoO₂ for Li-Ion Batteries</i> , 27 <i>Chemistry of Materials</i> , 1917–20 (2015)
1029	Aboulaich et al., <i>High Voltage Stability and Enhanced Electrochemical Performances of LiCoO₂ by CeF₃ Coating</i> , <i>IEEE</i> (2015)
1030	Bai et al., <i>Performance Improvement of LiCoO₂ by MgF₂ Surface Modification and Mechanism Exploration</i> , 134 <i>Electrochimica Acta</i> , 347–54 (2014)
1031	JP2012-018914 (“Patent Document 1”) (translated)
1032	JP2015-2013432 (“Patent Document 2”) (translated)
1033	Declaration of Jennifer Babbitt in Support of <i>Inter Partes</i> Review of U.S. Patent No. 10,741,828

I, Troy A. Hayes, hereby declare as follows:

I. ASSIGNMENT

1. I have been retained as an expert witness on behalf of Amperex Technology Ltd. (“Amperex” or “Petitioner”) to offer technical opinions in connection with the above-captioned Petition for Inter Partes Review (“IPR”) of U.S. Patent No. 10,741,828 (“the ’828 Patent”) (Ex. 1001).

2. I have been asked to provide my independent analysis of the ’828 Patent in light of the prior art patents and publications cited below.

3. I have been asked to provide my opinions regarding whether claims 1 through 8 of the ’828 Patent (the “Challenged Claims”) are invalid as being anticipated and/or obvious to a person having ordinary skill in the art (a POSITA) at the time of the alleged invention.

4. In preparing my Declaration, I reviewed the ’828 Patent, the file history of the patent (Ex. 1002), prior art references, technical references, and other publications from the time of the alleged invention, which are discussed herein.

5. For the purposes of my Declaration, I have been asked to assume that the priority date of the alleged invention recited in the ’828 Patent is July 5, 2016 (hereinafter the “Priority Date”).

6. I am not currently, and never have been, an employee of Amperex. I received no compensation for this Declaration beyond my normal hourly

compensation based on my time actually spent analyzing the '828 Patent, the prior art patents and publications cited below, and issues related thereto, and I will not receive any added compensation based on the outcome of any IPR or other proceeding involving the '828 Patent. I have no financial interest in Amperex.

II. BACKGROUND AND QUALIFICATIONS

7. I am over the age of 18 and am competent to write this Declaration. I have personal knowledge and expertise concerning the relevant technologies based upon my education, training, or experience. My relevant experience includes a deep understanding of the systems that we broadly refer to as Lithium-ion secondary (or rechargeable) batteries.

8. My complete qualifications and professional experience are described in my curriculum vitae, a copy of which is attached as Exhibit 1004. The following is a brief summary of my relevant qualifications and professional experience.

9. I have either trained or worked in the fields of mechanical engineering and materials science and engineering for approximately 32 years. I am currently a Principal Engineer at Exponent Inc. (Exponent), where I have worked since March 14, 2005.

10. While at Exponent, I have worked extensively on projects involving battery design evaluation, performance and safety testing, manufacturing, and failure analysis. I routinely perform failure analysis investigations on a variety of consumer

electronics as well as commercial and industrial products, including root cause analyses of products experiencing battery failures. In addition, I have conducted more than 80 on-site technical manufacturing line evaluations of battery cell and battery pack manufacturers worldwide. I also conduct design reviews of batteries and battery packs to evaluate the potential for safety issues or field failures, and help clients evaluate and choose vendors that have the capability to produce batteries meeting their quality and safety requirements.

11. I received my B.S. degree in Mechanical Engineering in 1995 from Oregon State University, my M.S. degree in Materials Science and Engineering in 1996 from Oregon State University, and my Ph.D. in Materials Science and Engineering in 2004 from the University of California, San Diego. I am registered in California as a Professional Metallurgical Engineer (#MT1938).

12. Cumulatively during the more than 80 lithium-ion battery cell and pack factory evaluations over the past 18 years, I have spent more than a year on the factory floor, evaluating battery designs, incoming quality control (IQC) specifications, testing and IQC reports, production processes and associated in-process quality control tests and reports, safety and reliability tests and reports all the way through outgoing quality control tests and reports. During these evaluations, I have worked extensively with battery cell manufacturing research and development engineers to discuss the details of battery design and manufacturing. These factory

evaluations also included extensive discussions with battery factory personnel related to battery design and manufacturing before and after the factory visits to drive corrective action in order to improve the battery design and manufacturing quality. These discussions and evaluations include the positive electrode material chemistry and composition, electrolyte composition and additives, as well as the separator design. My experience at the factory level began in 2005 and includes evaluations of factories and the associated existing and forward-looking battery technologies of manufacturers in China, Taiwan, Korea, Japan, Indonesia, the United States, France, Germany, and Switzerland. I developed Exponent's lithium-ion battery factory evaluation program and, through discussions with my teams, meetings with clients, and reviews of reports, have been directly involved in evaluations of many additional factories and associated battery designs. Through projects working with original equipment manufacturers (OEMs) and battery cell and pack manufacturers starting in 2005, I am also very familiar with the trends and motivations in the battery industry, including trends related to battery separator design as well as electrolyte chemistry and positive battery electrode materials. My projects involved discussions with OEMs and battery manufacturers related to improvements in battery design, including separator characteristics, electrolyte composition and additives, as well as positive electrode chemistry and design. This includes work with many consumer electronics OEMs and the associated battery manufacturing partners.

13. I have authored or coauthored nearly 30 publications in peer-reviewed journals and conference proceedings, including articles entitled “Key manufacturing practices to achieve high quality cells,” “From lithium plating to cell thermal runaway: A combustion perspective,” “Use of CT scanning for defect detection in lithium-ion batteries,” “Li-ion battery cell failure analysis: The significance of surviving features on copper current collectors in cells that have experienced thermal runaway,” and “Methodologies of identifying root causes of failures in lithium ion battery packs” for the 24th to 27th International Battery Seminar & Exhibit for Primary & Secondary Batteries, Small Fuel Cells, and Other Technologies conferences. I have also co-authored a book chapter titled “Methodologies for battery failure analysis” in the *Handbook of Batteries*, Fourth Edition.

14. I have presented at more than two dozen conferences, seminars, webinars, training courses, or workshops on various topics pertaining to battery design, manufacturing, and failure. Exemplary topics I have presented on include “How Do You Choose a Battery Manufacturer for Your Device? Part 2: Cell and Pack Manufacturing Audits”; “Catastrophic Thermal Battery Failures in Vaping Devices and in Pockets: Why Do They Happen?”; “Considerations When Choosing Batteries Made in China”; “Detecting Failures and Manufacturing Defects in Li-ion Batteries”; “Manufacturing, Testing and Recycling Safe EV Li-ion Batteries”; “Analyzing Electronic Product Battery Failures”; “Key Manufacturing Practices to

Achieve High Quality Cells”; “Design and Manufacturing of Safe Lithium Ion Batteries for EVs”; “Identifying a Non-safe Li-ion Battery in the Production Cycle to Avoid Safety Recalls and Maintaining Your Brand’s Trust”; “Methodologies of Identifying Root Causes of Failures in Lithium Ion Battery Packs”; and “Use of CT Scanning for Defect Detection in Lithium Ion Batteries.”

15. I am a member of various professional organizations pertaining to materials science and engineering, including the Materials Research Society; The Minerals, Metals and Materials Society; and ASM International.

16. My opinions set forth in this report are informed by my experience in the fields of mechanical engineering and materials science and engineering. Based on my above-described experience, I am an expert in the field. Also, based on my experiences, I understand and know of the capabilities of persons of ordinary skill in this field at least as early as the earliest alleged priority date of the asserted patents (i.e., July 5, 2016).

III. MATERIALS AND OTHER INFORMATION CONSIDERED

17. In forming the opinions expressed in this Declaration, I relied upon my education and many years of experience in the relevant field of the art and have considered the viewpoint of a person having ordinary skill in the art (a POSITA) as of the Priority Date of the ’828 Patent.

18. I have considered the materials referenced in this Declaration, including the '828 Patent, its file history, and other documents listed in the Exhibit List to the '828 Petition, reproduced above. In particular, I have considered the prior art references listed below.

19. Jae-Hyun Shim, et al., *Synergistic effects of coating and doping for lithium ion battery cathode materials: synthesis and characterization of lithium titanate-coated LiCoO₂ with Mg doping* (“Shim”) (Ex. 1005) was published in 2015, and is prior art under post-AIA 35 U.S.C. §102(a)(1).

20. Hye Jin Lee, et al., *Interface characterization of MgF₂-coated LiCoO₂ thin films* (“Lee”) (Ex. 1006) was published in 2013, and is prior art under post-AIA 35 U.S.C. §102(a)(1).

21. Jinpeng Yu, et al., *The investigation of Ti-modified LiCoO₂ materials for lithium ion battery* (“Yu”) (Ex. 1007) was published in 2014, and is prior art under AIA 35 U.S.C. §102(a)(1).

22. U.S. Patent No. 9,105,926 (“Fujiki”) (Ex. 1008), was filed on July 13, 2010—before the Priority Date—and is prior art under post-AIA 35 U.S.C. §102(a).

IV. UNDERSTANDING OF PATENT LAW

23. I understand that prior art to the '828 Patent includes patents and printed publications in the relevant art that predate the Priority Date of the '828 Patent.

24. I understand that claims in an IPR are given their plain and ordinary meaning as understood by a person of ordinary skill in the art in view of the specification and prosecution history, unless those sources show an intent to depart from such meaning.

25. I understand that a patent claim is invalid if it is anticipated or obvious. Anticipation of a claim requires that every element of a claim be disclosed expressly or inherently in a single prior art reference, arranged in the prior art reference as arranged in the claim. Obviousness of a claim requires that the claim be obvious from the perspective of a POSITA at the time of the alleged invention. I understand that a claim may be obvious in view of one reference or a combination of two or more prior art references.

26. I understand that an obviousness analysis requires an understanding of the scope and content of the prior art, any differences between the alleged invention and the prior art, and the level of ordinary skill in evaluating the pertinent art.

27. I understand that certain factors—often called “secondary considerations”—may support or rebut an assertion of obviousness of a claim. I understand that such secondary considerations include, among other things, commercial success of the alleged invention, skepticism of those having ordinary skill in the art at the time of the alleged invention, unexpected results of the alleged invention, any long-felt but unsolved need in the art that was satisfied by the alleged

invention, the failure of others to make the alleged invention, praise of the alleged invention by those having ordinary skill in the art, and copying of the alleged invention by others in the field. I further understand that there must be a nexus—a connection—between any such secondary considerations and the alleged invention. I also understand that contemporaneous and independent invention by others is a secondary consideration tending to show obviousness.

28. I further understand that a claim can be found obvious if it unites old elements with no change to their respective functions, or alters prior art by mere substitution of one element for another known in the field, with that combination yielding predictable results. While it may be helpful to identify a reason for this combination, there is no rigid requirement for a teaching, suggestion, or motivation to combine. When a product is available, design incentives and other market forces can prompt variations of it, either in the same field or different one. If a person having ordinary skill in the relevant art can implement a predictable variation, obviousness likely bars patentability. Similarly, if a technique has been used to improve one device, and a person having ordinary skill in the art would have recognized that the technique would improve similar devices in the same way, use of the technique is obvious.

29. I also understand that the following rationales may support a finding of obviousness:

- Combining prior art elements according to known methods to yield predictable results;
- Simple substitution of one known element for another to obtain predictable results;
- Use of known technique to improve similar devices (methods, or products) in the same way;
- Applying a known technique to a known device (method, or product) ready for improvement to yield predictable results;
- “Obvious to try” – choosing from a finite number of identified, predictable solutions, with a reasonable expectation of success;
- Known work in one field of endeavor may prompt variations of it for use in either the same field or a different one based on design incentives or other market forces if the variations are predictable to one of ordinary skill in the art;
- Some teaching, suggestion, or motivation in the prior art that would have led one of ordinary skill to modify the prior art reference or to combine prior art reference teachings to arrive at the claimed invention.

V. SUMMARY OF OPINIONS

30. It is my opinion that Claims 1, 4, 5, and 8 of the '828 Patent are rendered obvious by Shim, either alone or in combination with Lee.

31. It is further my opinion that Claims 2 and 6 of the '828 Patent are rendered obvious by Shim (either alone or in combination with Lee) in combination with Yu.

32. It is further my opinion that Claims 3 and 7 of the '828 Patent are rendered obvious by Shim in combination with Lee.

33. It is my opinion that Claims 1, 4, 5, and 8 of the '828 Patent are rendered obvious by Fujiki.

34. It is further my opinion that Claims 2 and 6 of the '828 Patent are rendered obvious by Fujiki in combination with Yu.

35. It is further my opinion that Claims 3 and 7 of the '828 Patent are rendered obvious by Fujiki in combination with Lee.

VI. OVERVIEW OF TECHNOLOGY AND OF THE '828 PATENT

A. Background

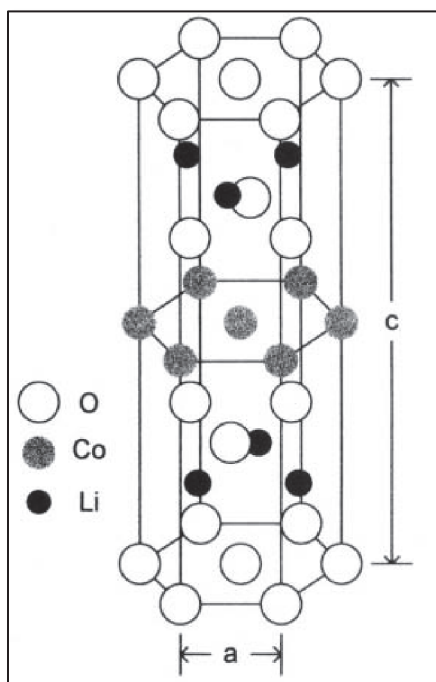
36. Early research on layered lithium-ion host materials in the 1970s and 1980s led to the commercialization of the lithium-ion battery by Sony in 1991. The lithium-ion battery first released by Sony used a carbon-based negative electrode, a lithium cobalt oxide-containing positive electrode, and an organic electrolyte. In comparison to existing rechargeable battery chemistries at the time, the lithium-ion battery offered a superior combination of energy density, power capability, and cycle life, enabling a wide range of portable consumer electronics devices among other battery-powered systems like power tools and electric vehicles. Since its first release, the lithium-ion battery has been the focus of continued research and development, with the constant push to improve the performance (i.e., energy density, power capability, and cycle life) guiding engineering innovations over the last several decades.

1. Lithium-Ion Battery Components and Operation

37. A lithium-ion battery has the basic components of a negative electrode (*i.e.*, anode) and a positive electrode (*i.e.*, cathode), a separator between the electrodes, and an electrolyte. The positive electrode typically consists of lithium metal oxide microparticles capable of reversible lithium-ion insertion/extraction, commonly lithium cobalt oxide (LiCoO_2) (also referred to as LCO), in batteries for consumer electronics, or an analogous material, coated on an aluminum foil current collector. The negative electrode typically consists of graphite microparticles capable of reversible lithium-ion insertion/extraction coated on a copper foil current collector. Both electrode coatings also typically contain conductive additives (*e.g.*, carbon black) and binder material (*e.g.*, polyvinylidene fluoride (PVDF) for the positive electrode and a mix of carboxymethyl cellulose (CMC) and styrene-butadiene rubber (SBR) for the negative electrode). The separator is typically a thin polymer membrane consisting of porous polyethylene and/or polypropylene, often combined with an inorganic heat resistant layer, that maintains electrical isolation between the electrodes but allows transport of lithium ions between the electrodes during charge/discharge. Electrolytes are typically liquid and are commonly mixtures of organic solvents (*e.g.*, carbonates) and a dissolved lithium salt, typically LiPF_6 . Various electrolyte additives may also be included to enhance the cell

performance and allow optimization for specific applications and specific electrode chemistries.

38. When a lithium-ion battery is charged, lithium ions are extracted from the positive electrode material, flow into the electrolyte, through the separator, and are inserted into the negative electrode material. In an analogous manner, during discharge, lithium ions are extracted from the negative electrode, flow into the electrolyte, through the separator, and are inserted into the positive electrode. A representation of the atomic structure of LiCoO_2 is shown below:



Ex. 1009, 143.

39. Manufacturing of lithium-ion pouch cells, for example, for consumer electronic applications typically involves these steps (at a minimum): (1) mixing of

electrode powder active materials, binders, and solvent (involves both wet and dry mixing steps); (2) coating the electrode layers onto foil current collectors; (3) drying the electrodes; (4) calendaring the electrodes; (5) cutting/slitting/notching the electrodes; (6) tab cutting and welding to the electrodes; (7) winding/stacking of the electrodes with the separator layer; (8) hot pressing of cell winding/stack; (9) packaging the electrode assembly in a pouch; (10) cell drying; (11) injecting electrolyte; (12) undergoing formation and aging; (13) degassing and final sealing; and (14) aging.

40. During the formation process that occurs near the end of the manufacturing process, cells are charged and discharged for several cycles in a controlled manner. In this process, the electrolyte (consisting of a mixture of solvents, salts, and additives) is known to react and generate a layer of decomposition products on the surface of the positive and negative electrode materials. The decomposition products are collectively referred to as the solid-electrolyte interphase (“SEI”) on the negative electrode surface and the cathode-electrolyte interphase (“CEI”) on the positive electrode surface. In a healthy battery, both of these layers allow for lithium ion transport and limit electron transport between the electrode active material and the electrolyte.

41. The SEI and CEI are critical to the longevity of the battery since they help shield the bulk electrode materials from further interactions with the electrolyte,

which effectively limits further electrolyte decomposition reactions. This self-passivating effect is due to the catalytic nature of the bare electrode active material towards the formation of the SEI and CEI. As the SEI and CEI grow thicker, less interaction exists between the electrode active material and the liquid electrolyte, which reduces the electrolyte decomposition reaction rate. Therefore, when the SEI and CEI are sufficiently thick, the reaction rate slows to a negligible amount, resulting in self-passivation. Importantly, if such layers were not present or not passivating, electrolyte solvent and salt species would continue to break down and form thick interfacial layers on the positive and negative electrode surfaces.

42. Importantly, if a fluorine-containing salt is used in the electrolyte, such as LiPF_6 —as is industry standard (*see* Ex. 1024, 1), the corresponding SEI and CEI layers are known to contain fluorine in various compounds such as lithium fluoride, fluorophosphates, and lithiated fluorophosphates. Of such species, lithium fluoride is a notable example of an electrochemically inactive compound under nominal lithium-ion battery operating conditions. *See id.* Although the initial formation reaction of lithium fluoride (during CEI or SEI formation) is believed to originate from LiPF_6 hydrolysis or the consumption of a small fraction of lithium from the positive electrode active material, the resulting lithium fluoride layer limits further electrolyte decomposition reactions from occurring as described in previous paragraphs. *See, e.g.,* Ex. 1021 (showing formation of LiF layer on cathode surface

due to LiPF_6 hydrolysis), Ex. 1022, 86 (describing fluorine “on the surface” of cathode particles due to “formation of LiF derivatives”); Ex. 1023, 12 (describing that XPS measurements on “ LiCoO_2 powder stored in LiPF_6 /alkyl-carbonate solutions always shows LiF peaks”); Ex. 1024, 1 (“Solid LiF . . . precipitates on the electrode particles[.]”); Ex. 1026, 6 (showing fluorine “detected at cracked particle surfaces, which suggests that the electrolyte invaded into the cracked gaps”).

2. Degradation in Common Commercial Lithium-Ion Battery Chemistries

43. As a lithium-ion battery is charged and discharged repeatedly, the achievable capacity generally decreases with increasing cycle number and time in the field. Early research efforts identified several mechanisms underlying this performance loss and were broadly related to degradation of the positive electrode material, negative electrode material, and electrolyte.

44. During charge and discharge, the positive and negative electrode materials typically experience volumetric changes as lithium is inserted (“intercalated”) and removed (“deintercalated”). As a result of the repeated stress related to lithium intercalation and deintercalation, the positive and negative electrode active material particles can become damaged over time through the formation of local crystal defects that interrupt the nominal periodicity of the atomic lattice. Examples of these crystal defects may include vacant sites in the lattice (vacancy defects), atoms residing on interstitial sites in the lattice (interstitial

defects), atoms residing in sites in the lattice where another atom is expected (substitutional defects), planes of atoms that have been displaced from their anticipated positions in a perfect lattice and form a discontinuity within the lattice (dislocations), or cracks. *See* Ex. 1010, 1–8; Ex. 1014 (“Fracture of grains was observed by TEM in the cathode layer”), Ex. 1015 (cracks forming on electrochemically cycled graphite).

45. The positive electrode, which is typically LiCoO_2 in batteries for consumer electronics, is well known to form cracks and crevices near the surface of the particles during cycling. The initial few cycles of a lithium-ion battery are performed during the formation process of manufacturing. With further cycling, a greater population of particles have been observed with cracks, which is consistent with fatigue expected for repeated volume changes. When cracks are generated during cycling, new active material surface area is exposed to the electrolyte. These regions are then passivated through further reactions with the electrolyte to form a new interphase layer at the crack location. This process leads to the consumption of electrolyte and the build-up of deposits on the positive electrode material, degrading the the performance of the cell.

46. Most commercial lithium-ion battery electrolyte chemistries utilize an LiPF_6 salt dissolved in a mixture of organic carbonate-based solvents and are susceptible to breakdown. Beyond the decomposition processes induced by

electrochemical cycling that result in the growth of interfacial layers on the positive and negative electrodes, common electrolytes utilizing LiPF_6 are also known to react with trace amounts of water in the electrolyte to form hydrofluoric acid (HF). The hydrolysis reaction of LiPF_6 was proposed as:



Ex. 1021, 6. It was widely understood that HF was detrimental to cathode stability and battery performance. Since positive active materials are often lithium transition metal oxides (e.g., LiCoO_2), they are highly susceptible to etching by acidic species, including HF. This degradation pathway results in the loss of active material and transition metal dissolution into the electrolyte. Ex. 1006, 3–4 (“It is well known that acid (such as HCl and **HF**) can be generated from the reaction between water and salts (such as LiClO_4 and **LiPF₆**) in the electrolyte. . . . The acid-containing electrolyte attacks the surface of the cathode during cycling.”) (emphases added); *see also* Ex. 1023, 2 (“a LiPF_6 solution always contains HF contamination.”); Ex. 1027, 1 (describing “capacity loss of LiCoO_2 ” “mainly due to the interfacial impedance growth between the cathode and electrolyte, which results from side reactions involving LiPF_6 ”).

3. Strategies to Improve LiCoO_2 Cathode Materials

47. Over the last several decades and across nearly all industry sectors that utilize lithium-ion batteries, four critical goals have guided battery engineering

efforts: (1) maximize energy density, (2) maximize power performance, (3) maximize cycle life, and (4) minimize safety concerns.

48. Although the theoretical capacity of LiCoO_2 is as high as 273.8 mAh/g, the maximum attainable practical capacity is far lower (approximately 137 mAh/g when cycled in the range of 3–4.2 V in a battery). To increase the cell-level energy density, a primary option is to increase the upper cut off voltage, which involves the extraction of more lithium ions from the positive electrode material. While this avenue provides an immediate increase in capacity, it subjects the electrodes to new degradation mechanisms that can compromise the long-term health of the cell. For example, charging LiCoO_2 above ~4.2 V vs. Li/Li^+ can cause a phase change in the LCO particles, resulting in a change in the unit cell dimensions, the generation of internal stress within the bulk particle, and an increase the likelihood of cracking. Cell operation at even higher voltages is known to result in increased dissolution of cobalt and can drive additional electrolyte decomposition, which could further compromise components of the cell. This process consumes additional lithium from the lattice to form new deposits of inorganic decomposition species. The thicker interfacial layer also presents a greater barrier to lithium-ion transport, increasing the impedance of the cell, which consequently degrades the power capability (limiting the potential discharge rates) and cycling performance. Additionally, charging a battery to where more than 50% of the lithium is removed from the

LiCoO₂ active material could increase the likelihood of battery degradation and possible thermal failures.

49. Doping has been a widely explored strategy to improve the electrochemical performance of LiCoO₂. Cobalt can be partially substituted with other inactive elements, such as magnesium, aluminum, titanium, and others, to improve both high-voltage and long-term cycling stability. *See* Ex. 1007; Ex. 1011, 5–8; Ex. 1012, 1–5. These dopant elements possess different atomic radii or valence states than cobalt, which can influence the local bonding structure, provide structural stability, and potentially advantageously modify the bulk material properties. *See, e.g.,* Ex. 1013, 2–4. To synthesize such materials, dopant precursors can be introduced simultaneously with cobalt and lithium precursors and subsequently calcined.

50. Another commonly employed strategy to improve the stability of LiCoO₂ is the creation of a protective surface coating. This strategy was first reported in by Cho (and colleagues) in the early 2000s, where the researchers (a collaboration between Seoul National University and Samsung SDI) reported the creation of a ~100 nm thick Al₂O₃ coating on LiCoO₂ formed through the use of a sol-gel process, demonstrating a significant improvement in the cycling performance of coated LiCoO₂ and proposing that the coating improved the structural stability of the material.

51. Following these early works, researchers went on to explore a wide range of different coatings. In particular, Fey (and colleagues) reported extensively on various coatings for LiCoO_2 , including silica, MgO , MgAl_2O_4 , B_2O_3 , TiO_2 , Al_2O_3 , ZrO_2 , ZrTiO_4 , and $\text{Li}_4\text{Ti}_5\text{O}_{12}$. *See generally* Ex. 1016. For example, Fey provides the TEM image below, showing a MgAl_2O_4 -coated LiCoO_2 particle.

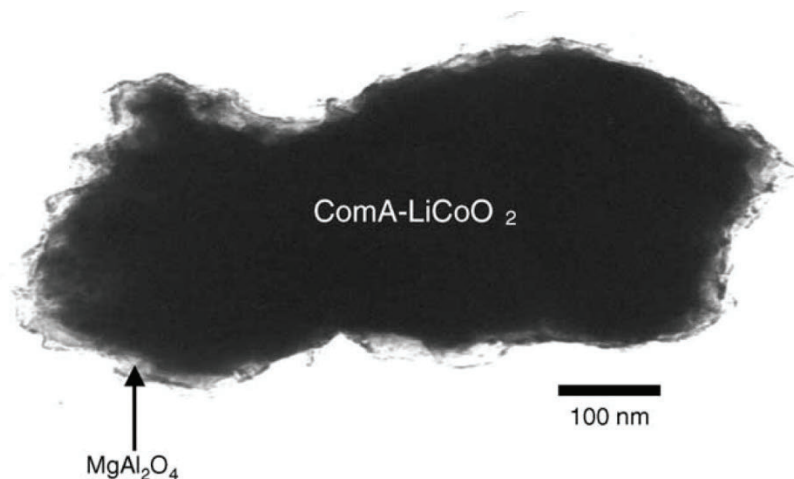


Fig. 2. TEM image of a 1.0 wt.% spinel-coated LiCoO_2 particle.

Ex. 1016, Fig. 2

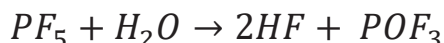
52. In many of their reports on coated LiCoO_2 , Fey proposed that “[i]t is inevitable that coated surfaces always have defects such as cracks and pinholes in them” and that “in the case of the coated cathodes, changes in the surface texture resulting from the contraction and expansion of the lattice during the cycling process could have enabled the particles of the coating material to become ingrained in the crevices and cracks on the cathode surface.” *Id.*, 4. This explanation (repeated by

several of Fey's papers) served to describe how the coatings continued to be beneficial after extended cycling.

53. Subsequent research explored the mechanisms by which exterior metal oxide coatings can impart chemical stability benefits. One proposed method is HF scavenging, where the metal oxide coating sacrificially reacts with HF present in the electrolyte such that the underlying active material is preserved. For example, alumina coatings can react with HF via the pathway:



The reaction pathway further shows that one of the reaction products is a metal fluoride, which would likely be located at the exterior surface of the positive active material particle. Ex. 1024, 1–2 (“[A] fluoride layer can form on top of an HF-scavenging oxide coating as a product of the HF-scavenging reaction.”). In this manner, the fluoride becomes another barrier that prevents the active material from subsequent reactions with HF. Although this reaction reduces the amount of HF present in a cell, as shown above, it results in the formation of water. This water can further hydrolyze the LiPF₆ electrolyte salt, generating more HF:



Ex. 1024, 1; *see also* Ex. 1028, 1 (noting that even “[s]mall levels of moisture” can react with lithium salts “to generate hydrofluoric acid (HF)”).

54. To this end, it was further understood in the art that metal fluoride coatings could be used directly (as opposed to being formed through scavenging reactions). Studies found that fluorine-containing coatings “serve[] as a protective layer, preventing direct contact with the reactive electrolyte.” Ex. 1006, 4; *see also id.*, 5 (“MgF₂ coating layer seemed to react with acidic electrolyte and incur damage on cycling instead of the electrode material.”), 6 (“[T]he MgF₂ coating layer was effective as a protective layer to prevent the dissolution of the transition metal . . . which may lead to enhanced electrochemical properties.”); Ex. 1028, 1 (“[M]etal fluoride coatings might provide better stability against the corrosive effects of HF.”); Ex. 1029, 2 (“In recent years, fluorine in the form of metal fluorides such as AlF₃ [22], CaF₂ [23], MgF₂ [24], LaF₃ [25], CeF₃ [26], FeF₃ [27] is considered to be one of the most promising coating materials to improve electrochemical performance of cathode, due to its high stability. This could be attributed to the reduction of the structural change of active material from HF attack by a thin coating layer preventing the direct contact between delithiated cathode and liquid electrolyte.”); Ex. 1030, 2 (finding that metal fluoride coating improved the thermal stability of LiCoO₂ electrodes); Ex. 1027, 1 (finding that metal fluoride coating protected LiCoO₂ from hydrofluoric acid attack).

55. Indeed, well before the '828 Patent, it was known to those of skill in the art that magnesium, titanium, and fluorine (in particular) could be used in the

coating layer—and would be beneficial in coatings on positive electrode active materials. *See* Ex. 1017, [0011]–[0013] (disclosing TiF, MgF₂, TiF₃, and TiF₄ coatings); Ex. 1018, 2:66–3:15 (disclosing various coatings, including TiF, MgF₂, TiF₃, and TiF₄); Ex. 1019, 1:24–31 (disclosing one element from the group “Ti, Zr and Hf” used with one element from the group “F or P”); Ex. 1020, [0022]–[0023], [0030] (disclosing surface oxides of “titanium, silicon (Si), magnesium, and zirconium”); Ex. 1008, 4:48–6:11 (disclosing coatings comprising at least one of “Na, Li, Mg, Zr, Zn, Ba, Bi, Y, Ti, Al and B”). Further, some studies indicated multiple coating layers could be layered on top of the bulk particle. *See* Ex. 1020, [0027]–[0032], [0077] (“[T]he surface layer is arranged on the coating layer”).

56. Additionally, a fluorine-containing coating was known to form directly on the LiCoO₂ surface upon contact with the electrolyte and also during electrochemical cycling. For example, XPS studies performed by Dedryvère in 2009 showed the formation of an LiF layer on the cathode surface from the hydrolysis of LiPF₆. Ex. 1021. This is shown in the formula below:



Ex. 1024, 1; *see also* Ex. 1022, 86 (describing fluorine present “on the surface” of the cathode particle “caused by the formation of LiF derivatives”); Ex. 1024 at 1 (“Solid LiF . . . precipitates on the electrode particles[.]”); Ex. 1023, 12 (describing that XPS measurements on “LiCoO₂ powder stored in LiPF₆/alkyl-carbonate

solutions always shows LiF peaks”). Muto specifically found that “small amounts of fluorine were mainly distributed along the grain boundaries of the primary particles” and “these were likely derived from the LiPF₆ electrolyte.” Ex. 1026, 6. Some studies found this formation of a fluorine-containing layer to beneficial. *See* Ex. 1025, 3 (concluding that a fluorine-containing solvent “provides an electrochemically stable surface film on a LiCoO₂ cathode and ***significantly improves the cycle performance*** under a high-voltage condition”).

57. Since doping and coating procedures are often not mutually exclusive, prior research has also explored how simultaneously doping and coating LiCoO₂ can be beneficial towards electrochemical performance. One example pathway to achieve this goal is to introduce dopant precursors during LiCoO₂ synthesis and subsequently coat the resulting doped LiCoO₂ particles through a second processing step. This combination-style approach has been previously explored in systems such as magnesium oxide coatings on magnesium-doped LiCoO₂, zirconium oxyfluoride coatings on magnesium-doped LiCoO₂, and aluminum oxide on aluminum-doped LiCoO₂.

4. X-ray Photoelectron Spectroscopy

58. X-ray photoelectron spectroscopy (“XPS”) is a characterization technique that provides elemental information approximately from the top 1–10 nm of a sample surface. The working principle is based on the photoelectric effect, in

which samples exposed to electromagnetic radiation emit electrons. The resulting electrons emitted have kinetic energies that are related to their “binding energies” that are unique for each element that is present in the sample. This phenomenon allows for elemental identification according to the following (simplified) expression:

$$E_B = hv - KE$$

where E_B is binding energy, $h\nu$ is the x-ray source energy, and KE is the measured kinetic energy. The source X-ray energy is known, and the kinetic energy of the emitted electron is measured, so the binding energy (each element’s unique “fingerprint”) can be determined by taking the difference between these two values.

59. The benefits of chemical identification and surface sensitivity are bolstered by the ability to perform quantitative compositional analysis and depth profiling with a possible lateral resolution on the order of microns. These features make XPS one of the most widely used characterization techniques for shallow surface analysis.

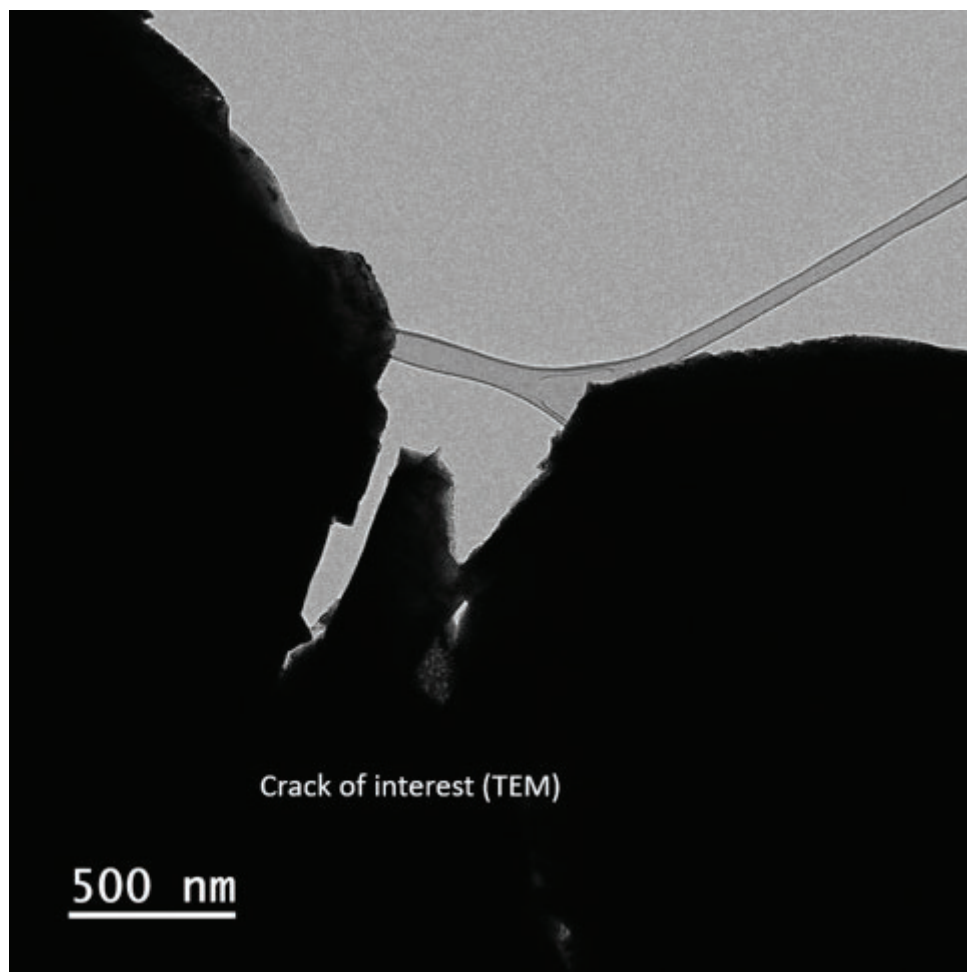
60. Due to its high surface sensitivity, XPS is often used to investigate the surface chemistry of battery materials. Notably, XPS can provide insight into the elemental composition of surface coatings on electrode particles, the presence of impurity species on electrode particles, and the composition of the SEI or the CEI. Furthermore, due to the inherently shallow photoelectron escape depth, XPS analysis

is commonly understood to be representative of a sample surface only, in contrast to other chemical and spectroscopic techniques that can probe elemental composition further into the bulk regions of a sample. Since the elemental bonding nature also influences the binding energy of emitted photoelectrons, XPS analysis can identify the presence and types of chemical bonds on a sample surface, provided that they are sufficiently spectrally resolved.

61. In lithium-ion battery research and development, XPS is often used to study the chemical compositions of the SEI and CEI after formation during manufacturing. XPS can identify both organic and inorganic electrolyte decomposition products and provide insight into the bonding nature of these species. For example, XPS analysis is capable of spectrally resolving carbon-oxygen bonds, which often are attributed to organic electrolyte decomposition products, from carbon-carbon bonds, which are often attributed to conductive carbon in the electrode. XPS can be used to determine the presence and relative amounts of inorganic species, such as lithium carbonate or lithium fluoride, to provide diagnostic information to understand long-term electrochemical performance. *See, e.g.,* Ex. 1006 (using XPS to examine the concentration-depth profile of the MgF_2 -coated LiCoO_2 film electrode before and after cycling); Ex. 1007 (using XPS to examine concentration ratios of Ti to Co).

5. Transmission Electron Microscopy

62. Transmission electron microscopy (“TEM”) is an advanced characterization technique used to study materials at the nanoscale or even down to the atomic scale (fractions of nanometers). TEM was first demonstrated in the 1930s, and has been regarded as an important tool for nanoscience for decades, including before July 5, 2016, which I understand to be the priority date of the ’828 Patent. TEM can provide extremely high-resolution analysis of a sample and can typically resolve features down to the nanometer length scale. With improved optics, appropriate electron sources, and care taken to control the sample and environmental conditions, TEM can provide resolution on the order of several angstroms (down to the atomic scale). A TEM operates by firing a beam of electrons into and through a sample to produce an image. The incident electrons interact with the sample and are sensitive to the thickness, local chemical composition, and local crystallinity. These electrons can scatter or transmit through the sample. The transmitted electrons are magnified and focused onto a detector. The resulting image contrast is accordingly dependent on the local intensity of the transmitted electrons. The scattered electrons may also be collected and analyzed since they can provide chemical and structural information about the sample. For instance, below is a TEM image taken (at my direction) showing a crack in a Samsung Galaxy A8 battery sample dated August 7, 2015 (discussed below in section VI.B):



In this bright-field image, the darkest colored portions correspond to solid regions of the sample which are sufficiently thick such that the incident electron beam is blocked and unable to reach the detector. Conversely, the lightest colored regions represent areas where the electron beam is not blocked by solid material (*i.e.*, areas that are absent of the sample).

63. Electron energy loss spectroscopy (EELS) or energy-dispersive X-ray spectroscopy (EDX) analyses, which are often conducted simultaneously with TEM imaging, provide chemical information that is mapped on an acquired image. TEM

can be used to examine electrode active material microparticles to interrogate atomic structure and chemistry. For LiCoO_2 microparticles imaged, TEM can provide information on particle size, grain sizes, and local structural ordering. TEM can also be used to image microcracks, voids, and other structural defects in prepared samples from both as-produced (after formation) batteries as well as those that may have experienced the accumulation of additional defects through cycling.

B. Testing Data (May 2023)

64. To demonstrate that a POSITA would have understood that TEM could be used to examine the “crack portions” of a positive electrode active material (PEAM) particle, and that fluorine and magnesium would be present in such a portion, I oversaw TEM, SEM, and EDS testing (conducted at my direction) on several battery samples harvested from commercial lithium-ion batteries manufactured prior to July 2016 (which I understand to be the priority date of the ‘828 patent). Although this testing was performed in May 2023, standard equipment and techniques that would have been available to a POSITA prior to July 2016 were utilized. To be clear, I am not relying on these particular products to demonstrate invalidity, as I understand that is not within the scope of an IPR. Instead, I provide these tests to confirm what was well known to a POSITA before July 2016: that a PEAM particle examined with TEM—a common observation technique—would

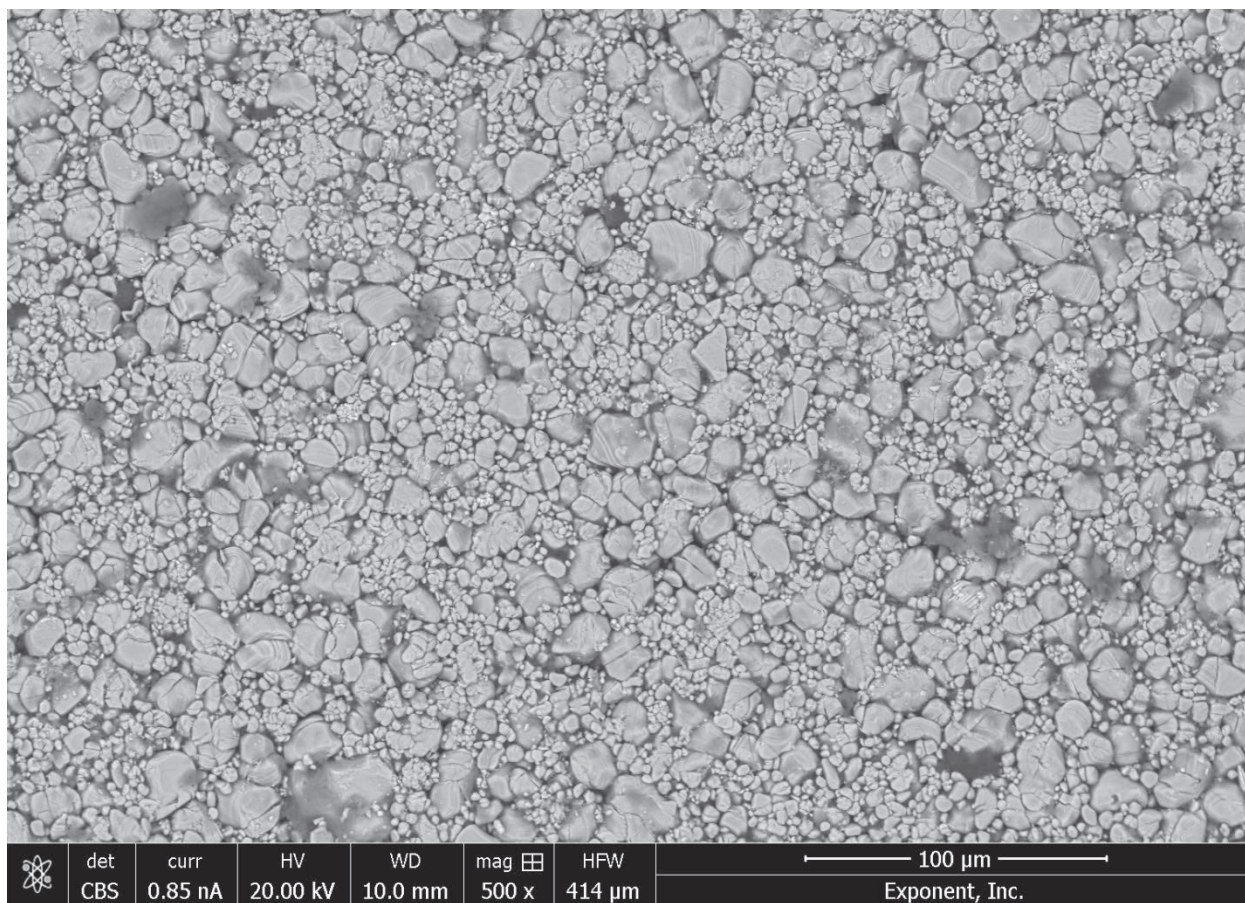
regularly have crack portions in the particle and that the cracks could be readily examined by TEM.

65. The first tested sample was a Samsung Galaxy A8 battery. This sample would have been available to a POSITA because the date of manufacture listed on the battery sample was “2015.08.07,” referring to August 7, 2015:

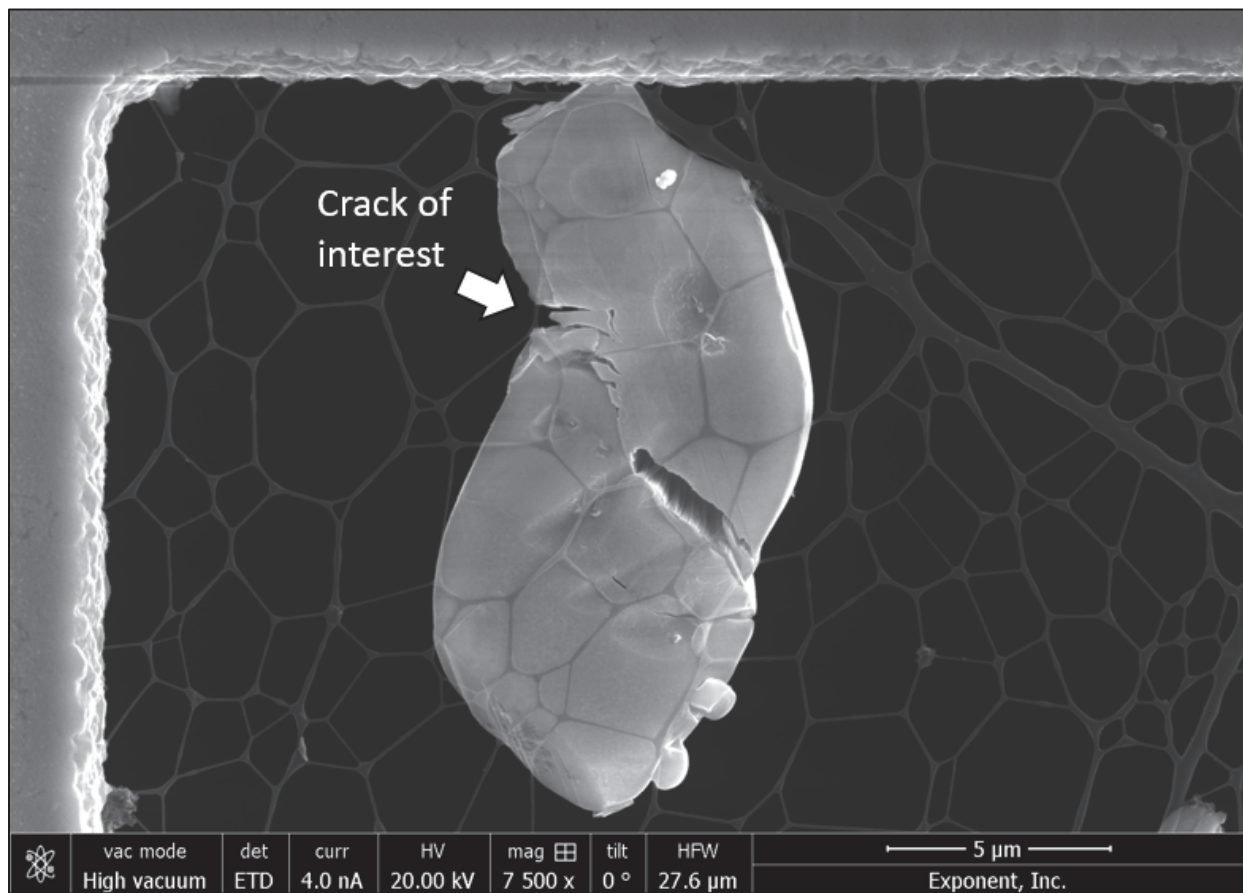


66. I first had SEM conducted under my direction to characterize the morphology of the PEAM particles. A POSITA would have understood that a commercial lithium-ion battery would contain cracked PEAM particles from the

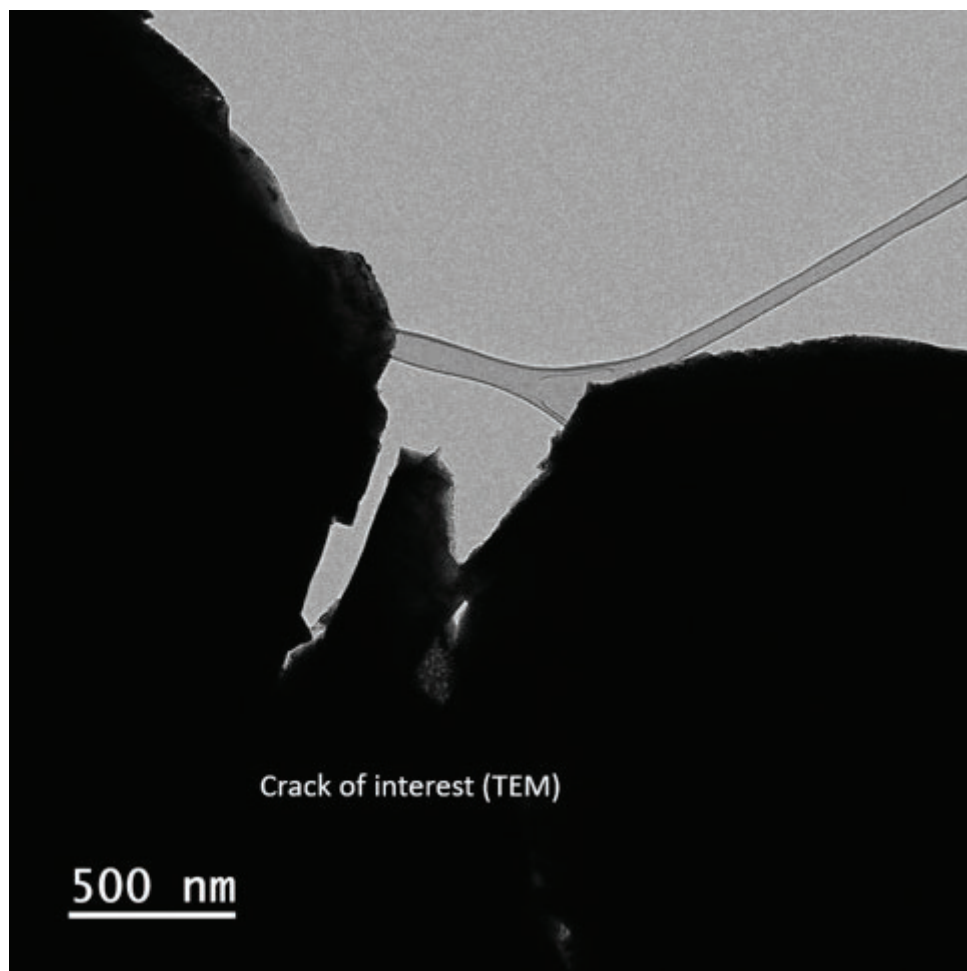
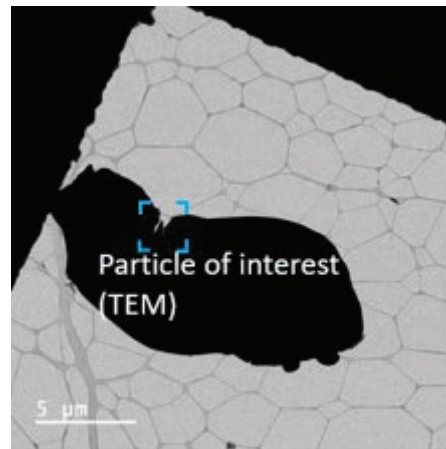
point of manufacturing, with the population of cracked particles growing in number with increasing cycles and aging time. Indeed, from an SEM image of the battery sample (as I show below), it is apparent that many of the PEAM particles exhibit cracks.



67. Next, a population of these particles was deposited on a lacey carbon grid for TEM analysis. As I show below, this grid was first imaged using SEM to identify a PEAM particle of interest with a prominent “crack portion” that could then be analyzed with TEM:



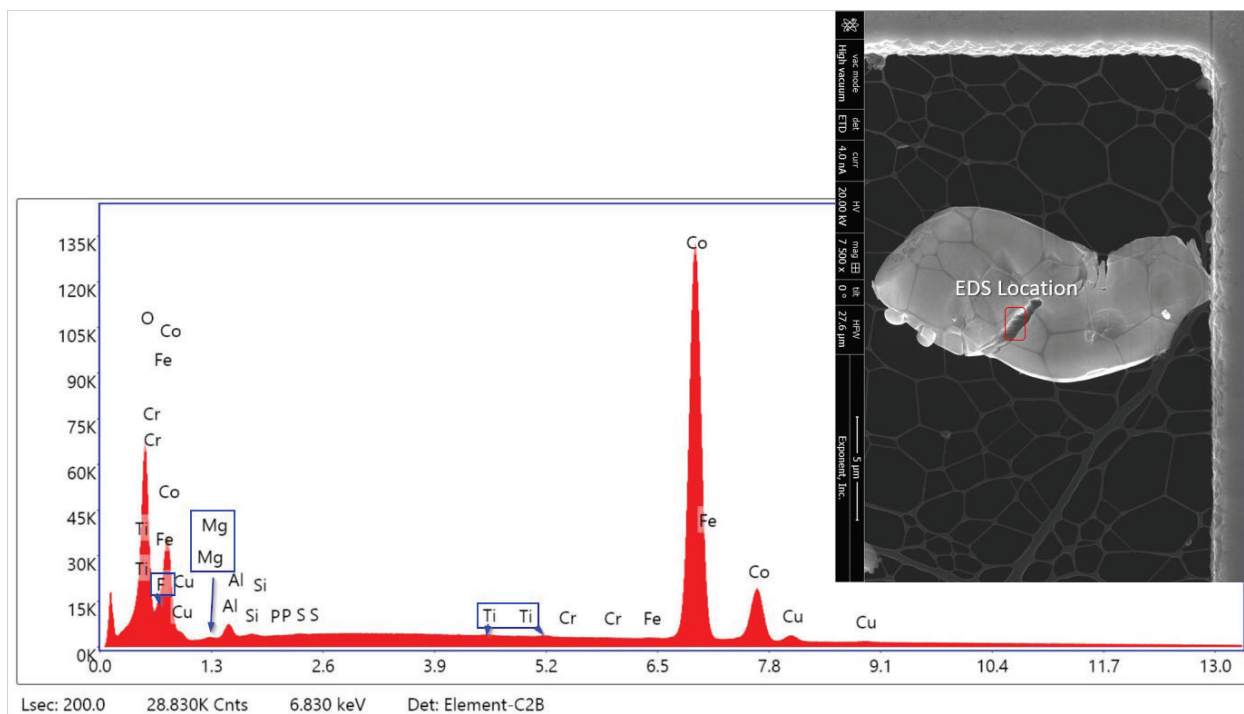
68. Then, under my direction, TEM imaging technology available before July 2016 (including the microscope itself) was used to analyze the crack of interest. TEM imaging showed that the “crack portion” on the same particle of interest was indeed visible, thereby confirming that a POSITA would have found obvious a “crack portion” observable by TEM:



69. In these bright field TEM images, the dark colored portions correspond to solid regions of the sample which are sufficiently thick such that the incident

electron beam is unable to reach the detector. Conversely, the lightest colored regions represent areas where the electron beam is not blocked by solid material (*i.e.*, is absent of PEAM). Thus, the light colored gap in the TEM image corresponds to a “crack portion” where the solid PEAM particle had fractured.

70. I directed EDS analysis to characterize the elemental composition of a “crack portion” of the same PEAM sample. The resulting EDS spectrum showed the presence of magnesium, titanium, and fluorine in the “crack portion” (annotated in blue):



71. A POSITA would have understood that the spectral intensities at approximately 0.68 eV, 1.25 eV, and 4.51 eV correspond to the characteristic K-alpha X-ray emission energies of fluorine, magnesium, and titanium, respectively—

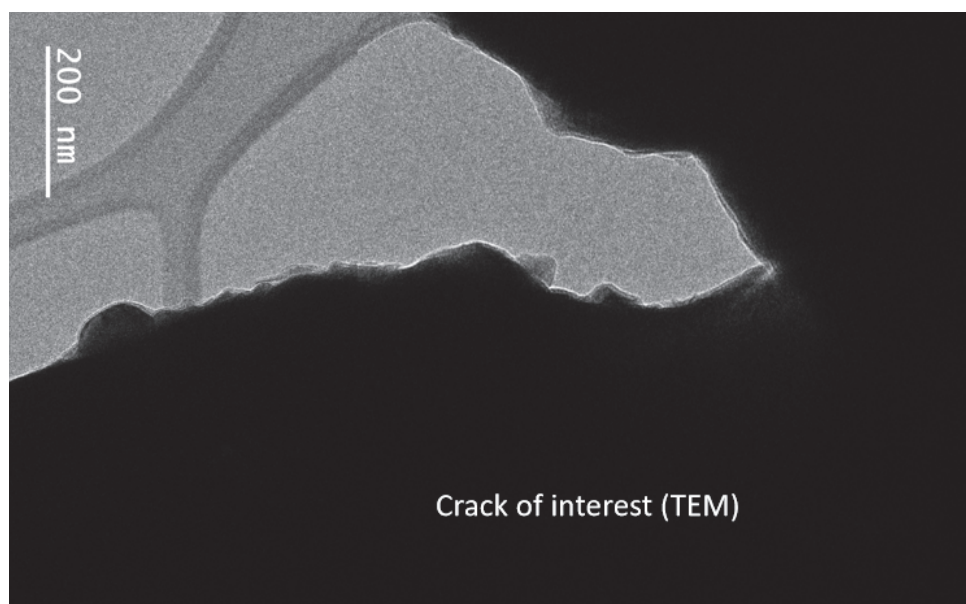
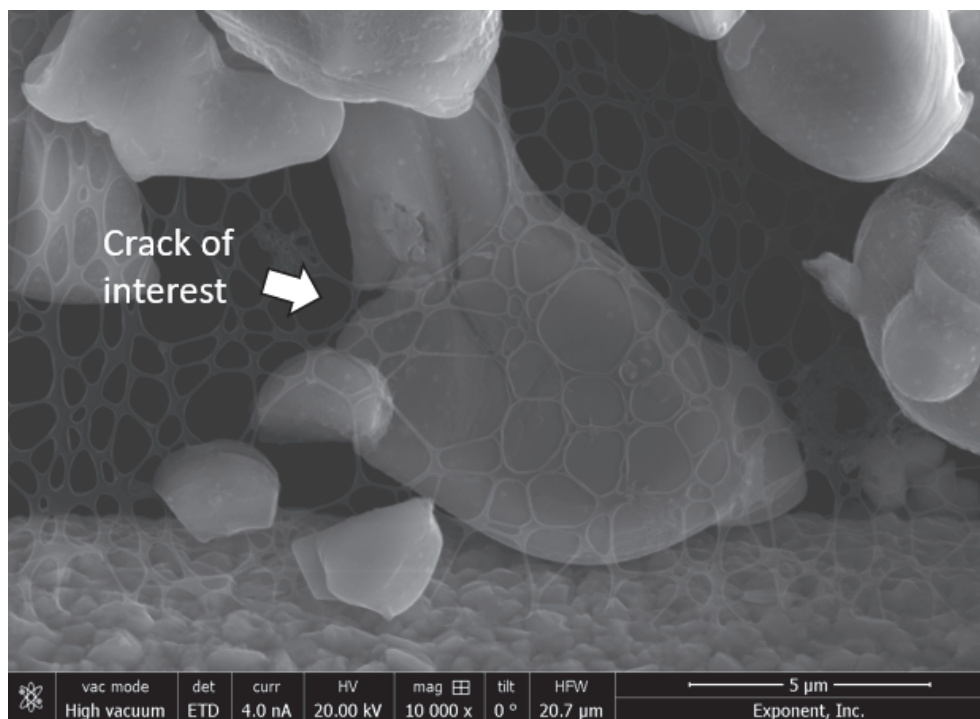
I have highlighted these elements on the above EDS spectrum by placing boxes around the labels. These spectral intensities are above the measured background, consistent with the presence of small amounts of fluorine, magnesium, and titanium in comparison to the cobalt and oxygen from the lithium cobalt oxide (note: the EDS detector does not detect lithium), confirming that the presence of fluorine, magnesium, and titanium in the “crack portion” of the PEAM sample would be well within the knowledge of a POSITA examining batteries using TEM at the time. A POSITA would further understand that if multiple elements have characteristic X-ray emission energies that are spectrally close to each other, and if those elements are present in a sample, then the measured EDS intensities may appear as combined peaks or as a smaller peak shoulder that is adjacent to a larger peak. For example, in the EDS map above, spectral intensities at 0.71 eV are observed, which are assigned to the L-alpha emission energies of iron. The presence of a shoulder at slightly lower energies (*e.g.*, 0.68 eV) is assigned to fluorine, suggesting that it is present in the sample.

72. The fact that the EDS peaks associated with “Mg,” “F,” and “Ti” are relatively weak in comparison to the “Co” and “O” signal would have indicated to a POSITA that such elements were present in small quantities in comparison to the lithium cobalt oxide base material, as would be expected in the case of, either as thin

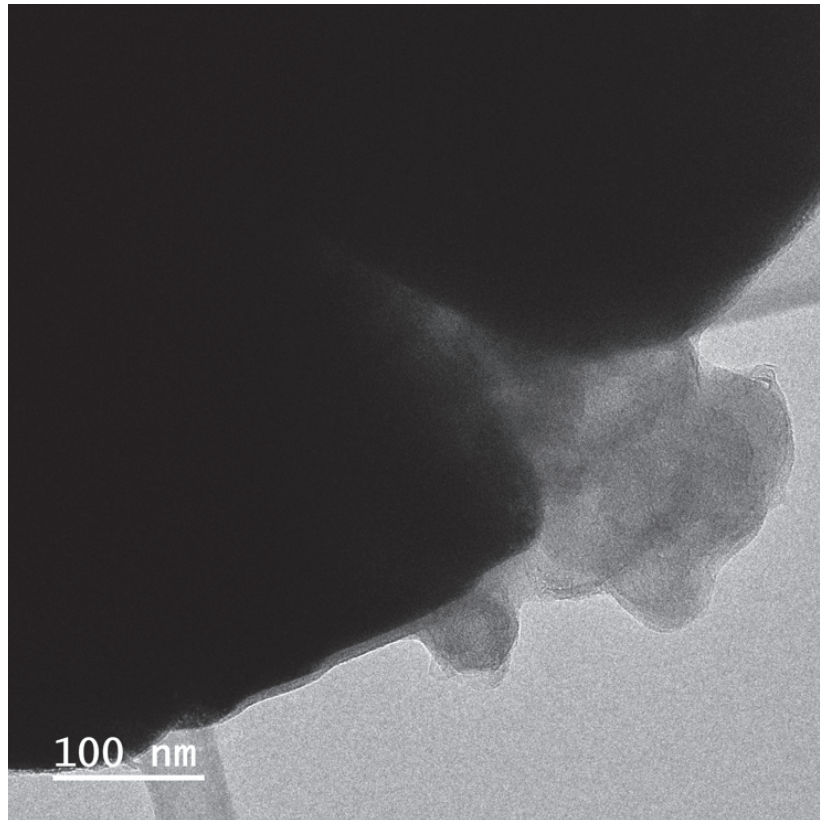
surface layers and/or dopants. Regardless, a POSITA would have understood that those elements were present in the sample.

73. In sum, my testing of a Samsung Galaxy A8 battery sample from 2015 confirms what a POSITA would have known: that a representative element and fluorine can be found in crack portions observed from a TEM image of the positive electrode active material particle.

74. To further support that a POSITA would have been aware that TEM could be used to observe crack portions in a PEAM particle, I oversaw additional testing (done at my direction) of other battery samples and observed similar results. For instance, the below two images are from a Samsung Galaxy Note 4 battery sample dated April 1, 2016. As shown below, SEM (top) and TEM (bottom) images show crack portions in a PEAM particle.



75. As a further example, the below TEM image shows a crack portion in a PEAM particle from an iPhone 5S battery sample dated October 2013.



C. Overview of the '828 Patent

76. I understand that the '828 Patent issued on August 11, 2020 from U.S. Application No. 15/638,449, filed on June 30, 2017. For purposes of this declaration, I have been asked to assume that the '828 Patent is entitled to a Priority Date of July 5, 2016.

1. Claims

77. The '828 Patent has 8 claims, including independent claims 1 and 5. The '828 Patent has 6 dependent claims. Claims 2–4 depend from claim 1 and claims 6–8 depend from claim 5. The Challenged Claims are claims 1, 2, 4, 5, 6, 7, and 8.

2. Summary of the Specification

78. Lithium-ion secondary batteries had been “actively developed” by the time the ’828 Patent was prosecuted, as the ’828 Patent itself admits. Ex. 1001, 1:32–45. The ’828 Patent is directed at addressing the “deformation of the particles of the positive electrode active materials, such as cracking or breaking” that might occur “[w]hen charge and discharge cycles are repeated”—a well-known issue of battery degradation. *Id.*, 4:1–3. The ’828 Patent proposes “prevent[ing] the deformation of the particles of the positive electrode active materials, such as cracking or breaking” by using a “coating film.” *Id.*, 4:10–12. By covering the surface of the PEAM with a coating film to protect the crystal structure, a “decrease in capacity due to charge and discharge cycles can be suppressed.” *Id.*, 3:54–61.

79. However, the ’828 Patent itself admits that such coating films were previously disclosed in the prior art. *See Id.*, 1:50–53 (“Thus, improvement of a positive electrode active material ***has been studied*** to increase the cycle performance and the capacity of the lithium ion secondary battery (Patent Document 1 and Patent Document 2)” (emphasis added)). The background art that the ’828 Patent discusses and cites to include JP2012-018914 (“Patent Document 1”), which teaches “[t]he formation of the ***surface layer*** of the positive electrode active material in a second region comprising iron can reduce the energy barrier in insertion and removal of lithium at the positive electrode active material surface.” Ex. 1031 (Abstract) (emphasis added). The ’828 Patent also cites to background art JP2015-2013432

(“Patent Document 2”), which teaches a PEAM with “a ***surface layer*** consisting of a layered structure of lithium cobalt oxide comprising element M (M is Mg and/or Al), and at least a layered structure of lithium cobalt oxide solidified by Ti.” Ex. 1032 (Abstract) (emphasis added).

80. The ’828 Patent additionally admits that the coating materials that it claims were in the prior art. Ex. 1001, 22:17–26 (noting that it was “generally known [] at the time” to “synthesiz[e] particles of the composite oxide containing ***lithium, cobalt, magnesium, and fluorine***[.]”) (emphasis added).

81. The ’828 Patent specification describes achieving an alleged improvement to the robust body of lithium-battery prior art by using different regions inside the PEAM. *Id.*, 2:24–30. Specifically, the ’828 Patent describes a PEAM comprising a first region (preferably of an oxide of lithium and a first transition metal); a second region (containing an oxide of a second transition metal, preferably titanium); and a third region (of a representative element, preferably magnesium oxide). *Id.*, 2:37–56. There may be overlap between the region of the second transition metal and the region of the representative element. *Id.*, 2:31–33. The ’828 Patent notes that if the composite oxide containing lithium and the first transition metal includes a crystal defect, or crack, the representative element and second transition metal will be present in the crack. *Id.*, 24:24–36. The specification

notes that, as a result, the active material particle will have increased chemical stability when in contact with the electrolyte solution. *Id.*

3. Summary of the Prosecution History

82. From my understanding, the application leading to the '828 Patent was originally filed with 22 claims (none of which issued). It is my understanding that the '828 Patent received rejections on the statutory grounds of non-patentable subject matter under 35 U.S.C. § 112, lack of novelty under 35 U.S.C. § 102, and obviousness under 35 U.S.C. § 103. Ex. 1002, Vol. 4, 823–37. I understand the Applicant ultimately made cancellations and amendments to the application that led to the '828 Patent's issuance. *Id.*, Vol. 5, 1336.

VII. LEVEL OF ORDINARY SKILL IN THE ART

83. I have been informed that a person of ordinary skill in the art is a hypothetical person who is presumed to have the skill and experience of an ordinary worker in the field at the time of the alleged invention.

84. I understand that there are multiple factors relevant to determining the level of ordinary skill in the pertinent art, including the educational level of active workers in the field at the time of the alleged invention, the sophistication of the technology, the type of problems encountered in the art, and the prior art solutions to those problems.

85. In determining the characteristics of a hypothetical person of ordinary skill in the art of the '828 Patent at the time of the claimed invention, I considered several things, including the type of problems encountered in this field, and the rapidity with which innovations were made. I also considered the sophistication of the technology involved, and the educational background and experience of those actively working in the field, and the level of education that would be necessary to understand the '828 Patent.

86. Finally, I placed myself back in the relevant period of time, and considered the state of the art and the level of skill of the engineers working in this field at that time.

87. I understand that patents and publications are to be understood from the perspective of a person having ordinary skill in the art ("POSITA").

88. I understand and have been informed that the factors I should consider in determining the ordinary level of skill in the art include: (i) the levels of education and experience of persons working in the field; (ii) the types of problems encountered in the field; and (iii) the sophistication of the technology. I understand that a POSITA is not a specific real individual, but rather a hypothetical individual having the qualities reflected by the factors above. This hypothetical person has knowledge of all prior art in the relevant field as if it were arranged on a workshop wall and takes from each reference what it would teach to a person having the skills

of a POSITA. Based on my education, experience, knowledge of problems encountered in the field, and the sophistication of the technology, the POSITA in the field of the asserted patents.

89. In my view, a POSITA with respect to the asserted patents would be an individual with at least a Bachelor's degree in chemical engineering, electrochemistry, mechanical engineering, materials engineering, or chemistry, with at least two years of experience in designing or developing lithium-ion batteries. A higher level of education related to the technology may require less experience, and vice versa.

90. Based on my professional experience, I have an understanding of the capabilities of a person of ordinary skill in the relevant field. Further, I myself was at least a POSITA as of the earliest alleged priority date of the asserted patents (i.e., July 5, 2016). At that time, I had a significantly higher level of education related to the technology, including a M.S. in Materials Science and Engineering (1996) and Ph.D. in Materials Science and Engineering (2004), and more than 11 years of relevant experience designing and developing lithium-ion batteries.

91. I also note that my opinions provided in this Declaration would not change in view of minor modifications to this level of ordinary skill.

VIII. CLAIM CONSTRUCTION

92. In my opinion, “crystal defect” (as used in claim 5) is the only term in the claims of the ’828 Patent requiring explicit construction. I note that the ’828 Patent defines “crystal defects” as follows:

[C]rystal defects refer to defects which can be observed from a TEM image and the like, that is, a structure in which another element enters crystal, a cavity, and the like.

Ex. 1001, 9:55–58.

93. Further, in my opinion, aside from the express definition of “crystal defect” provided by the ’828 Patent, the plain and ordinary meaning of “crystal defect,” as it would have been understood by a POSITA at the time of the ’828 Patent, would have been an “imperfection or interruption of regular arrangement of atoms in a crystalline solid.”

IX. DETAILED INVALIDITY ANALYSIS

A. Background on Prior Art References

1. Overview of Shim (Ex. 1005)

94. Shim is a peer-reviewed research article pertaining to the synthesis and testing of magnesium (Mg) doped lithium cobalt oxide with a lithium titanate (LTO) coating for use as a positive electrode material in rechargeable lithium-ion batteries. The work described was a collaboration between Samsung SDI’s Battery R&D Center and Gachon University, South Korea. Ex. 1005.

95. Shim motivates the effort to develop modified lithium cobalt oxide by describing the different degradation mechanisms that such materials suffer in lithium-ion batteries. In the introduction to the article, Shim describes the methods to improve LCO and the like as follows: “Coating is one of the simplest as well as most effective methods to be considered for minimization of the degradation and consequent capacity fade during cycling. The primary role of the coating layer is physical protection from chemical reaction with the electrolytes.” *Id.*, 1.

96. Shim reports a synthesis route for magnesium doped LCO with an LTO coating. Shim discloses the results of a range of material characterization techniques used to study the synthesized material, including nano secondary ion mass spectrometry (nano SIMS) and TEM. Analysis of the particle via nano SIMS reveals that the magnesium is uniformly distributed throughout the particle, as shown in the red box in the figure below. However, after the completion of the LTO coating procedure (see orange box in the figure below), Shim notes that “a small amount of Mg^{2+} ions in the Mg-doped LiCoO_2 has diffused to the coating layer” following the LTO coating process. Ex. 1005, 3. Shim continues: “The diffused Mg^{2+} ions enhance a role of the coating layer [~ 200 nm thick] to improve electrochemical performances[.]” *Id.*

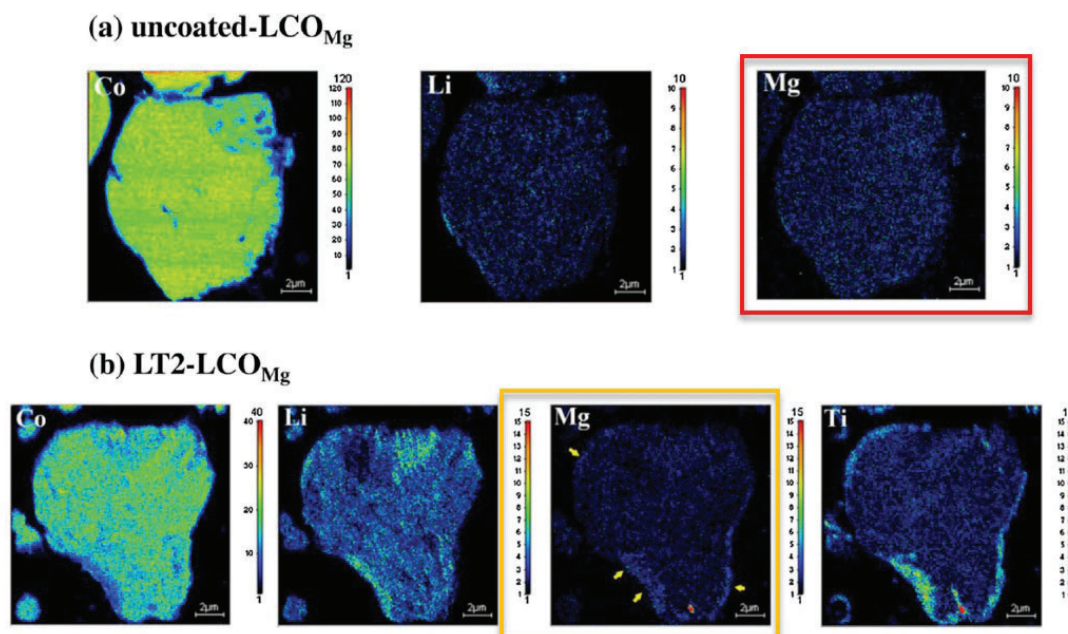


Fig. 2. Elemental distributions of metals on the cross sections of (a) uncoated-LCO_{Mg} and (b) LT2-LCO_{Mg} particles obtained by nano SIMS. (For interpretation of the references to color in text, the reader is referred to the web version of this article.)

Id., Fig. 2.

97. The performance of different iterations of the synthesized material is characterized through coin cell testing. Shim reports creating electrodes through the addition of carbon black and PVDF binder and the use of an electrolyte consisting of 1.15M LiPF₆ in a mixture of carbonate solvents. *Id.*, 2–3. Shim discloses cycling data, with charging up to 4.5 V vs Li/Li⁺, where the material that was synthesized with the highest concentration of Ti(OBu)₄ of lithium titanate-coated and magnesium-doped LCO (described in the paper as LT2-LCO_{Mg}) electrodes showed the highest cell capacity and best capacity retention. *Id.*, 5. In addition, LT2-LCO_{Mg} also showed better performance during fast charging-discharging. *Id.* LT2-LCO_{Mg} showed better electrochemical performance than LTO coated undoped LCO (using

the same synthesis parameters as LT2-LCO_{Mg}) and uncoated Mg-doped LCO, highlighting the importance of both LTO and Mg. Based on the findings of this study, Shim concludes that the creation of Mg-doped LTO on the surface of the LCO particles is the “origin of synergistic effects” that were observed. *Id.*, 8.

2. Overview of Lee (Ex. 1006)

98. Lee is a peer-reviewed research article pertaining to characterizing the electrochemical properties and interface reactions of LiCoO₂ film electrodes coated with MgF₂. Ex. 1006.

99. Lee notes that the cathode in a lithium ion battery affects the discharge capacity, cyclic performance, rate capability, and thermal stability of the cell. A known approach for improving the electrochemical properties of a cathode is coating the surface with stable materials, such as oxides, phosphates, or fluorides. Lee notes that using a coating layer has been found to stabilize the interface reaction and improve the electrochemical properties of the cathode. Lee aims to examine the effects on the interface reaction between the cathode and electrolyte when MgF₂, a “new fluoride coating material,” is used to coat the cathode. *Id.*, 1. Lee notes that Mg has been used as a dopant element to enhance the electrochemical properties of cathodes, as well as in an MgO coating material on LiCoO₂ to improve cyclic stability. Lee also takes into account studies showing that other fluorides, such as AlF₃, were very effective in modifying cathode surfaces. One motivation for Lee’s

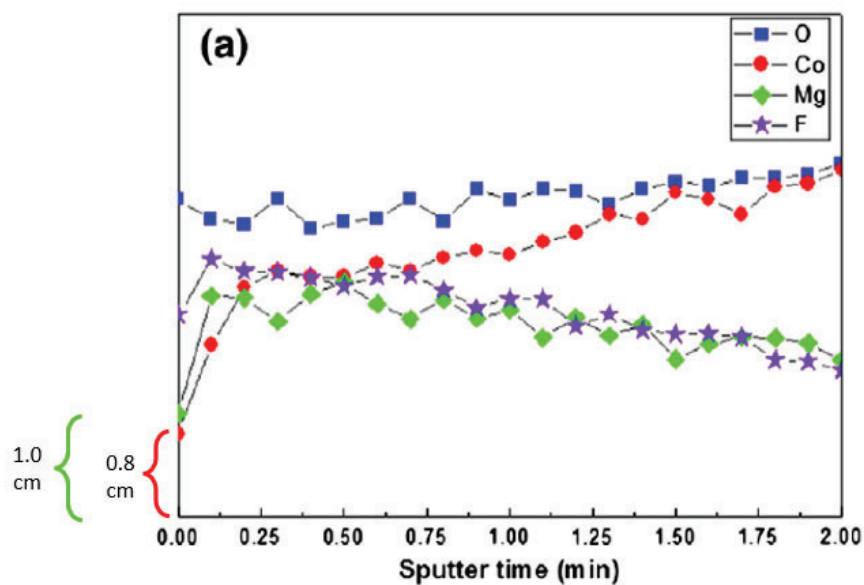
article (Ex. 1006) is to investigate and characterize the effect that an MgF_2 coating on the cathode has on battery cell performance.

100. Lee uses a LiCoO_2 thin film electrode to be coated and observed in detail. Lee notes that the surface of a thin film cathode may allow for more careful observation of the interface reaction between the cathode and electrolyte. Lee used field emission scanning electron microscopy (FE-SEM) to produce SEM images to observe the surface morphologies of both pristine and coated films.

101. Lee found that the MgF_2 -coated LiCoO_2 thin film delivered a higher discharge capacity than did the pristine film, showing that an MgF_2 coating enhances the rate capability of a LiCoO_2 electrode. Lee discloses that this is in part due to the coating hindering unwanted acid formation between the cathode and electrolyte. In particular, acid such as HCl or HF can be generated from the reaction between residual water within the electrodes and electrolytes and electrolyte salts (such as LiClO_4 and LiPF_6), which results in the acid-containing electrolyte attacking the surface of the cathode during cycling. As Lee notes, this leads to dissolution of transition metals, such as Co , and forms an unwanted interface layer that deteriorates the electrochemical properties of the pristine (uncoated) cathode. In contrast, when the surface of the cathode is covered with a coating layer, the coating material serves as a protective layer, preventing direct contact with the reactive electrolyte. Because this suppresses the dissolution of the transition metal and formation of an unwanted

interface layer, the coating protects the cathode from the reactive electrolyte, thereby enhancing electrochemical properties such as discharge capacity and rate capability. Lee also found that the MgF_2 coating layer enhanced the structural stability of the LiCoO_2 cathode.

102. Lee used XPS to examine the concentration-depth profile of the MgF_2 -coated LiCoO_2 film electrode before and after cycling. As shown below, Lee measured the concentrations of magnesium and cobalt throughout the particle:



Ex. 1006, Fig. 5(a) (annotated). Sputter time, on the X-axis, corresponds to how deep into the particle the measurement is being taken. Thus, at 0 minutes of sputter time, the XPS measurements are for concentrations at the surface of the particle. As sputter time increases, the measurements reflect concentrations deeper into the particle. Although the Y-axis of Lee's XPS figure is not labeled, a POSITA would have

understood the Y-axis to be linear. To determine the relative concentrations of Mg and Co in the particle, one can measure the relative magnitudes of the data in the plot. By measuring the data in the plot, I found that at a sputtering time of 0 minutes, the Co concentration measured 0.8 centimeters on the Y-axis, while the Mg concentration measured 1.0 cm on the Y-axis. Assuming that the Y-axis begins at 0, this would reflect a relative concentration of magnesium to cobalt (Mg/Co) of 1.2. A POSITA would also understand that if the Y-axis began at a value higher than 0, the Mg/Co ratio would decrease or converge to a minimum ratio of 1. For example, if the Y-axis of Figure 5(a) began at a value corresponding to 1.0 centimeters, the Mg/Co ratio would be $(1.0+1.0)/(1.0+0.8) = (2.0/1.8) = 1.11$. The higher the value that the Y-axis begins at, the closer the Mg/Co ratio would be to 1.0.

3. Overview of Yu (Ex. 1007)

103. Yu is a peer-reviewed research article pertaining to developing titanium (Ti) doped lithium cobalt oxide (LCO) as the positive electrode material for secondary lithium ion batteries. Yu's previous work developed a composite of Ti-containing LCO (denoted by $\text{LiCoO}_2/\text{LiCo}_{0.99}\text{Ti}_{0.01}\text{O}_2$) where a higher packing density of the positive electrode led to superior cycling performance. The improvement in cell performance is attributed to the partial doping of cobalt (Co) in LCO with Ti. A motivation for this article (Ex. 1007) is to better understand the structural configuration of Ti-doped LCO at both surface and bulk regions. This

work is led by a joint collaboration between Wuhan University and the National Key Laboratory of Power Source in China. Ex. 1007, 1.

104. Yu investigated three different positive electrode compositions which are comprised of LCO and Ti-doped LCO. Yu discloses the morphology of LCO and Ti-doped LCO composites via scanning electron microscopy (SEM). The surface and bulk concentration of the dopant element on LCO is measured by EDAX and XPS analysis. The battery performance of the synthesized materials is characterized by coin cell testing where the LCO composites are mixed with PTFE which is a fluorinated polymeric binder. The liquid electrolyte for the cell was prepared by using 1M LiPF₆ in a mixture of carbonate solvents. The voltage range for battery charging and discharging is 3–4.3 V vs. Li/Li⁺. *Id.*, 2–4.

105. From the depth profiling analysis via XPS, Yu further confirms the higher concentration (Ti/Co = 8.49%) of Ti in the surface layer of LCO, which gradually decreases toward the bulk regions. *Id.*, 3; Table 3. Yu further states that “adding a quite small amount of TiO₂ during the preparation cannot lead to the formation of LiCo_{0.998}Ti_{0.002}O₂ solid solution.” *Id.*, 2. The corresponding better cycling performance of Ti-doped LCO leads Yu to state “[t]he reason may be directly related to the distributed state with Ti element in LiCoO₂ particles for the improvement, even if its amount is very small.” *Id.*

106. Yu discloses the battery cycling data where both the Ti-doped LCO composites ($\text{LiCo}_{0.998}\text{Ti}_{0.002}\text{O}_2$ and $\text{LiCoO}_2/\text{LiCo}_{0.998}\text{Ti}_{0.002}\text{O}_2$) showed improved cycling performance and good capacity retention for 50 cycles. The improvement is attributed to the partial doping of Ti as Yu stated that doping leads to the formation of an interfacial protection layer that retains the stability of LCO during long-term cycling. *Id.*, 3. From the experimental results, Yu finally concludes that “[i]ntroducing a quite small amount of TiO_2 during preparation, the performance of the LiCoO_2 material is improved effectively.” *Id.*, 4.

4. Overview of Fujiki (Ex. 1008)

107. Fujiki is a U.S. Patent directed to develop the positive electrode material and non-aqueous electrolytes in lithium-ion batteries for high-capacity retention and improved cyclability.

108. Fujiki discloses various compositions for active materials where the principal oxide compound with metallic element A is covered by a secondary metallic oxide (containing element M1) and further coated by another metallic oxide (containing element M2). Ex. 1008, 3:15-20, 4:49-56. Fujiki states that, “in the case of compound oxide particles of which the surfaces have been modified, particularly where a metallic compound is present in the outside of the compound oxide particles, a high synergistic effect or a novel effect on enhancement of cell characteristics is generated.” *Id.*, 4:37–42.

109. Fujiki discloses that the M1 elements in the oxides are Mn, Fe, Mg, Al, Ni, and Co and M2 elements in the oxides are Na, Li, Mg, Zr, Zn, Ba, Bi, Y, Ti, Al, and B. *Id.*, 5:25-30.

110. As Fujiki explains, the positive electrodes in lithium-ion batteries generally suffer from stability issues over multiple cycles, resulting in lower capacity and charge/discharge efficiency. The positive electrode can also react with the liquid electrolyte and cause material degradation and unwanted byproduct formation which will further deteriorate the cell performance. Fujiki explains that coating the positive electrode active material surface with a metallic oxide or metallic fluoride has been widely adopted to overcome these stability and reactivity issues. *Id.*, 3:60–4:25. Fujiki also discloses the negative effect of a thick coating by stating, “when the coating amount is increased in order to obtain the desired effect, diffusion of lithium ions is impeded, so that it is very difficult to obtain a sufficient capacity at charging and discharging current values in practical-use ranges.” *Id.*, 2:38–41.

111. Fujiki describes using “particles of a lithium compound oxide which has a laminar rock salt structure” as the positive electrode active material. The compound oxide particles contain a principal transition metal A which is “selected from the group consisting of at least Ni, Co and Mn.” *Id.*, 4:48–67. Fujiki states that the metallic element M1, while coated on the surface of the active material (containing element A), suppresses the oxidation activity. Fujiki further discloses

that the optimized transition metal atomic ratio of M1 should be within the range of $0.001 < M1 < 0.2$. The metallic element M2 stabilizes the crystal structure of the compound oxides during charging. The optimized transition metal atomic ratio of M2 (in the form of oxides) should be within the range of $0.001 < M2 < 0.3$. *Id.*, 5:6–62.

112. By using the compound containing element M1 on the surface of lithium-ion active material particles (containing element A) and further coating these particles with the compound containing element M2, Fujiki discloses several fabricated positive electrode active material particles with stable crystal structures and reduced degradation over time (providing examples of coatings used with positive electrode active materials). *Id.*, 11:1–26:67.

113. In Example 1, Fujiki discloses a positive active material that utilizes Mg as element M1 and Li_3PO_4 as element M2. After characterizing the material with SEM and EDX, Mg is found to be uniformly distributed over the whole surface of the active material particle. *Id.*, 11:1–12:67.

114. In Example 14, Fujiki discloses using a lithium cobalt oxide (LCO) active material with magnesium (M1) coverage on the surface of the particle, which is further coated by titanium oxide (M2). The Mg:Co atomic ratio ($M1/M1+A$) is 0.01 and TiO_2 :Co atomic ratio ($M2/M2+A$) is 0.015. *Id.* 31:1–32:67; Table 1. SEM and EDX analysis reveal the uniform distribution of Mg over the entire surface of

LCO. Auger electron spectroscopic analysis reveals that the Mg concentration varies from the surface to the bulk portion of the particles. The presence of TiO₂ is revealed by X-ray diffraction analysis. *Id.*, 18:63–19:22.

115. In Example 23, Fujiki discloses the positive electrode active material lithium cobalt oxide (LCO) is coated with MgCO₃ and lithium fluoride (LiF). The Mg:Co atomic ratio (M1/M1+A) is 0.01 and LiF:Co atomic ratio (M2/M2+A) is 0.002. *Id.*, 32:1–32:67, Table 1. SEM and EDX analysis reveal the uniform deposition of LiF at the surface of LCO. Auger electron spectroscopic analysis reveals that the Mg concentration varies from the surface to the bulk portion of the particles. *Id.*, 22:57–23:17.

116. In Comparative example 2, Fujiki discloses the positive electrode active material lithium cobalt oxide (LCO) is coated with MgCO₃ with a Co:Mg atomic ratio (M1/M1+A) of 0.23. *Id.*, 32:1–32:67; Table 2. SEM and EDX analysis reveal the uniform deposition of Mg at the surface of LCO. Auger electron spectroscopic analysis reveals that the Mg concentration varies from the surface to the bulk portion of the particles. *Id.*, 26:26–50.

117. Fujiki discloses that the positive electrode consists of the coated active material, a conductive agent and a polymeric binder such as PVDF. In addition, lithium salts such as LiPF₆ can be used in organic solvents as liquid electrolytes. The

performance of the cells manufactured was evaluated at 45°C. The voltage range was 3–4.35V and the cells were charged/discharged for 300 cycles. *Id.*, 11:35–13:50.

118. Fujiki discloses the capacity retention (after 300 cycles) for all the combinations of positive electrode materials. Approximately 80% (or higher) of the initial capacity is retained for all the combinations of a coating that includes Mg. *Id.*, 32:1–32:67, Table 1. For example 14, the initial capacity of the cell is 8.9 Wh for a charging voltage of 4.35V. After 300 cycles, it shows 20% loss in initial capacity (80% retention). *Id.*, 32:1–32:67, Table 1. Compared to a control test described in comparative example 1, where no Mg coating (element M1) is used, the capacity retention decreases to 31% of its initial value. This shows that the surface coating with M1 element significantly improves the cell performance. *Id.*, 32:1–32:67, Table 2.

119. Comparative example 3 shows that the capacity retention for the positive electrode without any M2 element coating is only 50% after 300 cycles. This signifies the importance of the coating of a second compound (M2 compound) which stabilizes the crystal structure of the active material during cycling. *Id.* Comparative example 2 shows that if the Co:Mg atomic ratio (M1/M1+A) is increased to 0.23, the capacity retention decreases to 71%. *Id.* This signifies that the composition of the metallic element M1 should be within the preferable range for better cell performance.

120. Fujiki thus concludes “where the compound oxide particles coated with the metallic element having a composition in the preferable range...are used as the positive electrode active material, both a high initial capacity and a high capacity retention could be obtained simultaneously.” *Id.*, 34:8–13 (emphasis added).

B. Ground 1: Claims 1, 4, 5, and 8 Are Rendered Obvious by Shim Alone or in Combination with Lee

1. Motivation to Combine Shim with Lee

121. In my opinion, Shim, either alone or in combination with Lee, renders obvious the lithium-ion secondary battery described by the challenged claims. As explained in further detail below, both Shim and Lee disclose the use of a metal fluoride coating on LCO and use similar analytical tools (namely, electron microscopy and charge-discharge cycling) to both characterize a coating layer on LCO and evaluate its impact on cycling performance.

122. In my opinion, a POSITA would have been motivated to combine Shim with Lee for several reasons. Both Shim and Lee are focused on developing a lithium cobalt oxide positive electrode active material (PEAM) that enables a lithium-ion battery with improved energy density and improved cycle life—a constant aim of battery research and development. Both Shim and Lee demonstrate testing of their PEAM at charging voltages up to 4.5 V vs. Li/Li⁺, charging conditions that were understood by POSITAs to enable a higher energy densities. Both Shim and Lee were able to demonstrate improved cycling stability at elevated charging voltages

through the use of a protective coating layer on the PEAM. As Shim explains, “[c]oating is one of the simplest as well as most effective methods to be considered for minimization of the degradation and consequent capacity fade during cycling.” Ex. 1005, 1. Lee states that “[t]he coating layer has been reported to stabilize the interface reaction and improve the electrochemical properties of the cathode” and that “[o]ne approach for improving the electrochemical properties of a cathode is surface modification by coating with stable materials, such as . . . fluorides.” Ex. 1006, 1. In addition, both Shim and Lee specifically highlight the benefit of incorporating magnesium into this protective layer. Ex. 1005, 1; Ex. 1006, 1, 6. Accordingly, it would have been obvious to a POSITA that Shim and Lee both describe similar approaches, *i.e.*, protective coatings containing magnesium, to the same problem, *i.e.*, PEAM degradation during cycling at elevated charging voltages, and are not technically incompatible.

123. Moreover, through the teachings of Shim and Lee—which both teach the success of coating materials on LCO—a POSITA would have been motivated to explore combinations of coating materials including fluorine. As I explained previously, a POSITA would have understood that fluorine-containing electrolytes like LiPF_6 react with the cathode material to form a fluorine-containing film, or thin layer, on the surface of the cathode. *See, e.g.*, Ex. 1024, 1–2 (explaining that LiF “precipitates on the electrode particles”). A POSITA would have been aware that

studies describe a beneficial effect of this surface film, which can provide physical protection. *See* Ex. 1022, 35 (such surface layers “should protect the cathode from reactions with acidic species in the solution,” improving the lifespan and capacity of the battery); Ex. 1025, 3 (a fluorine-containing solvents “provide[] an electrochemically stable surface film on a LiCoO_2 cathode and significantly improve[] the cycle performance under a high-voltage condition.”).

124. Additionally, while a POSITA would understand that the surface film (in other words, the CEI) itself would contain fluorine, they also may take additional steps to use a fluorine-based coating to protect the cathode surface. A POSITA would also have been aware that other studies described a detrimental effect of LiPF_6 degradation, including the generation of hydrofluoric acid species that could degrade the cathode surface. Indeed, both Shim and Lee state intentions to prevent unwanted HF formation on the PEAM particles. *See* Ex. 1005, 2 (beneficial to prevent formation of HF); Ex. 1006, 3–4 (“It is well-known that acid (such as HCl and **HF**) can be generated from the reaction between water and salts (such as LiClO_4 and ***LiPF*₆**) in the electrolyte. . . . The acid-containing electrolyte attacks the surface of the cathode during cycling.”) (emphasis added); *see also* Ex. 1023, 1 (“[T]he apparent capacity fading of the LiCoO_2 electrodes is attributed mostly to changes on their surface the presence of HF in solutions may play a major negative role.”); Ex. 1024, 1 (beneficial to suppress HF formation); Ex. 1027, 1 (describing “capacity

loss of LiCoO_2 ” “mainly due to the interfacial impedance growth between the cathode and electrolyte, which results from side reactions involving LiPF_6 ”).

125. To this end, both Shim and Lee teach metal fluorides as candidate coating materials: Shim discloses AlF_3 as a primary example, which imparts stability since the “metal ion in the coating layer has only one stable valence state at ambient condition.” Ex. 1005, 1. Lee also explicitly mentions the same metal fluoride, stating that “our approach was inspired by the fact that other fluorides, such as AlF_3 , were very effective in modifying cathode surfaces,” and further discloses that, in particular, the “ MgF_2 coating layer was very effective in improving the electrochemical properties of the LiCoO_2 film electrode.” Ex. 1006, 1, 6. This teaching is corroborated by the prior art—numerous publications disclose that metal fluoride coating layers protect cathodes from the hydrofluoric acid generated by LiPF_6 degradation. *See* Ex. 1024, 1–2 (“This fluoride layer can provide an additional protection, forming a permanent physical barrier that restricts penetration of HF through the oxide coating.”); Ex. 1027, 1 (“The AlF_3 coating protected LiCoO_2 from the chemical attack by HF . . . and maintain[ed] the structural stability when cycled above 4.5V.”); Ex. 1029, 2 (“[F]luorine in the form of metal fluorides such as AlF_3 , CaF_2 , $\text{MgF}_{[2]}$, LaF_3 , CeF_3 , FeF_3 is considered to be one of the most promising coating materials to improve electrochemical performance of cathode, due to . . . preventing the direct contact between lithiated cathode and liquid electrolyte.”).

126. A POSITA would have known that Lee's MgF_2 coating would provide these exact benefits against cathode degradation. Lee teaches that "when the surface of the cathode is covered with a coating layer, the coating material serves as a protective layer, preventing direct contact with the reactive electrolyte." Ex. 1006, 4. Lee further notes that the " MgF_2 coating layer seemed to react with the acidic electrolyte and incur damage on cycling instead of the electrode material." *Id.*, 5; *see also id.*, 6 ("[T]he MgF_2 coating layer was effective as a protective layer to prevent the dissolution of the transition metal . . . which may lead to enhanced electrochemical properties.").

127. Beyond coating strategies alone, and considering that the key finding of Shim is the "synergetic effects" realized by combining magnesium doping with an LTO protective coating, a POSITA would have been motivated to further explore such a combination of materials. To this end, a POSITA would have found it obvious and desirable to apply the teachings of Shim to a standard LCO-base material, as opposed to specially synthesizing a magnesium-doped LCO as Shim did. This would have been appealing for a variety of reasons—including commercial applications—because standard LCO material was in widespread use at the time and was readily available. In this context, the MgF_2 coating reported in Lee would have been an attractive combination with Shim's LTO layer to ultimately create a Mg-rich LTO layer. This combination would lead to similar results to Shim's Mg-doped particle,

with the additive benefits of a fluorine-containing surface layer to better protect against attack from HF, as I explain above.

128. Furthermore, a POSITA would have been motivated to apply the surface characterization approach demonstrated in Lee with the PEAM teachings of Shim. Lee discloses XPS depth profiling as a method to characterize the elemental concentration in the surface layer of the PEAM. Lee highlights the findings of this approach both before and after charge-discharge cycling, demonstrating that XPS is a valuable method to better understand the relative concentrations of Mg, Co, F, and O near the surface of the PEAM. Ex. 1006 at 2–5. Applying this surface characterization method to Shim would have been obvious to a POSITA to gain further insight into the properties of the surface layer and how they change during cycling. This technique would complement the nano SIMS and electron microscopy techniques reported in Shim.

129. A POSITA would have had a reasonable expectation of success in combining the teachings of Lee and Shim. A POSITA would have understood that such teachings could be combined in a variety of ways. For example, prior publications disclosed that coating elements such as magnesium, titanium, and fluorine could be interchangeably combined in a coating layer or doped in the bulk of the PEAM. Ex. 1017, [0011]–[0013] (disclosing “a coating layer positioned on the lithium metal composite oxide core and including a fluorine compound” which

“may be one or more selected from the group” including TiF, MgF₂, TiF₃, and TiF₄); Ex. 1018, 2:66–3:15 (same); Ex. 1019 (Liu), 1:24–31 (“B is at least one element of Ti, Zr and HF, and C is F or P” and both B and C are “distributed on a surface of the positive electrode active material”); Ex. 1020, [0022]–[0023] [0030] (“[T]he cathode active material includes a surface layer made of an oxide including at least one surface element selected from the group consisting of titanium, silicon, magnesium, and zirconium.”); Ex. 1008 (Fujiki), 4:48–6:11 (“The metallic element M2 is particularly preferably at least one element selected from the group consisting of Na, Li, Mg, Zr, Zn, Ba, Bi, Y, Ti, Al and B.”).

130. Additionally, a POSITA would have also understood that multiple coating layers could also be layered on top of the PEAM. *See* Ex. 1020, [0027]–[0032], [0077] (“Thus, in the embodiment, the surface layer is arranged on the coating layer.”). Indeed, I understand that the Examiner read Ooyama (Ex. 1020) to include multiple coatings (including the “coating layer” and “surface layer”) arranged on a PEAM particle. Ex. 1002, Vol. 1, 140–41. Thus, a POSITA would have known that Lee’s MgF₂ coating layer could be added on top of the Shim’s lithium titanate, and would have had a reasonable expectation of success in doing so.

2. Independent Claim 1

- a. [Preamble]: A lithium-ion secondary battery comprising a positive electrode active material layer comprising:

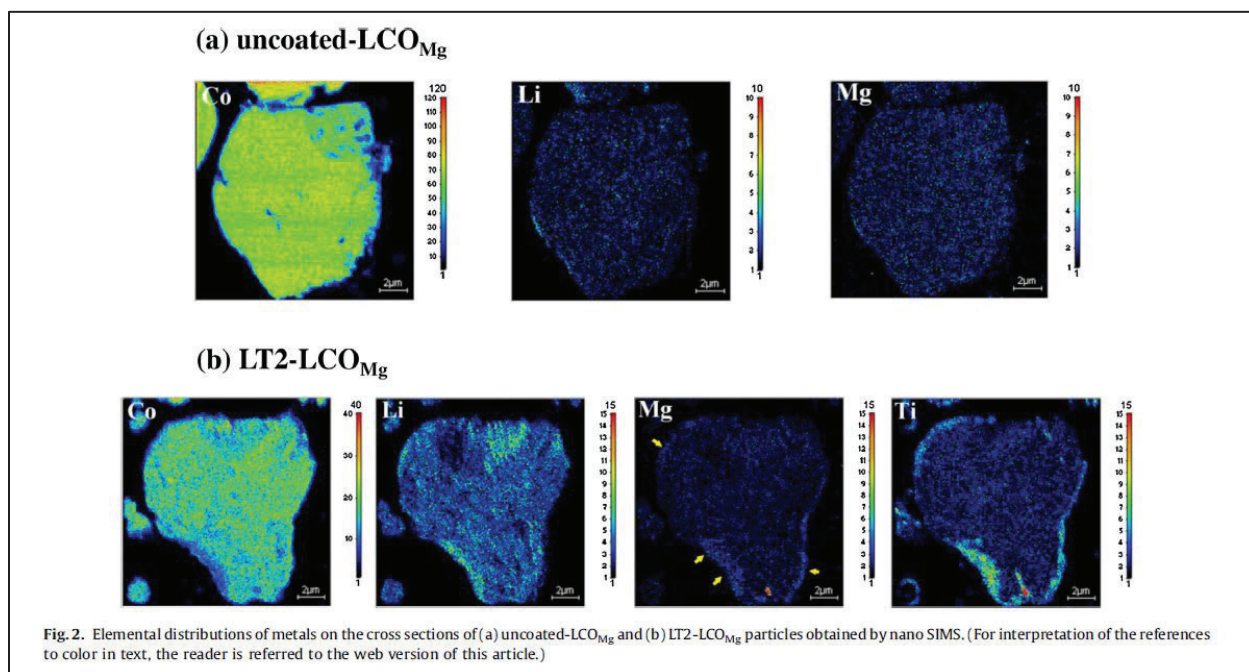
131. In my opinion, Shim discloses “[a] lithium-ion secondary battery comprising a positive electrode active material layer.”

132. As an initial matter, because Shim studies the charge-discharge behavior of lithium titanate-coated, Mg-doped LiCoO_2 , a POSITA would understand that Shim discloses rechargeable—or “secondary”—batteries. Ex. 1005, 3. Shim describes that the cathode of a lithium-ion battery is typically made of layered lithium transition metals, and that lithium transition metal oxides, such as LiCoO_2 , comprise the *active material of a cathode*. *Id.*, 1–2. Because the term “cathode” is used interchangeably with “positive electrode” in the battery field, a POSITA would have understood that Shim is a study on the “positive electrode active material” layer for application in a lithium-ion secondary battery. For instance, Shim describes the synthesis of a LCO-type material and the preparation of a positive electrode active material layer on an aluminum foil current collector. *Id.*, 3 (“First, the active materials, conducting materials (Super-P carbon black) and binders (polyvinylidene fluoride) were blended with a ratio of 96: 2:2 (in wt%) in N-methyl- 2-pyrrolidone. Then, the slurry was cast on aluminum foil and dried at 110 °C for 10 h in a vacuum oven.”).

b. [Element 1.1]: a positive electrode active material particle comprising

133. In my opinion, Shim discloses this element. Shim describes a lithium-ion battery comprising a positive electrode active material such as a lithium

transition metal oxide. *See* Ex. 1005, 1–2 (PEAMs can comprise LCO, including in the form of LiCoO_2). A POSITA would have understood that positive electrodes in lithium-ion secondary batteries contain microparticles of active material. For instance, Shim provides microscope images of such positive electrode active material particles:



Ex. 1005, 4, Fig. 2. Moreover, Shim describes a method of preparing “Mg-doped LiCoO_2 *particles*,” further indicating that Shim teaches PEAM *particles*. *Id.*, 2, Scheme 1 (emphasis added).

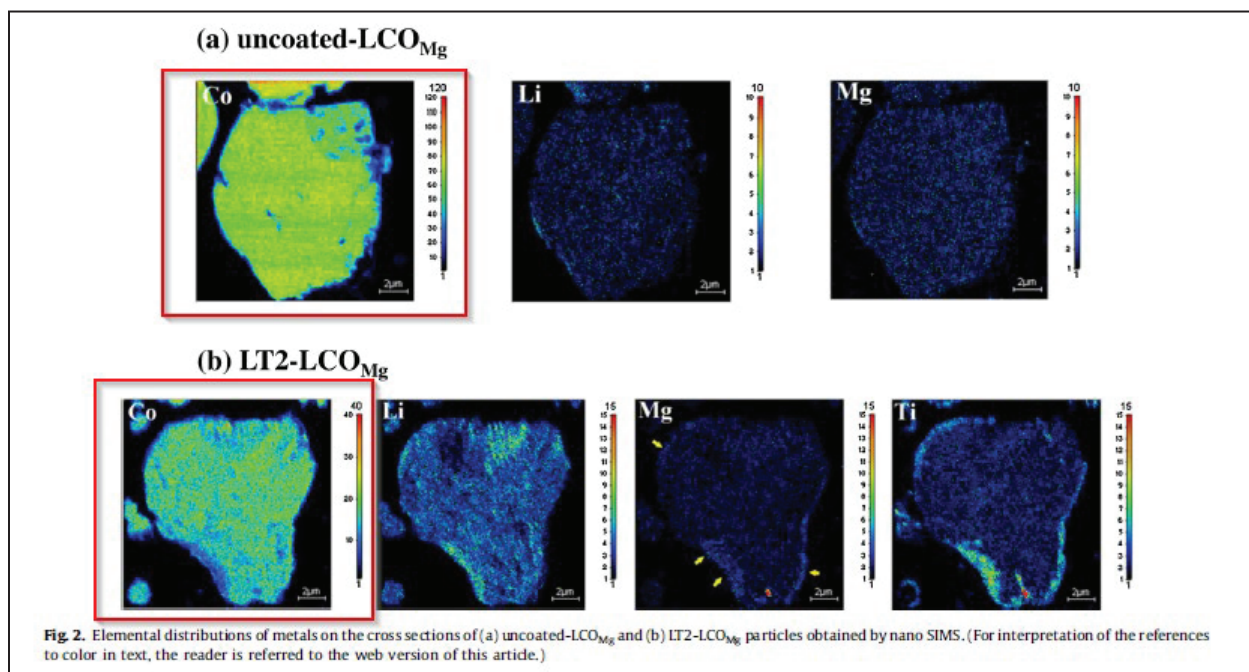
c. [Element 1.2]: a first transition metal,

134. In my opinion, Shim discloses this element. As an initial matter, a POSITA would have understood a transition metal to refer to an element that is

found in groups 3–12 in the periodic table. For example, cobalt (Co), nickel (Ni), and manganese (Mn) are examples of transition metals, as I show below in blue:

Group Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	* 71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	* 103 Lr	* 104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og
			* 57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb		
			* 89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No		

135. Shim discloses PEAM particles that comprise a first transition metal M, where M is cobalt in the form of Mg-doped LiCoO₂. Ex. 1005, 1. More specifically, in Figure 2, Shim depicts the elemental distribution of a first transition metal cobalt (cobalt spatial distribution map annotated in red boxes) in a positive electrode active material particle:



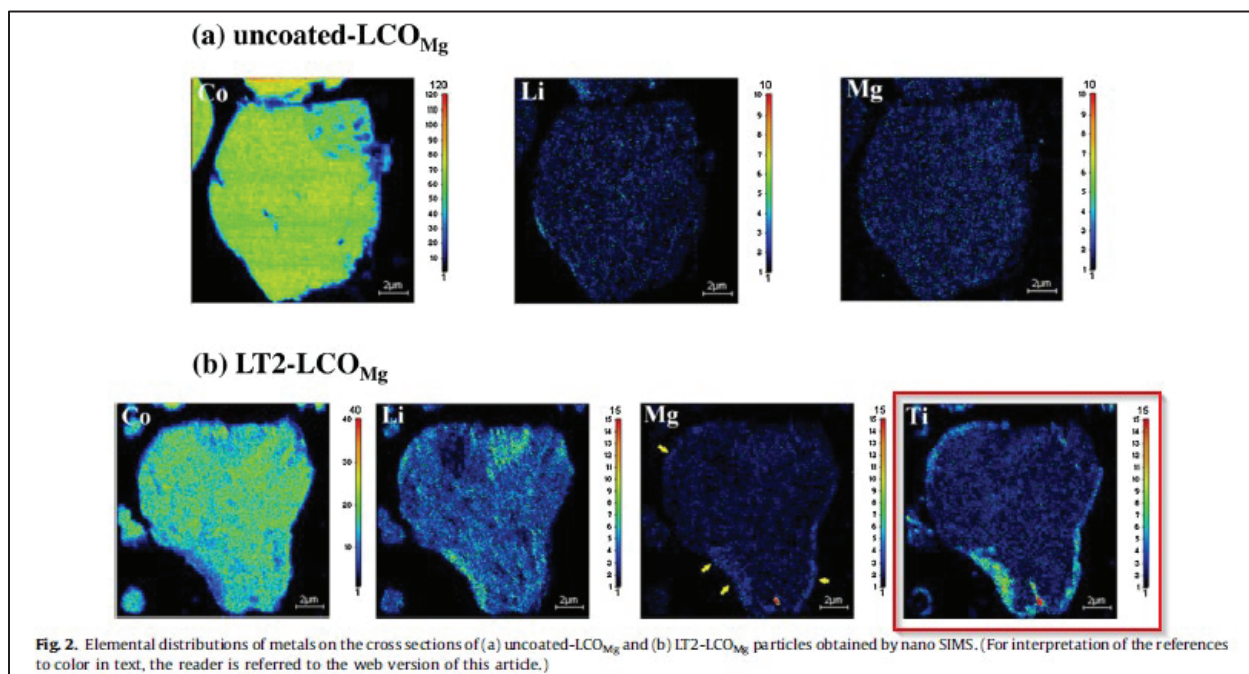
Id., Fig. 2 (annotated). Shim also describes the use of a broad range of PEAM particles in its introduction: “layered lithium transition metal oxides (LiMO₂, M: Co, Ni, Mn, and so forth) have been one of the most predominant cathode materials in LIBs, particularly, for portable electronics” *Id.*, 1 Shim also describes a cathode material degradation mechanism where such “**transition metal** ions” from the PEAM particle “dissol[ve]” into the electrolyte solution. *Id.* (emphasis added).

d. [Element 1.3]: a second transition metal,

136. In my opinion, Shim discloses a second transition metal. As I stated above, a POSITA would have understood a transition metal to mean an element that is found in groups 3–12 in the periodic table (shown in blue in the periodic table above).

137. As I explained previously, Shim discloses PEAM particles that comprise a first transition metal M, such as “Co, Ni, Mn, and so forth.” Ex. 1005, 1. Shim then discloses a second transition metal (different from the first) because it describes coating a lithium metal oxide active material with a suitable metal oxide, including oxides of zinc and titanium (which are transition metals). *Id.* Such oxide coatings were well known in the art. *See* Ex. 1016, 1 (“The other approach involves coating the cathode particles [e.g., LCO] with a thin layer of oxide materials such as Al_2O_3 , B_2O_3 , MgO , TiO_2 and ZrO_2 .”). Specifically, Shim discloses Mg-doped LiCoO_2 particles with a lithium titanate coating, including $\text{Li}_4\text{Ti}_5\text{O}_{12}$, where titanium is the second transition metal and cobalt is the first transition metal. Because zinc and titanium are transition metals, Shim discloses a “second transition metal” different from the “first transition metal” (including nickel, copper, and manganese).

138. Further, Shim specifically depicts the elemental distribution of titanium, a second transition metal (Ti spatial distribution map annotated in red boxes) in a PEAM by nano SIMS analysis:



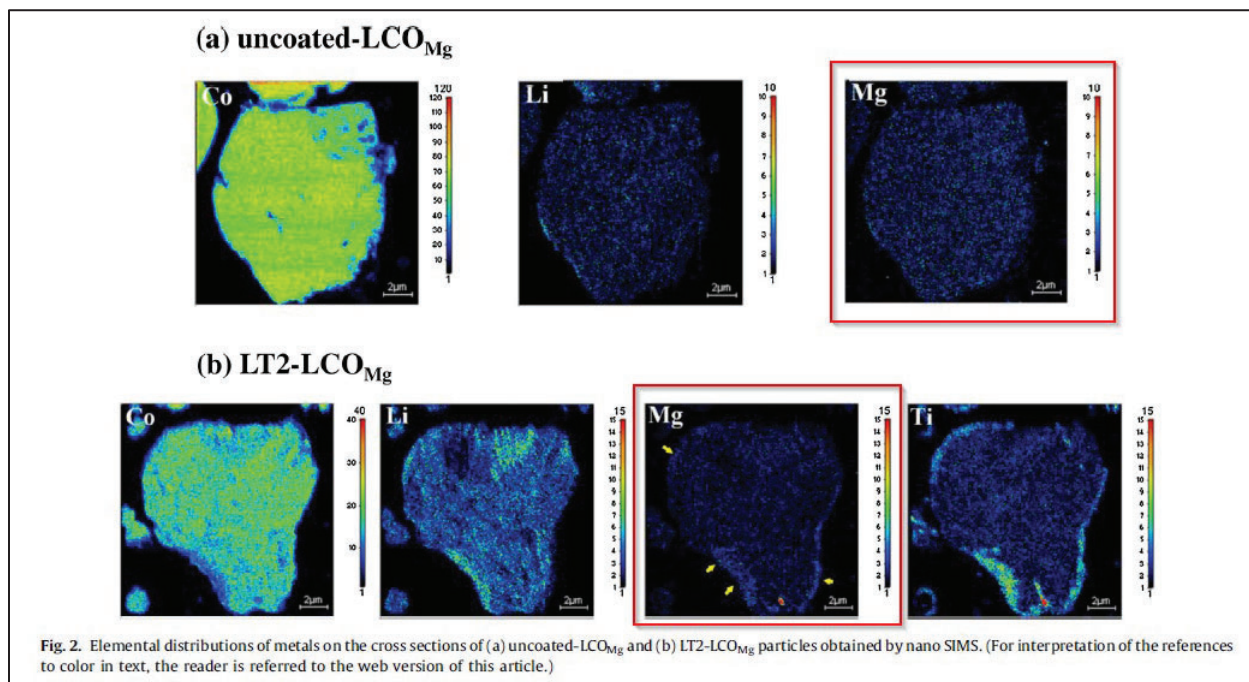
Ex. 1005, Fig. 2 (annotated); *see also id.*, Fig. 5 (showing the titanium-containing coating on Mg-doped LCO).

e. [Element 1.4]: a representative element, and

139. In my opinion, Shim discloses the use of magnesium with lithium cobalt oxide to create a magnesium-rich layer on the surface of the PEAM particles, where magnesium would satisfy the “representative element” as described by the ’828 Patent. The ’828 Patent lists the following examples of representative elements: magnesium, calcium, and sodium. Ex. 1001, 10:29–33, 11:65–12:4, cl. 4.

140. Shim describes doping PEAM particles with magnesium, a representative element. Ex. 1005, 1. Shim notes that magnesium is “distributed over the entire particle” of a PEAM, and may even “diffuse” into the coating layer to

“improve electrochemical performance.” *Id.* As I annotate below in red boxes, Shim depicts the elemental distribution of magnesium in a PEAM by nano SIMS analysis:



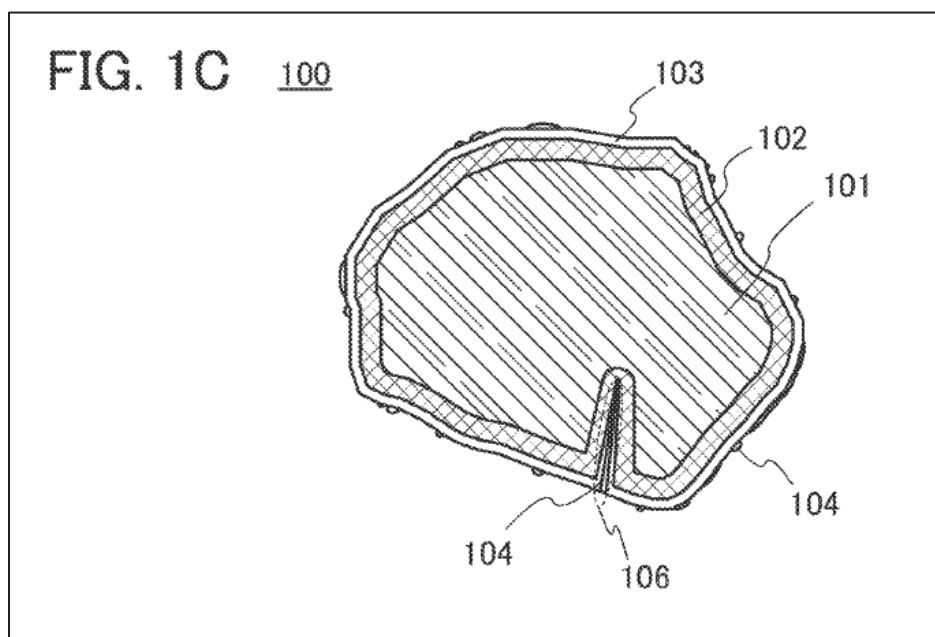
Ex. 1005, Fig. 2 (annotated).

f. [Element 1.5]: fluorine

141. In my opinion, Shim, either alone or in combination with Lee, renders obvious a positive electrode active material containing fluorine. Shim explicitly states that “[c]oating is one of the simplest as well as most effective methods to be considered for minimization of the degradation and consequent capacity fade during cycling,” and identifies aluminum fluoride (AlF₃) as an example of a “successful coating material.” Ex. 1005, 1; *see also* Ex. 1008, 6:3–11 (teaching similar use of metallic halides—especially metallic fluorides—to provide physical protection to PEAM particles). In light of Shim’s disclosure, a POSITA would have understood

that fluorine could be used to improve positive electrode active materials. While Shim specially focuses on the synergistic effects of coating and doping for LiCoO_2 cathodes, a POSITA would have also known from the prior art that metal fluorides were well-known *coating* materials for PEAM particles.

142. A POSITA would have understood this coating layer to be part of the PEAM particle. As I reproduce below, the '828 Patent illustrates that the PEAM includes both the bulk and coating of a particle:



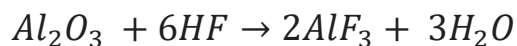
Ex. 1001, Fig. 1C; *see also id.*, Figs. 1A–B. Specifically, the '828 Patent states that “positive electrode active material 100” includes regions 102 and 103. *Id.*, 9:30–39. It also indicates that regions 102 and 103 are coating layers. *Id.*, 12:45–49. In light of these disclosures, a POSITA would have recognized that a coating layer is part of the PEAM, and would have understood that Shim’s disclosure of aluminum fluoride

(AlF₃) as an example of a “successful coating” to mean that fluorine could be present in a PEAM particle.

143. Beyond the explicit disclosure of an externally-applied fluorinated coating, a POSITA would have also understood that fluorine would be present in Shim’s PEAM particle because Shim uses a “1.15 M LiPF₆” electrolyte. Ex. 1005, 3. As I explained previously in section VI.A.3, a POSITA would have been aware that a fluorinated interfacial layer would form on the surface of PEAM particles due to the reaction of the LiPF₆ salts in the electrolyte with the cathode material. These electrolyte species were known to decompose to form fluorinated species as part of the CEI of the positive electrode active material. This mechanism was well documented; for example, in 2009, Dedryvère et al. performed XPS studies to show the formation of an LiF layer on the PEAM surface due to the hydrolysis of the LiPF₆ electrolyte solution. Ex. 1021; *see also* Ex. 1023, 2 (describing LiF and Li_xPF_y) Ex. 1022, 86 (describing the presence of fluorine “on the surface” of the cathode particle “caused by the formation of LiF derivatives”); Ex. 1024, 2 (“[A] fluoride layer can form on top of an HF-scavenging oxide coating as a product of the HF-scavenging reaction.”); Ex. 1026, 6 (“[S]mall amounts of fluorine were mainly distributed along the grain boundaries of the primary particles. These were likely derived from the LiPF₆ electrolyte Fluorine was also detected at cracked particle surfaces, which suggests that the electrolyte invaded into the cracked gaps.”). Moreover, as I

explained previously, some studies even showed that the formation of this fluorine-containing layer from LiPF_6 had a beneficial effect. *See* Ex. 1022, 35 (recognizing that such a surface layer “should protect the cathode from reactions with acidic species in the solution,” improving the lifespan and capacity of the battery); Ex. 1025, 3 (concluding that a fluorine-containing solvent “provides an electrochemically stable surface film on a LiCoO_2 cathode and significantly improves the cycle performance under a high-voltage condition.”). Since commercial lithium-ion batteries undergo a formation process during battery manufacturing, the presence of fluorine at the surface of PEAM particles would be well-known to a POSITA. Accordingly, a POSITA would have understood that a fluorine-containing species would be present on the surface of the Shim’s PEAM particles.

144. Moreover, Shim discloses the use of a lithium titanate coating on the exterior of LCO. As I describe above in Section VI.A.3, a POSITA would have understood that oxide coatings improve the cycle life of LiCoO_2 through the process of scavenging HF from the electrolyte. The scavenging reaction of HF results in the formation of metal fluoride surface compounds, according to the below reaction (using aluminum oxide and aluminum fluoride as examples):



Ex. 1024, 1–2. Accordingly, a POSITA reading Shim would have understood that fluorine could be present on the PEAM surface via the reaction of HF to form surface species in the form of transition metal fluorides, such as titanium fluorides, magnesium fluorides, etc.

145. Finally, a POSITA would have also found it obvious to add fluorine to Shim’s coating layer, including by combining Shim and Lee as I explained previously in section IX.B.1. Specifically, a POSITA would have been motivated to use a metal fluoride coating to protect PEAM particles, like the ones disclosed in Shim, from hydrofluoric acid—which was a well-known degradation byproduct of LiPF_6 . Since metal fluorides were known to shield the positive electrode active material from reactions with HF in the electrolyte (as I explain above in section VI.A.3), and possessed greater stability due to the metal ion having “only one stable valence state at ambient condition[s]” (Ex. 1005, 1), a POSITA therefore would have been motivated to consider such metallic fluoride coatings on positive electrode active materials. As Lee explains, its “ MgF_2 coating layer successfully protected the surface of the cathode from the reactive electrolyte, leading to enhanced electrochemical properties for MgF_2 -coated LiCoO_2 films.” Ex. 1006, 1. The fact that Shim expressly describes using LiPF_6 in its electrolyte solution would have motivated a POSITA to add fluorine (including by combining Shim with Lee’s

teachings) to protect Shim's PEAM material, as I explain above in section IX.B.1; Ex. 1005, 3; *see also* Ex. 1016, 2.

- g. [Element 1.6]: wherein the representative element and the fluorine are present in a crack portion observed from a TEM image in the positive electrode active material particle.

146. In my opinion, Shim (either viewed independently or in combination with Lee) would have rendered this claim element obvious to a POSITA.

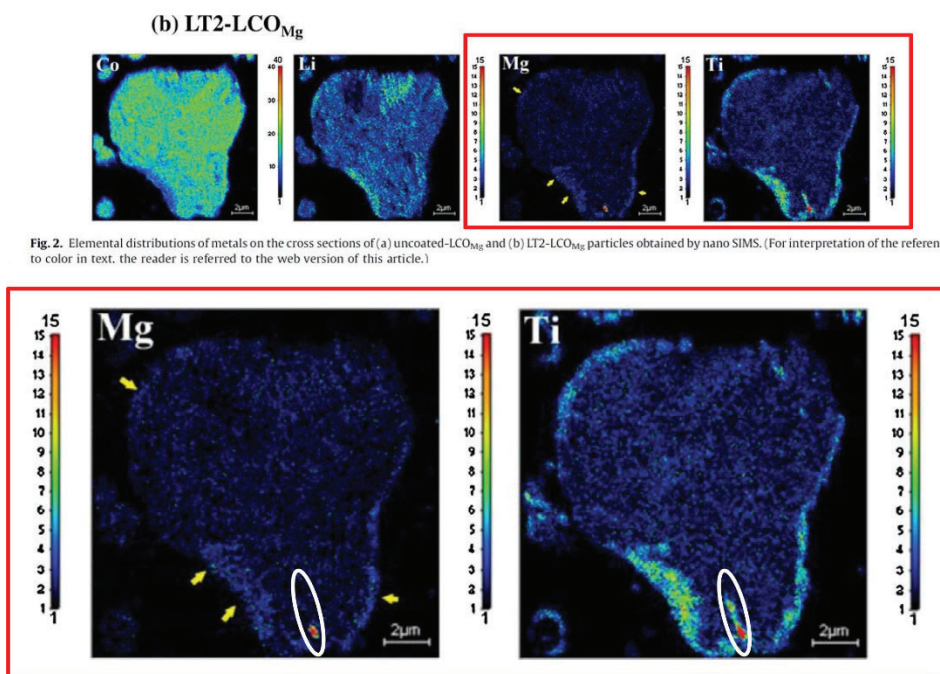
- i. Crack portion observed from a TEM image in the positive electrode active material particle

147. In my opinion, it would have been obvious to a POSITA that a “crack portion” would be present in Shim's PEAM particles.

148. Battery industry standard active material chemistries are based on lithium cobalt oxide (LiCoO₂), which is used in Shim's PEAM (in the form of Mg-doped LCO with a LTO coating). *See* Ex. 1005, 2. As I discussed previously in section VI.A.2, such PEAM materials are prone to cracking in part due to volume expansion and contraction during charge and discharge cycles that occur as early as the initial formation process. Indeed, Shim describes that its methods offer enhanced electrochemical performance with respect to “several *degradation mechanisms*”—specifically, “cathode degradation.” Ex. 1005, 1. A POSITA would have understood that the “cathode degradation” described by Shim would include cracks, or structural defects, within the cathode structure because LCO is well-known to form

microcracks on the particle surface as a result of charge and discharge, as I explain above in section VI.A.2.

149. In fact, Shim's Figure 2 shows evidence of an apparent crack in a sample PEAM particle where surface coating elements (Mg and Ti) are present along a seam extending from the surface into the core of the PEAM particle in the region I highlight below with white ovals.¹



Ex. 1005, Fig. 2 (annotated). As shown above, a crack in the cross section of the PEAM particle shows reveals lower concentrations of Co and Li (which comprise a majority of the bulk) but higher concentrations of Mg and Ti (which are expected to

¹ Note that the elemental maps were performed on a cross-sectioned PEAM particle.

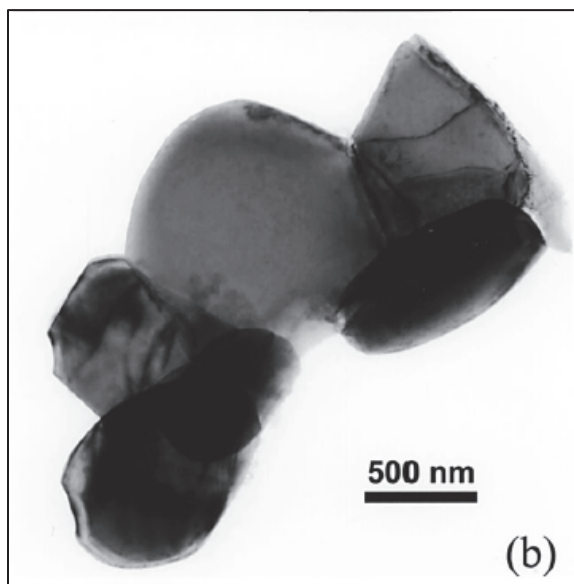
be found in the coating). Based on this figure and a POSITA's understanding of LCO-type materials, it would have been reasonable for a POSITA to conclude cracks were present in Shim's PEAM particles.

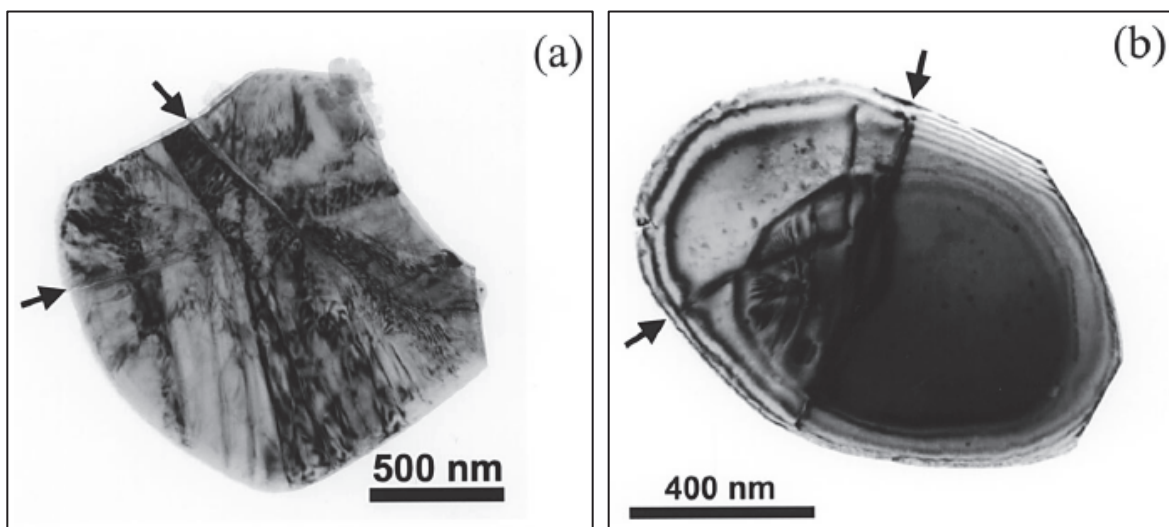
150. A POSITA would have been aware that TEM imaging could be and was used to characterize and better understand the apparent crack and other particle irregularities before and/or after cycling. A POSITA would have known that TEM was commonly used at the time of the earliest alleged priority date to characterize cracks and microscale/nanoscale defects. Indeed, Shim describes the use of TEM for imaging "structural changes of the materials after charging." Ex. 1005, 6. Further, Shim uses "TEM images" to "demonstrate the irreversible transformation of Li_2TiO_3 and the structure consistency of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ with cycling." *Id.*, 7, Figs. 5–6. Thus, a POSITA would have understood that TEM could have been used to see and examine cracks (*i.e.*, structural changes) within the PEAM particle.

151. In my opinion, a POSITA would have also found the "crack portion" obvious from reading Shim in light of his or her knowledge of the prior art. Prior publications showed that cracks in cathode particles like Shim's were inevitable and could be imaged using TEM. Specifically, Fey (published 10 years prior to Shim) describes that, "[i]t is *inevitable* that coated surfaces always have defects such as *cracks* and pinholes in them" and that such cracks "may form and close with the application of a load or upon thermal *cycling*." Ex. 1016, 4. Similarly, Wang

(published 16 years prior to Shim) describes that “[e]lectrochemical cycling creates a variety of defects in the LiCoO₂ particles in a typical composite cathode.” Ex. 1010, 8. Consistently, Shim describes “degradation and consequent capacity fade during *cycling*.” Ex. 1005, 1.

152. Further, a POSITA would have understood that such cracks in Shim’s PEAM could be observed using TEM imaging, as confirmed by prior publications. For instance, Fey also teaches that TEM can be used to “examin[e] . . . the microstructure of the coated particles.” Ex. 1016, 1, Fig. 2. And, as reproduced below, Wang also describes a “crack portion observed from a TEM image in the positive electrode active material particle” (Ex. 1010, 1, 3):

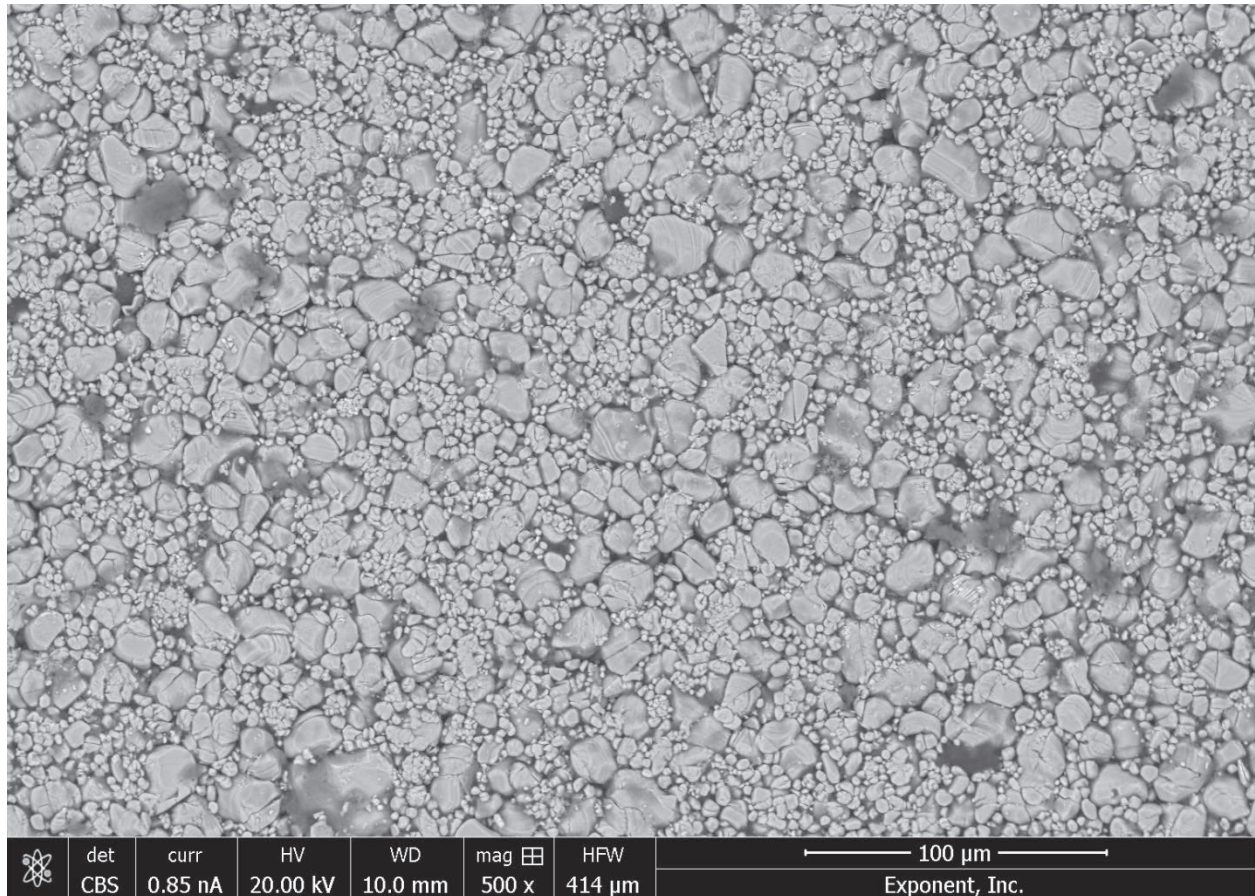




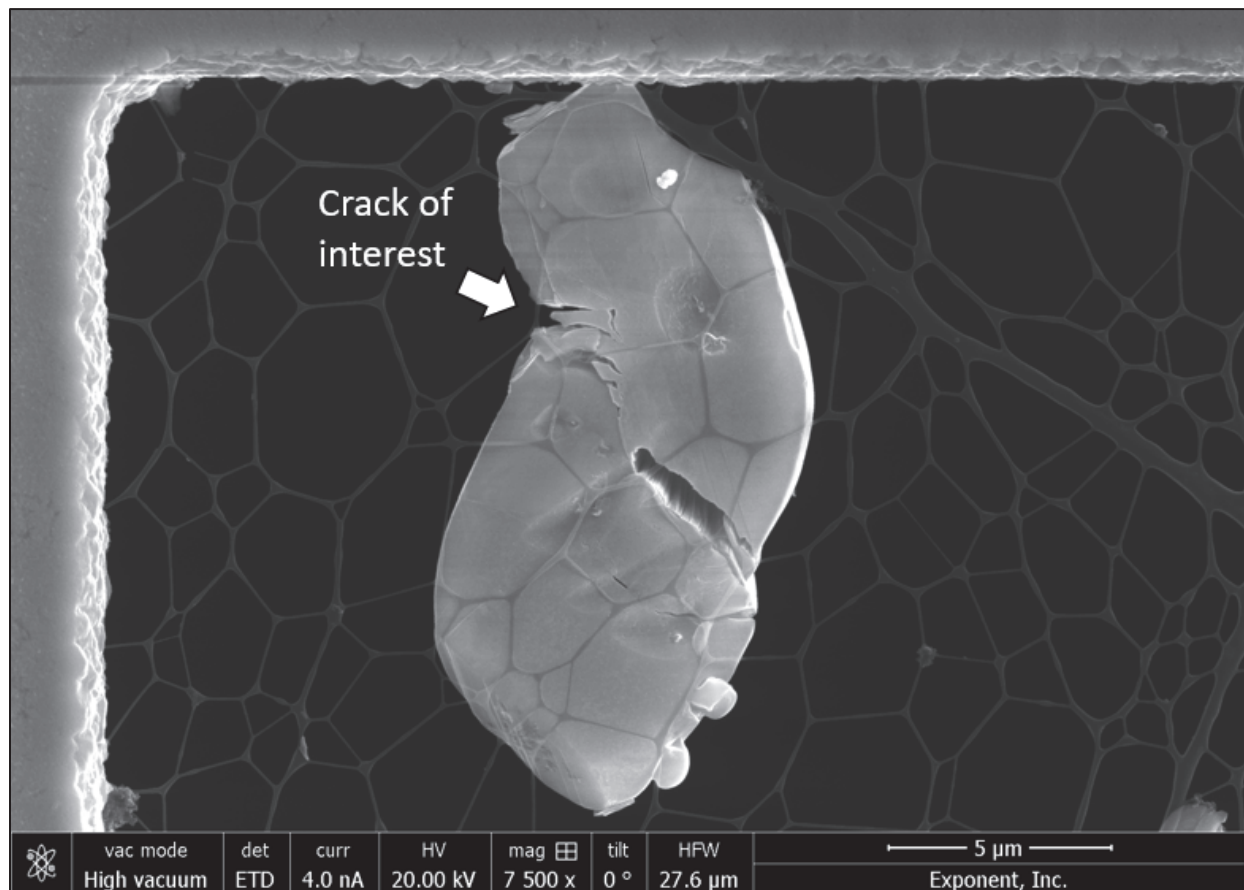
Id., Fig. 3(b), 5(b) (arrows indicating microfracture); *see also* Ex. 1026, 2 (noting that TEM image showed “fragment[s]” or “crack[s]” in cycled positive electrode grains).

153. Additionally, testing data conducted at my direction confirms, in my opinion, that a POSITA would have known Shim’s PEAM contains crack portions were observable from a TEM image, as I explain above in section VI.B. Specifically, testing of a Samsung Galaxy A8 battery sample (from August 7, 2015) revealed that

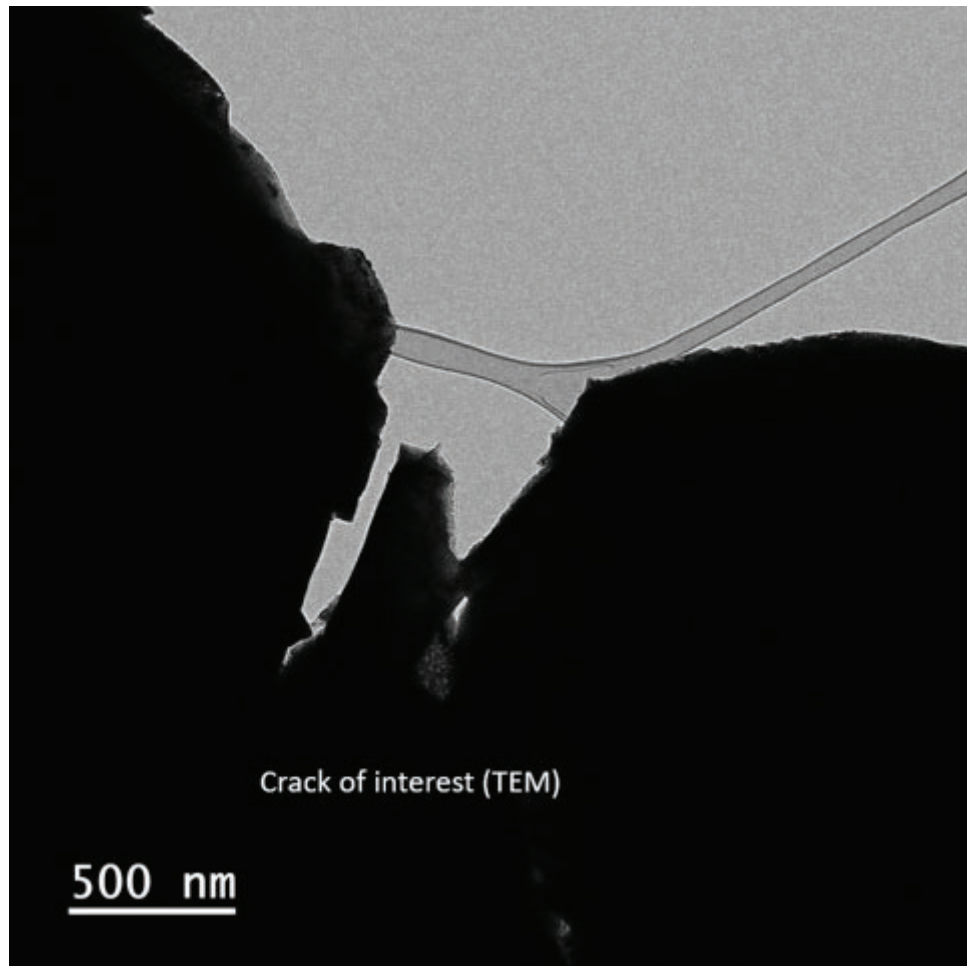
widespread cracks of the type expected from a cycled positive electrode were visible through SEM, as shown below:

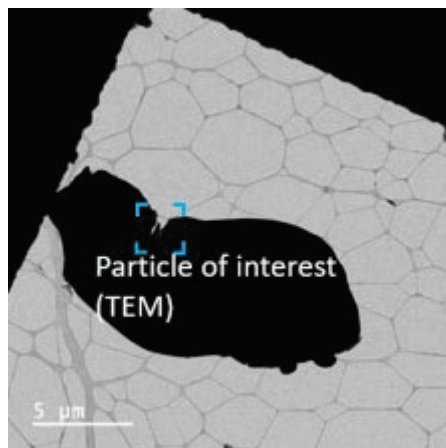


154. By isolating a single particle, the presence of cracks becomes more apparent. Before imaging with TEM, a PEAM particle of interest was first imaged using SEM, as shown below:



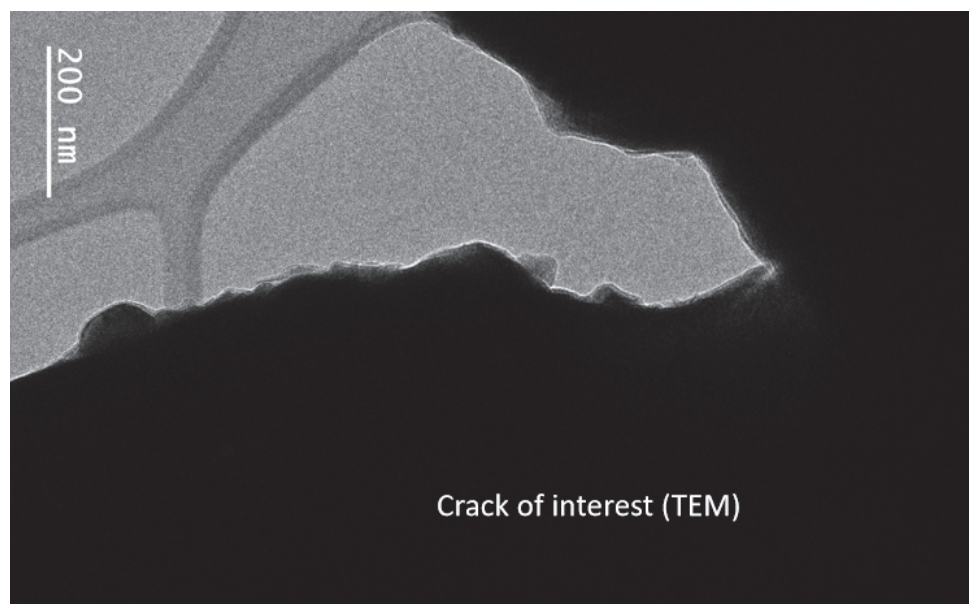
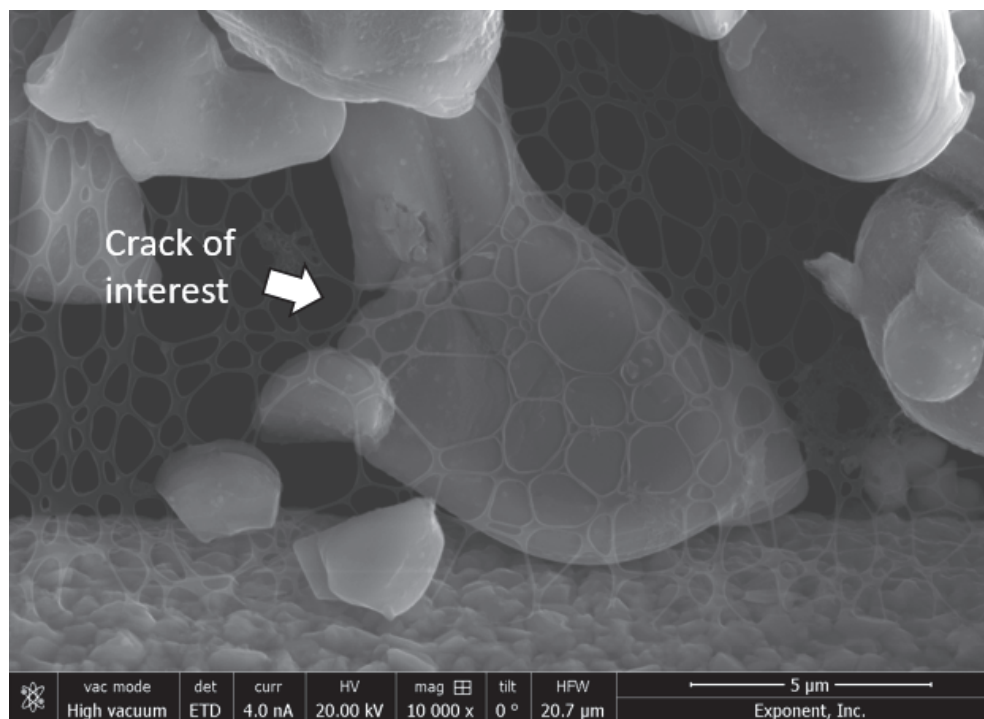
155. TEM imaging showed that the “crack portion” on the same particle of interest was indeed visible, thereby confirming that a POSITA would have found obvious a “crack portion” observable by TEM:



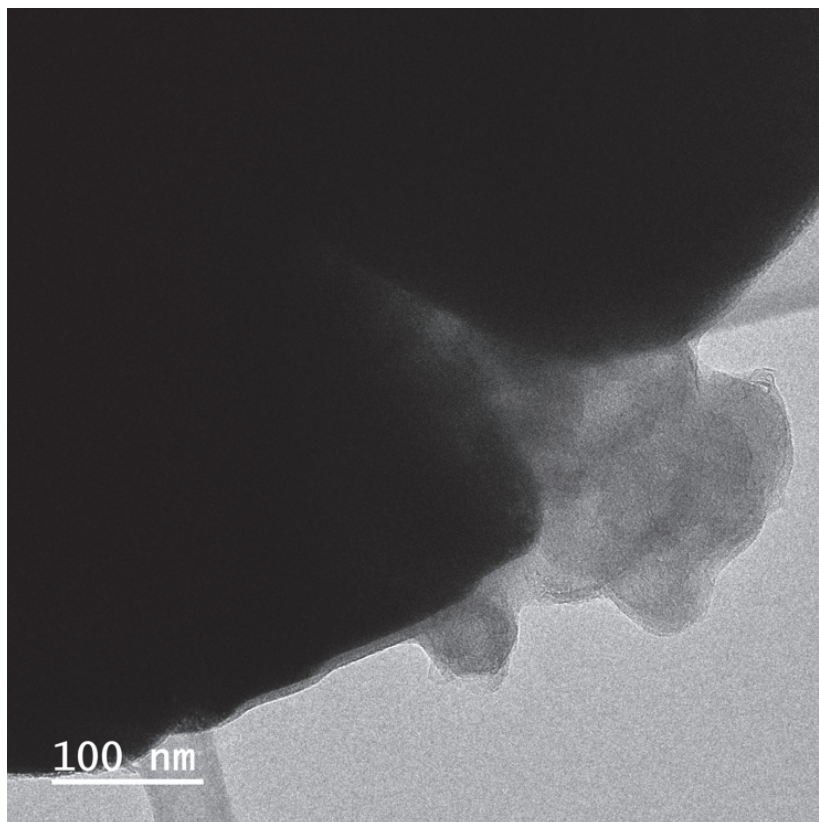


156. In these bright field TEM images, the dark colored portions correspond to solid regions of the sample which are sufficiently thick such that the incident electron beam is unable to reach the detector. Conversely, the lightest colored regions represent areas where the electron beam is not blocked by solid material (*i.e.*, is absent of PEAM). Thus, the light colored gap in the TEM image corresponds to a “crack portion” in the PEAM particle.

157. Additional samples tested (at my direction) offer similar results. For instance, the below two images from a Samsung Galaxy Note 4 battery sample (dated April 1, 2016) show SEM (top) and TEM (bottom) images revealing crack portions in a PEAM particle.



158. As a further example, the below TEM image shows a crack portion in a PEAM particle from an iPhone 5S battery sample dated October 2013.



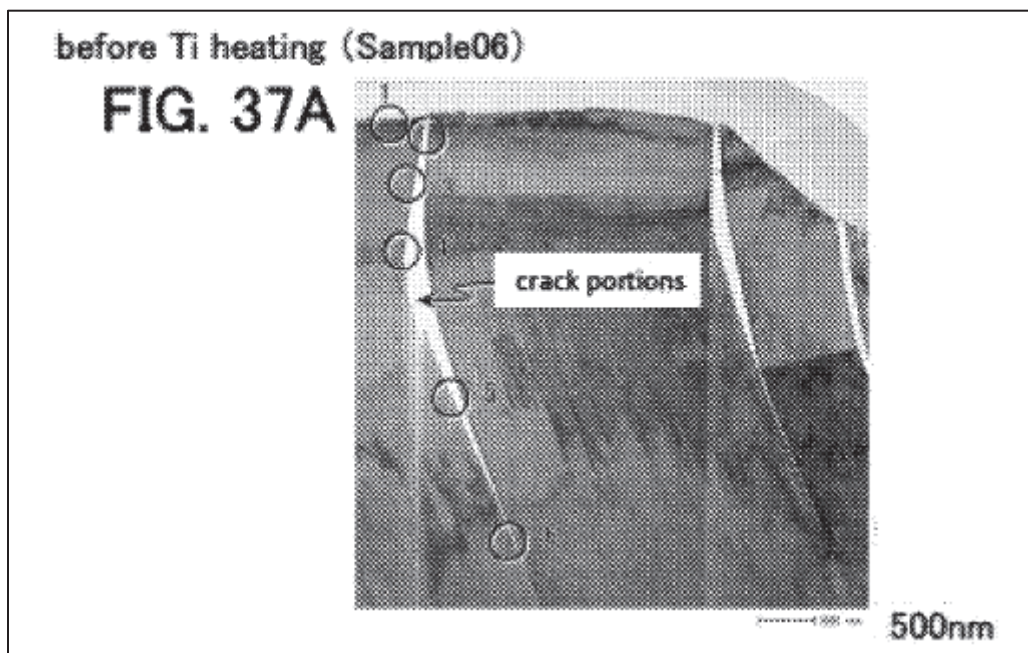
159. For the above reasons, and with reference to the testing I described above in section VI.B, in my opinion Shim renders obvious a crack portion observed from a TEM image.

ii. Representative element present in a crack portion

160. As I explained previously in section IX.B.2.f, Shim describes PEAM particles that contain the representative element magnesium. In my opinion, it would have been further obvious to a POSITA that magnesium would be present in the crack portion through at least two mechanisms.

161. First, Shim describes a method of doping the bulk of the PEAM particle with magnesium. *See* Ex. 1005, 3. Because the particle is doped with magnesium,

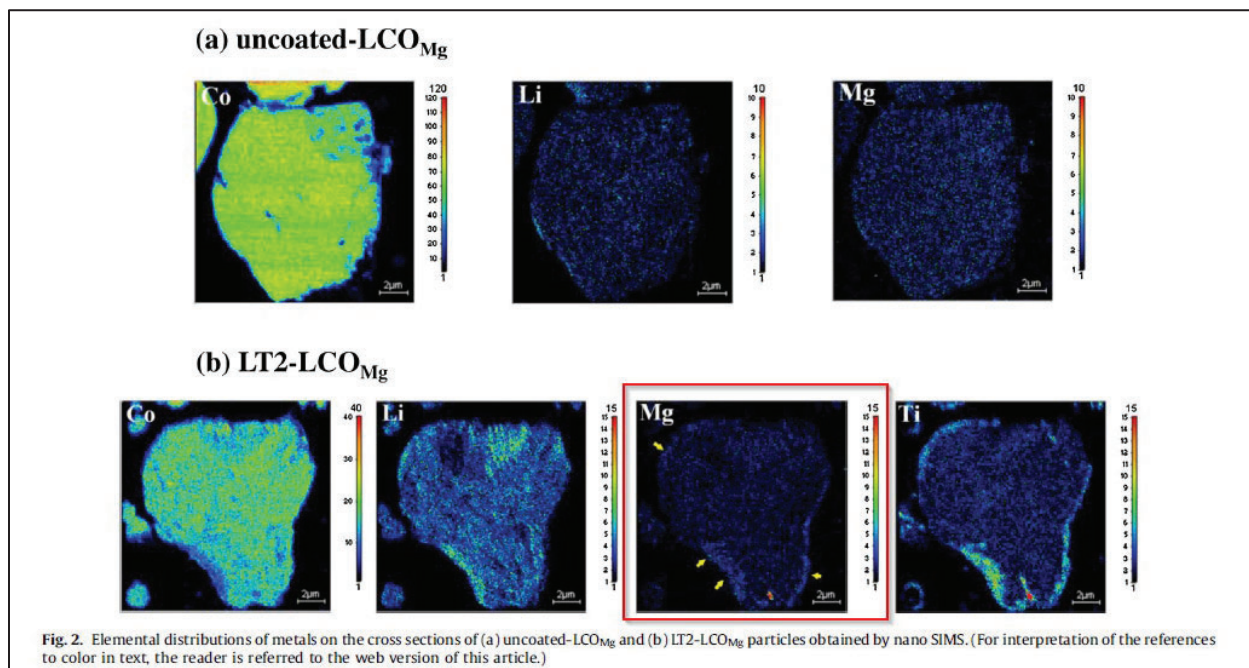
rather than merely coated with a magnesium-containing compound, magnesium is present throughout the entire particle. Thus, a POSITA would have understood that crack portions of Shim's PEAM particle that form in the bulk of the particle would necessarily contain magnesium. Further, Figure 37A of the '828 Patent confirms that the term "crack portions" (as used by the '828 Patent) encompasses portions of the bulk material of the PEAM:



Ex. 1001, Fig. 37A.

162. Second, magnesium in the bulk of Shim's particle diffuses into the coating, and elements within the coating will be found in crack portions. Shim discloses that the diffusion of magnesium into the coating is "clearly evidenced by nano SIMS analysis." Ex. 1005, 3. As I annotate below in red (in Shim's Figure 2), the elemental distribution of magnesium shows a larger concentration of magnesium

(lighter blue) at the edges of the particle (Shim's yellow arrows) than in the bulk of the particle (dark blue):



Ex. 1005, Fig. 2 (annotated). A POSITA would have understood that magnesium in Shim's coating would be ingrained in the crack portions of Shim's particle. *See* Ex. 1016, 4 (“[P]articles of the coating material [] become ingrained in the crevices and cracks on the cathode surfaces.”). As I previously discussed, evidence of an apparent crack in Shim's PEAM is visible in Figure 2, where the surface coating elements Mg and Ti are present along a seam extending from the surface into the core of the PEAM particle, in the region I highlight below with white ovals:

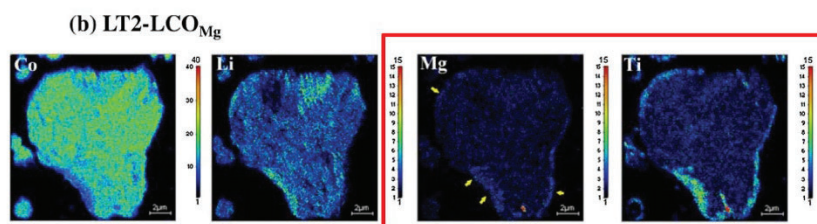
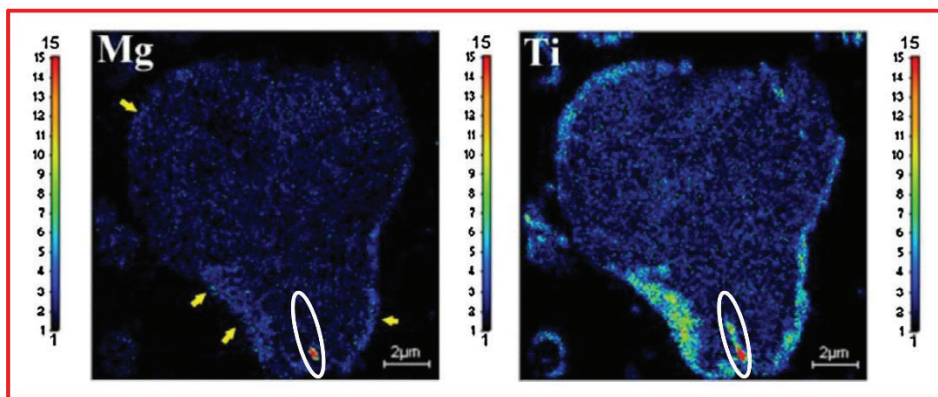


Fig. 2. Elemental distributions of metals on the cross sections of (a) uncoated-LCO_{Mg} and (b) LT2-LCO_{Mg} particles obtained by nano SIMS. (For interpretation of the references to color in text, the reader is referred to the web version of this article.)



Ex. 1005, Fig. 2 (annotated). As shown above, a crack in the cross section of the PEAM particle shows reveals lower concentrations of Co and Li but higher concentrations of Mg and Ti (which are expected to be found in the coating). Based on this figure and a POSITA's understanding of LCO-type materials, it would have been reasonable for a POSITA to conclude cracks were present in Shim's PEAM particles and that they contained magnesium.

163. Further, in my opinion, the combination of Shim and Lee I previously described renders this element obvious. Lee discloses the use of a MgF₂ exterior coating on LiCoO₂ PEAMs (where Mg is the representative element). As I explain above, if a crack forms in the PEAM as a result of electrochemical cycling, the outermost edge of the crack (where the crack extends through the surface coating)

would contain MgF_2 , implying that Mg would be therefore found in the crack portion. Moreover, in light of a POSITA's knowledge that coating materials could migrate and become embedded in the crack portion (Ex. 1016, 4 (“[P]articles of the coating material [] become ingrained in the crevices and cracks on the cathode surfaces.”)), it would have been reasonable for a POSITA to expect MgF_2 (in the case of Shim combined with Lee) to become lodged within the crack portion of the underlying PEAM.

iii. Fluorine

164. As I explained previously in section IX.B.2.f, Shim (either alone or in combination with Lee) discloses fluorine in Shim's PEAM particle—specifically, a POSITA would have understood fluorine to be present in the coating of the PEAM particle. And as I explained above in section IX.B.2.g.i, it would have been reasonable for a POSITA to expect that the coating material on the surface of the particle would become present in the crack portions of the particle. *See* Ex. 1005, Fig. 2(b) (showing high magnesium and titanium concentrations in a crack portion); Ex. 1016, 4 (“[T]he cycling process could have enabled the particles of the coating material to become ingrained in the crevices and cracks on the cathode surface.”). Thus, through the same mechanism that I previously described with respect to magnesium (in section IX.B.2.g.ii), fluoride in the coating layer would also become present in the “crack portion” of the PEAM particle.

165. In addition, it is my opinion that a POSITA would have found it obvious that fluorine-containing compounds would be found in Shim's crack portions due to the reaction of newly exposed electrode surface areas (in other words, the walls of the crack) with the electrolyte. As I explained previously in section VI.A.2, an LiPF_6 -containing electrolyte (such as the one disclosed by Shim) decomposes to form a fluorine-containing layer on the LCO surface area, which includes cracks.

3. Dependent Claim 4

- a. [Preamble]: The lithium-ion secondary battery according to claim 1,

166. As I explained previously in section IX.B.2, in my opinion Shim (either alone or in combination with Lee) renders obvious the lithium-ion secondary battery according to claim 1.

- b. [Element 4.1]: wherein the first transition metal is cobalt,

167. As I explained previously in section IX.B.2.c, in my opinion Shim discloses the use of lithium cobalt oxide where cobalt would satisfy the first transition metal as described by the '828 Patent.

- c. [Element 4.2]: wherein the second transition metal is titanium, and

168. As I explained previously in section IX.B.2.d, in my opinion Shim discloses the use of magnesium with titanium and lithium cobalt oxide to create lithium titanate-coated, Mg-doped LCO, a material that contains cobalt, titanium,

and magnesium. Here, the titanium would satisfy the second transition metal as described by the '828 Patent.

- d. [Element 4.3]: wherein the representative element is magnesium.

169. As I explained previously in section IX.B.2.e, in my opinion Shim discloses the use of magnesium with titanium and lithium cobalt oxide to create lithium titanate-coated, Mg-doped LCO, a material that contains cobalt, titanium, and magnesium. Here, the magnesium would satisfy the representative element as described by the '828 Patent.

4. Independent Claim 5

- a. [Preamble]: A lithium-ion secondary battery comprising a positive electrode active material layer comprising:

170. As I explained previously in section IX.B.2.a, in my opinion Shim discloses this element.

- b. [Element 5.1]: a positive electrode active material particle comprising:

171. As I explained previously in section IX.B.2.b, in my opinion Shim discloses this element.

- c. [Element 5.2]: a first transition metal,

172. As I explained previously in section IX.B.2.c, in my opinion Shim discloses this element.

- d. [Element 5.3]: a second transition metal,

173. As I explained previously in section IX.B.2.d, in my opinion Shim discloses this element.

e. [Element 5.4]: a representative element, and

174. As I explained previously in section IX.B.2.e, in my opinion Shim discloses this element.

f. [Element 5.5]: fluorine,

175. As I explained previously in section IX.B.2.f, in my opinion Shim (either alone or in combination with Lee) renders this element obvious.

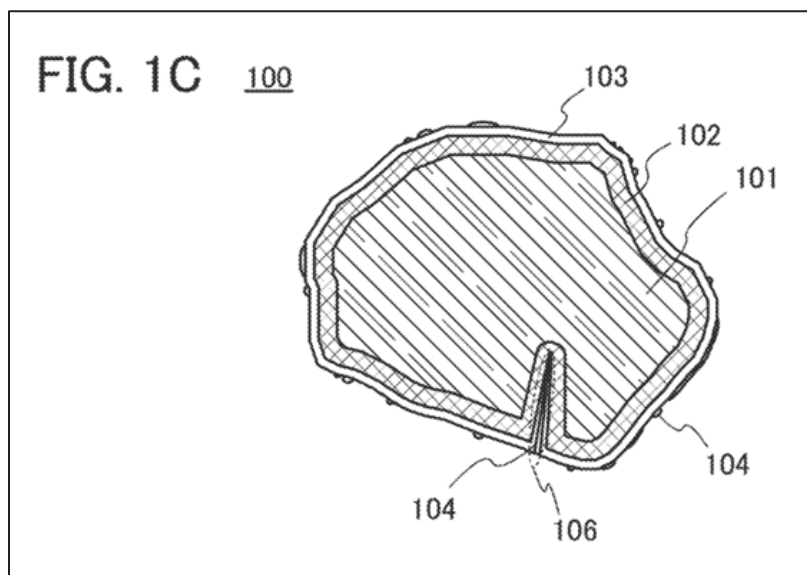
g. [Element 5.6]: wherein the representative element and the fluorine are present in a portion which includes crystal defects observed from a TEM image in the positive electrode active material particle.

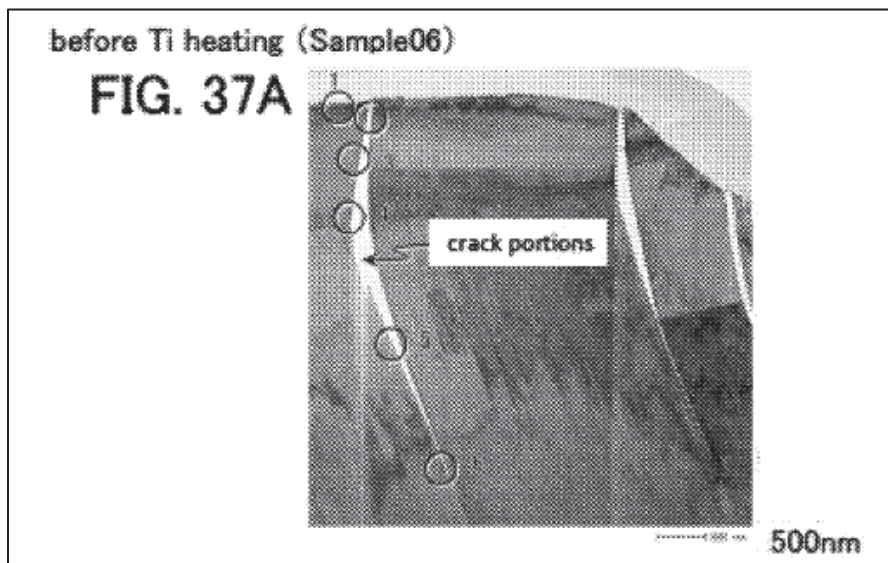
176. In my opinion, Shim (either alone or in combination with Lee) renders this element obvious because a POSITA would have known that “crystal defects” encompasses a variety of imperfections that can interrupt the periodicity of the atomic lattice.

177. As I explained above in section VIII, the '828 Patent defines a crystal defect as a “defect[] which can be observed from a TEM image and the like,” and specifically includes “a structure in which another element enters crystal, *a cavity*, and the like.” Ex. 1001, 9:55–58. Alternatively, a POSITA would have known that the plain and ordinary meaning of “crystal defects” is an “imperfection or interruption of regular arrangement of atoms in a crystalline solid.” In my opinion,

Shim (either alone or in combination with Lee) renders this element obvious under both definitions of “crystal defect.”

178. Under the '828 Patent's express definition that crystal defects are “defects which can be observed from a TEM image and the like,” cracks are a three-dimensional type of crystal defect. This is confirmed by independent claim 1 of the '828 Patent, which describes “a crack portion observed from a TEM image.” Ex. 1001, cl. 1. Further, the '828 Patent expressly states that a “cavity” is an example of a crystal defect. As shown below in the patent's Figure 1C, a crack portion (106) is a cavity within a PEAM particle.





Ex. 1001, Figs. 1C, 37A.

179. Similarly, Shim also discloses a crystal defect under its plain and ordinary meaning of an “imperfection or interruption of regular arrangement of atoms in a crystalline solid.” A POSITA would understand a “crack portion” to be an example of such a “crystal defect” because the crack itself interrupts the periodicity of the atomic lattice. That is, cracking necessarily causes interruptions of the regular arrangement of atoms in the crystalline solid because there is an absence of material within the crack where the atomic lattice would otherwise be expected. Moreover, the formation of a crack is known to stress and distort the atomic lattice at and near the crack tip, forming crystal defects at such “crack portions.” As I explained previously in section IX.B.2.g, because a POSITA would have understood that magnesium and fluorine would be present in a “crack portion” observable from a TEM, the same would be true for a “crystal defect.”

180. A POSITA would have also known that another such class of defects are point defects, which includes substitutional defects (such as intentional “dopants”). With respect to substitutional defects, one element takes the place of one (or more) of the native elements making up the crystal lattice. As the substitutional atom will not have the same atomic radius as the original atom, for example, this will result in local distortions (expansion or contraction) of the lattice. Indeed, Fey cites the established importance of doping LCO since “the structural characteristics of LiCoO_2 could be improved by doping with non-transition metal ions.” Ex. 1016 at 245. A POSITA would have further understood that substitutional defects generated via doping would, by definition, be considered crystal defects. Furthermore, a POSITA would have also understood that dopant elements are often distributed throughout a particle, both in the particle interior, as well as at the surface.

181. Consistently, Shim discloses a PEAM particle with a lattice structure that experiences a variety of “degradation mechanisms,” including where the “mechanically vulnerable charged form of the materials with the *expanded lattice* suffers from oxygen loss and side reactions with electrolytes at the highly oxidized surfaces.” See Ex. 1005, 1. A POSITA would have understood that the atomic lattice across the PEAM particle is susceptible to these “degradation mechanisms,” including portions of the atomic lattice in the vicinity of the crystal defects. Thus,

Shim additionally renders obvious the presence of fluorine within these crystal defect portions with these disclosures.

5. Dependent Claim 8

- a. [Preamble]: The lithium-ion secondary battery according to claim 5,

182. As I explained previously in section IX.B.4, in my opinion Shim (either alone or in combination with Lee) renders obvious the lithium-ion secondary battery according to claim 5.

- b. [Element 8.1]: wherein the first transition metal is cobalt,

183. As I explained previously in section IX.B.2.a, in my opinion, Shim discloses the use of lithium cobalt oxide where cobalt would satisfy the first transition metal as described by the '828 Patent.

- c. [Element 8.2]: wherein the second transition metal is titanium, and

184. As I explained previously in section IX.B.2.d, Shim discloses the use of magnesium with titanium and lithium cobalt oxide to create lithium titanate coated, magnesium-doped LCO, a material that contains cobalt, titanium, and magnesium. Here, the titanium would satisfy the second transition metal as described by the '828 Patent.

- d. [Element 8.3]: wherein the representative element is magnesium.

185. As I explained previously in section IX.B.2.e, Shim discloses the use of magnesium with titanium and lithium cobalt oxide to create lithium titanate-coated, magnesium-doped LCO, a material that contains cobalt, titanium and magnesium. Here, the magnesium would satisfy the representative element as described by the '828 Patent.

C. Ground 2: Claims 2 and 6 are Rendered Obvious by Shim (Either Alone or in Combination with Lee) in View of Yu

1. Motivation to Combine Shim and Yu

186. A POSITA would have been motivated to further combine the teachings of Yu with Shim (either alone or in combination with Lee). Both Yu and Shim highlight the advantage of using a protective surface coating rich in titanium on the surface of LCO PEAM particles. Specifically, Yu teaches adding titanium to the LCO particles and using XPS to measure a resulting concentration ratio of titanium to cobalt equaling 8.49% at the surface of the particle. Ex. 1007, 3, Table 3. It is therefore logical that a POSITA aiming to improve the cycling performance of lithium cobalt oxide would have recognized the conceptual similarities of Yu and Shim, as both describe the formation of a titanium-enriched exterior layer on LCO PEAMs to improve the cycling performance, as well as cell testing techniques using electrolytes consisting of LiPF_6 and carbonates. In my opinion, this combination would have involved routine implementation with no apparent technical risks, given the references' complementary teachings.

187. As an initial matter, Shim and Yu are directed at the same problem of improving the stability of PEAM particles to address battery degradation. Specifically, both references aim to remedy the issue of capacity fade during cycling through the formation of a metal oxide coating the PEAM particles. *See* Ex. 1005, 1 (“Coating is one of the simplest as well as most effective methods to be considered for minimization of the degradation and consequent capacity fade during cycling.”); Ex. 1007, 1 (“[The titanium] is more inclined to enrich in the particle surface”). A POSITA would have understood that these methods are similar and compatible because both Shim and Yu disclose approaches to create a titanium-rich surface layer on LCO PEAM particles to protect the particles during cycling. Ex. 1005, 2; Ex. 1007, 1.

188. Moreover, a POSITA would have recognized express motivations to combine Shim and Yu, who both disclose methods and performance assessments of LiCoO_2 PEAM particles containing an exterior titanium-enriched coating. These motivations for pursuing such a combination would have been obvious to a POSITA given the known degradation associated with hydrofluoric acid species in LiPF_6 -based electrolytes. A POSITA would have recognized that a titanium-enriched exterior layer can scavenge such acidic species and act as a physical protection layer for the LCO in the bulk of the PEAM particle. Furthermore, Shim and Yu both provide express teachings about dopant elements in LCO, which would have led a

POSITA to consider implementing teachings from Shim and Yu together. Dopants were well-known to impart additional structural stability to the LCO. Indeed, Yu states it was “[a]s well as known [that] partial doping of cobalt with metal ions leads to extended cyclability and increase in capacity of lithium ion cells by enhancing structural stability.” Ex. 1007, 1. To this end, Yu discloses an intent to use Ti as a dopant, while Shim elected to use Mg as a dopant. Additionally, a POSITA would have understood that, “because Mg has a valence state of 2^+ rather than Co (3^+) or Li (1^+),” using “ Ti^{4+} ” as a dopant could improve cell performance by improving the structural stability of the lattice. Ex. 1005, 2.

189. Finally, Shim discloses another express motivation to combine. Shim notes that the “[t]hickness of the coating layer is too thin (200 nm) to characterize its detailed structure by XRD.” Ex. 1005, 3. A POSITA would have understood that Yu teaches a method of characterizing this thin coating layer—Yu teaches using both energy dispersive X-ray spectroscopy (EDX) and XPS to measure Ti-to-Co ratios in the synthesized samples. Ex. 1007, 3. Importantly, the XPS depth profiling analysis in Yu provides the Ti-to-Co ratio at the surface and at progressive depths into the interior of the sample. *Id.* Since this type of surface analysis was not reported by Shim, it is reasonable that a POSITA attempting to improve Shim would consider Yu—specifically the results of the presented XPS analysis—to gain a better understanding of the composition and thickness of the surface coating on the lithium

cobalt oxide PEAMs. For instance, the surface measurement techniques of Yu could be used to better understand the extent of magnesium diffusion into the coating layer described by Shim. *See* Ex. 1005, 3.

190. In my opinion, the combination of Yu and Shim would simply require using a known technique to improve similar devices and methods in the same way, which I understand is a way of showing that a POSITA would have been motivated to combine these references. That is, a POSITA could apply Yu's known technique of XPS characterization to Shim's PEAM particles (which are a known device ready for improvement) in view of a POSITA's understanding of inevitable cracks, to yield predictable results. Furthermore, as disclosed in Yu, a POSITA would have understood that adding some amount of titanium as a dopant would improve the electrochemical performance (*i.e.*, cyclability) of the LiCoO_2 . However, since titanium dopant atoms are inactive under a typical operating voltage window of an LiCoO_2 -based cell (in other words, substituting titanium for cobalt theoretically results in a small loss of capacity), a POSITA would also have been motivated to use as little titanium as necessary to stabilize the structure. With these competing factors in mind, a POSITA would have been motivated to determine an optimum range of titanium concentration (and by extension, Ti:Co concentration) through routine experimentation.

2. Dependent Claim 2

- a. [Preamble]: The lithium-ion secondary battery according to claim 1,

191. As I explained previously in section IX.B.2.a, in my opinion Shim (either alone or in combination with Lee) renders obvious the lithium-ion secondary battery according to claim 1.

- b. [Element 2.1]: wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

192. In my opinion, Shim and Yu render this limitation obvious. As I explained previously in sections IX.B.3.b–IX.B.3.c, the first transition can be cobalt and the second transition metal can be titanium.

193. Yu teaches measuring the relative concentrations of cobalt and titanium using “X-ray photoelectron spectroscopy (XPS)” and “Ar ion etching.” Ex. 1007, 1, 2 (“Fig. 2 displays the Ti2p and Co2p [XPS] spectrum of [lithium cobalt titanium oxide]. Ar ion etching was used to examine the concentration depth profiles of the [lithium cobalt titanium oxide] powder.”). A POSITA would have known that Ar ion etching is a technique used in conjunction with XPS analysis to determine the depth-dependent elemental concentration profile of a sample. Specifically, as the argon ions are “sputtered” (or etched) into the sample, subsurface regions are uncovered, which allows the incident X-ray beam to interact with new regions of the

sample. By alternating Ar etching and XPS scanning, the elemental concentration at various depths of the sample can be measured. A sputtering time of zero corresponds to the surface of the sample, and as the sputtering time increases, the analysis depth likewise increases.

194. Yu teaches a decreasing concentration gradient of titanium through the PEAM particle. After 4000 seconds of sputtering, Yu notes that “[t]he intensity for Ti becomes lower gradually” and eventually “hardly visible.” *Id.* By contrast, “the intensity for Co becomes obviously higher with the increasing sputtering time.” *Id.* Yu discloses measured Ti/Co concentrations of a PEAM at different sputtering times in Table 3:

Table 3 The Ti/Co atomic ratio of $\text{LiCo}_{0.998}\text{Ti}_{0.002}\text{O}_2$ powders with different sputtering times.					
Sputtering time (s)	0	500	1000	2500	4000
Ti/Co (%)	8.49	3.02	2.17	1.25	0.75

Ex. 1007, 3, Table 3 (annotated). The spectral data underlying the results in Table 3 are presented in Figure 2. *Id.*, 2, Fig. 2.

195. As I annotated in the red box above, at 0 seconds of sputtering time—the surface of the sample—the ratio of titanium to cobalt is 8.49%, or 0.0849. Ex. 1007, 3, Table 3. At this surface point, when the concentration of cobalt (the first transition metal) is defined as 1, the concentration of titanium (the second transition

metal) is 0.0849, and therefore falls in the range of 0.05 to 0.4, as required by the claim. Thus, in my opinion, Yu discloses this element.

196. Table 3 also shows the highest Ti-to-Co ratio at a sputtering time of 0, with this concentration steadily decreasing at greater depths from the particle surface. From Table 3, a POSITA would understand that when a PEAM is treated with titanium in the method described by Yu, the concentration of titanium will be the highest at the surface of the particle. Indeed, Yu confirms that “the added Ti is more likely to be enriched in the surface layer.” Ex. 1007, 2. In the context of the distribution of titanium in the particle, Yu’s effect of Ti enrichment at the surface is similar to Shim’s titanium coating layer. Thus, POSITA would have understood that PEAM particles prepared as described by Yu, containing a titanium coating layer as taught by Shim, or a combination of the strategies would yield a Ti-to-Co concentration measurement at the surface within the range of 0.05 to 0.4 when measured by XPS. Thus, in my opinion, a combination of Shim and Yu would also render this element obvious.

197. As I explained previously in section IX.C.1, a POSITA would have been motivated to combine the teachings of Yu with Shim. Specifically, a POSITA would have understood that Shim and Yu address the same problem of structural degradation within the same lithium cobalt oxide battery cathode, and that using titanium as a dopant would have increased the battery’s performance.

3. Dependent Claim 6

- a. [Preamble]: The lithium-ion secondary battery according to claim 5,

198. As I explained previously in section IX.B.2.a, Shim (either alone or in combination with Lee) renders obvious the lithium-ion secondary battery according to claim 5.

- b. [Element 6.1]: wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

199. As I explained previously in section IX.C.3.b, Shim (either alone or in combination with Lee) in combination with Yu would have rendered this element obvious to a POSITA.

D. Ground 3: Claims 7 are Rendered Obvious by Shim in Combination with Lee

1. Dependent Claim 3

- a. [Preamble]: The lithium-ion secondary battery according to claim 1

200. As I explained previously in section IX.B.2, in my opinion Shim (either alone or in combination with Lee) renders obvious the lithium-ion secondary battery according to claim 5.

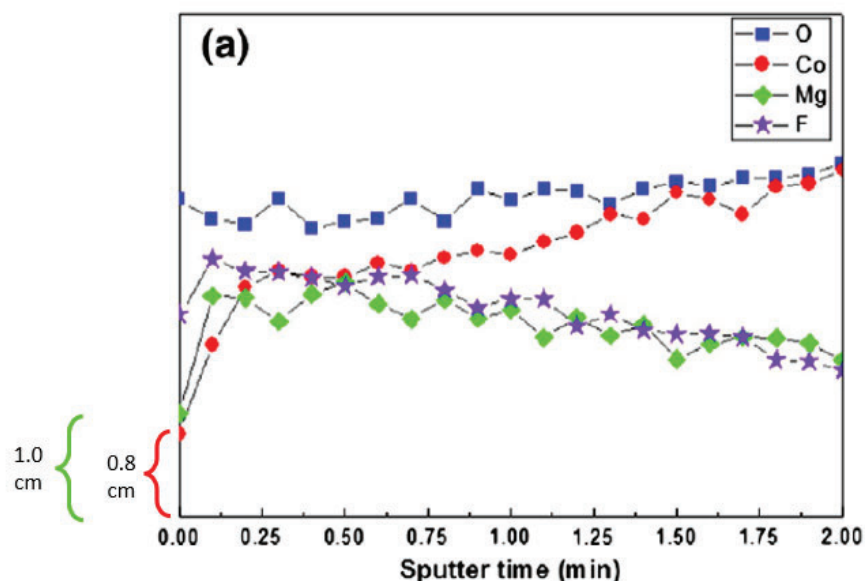
- b. [Element 3.1]: wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the

positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

201. In my opinion, Shim and Lee render this limitation obvious. As I explained previously in sections IX.B.2.c–e, the first transition metal can be cobalt and the representative element can be magnesium. Lee teaches a lithium cobalt oxide material coated with MgF_2 and a method of measuring the “depth-concentration profiles of the elements (Co, O, Mg, and F)” using “XPS, which is one of the efficient surface analysis tools.” Ex. 1006 at 86–87, 90.

202. A POSITA would have understood that XPS was a well-known method of surface concentration analysis. As I explained previously in section VI.A.4 and IX.C.2.b, in an XPS-based depth analysis, a sample is sputtered, or etched (e.g., by argon ions) while XPS measurements are conducted intermittently. As the sample is etched, subsurface regions of the sample are revealed, enabling the composition to be measured at various depths. That is, a longer etching time correlates with a measurement deeper into the sample, while an etching time of 0 corresponds to a measurement at the surface of the sample.

203. Figure 5(a) in Lee depicts the result XPS analysis of its MgF_2 -coated lithium cobalt oxide material:



Ex. 1007, Fig. 5(a) (y-axis annotations added).

204. To determine the relative concentrations of Mg and Co in the particle, a POSITA could have measured the relative magnitudes of the data in the plot. In doing so, a POSITA would have found that at a sputtering time of 0 minutes, the Co concentration measured 0.8 centimeters on the Y-axis, while the Mg concentration measured 1.0 cm on the Y-axis. Assuming that the Y-axis begins at 0, this would reflect a relative concentration of magnesium to cobalt (Mg/Co) of 1.2. A POSITA would also understand that if the Y-axis began at a value higher than 0, the Mg/Co ratio would decrease or converge to a minimum ratio of 1. For example, if the Y-axis of Fig. 5(a) began at a value corresponding to 1.0 centimeters, the Mg/Co ratio would be $(1.0+1.0)/(1.0+0.8) = (2.0/1.8) = 1.11$. The higher the value that the Y-axis begins at, the closer the Mg/Co ratio would be to 1.0.

205. Further, as shown above, within 0.25 min, or 15 seconds of sputtering time (*i.e.*, close to the surface of the sample), the relative concentrations of cobalt and magnesium are approximately equal. The concentrations are equal when the traces for Co and Mg intersect. When the concentrations are equal, the relative concentration of magnesium to cobalt is 1.0. Thus, Lee discloses a relative concentration of magnesium to cobalt at the surface of the PEAM within the claimed range of 0.4 to 1.5.

206. For the reasons I described previously in section IX.B.2, a POSITA would have been motivated to combine the teachings of Lee with Shim. Further, a POSITA would have understood that Lee's method of more precisely measuring the composition at the surface layer could be applied to Shim. *See* Ex. 1005, 3 (noting that the "[t]hickness of the coating layer is too thin (200 nm) to characterize its detailed structure by XRD"). Additionally, a POSITA would have understood that adding magnesium as a dopant improves the electrochemical performance (*i.e.*, electrical conductivity) of the LiCoO_2 . However, since magnesium dopant atoms are inert within a typical operating voltage window of an LiCoO_2 -based cell (in other words, substituting magnesium for cobalt theoretically results in a small loss of capacity), a POSITA would also have been motivated to use as little magnesium as necessary to stabilize the structure. With these competing factors in mind, a POSITA would have been motivated to determine an optimum range of magnesium

concentration (and by extension, Mg:Co concentration) through routine experimentation.

2. Dependent Claim 7

- a. [Preamble]: The lithium-ion secondary battery according to claim 5,

207. As I explained previously in section IX.B.4, in my opinion, Shim (either alone or in combination with Lee) renders obvious the lithium-ion secondary battery according to claim 5.

- b. [Element 7.1]: wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

208. As I explained previously in section IX.D.1.b, Shim and Lee render this limitation obvious.

E. Ground 4: Claims 1, 4, 5, and 8 Are Rendered Obvious by Fujiki

1. Independent Claim 1

- a. [Preamble]: A lithium-ion secondary battery comprising a positive electrode active material layer comprising:

209. In my opinion, Fujiki discloses this element. A POSITA would have understood that Fujiki describes strategies for improving the positive electrode active material in lithium-ion secondary batteries. Fujiki discusses a “positive electrode and a nonaqueous electrolyte secondary cell”—*i.e.*, a lithium-ion

secondary battery. Ex. 1008, 1:14–17. Fujiki notes that “secondary cells and batteries as driving power sources for” electronic apparatuses “have necessarily been desired to show an enhanced energy density” and “a prolonged cycle life.” *Id.*, 1:32–34. To meet these needs, Fujiki describes a “positive electrode active material” with “a high capacity and excellent charge-discharge cycle characteristics,” with “little” “deterioration during use in high-temperature environments.” *Id.*, 2:62–67.

210. In particular, Fujiki discloses a “positive electrode active material,” where a “lithium-containing transition metal oxide” forms the active material layer. Ex. 1008, Abstract, 3:1–9, 3:10–17, 3:18–30. Fujiki further discloses fabricating a positive electrode by admixing lithium cobaltate particles with magnesium carbonate in a way that deposits magnesium carbonate onto the surfaces of the lithium cobaltate particles. *Id.*, 11:53–64. Fujiki teaches that the PEAM may be prepared by “various dry-type, wet-type or other known methods” in order for metallic element M1 (generally magnesium) to be present at the surfaces of the compound oxide particles. *Id.*, 10:27–32. The resulting particles are then admixed with a different metallic element, M2, which may be a metallic salt like lithium phosphate or titanium oxide. *Id.*, 11:65–12:8, 18:61–19:3. The resulting fabricated “lithium-transition metal compound oxide” is used by Fujiki “as a positive electrode active material” in a secondary battery cell. *Id.*, 12:29–34, 13:52–34:13.

b. [Element 1.1]: a positive electrode active material particle comprising

211. In my opinion, Fujiki discloses a PEAM particle, because it would have been obvious to a POSITA that positive electrode active materials are microparticles. Additionally, Fujiki addresses a known problem in lithium-ion batteries where “compound oxide particles” (*i.e.*, lithium-containing transition metal oxide particles, or PEAM particles) deteriorate with use. Ex. 1008, 3:64–4:47. Fujiki discloses combating this deterioration by modifying the surfaces of these “compound oxide particles,” *i.e.*, PEAM particles, to improve the cycling stability of the battery. *Id.*, 4:35–47.

212. Fujiki discloses treating PEAM particles with various metallic elements to improve battery capacity after charge and discharge cycles. *Id.*, 13:54–34:13. In particular, Fujiki discloses admixing the PEAM particles with a metallic element M1 such that as a result, M1 is “present at the surfaces of the compound oxide particles.” *Id.*, 5:10-12, 10:27–32. For example, Fujiki discloses admixing the PEAM particles with magnesium carbonate such that magnesium carbonate is present on the surface of the PEAM (*i.e.*, lithium-containing transition metal oxide) particles. *Id.* Fujiki discloses numerous exemplary embodiments of a lithium-ion battery, each of which includes a PEAM particle. *See generally id.*, 13:54–30:17; Table 1, Table 2.

c. [Element 1.2]: a first transition metal,

213. In my opinion, Fujiki discloses this element. Fujiki discloses a PEAM particle comprising cobalt as a first transition metal. Fujiki discloses active material particles that include “a lithium-containing transition metal oxide containing the principal transition metal element A.” Ex. 1008, Abstract, 3:1–9, 3:10–17, 3:18–30, 34:30–34. Fujiki’s active material particles are “particles of a lithium compound oxide which has a laminar rock salt structure and in which the principal transition metal element A is at least one selected from the group consisting of at least Ni, Co and Mn.” *Id.*, 4:57–67. Nickel, cobalt, and manganese are all transition metals, as I show below in groups 3–12 of the periodic table:

Group →	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
Period ↓	1 1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	* 71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	* 103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og
			* 57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb		
			* 89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No		

Fujiki explains these transition metals are “preferable, since the lithium compound oxide particles make it possible to obtain a high capacity.” Ex. 1008, 4:57–67. Fujiki also discloses an active material particle made of lithium cobaltate (LiCoO_2) which

contains cobalt, a transition metal. *Id.*, 11:56–64, 24:14–20, 29:56–62, 30:10–17.

The fabricated lithium-transition metal compound oxide is then used “as a positive electrode active material” in a secondary cell. *Id.*, 12:29–34. Thus, Fujiki discloses a PEAM particle comprising a first transition metal.

d. [Element 1.3]: a second transition metal,

214. In my opinion, Fujiki discloses this element. Fujiki discloses a PEAM particle comprising titanium, a second transition metal. In particular, Fujiki discloses a second transition metal within “a compound of metallic element M2.” Ex. 1008, Abstract, 3:1–9, 3:10–17, 3:18–30, 4:49–56. Fujiki discloses that metallic element M2 is preferably at least one element selected from a group including magnesium (Mg), titanium (Ti), and aluminum (Al); titanium is a transition metal. *Id.*, 5:29–42. In this way, Fujiki discloses a second transition metal M2 (preferably Mg, Ti, or Al) different from the first transition metal A (preferably Co, Ni, or Mn).

215. Fujiki further discloses that metallic element M2 “stabiliz[es] the crystal structure of the compound oxide particles at the time of charging.” Ex. 1008, 5:29–42. Fujiki discloses preferably using the metallic element M2 as a coating on the surface on the active material particles, in the form of a metallic salt, a metallic oxide (such as titanium oxide), or a metallic halide (such as titanium fluoride). Ex. 1008, 5:29–42, 5:46–61, 5:62–6:2, 34:1–7.

216. Fujiki illustrates, in an Example 14, the use of titanium oxide (TiO_2), a transition metal compound, as M2 in the PEAM. Ex. 1008, 18:61–19:22. Fujiki further teaches observing the particles treated with titanium oxide with “a scanning electron microscope (SEM) equipped with an energy dispersive X-ray analyzer (EDX)” to confirm that “titanium oxide was present in the state of being deposited on the surfaces of the particles of the lithium-transition metal compound oxide or present between the particles.” *Id.*, 12:9–18, 19:4–11. Fujiki’s use of titanium oxide as M2 in secondary cells resulted in a battery that retained 80% of its initial capacity after 300 cycles. *See id.*, Table 1 (Example 14).

e. [Element 1.4]: a representative element, and

217. In my opinion, Fujiki discloses this element. Fujiki discloses 30 separate examples of coated LCO PEAMs containing magnesium, where magnesium would satisfy the representative element as described by the ‘828 Patent. The ‘828 Patent, for example, states that representative elements are electrochemically stable, and lists magnesium, calcium, and sodium as examples of representative elements. *Id.*, 10:29–33, 11:65–12:4. Accordingly, the presented PEAMs, such as Example 14 which consists of LCO with magnesium (M1) and a TiO_2 (M2) coating, satisfy the representative element as described by the ‘828 Patent.

218. As discussed above, Fujiki discloses admixing “metallic element M1,” the representative element, with lithium compound oxide particles so that M1 is “present at the surfaces of the compound oxide particles” after fabrication. Ex. 1008, 10:26-50, 11:65–12:8. Fujiki describes M1 to preferably be selected from the elements Mn, Fe, Mg, Al, Ni, and Co. *Id.*, 5:6–19. The use of M1 “suppress[es] an oxidation activity, particularly at the time of charging, at the surfaces of the compound oxide particles.” *Id.*, 5:20–28.

219. Fujiki further teaches fabricating a positive electrode by admixing lithium cobaltate with magnesium carbonate. *Id.*, 10:27–50, 11:56–64. Fujiki discloses heating and slowly cooling the admixture (of lithium cobaltate with magnesium carbonate) to obtain active material particles “in which magnesium (Mg) was uniformly distributed on surfaces of lithium cobaltate particles.” *Id.*, 11:65–12:8. SEM/EDX may be used to observe the lithium cobaltate particles and confirm that “magnesium was uniformly distributed over the whole surfaces of the particles of the lithium-transition metal compound oxide.” *Id.*, 12:9–28. Fujiki further discloses that PEAM particles fabricated without the addition of M1 (magnesium) have worse capacity retention after 300 charging cycles, as I annotate in the table below.

TABLE 2

Positive electrode active material								Capacity	
A	M1	M1/(M1 + A)	M2 compound	Average particle diameter [μm]	M2/(M2 + A)	Charging voltage [V]	Initial capacity [Wh]	retention after 300 cycles [%]	
Example 26	Ni, Co, Mn	Mg	0.01	Li ₃ PO ₄	0.8	0.02	4.35	8.2	85
Example 27	Co	Mg	0.01	LiCl	1.1	0.02	4.35	8.9	76
Example 28	Co	Mg	0.01	AlCl ₃	1.2	0.02	4.35	8.8	77
Example 29	Co	Mg	0.01	ZrCl ₄	1.1	0.02	4.35	9.0	74
Example 30	Co	Mg	0.01	Li ₃ PO ₄	0.8	0.02	4.20	8.0	85
Example 31	Co	Mg	0.01	Li ₃ PO ₄	0.8	0.02	4.25	8.4	84
Example 32	Co	Mg	0.01	Li ₃ PO ₄	0.8	0.02	4.40	9.4	80
Example 33	Co	Mg	0.01	Li ₃ PO ₄	0.8	0.02	4.50	10.0	68
Comp. Ex. 1	Co	—	—	Li ₃ PO ₄	0.8	0.02	4.35	9.1	31
Comp. Ex. 2	Co	Mg	0.23	Li ₃ PO ₄	0.8	0.02	4.35	6.3	71
Comp. Ex. 3	Co	Mg	0.01	—	—	—	4.35	9.0	50
Comp. Ex. 4	Co	Mg	0.01	Li ₃ PO ₄	0.8	0.3	4.35	7.8	61
Comp. Ex. 5	Co	Mg	0.01	Li ₃ PO ₄	35	0.02	4.35	9.2	56
Comp. Ex. 6	Co	—	—	ZrO ₂	0.7	0.015	4.35	9.0	48
Comp. Ex. 7	Co	Mg	0.01	ZrO ₂	0.7	0.2	4.35	7.5	55
Comp. Ex. 8	Co	Mg	0.01	ZrO ₂	28	0.015	4.35	9.0	38
Comp. Ex. 9	Co	Mg	0.01	LiF	1.0	0.2	4.35	7.7	52
Comp. Ex. 10	Co	Mg	0.01	LiF	45	0.02	4.35	8.5	41
Comp. Ex. 11	Ni, Co, Mn	—	—	Li ₃ PO ₄	0.8	0.02	4.35	8.2	43
Comp. Ex. 12	Ni, Co, Mn	Mg	0.01	—	—	—	4.35	8.0	53
Comp. Ex. 13	Co	—	—	Li ₃ PO ₄	0.8	0.02	4.20	8.0	52
Comp. Ex. 14	Co	—	—	Li ₃ PO ₄	0.8	0.02	4.25	8.3	40
Comp. Ex. 15	Co	—	—	Li ₃ PO ₄	0.8	0.02	4.40	9.2	21
Comp. Ex. 16	Co	—	—	Li ₃ PO ₄	0.8	0.02	4.50	9.9	4

Id., Table 2 (Comparative Examples 1, 6, 11, 13–16) (annotated).

f. [Element 1.5]: fluorine,

220. In my opinion, Fujiki discloses this element. Fujiki discloses a PEAM particle comprising fluorine, including by noting that metallic element M2 may take the form of a metallic halide or metallic fluoride. Ex. 1008, 6:3–11 (emphasis added) (noting that when M2 is a metallic halide, it is particularly preferable that M2 is a metallic “*fluoride*”). A POSITA would have understood metallic halides include various types of fluorides which contain the element fluorine.

221. Fujiki also discloses the use of LiPF_6 -based electrolytes, which (as I explain above in section VI.A.2) are known to decompose during charge and discharge, including the formation process that takes place during battery manufacturing, to form fluorinated species as part of the CEI of the positive electrode active material. Accordingly, a POSITA would have understood that fluorine would be present on the positive electrode active material surface in Fujiki's positive electrode active material particle.

222. Moreover, Fujiki discloses an extensive list of various coating layers paired with Mg-containing LiCoO_2 , which includes fluorinated coatings such as aluminum fluoride (AlF_3), yttrium fluoride (YF_3), and lithium fluoride (LiF). *Id.*, 21:36–44, 21:66–22:6, 23:21–29. Specifically, Fujiki's Example 22 discloses a PEAM comprising a first transition metal cobalt (*i.e.*, transition metal element A), a representative element magnesium (*i.e.*, metallic element M1), and a second transition metal, yttrium fluoride (*i.e.*, compound of metallic element M2). *Id.*, 22:28–53. The resulting fabricated particles have yttrium fluoride “deposited on the surfaces of the particles of the lithium-transition metal compound oxide.” *Id.*, 22:39–43. Thus, Fujiki discloses a PEAM particle with fluorine present.

223. Fujiki further discloses that using a metallic fluoride as the compound of metallic element M2 significantly improves the capacity of a battery over several cycles. Ex. 1008, Table 1. In Fujiki's experiments, active material particles that used

yttrium fluoride retained 79% of the original capacity after 300 cycles (Example 22), whereas active material particles without such a compound retained only 50% of the original capacity after 300 cycles (Comparative Example 3).

TABLE 1

	Positive electrode active material						Capacity		
	A	M1	M1/(M1 + A)	M2 compound	Average particle diameter [μm]	M2/(M2 + A)	Charging voltage [V]	Initial capacity [Wh]	retention after 300 cycles [%]
Example 22	Co	Mg	0.01	YF ₃	1.1	0.02	4.35	8.9	79

TABLE 2

	Positive electrode active material						Capacity		
	A	M1	M1/(M1 + A)	M2 compound	Average particle diameter [μm]	M2/(M2 + A)	Charging voltage [V]	Initial capacity [Wh]	retention after 300 cycles [%]
Comp. Ex. 3	Co	Mg	0.01	—	—	—	4.35	9.0	50

Id., Tables 1 and 2 (excerpted and annotated). Fujiki also analyzes other particles fabricated with metallic fluorides, thereby further demonstrating the presence of fluorine in the PEAM, as well as the improvement in capacity in the particles from the addition of a metallic fluoride. *Id.*, 21:36–62, 22:57–23:17, 23:51–24:10, Table 1 (Examples 20–25).

- g. [Element 1.6]: wherein the representative element and the fluorine are present in a crack portion observed from a TEM image in the positive electrode active material particle.

224. In my opinion, Fujiki renders this element obvious. As I explained above in section IX.B.2.g (with respect to Shim), a POSITA would have known that coated surfaces will inevitably have defects, including cracks, and would have further understood that defects or cracks in the coating would contain particles of the coating material that had become ingrained into the cracks. *See* Ex. 1016, 4 (“It is inevitable that coated surfaces always have defects such as cracks and pinholes in them” and that these defects “may form and close with the application of a load or upon thermal cycling.”); *id.* (“In the case of the coated cathodes . . . the cycling process could have enabled the particles of the coating material to become ingrained in the crevices and cracks on the cathode surface.”).

225. Furthermore, a POSITA would have known that the LCO positive electrode active material is intrinsically susceptible to microcracking and the formation of crystal defects due to cycling. In light of both Fujiki, which previously disclosed the role of the representative element, and the background knowledge of a POSITA, who would have recognized the ubiquity of fluorine on a positive electrode active material surface—either from an externally applied metal fluoride coating, a fluorinated surface layer on the PEAM particles due to decomposition of the LiPF_6 salt, or the formation of surface metal fluorides due to the reaction of metal oxides with HF in the electrolyte—a POSITA would have concluded that the representative

element and fluorine would therefore be present in a crack or other crystal defect of a positive electrode active material particle.

i. *Crack portion observed from a TEM image in the positive electrode active material particle*

226. In my opinion, Fujiki renders obvious a crack portion observed from a TEM image. Fujiki discussed reducing the “deterioration of a positive electrode active material” as a means of improving performance of lithium-ion batteries, in particular by attaining “a high cell capacity and excellent charge-discharge cycle characteristics.” Ex. 1008, 3:34–39, 3:64–4:7 (describing “deterioration of the active material” in LiCoO₂-based batteries), 4:28–34 (noting that raising the charging voltage accelerates “deterioration of the compound oxide [*i.e.*, LiCoO₂] particles used as the active material”), 5:32–35 (describing “stabilizing the crystal structure of the compound oxide particles [*i.e.*, LiCoO₂] at the time of charging”). In my view, a POSITA would have understood that Fujiki’s references to deterioration of the LiCoO₂ active material includes the generation of cracks or other defects in the active material. Further, a POSITA would have understood that Fujiki’s discussion of the need to “stabiliz[e] the crystal structure” of LiCoO₂ also includes cracking of the active material (*i.e.*, PEAM particle).

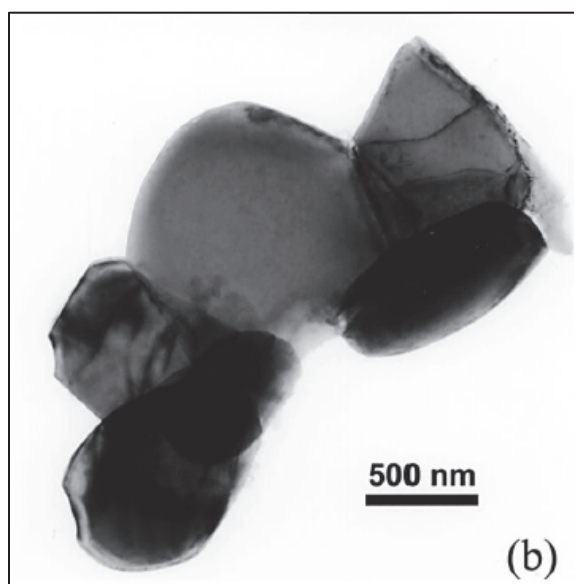
227. In my opinion, Fujiki also renders obvious observing a crack portion from a TEM image. Fujiki describes several methods or technologies for observing fabricated active material particles. Fujiki describes producing a powder by

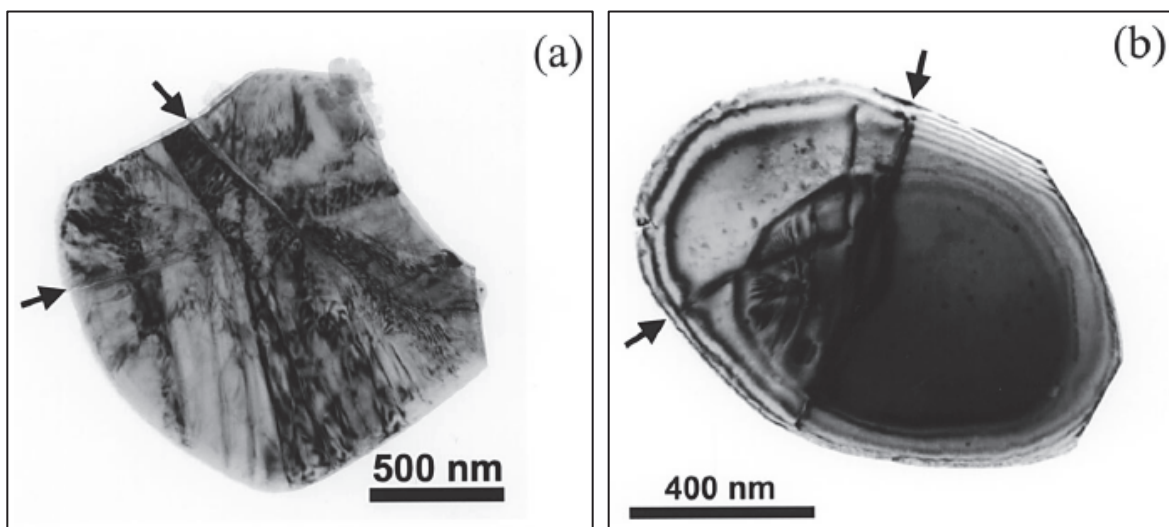
admixing PEAM particles (comprising lithium, principal transition element A, and PEAM M1) with a compound of metallic element M2, and observing the powder by using “a scanning electron microscope (SEM) equipped with an energy dispersive X-ray analyzer (EDX).” Ex. 1008, 12:9–11. A POSITA would have understood that both TEM and SEM images could have been used to observe a crack portion in an active material particle. A POSITA would therefore have found it obvious to use either SEM or TEM to make such observations of cracks occurring on the surface of active material particles. Ex. 1010, 3–4; Ex. 1026, 2.

228. To the extent that the Board finds the crack portion is not expressly disclosed by Fujiki, it is my opinion that the crack portion would have been obvious to a POSITA in light of his or her knowledge. Before the priority date of the '828 Patent, it was well-known in the art that cathodes such as Fujiki's would inevitably have crack portions that could be observed with TEM imaging. For example, over a decade before the '828 Patent, it was known that “[i]t is inevitable that coated surfaces always have defects such as cracks and pinholes in them,” which “may form and close with the application of a load or upon thermal cycling.” Ex. 1016, 4. As another example, at least 17 years before the '828 Patent, it was well-known that “[e]lectrochemical cycling creates a variety of defects in the LiCoO_2 particles in a *typical composite cathode*.” Ex. 1010, 8. A POSITA would have been aware of the inevitability of these defects, or cracks, in cathodes. Fujiki also recognized this well-

known general concept in its discussion of “deterioration of the active material” in lithium cobalt oxide cathodes. Ex. 1008, 3:64–4:7. Thus, in my opinion, a POSITA would have understood Fujiki’s discussion of deterioration of the active material to include inevitable cracks within PEAM particles.

229. Furthermore, to the extent that the Board finds that Fujiki does not expressly disclose observing the cracks in Fujiki’s PEAM particles using TEM imaging, it is my opinion that this would have been obvious to a POSITA in light of his or her knowledge. As an example, and as shown by Fey, it was well-known to use TEM to “examin[e] . . . the microstructure of the coated particles,” which would necessarily include observing any cracks in the particles by TEM imaging. Ex. 1016, 1, Fig. 2. As another example of a POSITA’s knowledge, Wang describes observing via TEM imaging “strain contours, dislocations and other extended defects, and microcracks” in LiCoO_2 particles (Ex. 1010, 1, 3):

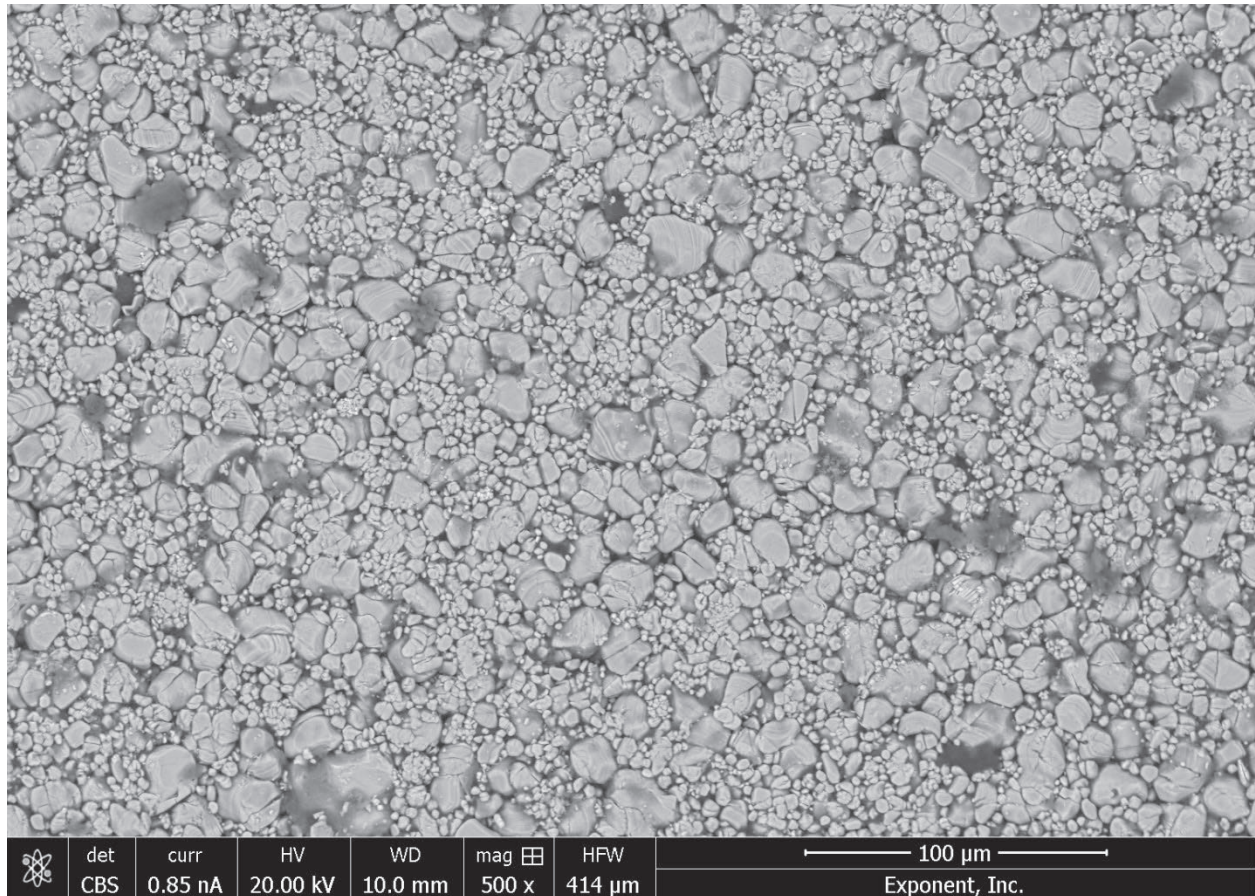




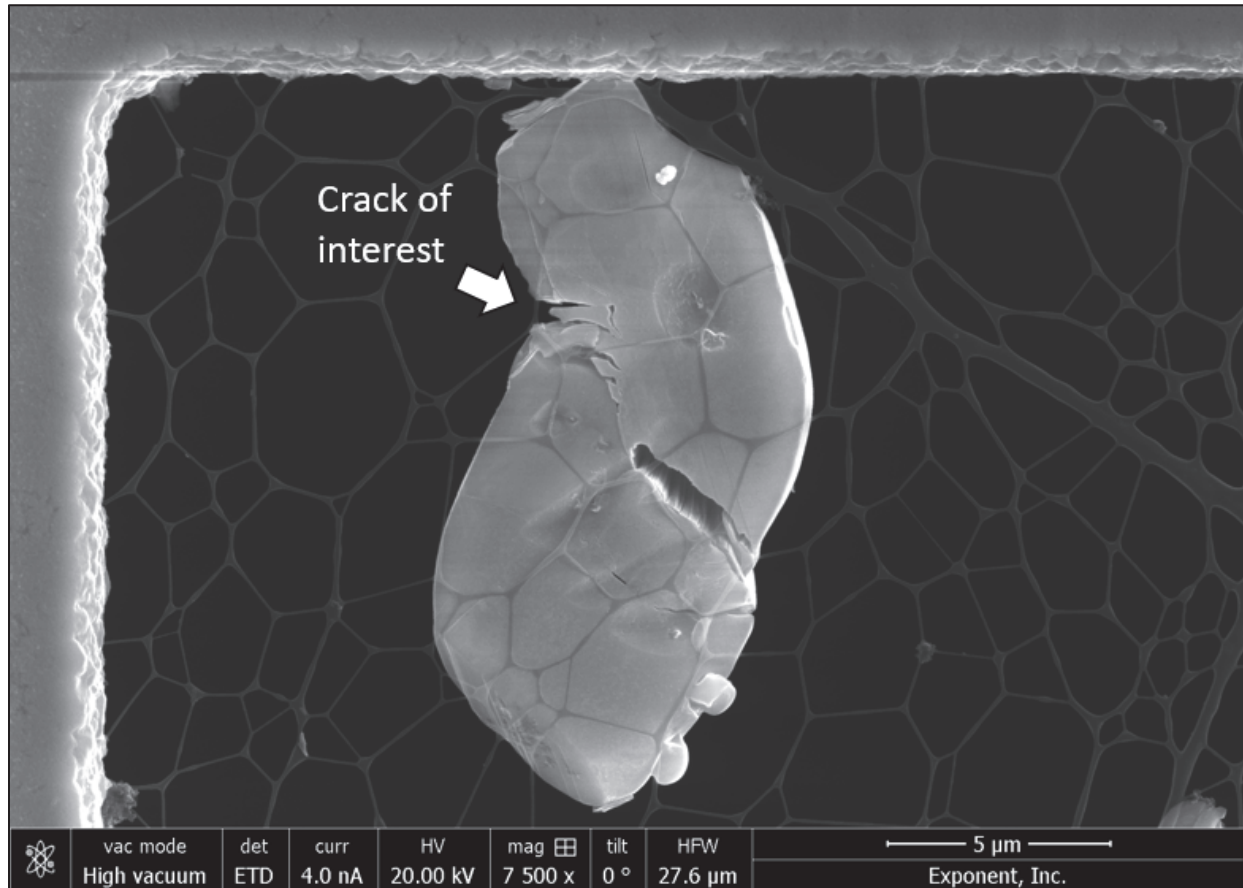
Id., Fig. 3(b) (“TEM bright-field image”); 5(a), 5(b) (“LiCoO₂ particles from a cycled cathode showing microfracture (arrows)”); *see also* Ex. 1026, 2 (pointing out “fragment[s]” or “crack[s]” in cycled positive electrode grains shown in TEM imaging).

230. Additionally, testing data conducted at my direction confirms, in my opinion, that a POSITA would have known Shim’s PEAM contains crack portions observable from a TEM image, as I explain above in section VI.B. Specifically,

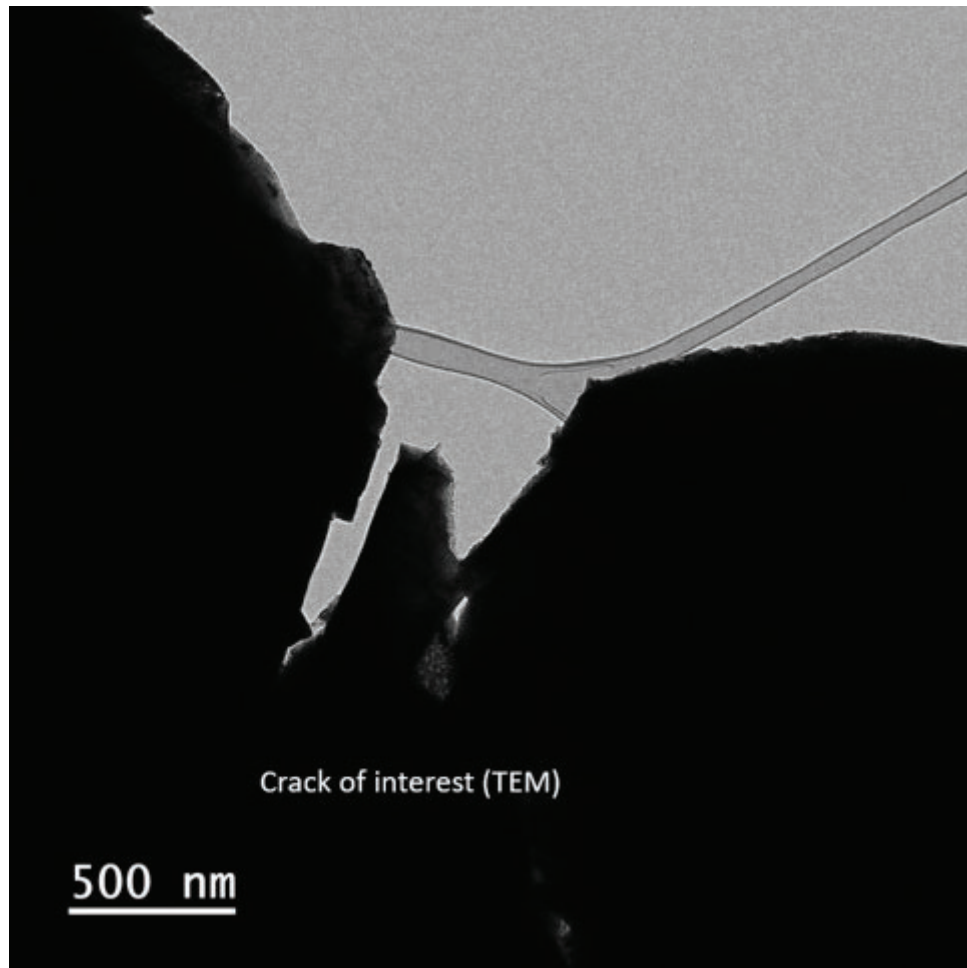
testing of an Samsung Galaxy A8 battery sample (from August 7, 2015) revealed widespread cracks visible through SEM, as shown below:

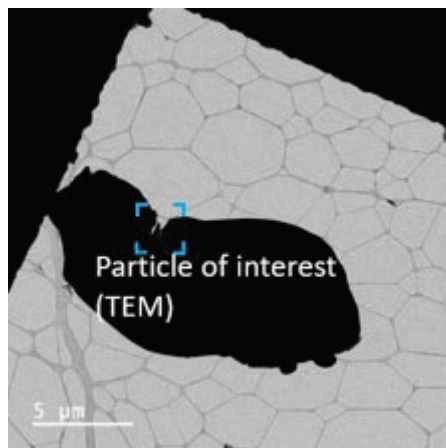


231. Consistently, further SEM analysis revealed crack portions in these particles, as shown below:



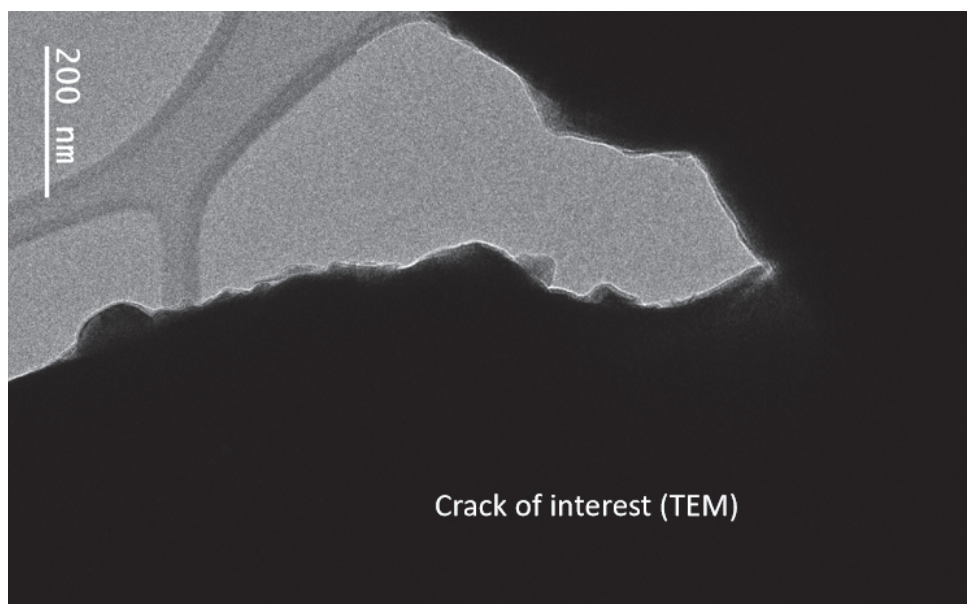
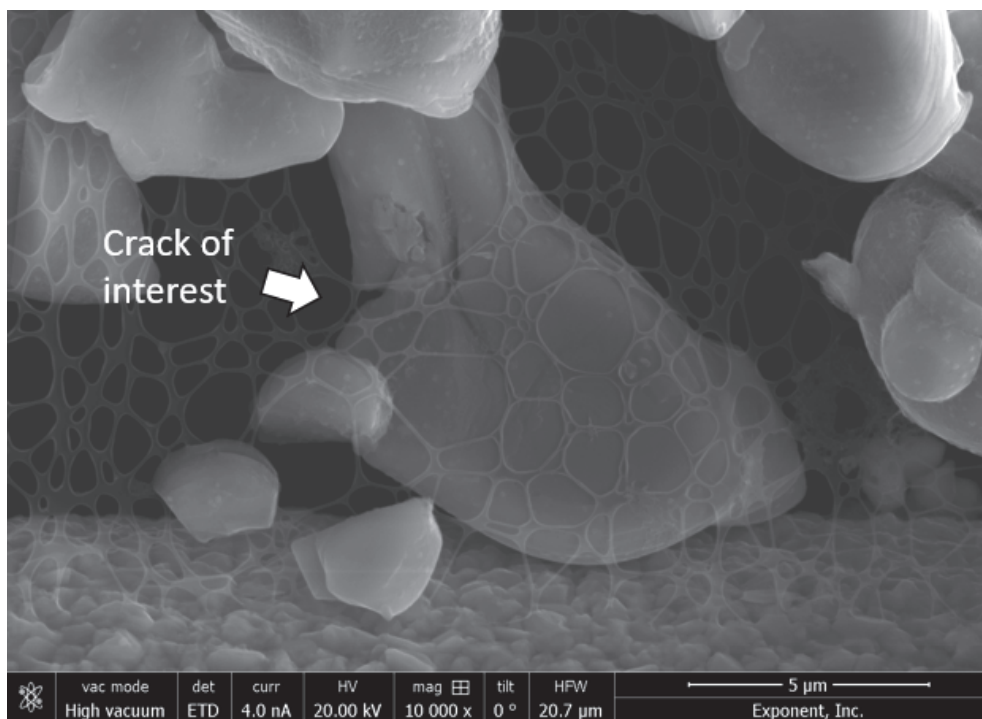
232. TEM imaging showed that the “crack portion” on the same particle of interest was indeed visible, thereby confirming that a POSITA would have found obvious a “crack portion” observable by TEM:



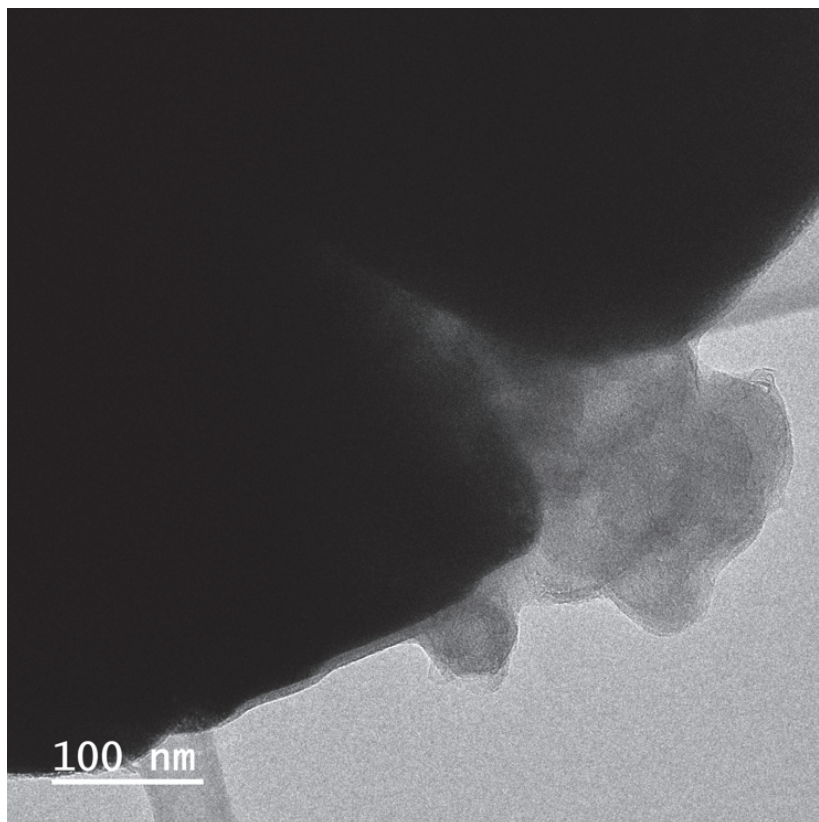


233. In these bright field TEM images, the dark colored portions correspond to solid regions of the sample which are sufficiently thick such that the incident electron beam is unable to reach the detector. Conversely, the lightest colored regions represent areas where the electron beam is not blocked by solid material (*i.e.*, is absent of PEAM). Thus, the light colored gap in the TEM image corresponds to a “crack portion” in the PEAM particle.

234. Additional samples tested (at my direction) offer similar results. For instance, the below two images from a Samsung Galaxy Note 4 battery sample (dated April 1, 2016) show SEM (top) and TEM (bottom) images revealing crack portions in a PEAM particle.



235. As a further example, the below TEM image shows a crack portion in a PEAM particle from an iPhone 5S battery sample dated October 2013.



236. For the above reasons, and with reference to the testing I described above in section VI.B, in my opinion Shim renders obvious a crack portion observed from a TEM image.

ii. *Representative element present in a crack portion*

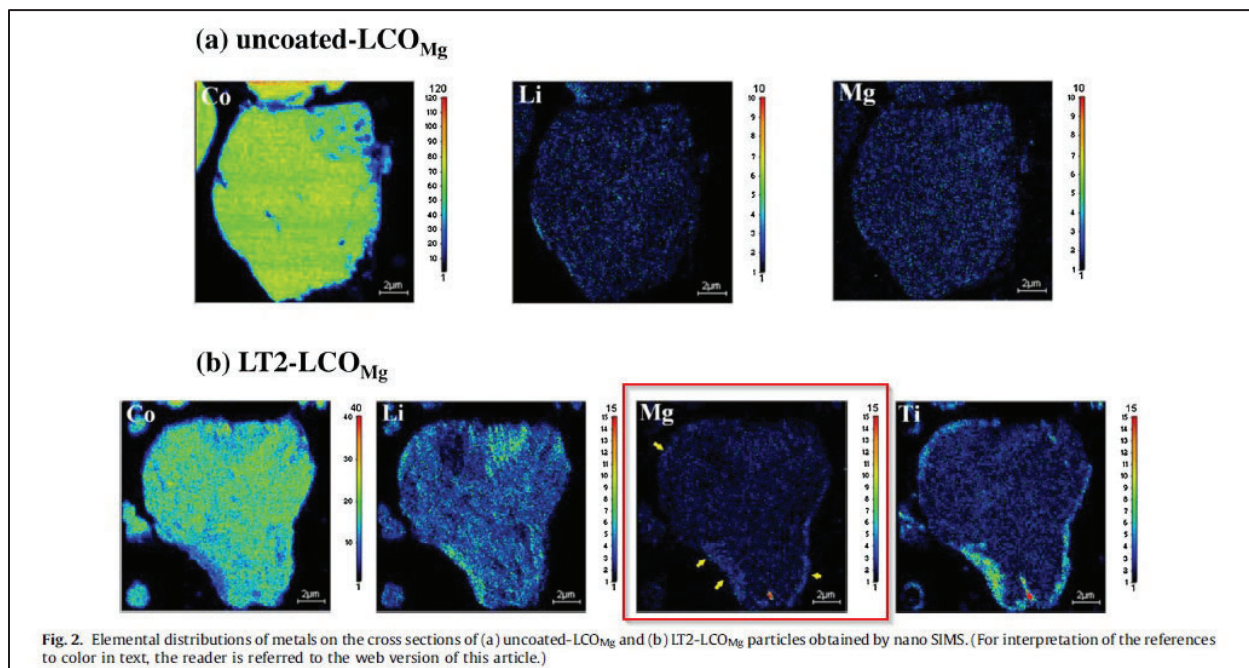
237. As I explained above in Section IX.E.1.e, Fujiki teaches a PEAM particle comprising a representative element (*i.e.*, magnesium). In my opinion, a POSITA would have understood that, because Fujiki teaches that magnesium is present throughout the particle (that is, from the surface to the interior of the particle), magnesium would also be present in a crack portion in the particle, as observed from a TEM image. In particular, Fujiki teaches that metallic element M1

(*i.e.*, the representative element, such as magnesium or aluminum) should, for “effectiveness,” be “diffused from the surfaces *into inside portions* near the surfaces of the compound oxide particles so as to show a continuous concentration gradient *toward the centers* of the [PEAM] particles.” Ex. 1008, 5:12–19 (emphasis added). This diffusion of magnesium results in the magnesium concentration “varying continuously from the surface toward the inside of the particle.” *Id.* 12:19–28. Thus, a POSITA would have understood that the crack portions formed in the bulk of Fujiki’s PEAM particle contain magnesium because magnesium is present between the surface and the center (*i.e.*, into the interior or bulk) of the particle.

iii. *Fluorine present in a crack portion*

238. Fujiki further discloses a PEAM particle with fluorine present in a crack portion. As I discussed above, Fujiki teaches coating PEAM particles with metallic fluorides, which contain fluorine. In my opinion, a POSITA would have understood that Fujiki’s metallic fluoride coating of a PEAM particle would result in fluorine being present in crack portions of the particle. Fujiki discloses coating the surface of an active material particle using a compound of metallic element M2 for the purpose of stabilizing the crystal structure of the lithium-ion particles during charging. Ex. 1008, 4:48–56, 5:29–42. As I discussed above in Section IX.E.1.f, Fujiki teaches that when element M2 is a metallic halide, M2 is preferably a metallic fluoride. *Id.*, 6:3–11, 22:28–53. A POSITA would have understood that the fluorine in the

metallic fluoride in Fujiki's coating would be present in the crack portions because it was well-known that coating materials would be present at the exterior edge of the crack wall, as I explain above in section IX.B.2.g (with reference to Shim). Moreover, as previously mentioned, a POSITA would have been aware that coating materials could become embedded or lodged inside such cracks. *See* Ex. 1016, 4 (“[T]he cycling process could have enabled the particles of the coating material to become ingrained in the crevices and cracks on the cathode surface.”). This is supported by Shim's Figure 2, which (as I discussed above in section IX.B.2.g.ii), shows a high concentration of magnesium and titanium (two elements present in the coating layer) in an apparent crack portion:



Ex. 1005, Fig. 2 (annotated).

2. Dependent Claim 4

- a. [Preamble]: The lithium-ion secondary battery according to claim 1,

239. As I explained previously in section IX.E.1, Fujiki renders obvious the lithium-ion secondary battery according to claim 1.

- b. [Element 4.1]: wherein the first transition metal is cobalt,

240. As I explained previously in section IX.E.1.c, Fujiki discloses wherein the first transition metal is cobalt.

- c. [Element 4.2]: wherein the second transition metal is titanium, and

241. As I explained previously in section IX.E.1.d, Fujiki discloses wherein the second transition metal is titanium.

- d. [Element 4.3]: wherein the representative element is magnesium.

242. As I explained previously in section IX.E.1.e, Fujiki discloses wherein the representative element is magnesium.

3. Independent Claim 5

- a. [Preamble]: A lithium-ion secondary battery comprising a positive electrode active material layer comprising:

243. As I explained previously in section IX.E.1.a, in my opinion Fujiki discloses this element.

- b. [Element 5.1]: a positive electrode active material particle comprising

244. As I explained previously in section IX.E.1.b, in my opinion Fujiki discloses a positive electrode active material particle.

c. [Element 5.2]: a first transition metal,

245. As I explained previously in section IX.E.1.c, in my opinion Fujiki discloses a PEAM particle comprising a first transition metal.

d. [Element 5.3]: a second transition metal,

246. As I explained previously in section IX.E.1.d, in my opinion Fujiki discloses a PEAM particle comprising a second transition metal.

e. [Element 5.4]: a representative element, and

247. As I explained previously in section IX.E.1.e, in my opinion Fujiki discloses a PEAM particle comprising a representative element.

f. [Element 5.5]: fluorine,

248. As I explained previously in section IX.E.1.f, in my opinion Fujiki discloses a PEAM particle comprising fluorine.

g. [Element 5.6]: wherein the representative element and the fluorine are present in a portion which includes crystal defects observed from a TEM image in the positive electrode active material particle.

249. In my opinion, Fujiki renders this limitation obvious, similar to my discussion above in section IX.E.1.g. Before the priority date of the '828 Patent, a POSITA would have understood a "crack portion" to be an example of a crystal defect under the '828 Patent's express definition. The '828 Patent unambiguously

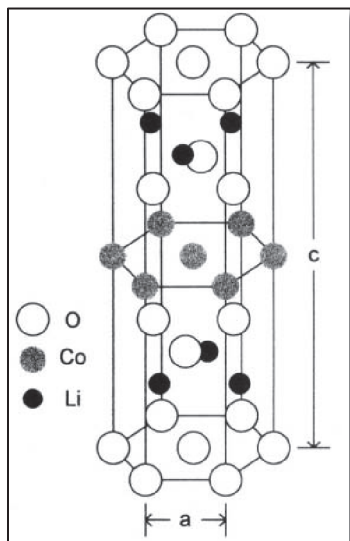
defines “crystal defects” as “defects which can be observed from a TEM image and the like, that is, a structure in which another element enters crystal, a cavity, and the like.” Ex. 1001, 9:55–58. A POSITA would have understood that a “crack portion” is a type of crystal defect.

250. Furthermore, a POSITA would have understood a crystal defect by its plain and ordinary meaning, which is an imperfection or interruption of a regular arrangement of atoms in a crystalline solid. A POSITA would have known that “crystal defects” encompasses a variety of imperfections that can interrupt the periodicity of the atomic lattice. A POSITA would understand a “crack portion” to be an example of such a “crystal defect” because the crack itself interrupts the periodicity of the atomic lattice. That is, cracking necessarily causes interruptions of the regular arrangement of atoms in the crystalline solid because there is an absence of material within the crack where the atomic lattice would otherwise be expected. Moreover, the formation of a crack is known to stress and distort the atomic lattice at and near the crack tip, forming crystal defects at such “crack portions.” Thus, because Fujiki renders obvious a crack portion observed from a TEM image (as I explained in section IX.E.1.g), it also renders obvious a crystal defect observed from a TEM image in view of the general knowledge of the POSITA.

251. A POSITA would have also known that another such class of defects are point defects, which includes substitutional defects (such as intentional

“dopants”). With respect to substitutional defects, one element takes the place of one (or more) of the native elements making up the crystal lattice. As the substitutional atom will not have the same atomic radius as the original atom, for example, this will result in local distortions (expansion or contraction) of the lattice. The dopant elements therefore interrupt the nominal periodicity of the atomic lattice. Fujiki’s disclosure of dopant elements renders obvious a crystal defect, as such dopants would be present throughout the PEAM particle.

252. Consistently, Fujiki notes that its lithium compound oxide (*i.e.*, LiCoO_2) active material has a “crystal structure” or a “laminar rock salt structure.” Ex. 1008, 5:32–35 (describing “the crystal structure of the compound oxide particles”); 12:19–24 (noting that X-ray diffraction demonstrated “a diffraction peak corresponding to lithium cobaltate having a laminar rock salt structure”). A crystal structure is a description of an ordered arrangement of atoms in a crystalline solid. A POSITA would have understood that an example of such crystal structures include the “laminar rock salt structure” of Fujiki’s LiCoO_2 active material (which was also known as a “layered” structure) because such structures have alternating planes of lithium, cobalt, and oxygen atoms as shown in the schematic below:



Ex. 1009, 143. Fujiki also teaches the use of metallic element M2 (such as a metallic fluoride and/or titanium) to “stabiliz[e] the crystal structure of the compound oxide particles at the time of charging.” Ex. 1008, 5:32–35. Fujiki’s teachings thus recognize that there are defects in the crystal structure of LiCoO_2 and teach ways (such as the use of M2) to limit the negative effects of the defects.

253. As I explained above in section IX.E.1.g, Fujiki also renders obvious that the representative element (*i.e.*, magnesium) and fluorine are present in a crack portion observed from a TEM image, where the crack portion includes such crystal defects.

4. Dependent Claim 8

- a. [Preamble]: The lithium-ion secondary battery according to claim 5,

254. As I explained previously in section IX.E.3, in my opinion Fujiki renders obvious the lithium-ion secondary battery according to claim 5.

b. [Element 8.1]: wherein the first transition metal is cobalt,

255. As I explained previously in section IX.E.1.c, in my opinion Fujiki discloses wherein the first transition metal is cobalt.

c. [Element 8.2]: wherein the second transition metal is titanium, and

256. As I explained previously in section IX.E.1.d, in my opinion Fujiki discloses wherein the second transition metal is titanium.

d. [Element 8.3]: wherein the representative element is magnesium.

257. As I explained previously in section IX.E.1.e, in my opinion Fujiki discloses wherein the representative element is magnesium.

F. Ground 5: Claims 2 and 6 are Rendered Obvious by Fujiki in View of Yu

1. Motivation to Combine Fujiki and Yu

258. A POSITA would have been motivated to combine Fujiki and Yu due to their complementary teachings. As discussed above, Fujiki teaches a LiCoO_2 PEAM particle. Yu teaches adding titanium to a LiCoO_2 PEAM particle such that the concentration ratio of titanium to cobalt at the surface of the particle measures 8.49%, or 0.0849. *See* Ex. 1009, 142, Table 3. Given the references' complementary teachings, combining Fujiki and Yu would have involved routine implementation with no apparent technical risks.

259. As an initial matter, a POSITA would have understood that Fujiki and Yu are both directed to improving PEAM particles to enhance battery performance. Both references describe lithium-ion batteries with LiCoO_2 cathode particles treated with the transition metal titanium. Ex. 1008, 5:29–42, 18:63–19:22; Ex. 1009, 140. Both references also discuss addressing the issue of capacity fade during cycling due to degradation of the cathode structure by coating the core (bulk) PEAM particle with a transition metal-containing surface layer. *See* Ex. 1008, 34:8–13 (“Thus, where the compound oxide particles coated with the metallic element . . . are used as the positive electrode active material, both a high initial capacity and a high capacity retention could be obtained simultaneously.”). Ex. 1007, 1 (“As well as known, partial doping of cobalt with metal ions leads to ***extended cyclability and increase in capacity of lithium ion cells*** by enhancing structural stability. According to our previous conjecture, the excellent electrochemical properties of the composite would be ascribed to the protection and enhancement of the shell $\text{LiCo}_{0.99}\text{Ti}_{0.01}\text{O}_2$ material[.]”) (emphasis added).

260. A POSITA aiming to improve the cycling performance of LCO would have recognized the conceptual similarities of Yu and Fujiki, as both describe the generation of a titanium-enriched layer on LCO that contributes to enhanced cycling performance. The motivations for pursuing such a combination would have been obvious to a POSITA given the known degradation associated with hydrofluoric acid

species in LiPF_6 -based electrolytes. A POSITA would have recognized that a titanium-enriched exterior layer can scavenge such acidic species and act as a physical protection layer for the LCO in the bulk of the PEAM particle. Importantly, both Fujiki and Yu disclose similar methods of fabricating titanium-enriched surface layers on their respective PEAMs. Both references disclose the same compound, TiO_2 , as a preferred titanium source. The TiO_2 is further incorporated using similar methods in both disclosures – namely, homogeneous mechanical mixing with the lithium and cobalt containing precursors, followed by a high-temperature heating step (above 800°C). Accordingly, a POSITA could conclude that these similar procedures would produce similar PEAM particles. Yu discloses that the “added Ti tends to stay on the particle surface in the form of TiO_2 or some unidentified impurity,” (Ex. 1007, 2) while Fujiki states “Also, it was confirmed that titanium oxide was present in the state of being deposited on the surfaces of the particles of the lithium transition metal compound oxide or present between the particles.” Ex. 1008, 19:4–11.

261. In addition to Fujiki and Yu being directed at the same problems in enhancing battery performance, Fujiki and Yu both provide express teachings about dopant elements in LCO, which would have led to a POSITA to consider implementing teachings from Fujiki and Yu together. Dopants were well-known to impart additional structural stability to the LCO. Indeed, Yu teaches it was “[a]s well

as known [that] partial doping of cobalt with metal ions leads to extended cyclability and increase in capacity of lithium ion cells by enhancing structural stability.” Ex. 1007, 1. To this end, Yu uses titanium as a dopant, while Fujiki discloses multiple M1 elements, such as magnesium, that would be expected to diffuse into the core of the LCO particles to effectively dope the particles. Ex. 1008, 5:6–19 (“The metallic element M1 is at least one element which can be substituted for the principal transition metal element A on a solid solution basis, more preferably at least one element selected from the group consisting of Mn, Fe, Mg, Al, Ni, and Co.”). Moreover, Yu expressly describes the benefits doping a PEAM particle with titanium, noting that “[it] is found that even a very small amount of Ti addition . . . can effectively improve the electrochemical performance of the LiCoO_2 material.” Ex. 1007, 4.

262. Fujiki and Yu share further similarities in their disclosures of material and electrochemical characterization techniques. Both references use cell testing using electrolytes consisting of LiPF_6 and carbonates. Fujiki describes the use of energy dispersive X-ray spectroscopy (EDX) to qualify the modified LCO. *Id.*, 12:9–18. Similarly, Yu presents both EDX and XPS to characterize the Ti-to-Co ratios in the synthesized samples. The XPS depth profiling analysis in Yu provides the Ti-to-Co ratio at the surface and at progressive depths into the interior of the sample. *See generally* Ex. 1007. A POSITA attempting to develop an improved battery for

consumer electronics using Fujiki's PEAM particle would have been motivated to incorporate Yu (specifically, the Yu's methods of XPS analysis), to gain a better understanding of the composition of the LCO surface coatings containing Ti.

263. Fujiki additionally discloses the use of Auger electron spectroscopy (AES) to determine the depth-dependent elemental concentration of magnesium, stating "a particle section was cut, and the distributions of elements in the radial direction were measured by Auger electron spectroscopy, upon which it was observed that the magnesium concentration was varying continuously from the surface to the inside of the particle." Ex. 1008, 12:19–28. A POSITA would have recognized the conceptual similarities between AES and XPS, as well as the expressed intent of the measurement by Fujiki. Specifically, a POSITA would have understood that both techniques excite core electrons from a sample, must be conducted in ultrahigh vacuum conditions, use similar electron analyzers and hardware, and are highly sensitive methods to interrogate material composition within nanometers of the surface. Given the complementary nature of AES and XPS, a POSITA would have considered XPS, as disclosed in Yu, to investigate the depth-dependent surface chemistry of Fujiki's PEAM particles.

264. The combination of Fujiki and Yu would simply require using a known technique to improve similar devices and methods in the same way, which I understand is a way of showing that a POSITA would have been motivated to

combine these references. That is, a POSITA could apply Yu's known technique of XPS characterization to Fujiki's particles (which are a known device ready for improvement) to yield predictable results. Furthermore, as disclosed in Yu, a POSITA would have understood that adding some amount of titanium as a dopant would improve the electrochemical performance (*i.e.*, cyclability) of the LiCoO_2 . However, since titanium dopant atoms are inactive under a typical operating voltage window of an LiCoO_2 -based cell (in other words, substituting titanium for cobalt theoretically results in a small loss of capacity), a POSITA would also have been motivated to use as little titanium as necessary to stabilize the structure. With these competing factors in mind, a POSITA would have been motivated to determine an optimum range of titanium concentration (and by extension, Ti:Co concentration) through routine experimentation.

2. Dependent Claim 2

- a. [Preamble]: The lithium-ion secondary battery according to claim 1,

265. As I explained previously in section IX.E.1, Fujiki renders obvious the lithium-ion secondary battery according to claim 1.

- b. [Element 2.1]: wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

266. In my opinion, Fujiki and Yu render this limitation obvious. As I explained above in Section IX.C.2.b (with reference to the combination of Shim and Yu), at the surface of Yu's PEAM particle, the concentration ratio of titanium to cobalt is 8.49%, which equals 0.0849. Ex. 1007 at 138, Table 3. This measured ratio falls within the claimed range of 0.05 to 0.4.

267. In my opinion, a POSITA would have been motivated to combine Fujiki and Yu for all the reasons I discussed above in Section IX.F.1. In particular, a POSITA would have understood that Fujiki and Yu are directed to improving the same degradation problem within the same battery cathode, and that Yu teaches that doping a lithium cobalt oxide PEAM particle with titanium would have enhanced the battery's performance and capacity.

3. Dependent Claim 6

- a. [Preamble]: The lithium-ion secondary battery according to claim 5,

268. As I explained previously in section IX.E.3, in my opinion Fujiki renders obvious the lithium-ion secondary battery according to claim 5.

- b. [Element 6.1]: wherein a relative value of a concentration of the second transition metal is greater than or equal to 0.05 and less than or equal to 0.4 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

269. As I explained previously in section IX.F.2.b, in my opinion Fujiki and Yu render this element obvious.

G. Ground 6: Claims 3 and 7 are Rendered Obvious by Fujiki in Combination with Lee

1. Motivation to Combine Fujiki and Lee

270. A POSITA would have been motivated to combine Fujiki and Lee due to their complementary teachings. As discussed above, Fujiki teaches a LiCoO_2 PEAM particle. Lee teaches coating a LiCoO_2 PEAM (such as the one taught by Fujiki) with MgF_2 such that the concentration ratio of magnesium to cobalt at the surface of particle measures approximately 1.0 to 1.2. *See* Ex. 1007, Fig. 5(a). Given the references' complementary teachings, combining Fujiki and Lee would have involved routine implementation with no apparent technical risks.

271. Fujiki and Lee share further similarities in their disclosures of material and electrochemical characterization techniques. A POSITA would have understood that Fujiki and Lee are both directed to improving PEAM particles to enhance battery performance. Both references describe lithium-ion batteries with a LiCoO_2 cathode. Furthermore, both references characterize properties of the cathode by using similar analytical methods, such as SEM/EDS. Ex. 1008, 12:9–18; Ex. 1006, 1, 5. Both references also disclose the fabrication of secondary batteries and subsequent electrochemical characterization, including charge-discharge testing. *See generally* Ex. 1006; Ex. 1008.

272. Both Fujiki and Lee also discuss improving the structural integrity of the PEAM particles during cycling by coating the bulk PEAM material—in particular by using metallic fluorides. *See* Ex. 1008, 5:32–35 (“The metallic element M2 is an element which has the function of stabilizing the crystal structure of the compound oxide particles at the time of charging. It is preferable that the metallic element M2 is in present in the state of coating[.]”), 6:3–11 (“In the case where the metallic element M2 is present in the form of a metallic halide particularly preferable is a metallic fluoride.”); Ex. 1006, 6 (“The MgF_2 coating layer was very effective in improving the electrochemical properties of the LiCoO_2 film electrode.”). Lee’s coating of MgF_2 is a “metallic fluoride,” which Fujiki notes is a “particularly preferable” coating for stabilizing the crystal structure of the PEAM particles.

273. A POSITA would also have understood that combining Lee’s MgF_2 coating with Fujiki would simply require using similar techniques to improve similar devices and methods in the same way and be an example of a combination of known prior-art elements according to known methods to yield predictable results, which I understand are ways of showing that a POSITA would have been motivated to combine these references. Fujiki expressly teaches that the electrolyte can contain lithium salts, among which “lithium hexafluorophosphate (LiPF_6) is preferable.” Ex. 1008, 9:37–51. Furthermore, as I discussed above, a POSITA would have

understood that a fluorine-containing electrolyte (such as LiPF_6) degrades to form hydrofluoric acid, which detrimentally affects battery performance. *See* Ex. 1006, 3–4; Ex. 1023, 1 (HF degrades cathode particles); Ex. 1024, 1 (beneficial to suppress HF formation); Ex. 1027, 1 (describing “capacity loss of LiCoO_2 ” “mainly due to the interfacial impedance growth between the cathode and electrolyte, which results from side reactions involving LiPF_6 ”).

274. To address this problem, a POSITA would have been motivated to use Lee’s MgF_2 coating. Lee teaches that “when the surface of the cathode is covered with a coating layer, the coating material serves as a protective layer, preventing direct contact with the reactive electrolyte.” Ex. 1006, 4. Thus, Lee’s MgF_2 coating could “react with the acidic electrolyte and incur damage on cycling instead of the electrode material.” *Id.*, 5. This could lead to enhanced electrochemical properties due to “the MgF_2 coating layer [being] effective as a protective layer to prevent the dissolution of the transition metal.” *Id.* Furthermore, the fluoride layer would protect the particle from damage caused by exposure to HF acid. *See* Ex. 1024, 1–2 (“This fluoride layer can provide an additional protection, forming a permanent physical barrier that restricts penetration of HF through the oxide coating.”); Ex. 1027, 1 (“The AlF_3 coating protected LiCoO_2 from the chemical attack by HF . . . and maintain[ed] the structural stability when cycled above 4.5V.”).

275. Additionally, a POSITA would have understood that adding magnesium as a dopant improves the electrochemical performance (*i.e.*, electrical conductivity) of the LiCoO_2 . However, since magnesium dopant atoms are inactive within a typical operating voltage window of an LiCoO_2 -based cell (in other words, substituting magnesium for cobalt theoretically results in a small loss of capacity), a POSITA would also have been motivated to use as little magnesium as necessary to stabilize the structure. With these competing factors in mind, a POSITA would have been motivated to determine an optimum range of magnesium concentration (and by extension, Mg:Co concentration) through routine experimentation. Additionally, Fujiki discloses the use of Auger electron spectroscopy (AES) to determine the depth-dependent elemental concentration of the disclosed examples utilizing metal fluoride coatings, stating “a particle section was cut, and the distributions of elements in the radial direction were measured by Auger electron spectroscopy, upon which it was observed that the magnesium concentration was varying continuously from the surface to the inside of the particle.” *E.g.*, Ex. 1008, 12:19–28 (and describing AES procedures similarly with respect to *all* example disclosures). A POSITA would have recognized the conceptual similarities between AES and XPS (as disclosed in Lee), as well as the expressed intent of the measurement by Fujiki. Specifically, a POSITA would have understood that both techniques excite core electrons from a sample, must be conducted in ultrahigh vacuum conditions, use

similar electron analyzers and hardware, and are highly sensitive methods to interrogate material composition within nanometers of the surface. Given the complementary nature of AES and XPS, a POSITA would have considered XPS, a POSITA would have considered XPS, as disclosed in Lee, to investigate the depth-dependent surface chemistry of Fujiki's PEAM particles.

2. Dependent Claim 3

- a. [Preamble]: The lithium-ion secondary battery according to claim 1,

276. As I explained previously in section IX.E.1, Fujiki renders obvious the lithium-ion secondary battery according to claim 1.

- b. [Element 3.1]: wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

277. In my opinion, Fujiki and Lee render this limitation obvious. As explained above in Section IX.D.1.b (with respect to the combination of Shim and Lee), at the surface of Lee's PEAM particle, the concentration ratio of magnesium to cobalt measures between 1.0 and 1.25. Ex. 1006, 1–2, 90, Fig. 5(a). This range of ratios falls fully within the claimed range of 0.4 to 1.5.

278. In my opinion, a POSITA would have been motivated to combine Fujiki and Lee for all the reasons I discussed above in Section IX.G.1. In particular,

a POSITA would have understood that Fujiki and Lee are directed to improving the same structural degradation problem within the same battery cathode. A POSITA would have further understood that Fujiki expressly teaches using a “metallic fluoride,” and that Lee teaches that using MgF_2 (a metallic fluoride) as a coating would have enhanced the battery’s performance, particularly in light of Fujiki’s preference for LiPF_6 used as the electrolyte salt.

3. Dependent Claim 7

- a. [Preamble]: The lithium-ion secondary battery according to claim 5,

279. As I explained previously in section IX.E.3, in my opinion Fujiki renders obvious the lithium-ion secondary battery according to claim 5.

- b. [Element 7.1]: wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of the first transition metal is defined as 1.

280. As I explained previously in section IX.G.b, in my opinion Fujiki and Lee render this element obvious.

X. SECONDARY CONSIDERATIONS

281. I have seen no evidence of, and I am not aware of any, secondary considerations that would tend to show non-obviousness of the Challenged Claims of the ’828 Patent, including commercial success, long-felt need, failure of others,

skepticism, praise, teaching away, recognition of a problem or copying by competitors. However, should Patent Owner assert any secondary considerations of non-obviousness, I reserve the right to respond.

XI. CONCLUSION

282. I declare under penalty of perjury under the laws of the United States of America that the foregoing is true, and that all statement made of my own knowledge are true, and that all statements made on information and belief are believed to be true. I understand the willful false statements are punishable by fine or imprisonment or both. *See* 18 U.S.C. §1001.

Dated: June 4, 2023



Dr. Troy A. Hayes