

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent of: Ochiai et al.

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

Attorney Docket No.: 12732-2330IP1

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Title: POSITIVE ELECTRODE ACTIVE MATERIAL
INCLUDING LITHIUM COBALTATE COATED
WITH LITHIUM TITANATE AND
MAGNESIUM OXIDE

DECLARATION OF DR. BRETT LUCHT

I currently hold the opinions expressed in this declaration. But my analysis may continue, and I may acquire additional information and/or attain supplemental insights that may result in added observations.

I hereby declare that all statements made of my own knowledge are true and that all statements made on information and belief are believed to be true. I further declare that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.

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I, BRETT LUCHT, PH.D., hereby declare the following:

I. QUALIFICATIONS AND BACKGROUND INFORMATION

1. My education and experience are described more fully in the attached curriculum vitae (Ex. 2002). For ease of reference, I have highlighted certain information below.

2. I received my bachelor's degree in 1991 from the University of Puget Sound in Tacoma, WA. I received my doctorate (Ph.D.) in 1996 from Cornell University in Ithaca, NY. I arrived at the University of Rhode Island in 1998, was promoted to Associate Professor with tenure in 2002 and Professor in 2006.

3. My academic research is focused on novel electrolytes and electrolyte electrode interfaces for lithium-ion batteries for electric vehicle applications which include extending the calendar life of lithium-ion batteries, improving the performance of novel high-capacity anodes such as lithium metal or silicon and improving the performance of high voltage cathode materials. My research has been funded by several United States Government agencies including the Department of Energy, National Aeronautics and Space Administration, National Science Foundation, and Department of Defense and several companies including BASF, Samsung, Duracell, and Volkswagen. I have received over \$14 M in external research grants and contracts in support of my research since 1998.

4. I have mentored more than 100 students in the research laboratory including over 40 graduate students, 30 undergraduate students, 20 postdoctoral fellows, along with several visiting students.
5. I have numerous awards for research and intellectual property at the University of Rhode Island, including the 2016 URI Foundation Scholarly Excellence Award.
6. I am currently an Associate Editor for the Journal of Electrochemical Society, Chair of the Battery Division of the Electrochemical Society, and a Fellow of the Electrochemical Society.
7. I have published over 180 manuscripts in peer reviewed journals, my manuscripts have been referenced over 14,000 times, and I have an h index of 66. I have also published two book chapters and am listed as an inventor on seven patents. I have been an invited or keynote speaker at over 100 companies, universities, national laboratories, and national or international conferences. My invited and keynote lectures include many of the most prestigious battery conferences including the International Meeting for Lithium Batteries (Kyoto 2018, Sydney 2022), International Battery Association meeting (Barcelona, 2013; Nantes, France, 2016; Juju, Korea, 2018; Bled, Slovenia, 2022), Japan Battery Symposium (Kyoto 2019), Munich Battery Discussions (Munich 2022) and the Advanced Automotive Battery Conference (Orlando 2012; Atlanta 2014; Detroit 2016; Mainz, Germany 2018).

8. I have been retained on behalf of SEMICONDUCTOR ENERGY LABORATORY CO., LTD. (“SEL” or “Patent Owner”) to offer technical opinions related to U.S. Patent No. 10,741,828 (“the ’828 Patent”) (Ex. 1001) and alleged prior art references relating to its subject matter. I have reviewed the ’828 Patent and its accompanying prosecution history (Ex. 1002). Additionally, I have reviewed materials cited in the Petition in addition to the materials cited in the present declaration. For example, I have reviewed at least the following references:

- Declaration of Dr. Troy Hayes in Support of *Inter Partes Review* of U.S. Patent No. 10,741,828 (Ex. 1003);
- *Curriculum Vitae* of Dr. Troy Hayes (Ex. 1004);
- 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*, 186 *Electrochimica Acta*, 201–08 (2015) (Ex. 1005);
- Lee & Park, *Interface Characterization of MgF₂-Coated LiCoO₂ Thin Films*, 230 *Solid State Ionics*, 86–91 (2013) (Ex. 1006);
- Yu et al., *The Investigation of Ti-Modified LiCoO₂ Materials for Lithium Ion Battery*, 262 *Journal of Power Sources*, 136–139 (2014) (Ex. 1007);
- U.S. Patent No. 9,105,926 (“Fujiki”) (Ex. 1008);

- Li et al., *Fracture Behavior in Battery Materials*, J. Phys. Energy 2 (2020) (Ex. 2003);
- Li et al., *Cathode Materials Modified by Surface Coating for Lithium Ion Batteries*, Electrochimica Acta 51, 3872–83 (2006) (Ex. 2004);
- Takada, *Progress and Prospective of Solid-State Lithium Batteries*, Acta Materiala 61, 759–70 (2013) (Ex. 2005);
- Xu, et al., *Lithium Transport Through Lithium-Ion Battery Cathode Coatings*, J. Mater. Chem. A. 3, 17248–72 (2015) (Ex. 2006);
- Jayasree, et al., *Nanoscale-Engineered LiCoO₂ as a High Energy Cathode for Wide Temperature Lithium-Ion Battery Applications—Role of Coating Chemistry and Thickness*, Nanotechnology 33, 1–19 (2022) (Ex. 2007);
- Ahaliabadeh, et al., *Extensive Comparison of Doping and Coating Strategies for Ni-Rich Positive Electrode Materials*, J. Power Sources 540, 1–23 (2022) (Ex. 2008);
- Han, et al., *Influence of Coating Protocols on Alumina-Coated Cathode Material: Atomic Layer Deposition versus Wet-Chemical Coating*, J. Electrochem. Soc. 166, A3679–84 (2019) (Ex. 2009);

- Yamamoto, et al., *Stabilization of the Electronic Structure at the Cathode/Electrolyte Interface via MgO Ultra-thin Layer during Lithium-ions Insertion/Extraction*, *Electrochemistry* 82, 891–96 (2014) (Ex. 2010).

9. I have also reviewed various supporting references and other documentation (including Ex. 1016: Fey et al., *MgAl₂O₄ Spinel-Coated LiCoO₂ as Long-Cycling Cathode Materials*, *J. Power Sources* 146, 245–49 (2005)) as further noted in my opinions below.

10. Counsel has informed me that I should consider these materials through the lens of one of ordinary skill in the art related to the '828 Patent at the time of the earliest possible priority date of the '828 Patent. The '828 Patent was filed on June 30, 2017 and claims priority to several Japanese applications, the earliest of which was filed on July 5, 2016 (“the Critical Date”). Counsel has informed me that the Critical Date represents the earliest possible priority date to which the challenged claims of '828 Patent are entitled, and I have therefore used that Critical Date in my analysis below.

11. I have no financial interest in the parties or in the outcome of this proceeding. I am being compensated for my work as an expert on an hourly basis. My compensation is not dependent on the outcome of these proceedings or the content of my opinions.

12. In writing this declaration, I have considered the following: my own knowledge and experience, including my work experience in the fields of chemistry, energy storage devices and materials science and teaching those subjects; and my experience in working with others involved in those fields. In addition, I have analyzed various publications and materials, in addition to other materials I cite in my declaration.

13. My opinions, as explained below, are based on my education, experience, and expertise in the fields relating to the '828 Patent. Unless otherwise stated, my testimony below refers to the knowledge of one of ordinary skill in the art as of the Critical Date, or before. Any figures that appear within this document have been prepared with the assistance of Counsel and reflect my understanding of the '828 Patent and the prior art discussed below.

II. OVERVIEW OF CONCLUSIONS FORMED

14. This declaration explains the conclusions that I have formed based on my analysis. To summarize those conclusions, based on my knowledge and experience and my review of the prior art publications listed above, I believe that:

- The prior art references relied on in Grounds 1–6 do not teach or suggest “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,” as recited in independent claim 1; and

- The prior art references relied on in Grounds 1–6 do not teach or suggest “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,” as recited in independent claim 5.

III. LEGAL STANDARDS

A. Legal Standards for Anticipation

15. I have been informed by Counsel and understand that documents and materials that qualify as prior art can render a patent claim unpatentable as anticipated. I have been informed by Counsel and understand that all prior art references are to be looked at from the viewpoint of a person of ordinary skill in the art.

16. I have been informed by Counsel and understand that a challenged claim is unpatentable as “anticipated” under 35 U.S.C. § 102 if it is determined that all the limitations of the claim are described in a single prior art reference. I have been informed by Counsel and understand that, to anticipate a claim, a prior art reference must disclose, either expressly or inherently, each and every limitation of that claim and enable one of ordinary skill in the art to make and use the invention.

17. I have been informed by Counsel and understand that in an *inter partes* review, “the petitioner shall have the burden of proving a proposition of unpatentability,” including a proposition of anticipation, “by a preponderance of the evidence.” 35 U.S.C. § 316(e).

B. Legal Standards for Obviousness

18. I have been informed by Counsel and understand that documents and materials that qualify as prior art can render a patent claim unpatentable as obvious. I have been informed by Counsel and understand that all prior art references are to be looked at from the viewpoint of a person of ordinary skill in the art at the time of the invention, and that this viewpoint prevents one from using his or her own insight or hindsight in deciding whether a claim is obvious.

19. I have been informed by Counsel and understand that a claim is unpatentable for obviousness under 35 U.S.C. § 103 “if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains.” I have been informed by Counsel and understand that obviousness may be based upon a combination of references. I have been informed by Counsel and understand that the combination of familiar elements according to known methods is likely to be obvious when it does no more than yield predictable results. However, I have been informed by Counsel

and understand that a patent claim composed of several elements is not proved obvious merely by demonstrating that each of its elements was, independently, known in the prior art.

20. I have been informed by Counsel and understand that when a patented invention is a combination of known elements, a court must determine whether there was an apparent reason to combine the known elements in the fashion claimed by the patent at issue by considering the teachings of prior art references, the effects of demands known to people working in the field or present in the marketplace, and the background knowledge possessed by a person having ordinary skill in the art.

21. I have been informed by Counsel and understand that a patent claim composed of several limitations is not proved obvious merely by demonstrating that each of its limitations was independently known in the prior art. I have been informed by counsel for the Patent Owner and understand that identifying a reason those elements would be combined can be important because inventions in many instances rely upon building blocks long since uncovered, and claimed discoveries almost of necessity will be combinations of what, in some sense, is already known. I have been informed by Counsel and understand that it is improper to use hindsight in an obviousness analysis, and that a patent's claims should not be used as a "roadmap."

22. I have been informed by Counsel and understand that an obviousness inquiry requires consideration of the following factors: (1) the scope and content of the prior

art; (2) the differences between the claims and the prior art; (3) the level of ordinary skill in the pertinent art; and (4) any objective indicia of non-obviousness, such as commercial success, long-felt but unresolved need, failure of others, industry recognition, copying, and unexpected results. I understand that the foregoing factors are sometimes referred to as the “Graham factors.”

23. I have been informed by Counsel and understand that an obviousness evaluation can be based on a combination of multiple prior art references. I understand that the prior art references themselves may provide a suggestion, motivation, or reason to combine, but that the nexus linking two or more prior art references is sometimes simple common sense. I have been informed by Counsel and understand that obviousness analysis recognizes that market demand, rather than scientific literature, often drives innovation, and that a motivation to combine references may be supplied by the direction of the marketplace.

24. I have been informed by Counsel and understand that if a technique has been used to improve one device, and a person of ordinary skill at the time of invention would have recognized that it would improve similar devices in the same way, using the technique is obvious unless its actual application is beyond his or her skill.

25. I have been informed by Counsel and understand that practical and common sense considerations should guide a proper obviousness analysis, because familiar items may have obvious uses beyond their primary purposes. I have been informed

by Counsel and understand that a person of ordinary skill looking to overcome a problem will often be able to fit together the teachings of multiple prior art references. I have been informed by Counsel and understand that obviousness analysis therefore takes into account the inferences and creative steps that a person of ordinary skill would have employed at the time of invention.

26. I have been informed by Counsel and understand that a proper obviousness analysis focuses on what was known or obvious to a person of ordinary skill at the time of invention, not just the patentee. Accordingly, I understand that any need or problem known in the field of endeavor at the time of invention and addressed by the patent can provide a reason for combining the elements in the manner claimed.

27. I have been informed by Counsel and understand that a claim can be obvious in light of a single reference, without the need to combine references, if the elements of the claim that are not found explicitly or inherently in the reference can be supplied by the common sense of one of skill in the art.

28. I have been informed by Counsel and understand that secondary indicia of non-obviousness may include (1) a long felt but unmet need in the prior art that was satisfied by the invention of the patent; (2) commercial success of processes covered by the patent; (3) unexpected results achieved by the invention; (4) praise of the invention by others skilled in the art; (5) taking of licenses under the patent by others; (6) deliberate copying of the invention; (7) failure of others to find a solution to the

long felt need; and (8) skepticism by experts. I understand that evidence of secondary indicia of non-obviousness, if available, should be considered as part of the obviousness analysis.

29. I have been informed by Counsel and understand that there must be a relationship between any such secondary considerations and the invention, and that contemporaneous and independent invention by others is a secondary consideration supporting an obviousness determination.

30. In sum, my understanding is that prior art teachings are properly combined where one of ordinary skill having the understanding and knowledge reflected in the prior art and motivated by the general problem facing the inventor, would have been led to make the combination of elements recited in the claims. Under this analysis, the prior art references themselves, or any need or problem known in the field of endeavor at the time of the invention, can provide a reason for combining the elements of multiple prior art references in the claimed manner.

31. I have been informed by Counsel and understand that in an *inter partes* review, “the petitioner shall have the burden of proving a proposition of unpatentability,” including a proposition of obviousness, “by a preponderance of the evidence.” 35 U.S.C. § 316(e).

C. Level Of Ordinary Skill in The Art

32. I understand that Petitioner contends (in contradiction with Petitioner's own expert) that a person of ordinary skill in the art as of the Critical Date of the '828 Patent (hereinafter a "POSITA") would have at least a bachelor's degree in chemistry, chemical engineering, or materials science and at least five years of experience in the field of batteries and battery materials, and individuals with different education and additional experience could still be of ordinary skill in the art if that additional experience compensates for a deficit in their education and experience. Pet., 33. For the purpose of the analysis provided herein, Counsel has instructed me to adopt and apply the Petitioner's definition of a POSITA, and I have done so in my analysis in this declaration.

33. Based on my knowledge, skill, and experience, I have an understanding of the capabilities of a POSITA as defined by the Petitioner.

IV. OVERVIEW OF THE '828 PATENT

34. Lithium-ion batteries (LIBs) have been the subject of research and development since at least the 1970s for their power storage capabilities. LIBs are rechargeable batteries that include a positive electrode (made of a "positive electrode active material" or "PEAM"), a negative electrode, an electrolyte, and a separator. As a LIB discharges, lithium ions move from the negative electrode to the positive

electrode through the electrolyte and the separator. On the other hand, as the LIB charges, lithium ions move back to the negative electrode from the positive electrode.

35. As the '828 Patent describes, the performance of LIBs can be characterized based on “charge and discharge characteristics, cycle characteristics, reliability, safety, cost, and the like.” Ex. 1001, 1:66–2:3. At the time of the invention of the '828 Patent, a known issue with LIBs was the reduction in capacity due to repeated charge and discharge cycles. *Id.*, 2:4–14. It was understood that this degradation of a LIB’s cycle characteristics and charge and discharge characteristics was caused, at least in part, by the cracking or breaking of PEAM particles, which exposes new surfaces of the PEAM particles to electrolyte solution and can result in a decomposition reaction. *See id.*, 4:1–9. The '828 Patent presents a then-novel solution to this degradation problem that suppresses capacity fade and yields improved battery performance. *Id.*, 2:4–14.

36. Other LIB researchers at the time had explored surface modification of PEAMs using coatings such as metal oxides (e.g., Al_2O_3 , MgO , ZrO_2 , etc.) and metal fluorides (e.g., AlF_3) to stabilize LIBs operating at high voltages. Ex. 2006, 1. However, as late as 2015, “the coating functionalities and the stabilizing mechanisms [were] still not fully understood and [were] a subject of intensive research.” *Id.* At the time, researchers had proposed several hypotheses for how

coatings improved PEAM particle performance, including (i) the facilitation of electron transport, (ii) the alteration of the chemical properties of the cathode surface to improve stability and performance, (iii) the local reduction of acidity of the electrolyte near the cathode surface, and (iv) physical protection of the PEAM particle from oxidation and corrosion. *Id.* A number of different coating techniques for PEAM particles had (and have since) also been explored including solid state approaches, co-precipitation approaches, sol-gel approaches, hydro-thermal approaches, combustion approaches, and spray-drying approaches—with “[d]ifferent synthesis methods [having] important effects on the morphology and structure properties of final products.” Ex. 2008, 5. As the ’828 Patent identified, existing coating techniques suffered from the drawback that they were not sufficiently able to protect all solvent-exposed surfaces of the PEAM. *See, e.g.*, Ex. 1001, 4:14–18.

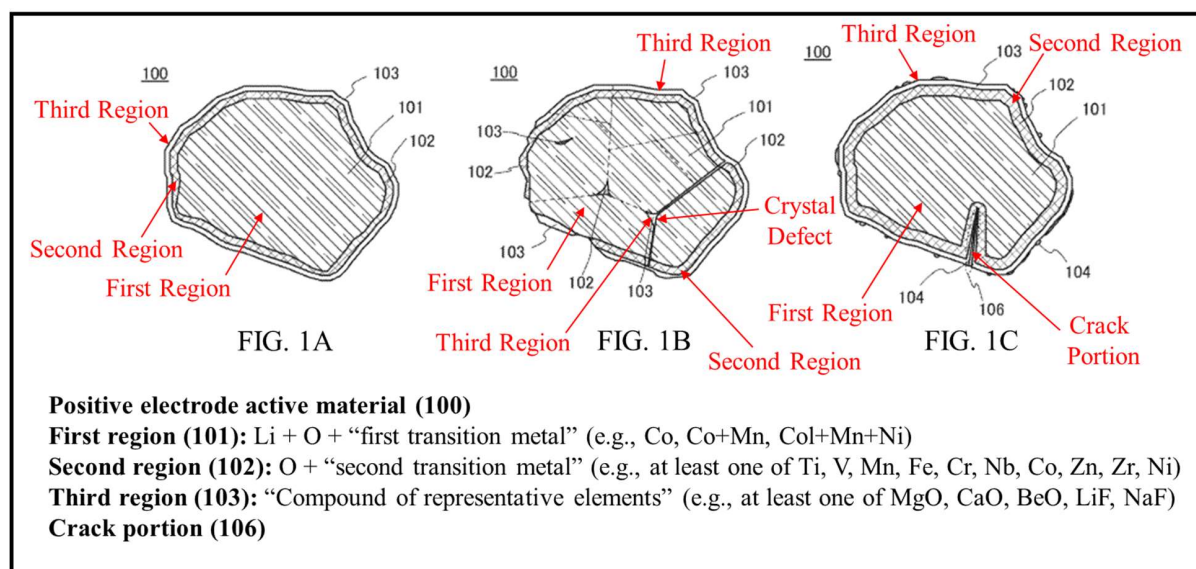
37. The ’828 Patent describes and claims PEAM particles with coatings that were a substantial and unexpected improvement compared to prior coating materials and techniques. Rather than using conventional approaches to coat the outside of the particle but exclude surfaces of crack portions, the inventors of the ’828 Patent instead described forming a protective layer over surface regions of the PEAM particle—including surfaces of crack portions—by segregating protective components from within the particle and bringing those components to surface

regions of the particle to form a protective layer. *Id.*, 23:60–24:5, 24:24–36. Another added benefit of this process is that it can also segregate the protective components into the vicinity of crystal defects within the PEAM particle, further improving the stability of the crystal structure of the PEAM. *Id.*, 24:11–23, Claim 5.

38. As an example, the '828 Patent describes a PEAM particle 100 having a first region 101, a second region 102, and a third region 103. *See id.*, 9:30–39. The first region 101 is described as containing “a composite oxide of lithium and a first transition metal,” wherein the first transition metal can be cobalt only (Co); cobalt and manganese (Mn); or cobalt, manganese, and nickel (Ni). *Id.*, 10:5–12. The second region 102 is described as containing “an oxide of a second transition metal,” wherein the second transition metal can be a non-stoichiometric metal such as at least one of titanium (Ti), vanadium (V), manganese, iron (Fe), chromium (Cr), niobium (Nb), cobalt, zinc (Zn), zirconium (Zr), nickel, and the like. *Id.*, 10:52–62. The third region 103 is described as containing “a compound of representative elements” such as at least one of magnesium oxide (MgO), calcium oxide (CaO), beryllium oxide (BeO), lithium fluoride (LiF), and sodium fluoride (NaF). *Id.*, 11:64–12:4. As described in the '828 Patent, the third region 103—which can include fluorine and a representative element such as magnesium—is an example of a protective layer of the PEAM particle 100. *See id.*, 12:7–11 (“[F]or the third region 103, a material which is hardly changed electrochemically in the process of charging

and discharging and is not easily transformed by contact with the electrolyte solution is preferably used.”); *id.*, 12:14–17 (“The positive electrode active material 100 includes the third region 103 in a superficial portion to improve stability in charging and discharging of the secondary battery.”); *see also* Figs. 5C, 6D (showing that the third region 103 includes fluorine and magnesium).

39. This information is summarized in annotated Figs. 1A-1C of the ’828 Patent, reproduced below.



Ex. 1001, Figs. 1A–1C (annotated)

40. According to the ’828 Patent (e.g., as shown in Figs. 1A–1C), “the third region 103 may be present in a portion which includes crystal defects or a crack portion in the positive electrode active material 100 or in the vicinity thereof.” *Id.*, 9:51–54. In other words, the ’828 Patent describes that the third region 103 forms not only on an

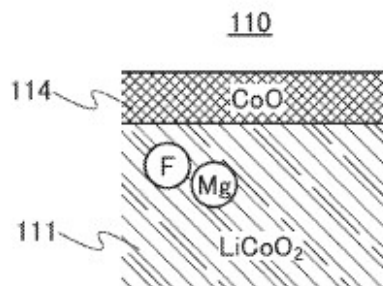
exterior surface of a PEAM particle but also in crack portions and portions which include crystal defects, thereby providing protection in these areas where conventional coatings did not. Conventional coatings could not have provided protection in these areas because coating techniques such as co-precipitation method, sol-gel methods, spray-drying methods, etc. provide external coatings and simply could not have resulted (or at least would not have reliably resulted) in coating materials becoming present in internal portions of the PEAM such as the crack portion 106 (shown in Fig. 1C) or the entirely internal crystal defect shown in Fig. 1B.

41. The segregation phenomenon described in the '828 Patent was unlike conventional coating techniques being implemented at the time of the invention since the protective components were brought to the surface region of the PEAM particle from within the particle itself. For example, Figs. 5A–5C and 6A–6D of the '828 Patent illustrate a segregation model of elements contained in a PEAM, showing how during a manufacturing process that can involve heating (e.g., at 500°C to 1200°C), representative elements such as magnesium (Mg) and fluorine (F) can unexpectedly segregate towards a superficial portion of the PEAM to form the third region 103. *Id.*, 23:36–24:5, 10:29–33, 27:1–28:17.

42. In more detail, and starting, for example, with Fig. 5A of the '828 Patent, a particle 110 is shown as starting with a cobalt oxide layer 114 in contact with a

region 111 containing lithium cobaltate, magnesium, and fluorine. *Id.*, 22:33–37, 22:52–54.

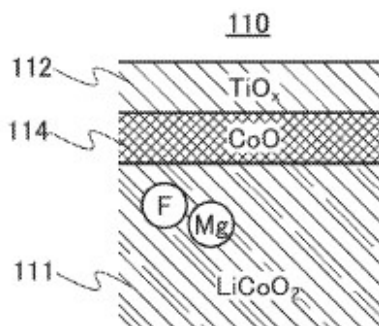
FIG. 5A



Ex. 1001, Fig. 5A

43. The particle 110 is then described as being coated with a material containing titanium (e.g., a titanium oxide) using a sol-gel method to produce the state of the particle 110 shown in Fig. 5B. *Id.*, 22:38–43.

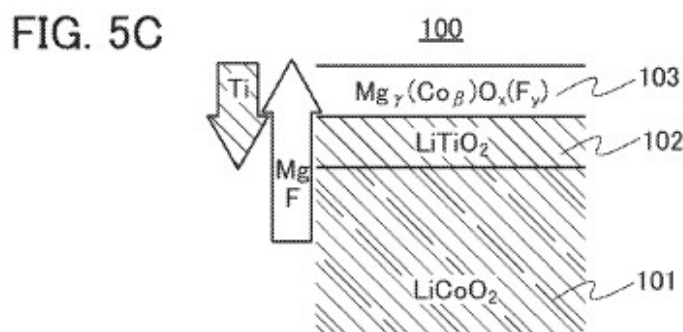
FIG. 5B



Ex. 1001, Fig. 5B

44. Then, the particle 110 coated with the layer 112 containing titanium is heated, for example, at 800°C for two hours to form the PEAM 100 shown in Fig. 5C. *Id.*, 22:44–54. According to the '828 Patent, the heating can cause the titanium in the

layer 112 to diffuse into the inside of the particle 110, and simultaneously causes the magnesium and fluorine contained in the region 111 to segregate on a surface region of the particle (not only on an exterior surface of the particle, but also “in the vicinity of crystal defects” and “in [a] crack portion”). *Id.*; *see also id.*, 24:11–36. This heating process can form the second region 102 and the third region 103 of the PEAM 100. *Id.*, 24:43–51. As described above, the third region 103 can serve as a protective layer, “improv[ing] stability in charging and discharging of the secondary battery.” *Id.*, 12:14–17.



Ex. 1001, Fig. 5C

45. For example, a comparison of Figs. 39A, 39B1, 39B2, 39C1, 39C2, 39D1, 39D2, 39E1, and 39E2 with Figs. 40A, 40B1, 40B2, 40C1, 40C2, 40D1, 40D2, 40E1, and 40E2 of the '828 Patent further provides empirical evidence that through heating, Mg can become segregated in a crack portion of the PEAM observed from a TEM image. *See id.*, 63:1–32 (“[I]n Sample 06 before heating, segregation of magnesium is not observed on the surfaces of the particles or in the crack portion.

In contrast, in Sample 07 after heating, segregation of magnesium is observed both on the surfaces of the particles and in the crack portion.”). In my opinion, these figures demonstrate the unique segregation process described in the ’828 Patent and the efficacy of this process for ensuring that crack portions and portions of the PEAM particle which include crystal defects include the presence of protective components such as fluorine and magnesium (which conventional coating approaches may not afford). The ’828 Patent further demonstrates the utility of this segregation approach by demonstrating that a PEAM particle formed through this process showed “excellent initial charge and discharge capacity, rate characteristics, and cycle characteristics.” *Id.*, 65:29–33.

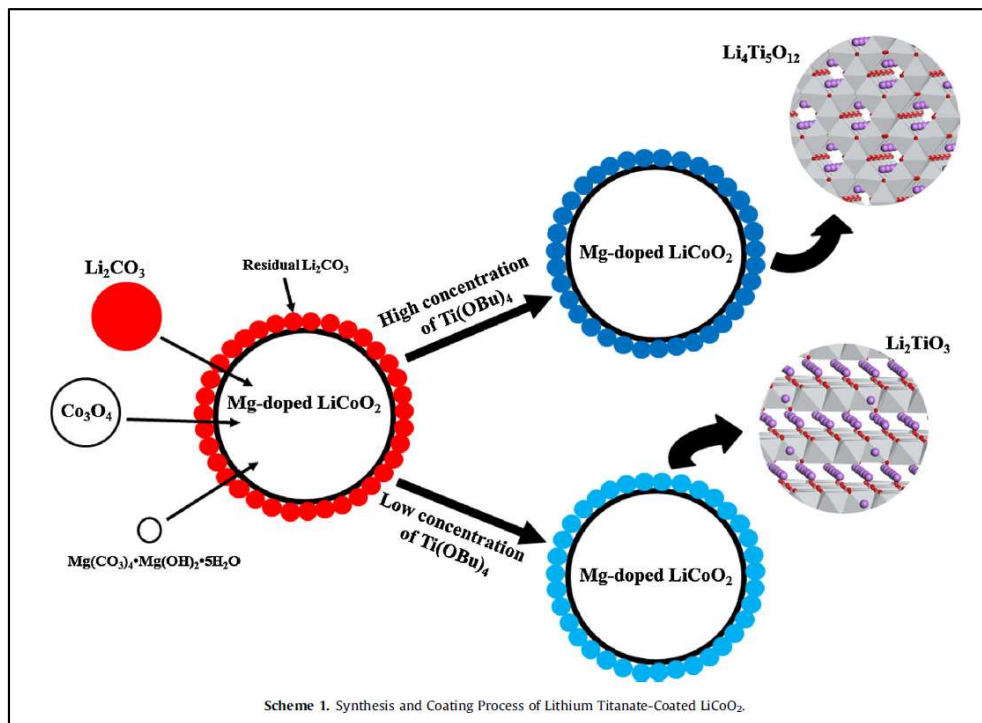
V. OVERVIEW OF THE CITED REFERENCES

A. Shim

46. I have been informed by Counsel and understand that Petitioner relies on Shim in Grounds 1–3 of the Petition to argue that the invention of the ’828 Patent would have been obvious to a POSITA. Shim is a research article that describes an investigation of the performance of lithium titanate coatings for LiCoO_2 cathode materials with Mg doping. Ex. 1005, 1. Shim recognized that successful coatings for cathode materials such as ZrO_2 , SiO_2 , ZnO , Al_2O_3 , $\text{Al}(\text{OH})_3$, AlPO_4 , and AlF_3 have only one stable valence state at ambient condition, enabling active materials to

obtain “enhanced stability” (e.g., minimizing degradation and consequent capacity fade during cycling). *Id.* However, Shim noted that these coating materials “generally exhibit poor lithium ion conduction,” and therefore opted to study coatings made of “inorganic materials containing lithium ions” such as Li_2TiO_3 and $\text{Li}_4\text{Ti}_5\text{O}_{12}$ coatings. *Id.*

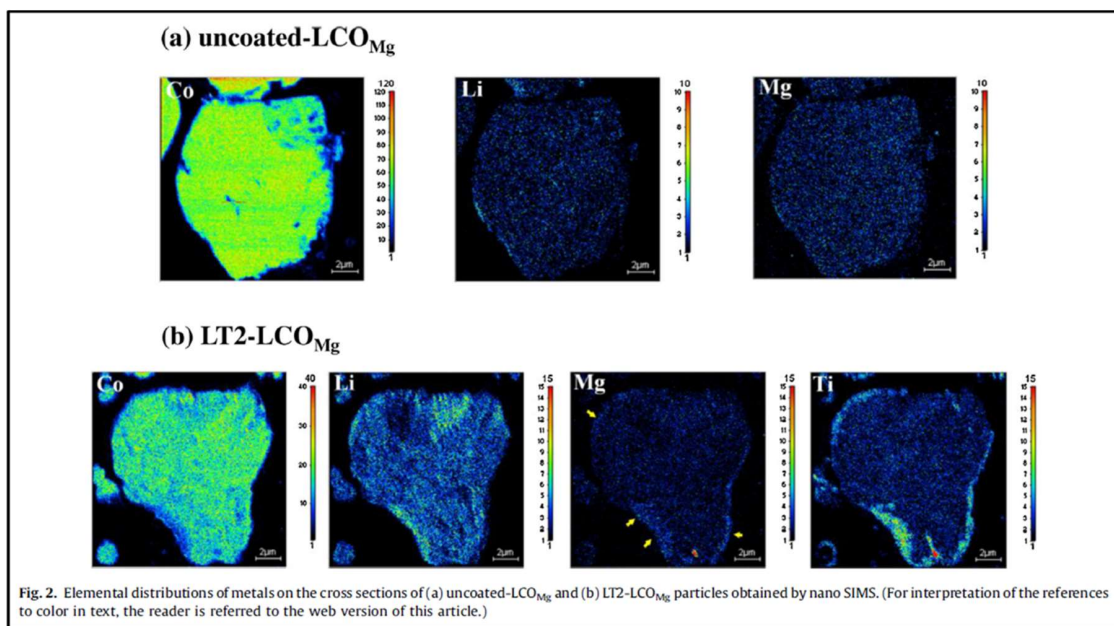
47. Shifting away from traditional coatings with low Li ion conduction, Shim discloses (i) manufacturing cathode materials by coating Mg-doped LiCoO_2 with lithium titanate using a sol-gel technique, (ii) characterizing the resulting materials, and (iii) measuring the electrochemical performance of cells that were prepared using the manufactured cathode materials. *Id.*, 2–3. A diagram of the cathode materials manufactured in Shim is reproduced below.



Ex. 1005, 2 (“Scheme 1”)

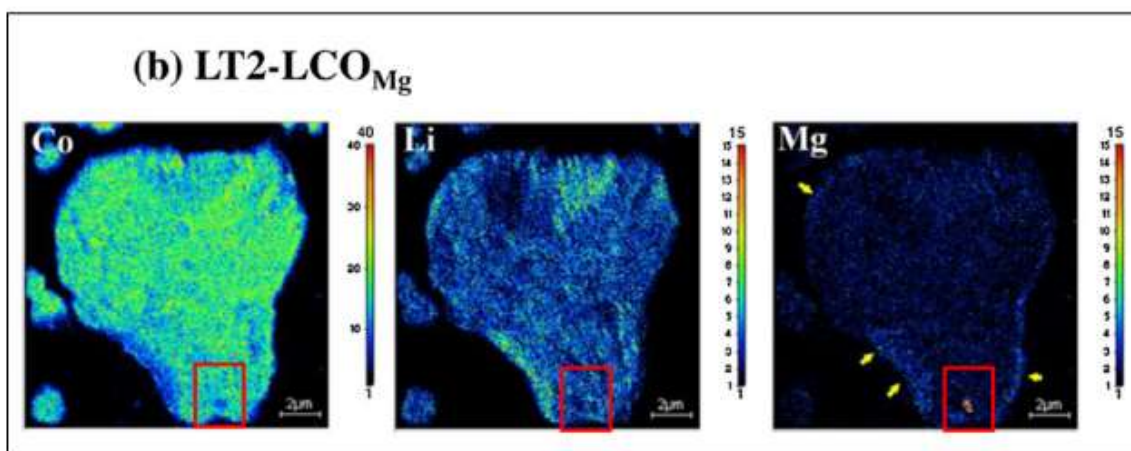
48. One thing that Shim does not disclose, is a PEAM with “crack portions” or portions that include crystal defects. Shim mentions “degradation mechanisms” of layered cathode materials including “the dissolution of transition metal ions into the electrolytes when the electrode is delithiated,” the shearing of the MO_2 layer at the delithiated state from the electrode surface, oxygen loss, and side reactions with electrolytes. *Id.*, 1. However, none of these degradation mechanisms are the same as a crack portion or a portion that includes crystal defects. Shim also makes reference to “structural changes of the [coating] materials after charging” (*id.*, 6) and “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). However, these are not crack portions or portions that include crystal defects either. Shim’s broad disclosure and general description of “degradation mechanisms” lack any particular reference to cracks and crystal defects—not to mention crack portions of a PEAM particle or portions that include crystal defects.

49. Finally, Shim discloses elemental distributions of metals on the cross-section of its uncoated- LCO_{Mg} and coated LT2- LCO_{Mg} PEAM particles obtained by nanoscale secondary ion mass spectrometry (nano SIMS):



Ex. 1005, Fig. 2

50. Petitioner identifies and annotates a feature shown in Fig. 2(b) of Shim (reproduced below) and alleges that this feature constitutes a “crack portion” that contains a high concentration of magnesium. *See* Pet., 53–55; Ex. 1003, ¶ 162.



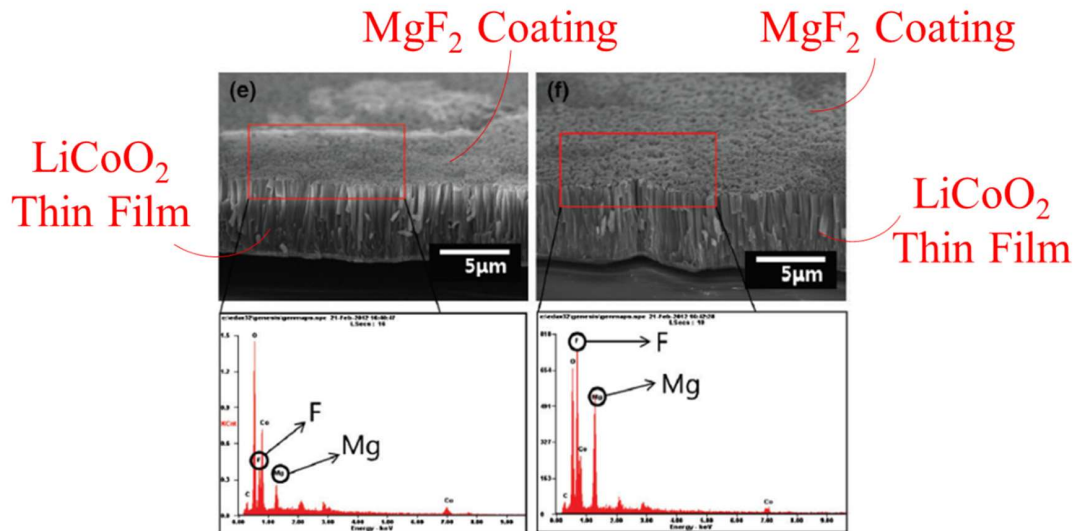
Ex. 1005, Fig. 2(b) (annotated by Petitioner)

However, I disagree. The region pointed out by Petitioner is never mentioned by Shim as an area of interest, and it is certainly not identified as a crack portion. Nor is it a crack portion “observed from a TEM image,” since Fig. 2(b) of Shim was obtained using nano SIMs. In my opinion, the region pointed out by Petitioner could be any number of features including a manufacturing impurity caused by poor mixing of the starting materials, and the Petitioner has provided no evidence to suggest it is indeed a crack portion.

B. Lee

51. I have been informed by Counsel and understand that Petitioner relies on Lee as a secondary reference in Grounds 1–3 of the Petition to argue that the invention of the ’828 Patent would have been obvious to a POSITA. Lee is a research article that describes investigating MgF_2 as a coating material to improve the electrochemical properties of a LiCoO_2 film cathode. *See* Ex. 1006, 1. In particular, Lee states that the inspiration for using a MgF_2 coating came from “the fact that other fluorides, such as AlF_3 , were very effective in modifying cathode surfaces.” *Id.* In the study, Lee discloses (i) manufacturing cathode materials by coating pristine LiCoO_2 thin film with MgF_2 using a spin-coating technique, (ii) observing the coating layers, and (iii) measuring the electrochemical properties of the coated LiCoO_2 thin films “to investigate the effectiveness of the MgF_2 surface

modification.” *Id.*, 1–2. Imagery of the MgF_2 -coated cathode materials tested by Lee is shown in annotated Figs. 1(e) and 1(f) of Lee (reproduced below).



Ex. 1006, Figs. 1(e) and 1(f) (annotated)

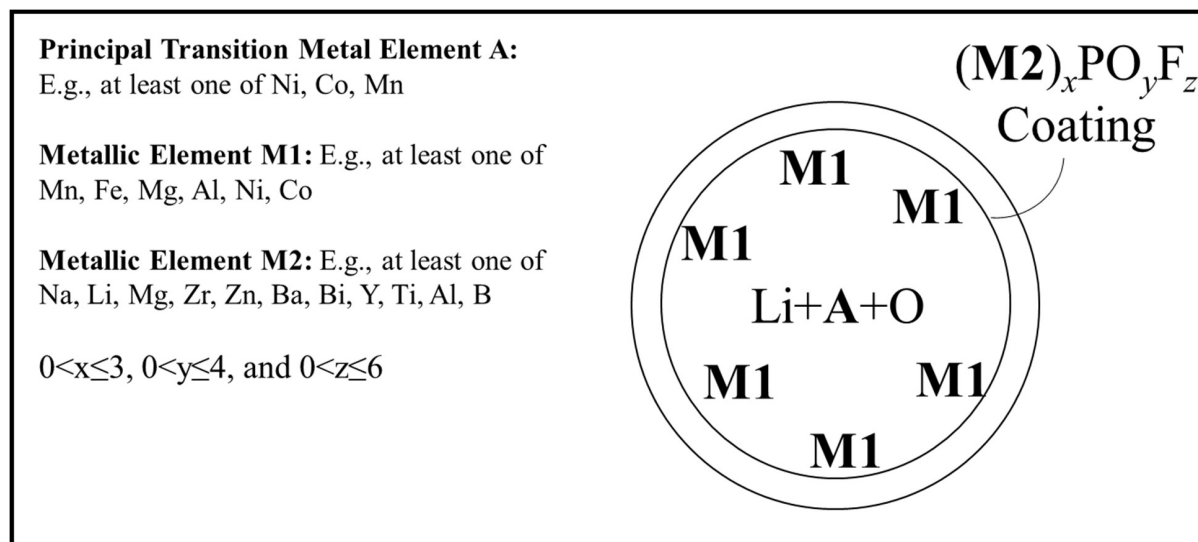
52. Like Shim, Lee never discloses PEAMs with crack portions or portions that include crystal defects. Lee merely describes the problem of “acid-containing electrolyte attack[ing] the surface of the cathode during cycling,” (*id.*, 4), which is a distinct issue that can lead to dissolution of transition metals and the formation of an unwanted interface layer that can deteriorate the electrochemical properties of a cathode.

C. Fujiki

53. I have been informed by Counsel and understand that Petitioner relies on Fujiki as the primary reference in Grounds 4–6 of the Petition to argue that the

invention of the '828 Patent would have been obvious to a POSITA. Fujiki is a patent reference describing a PEAM that includes (i) a “principal transition metal element A,” (ii) “at least one metallic element M1” that is different from principal transition metal element A and is “present at least at part of a surface of a lithium-containing transition metal oxide containing the principal transition metal element A,” and (iii) “a compound of a metallic element M2 [that is] different from the metallic element M1 [and] is present in a coating manner or in the vicinity of the lithium-containing transition metal oxide.” *See* Ex. 1008, Abstract. Fujiki says that the principal transition metal element A can be “at least one selected from the group consisting of at least Ni, Co and Mn.” *Id.*, 4: 60–61. Fujiki also states that the metallic element M1 can be “at least one element which can be substituted for the principal transition metal element A on a solid solution basis” such as “at least one element selected from the group consisting of Mn, Fe, Mg, Al, Ni, and Co.” *Id.*, 5:6–10. And finally, Fujiki states that the metallic element M2 can be “at least one element selected from the group consisting of Na, Li, Mg, Zr, Zn, Ba, Bi, Y, Ti, Al, and B,” which “has the function of stabilizing the crystal structure of the compound oxide particles at the time of charging.” *Id.*, 5:29–35. For example, Fujiki discloses that it is preferable that the metallic element M2 is “present in the state of a coating” and is “in the form of a metallic salt, a metallic oxide or a metallic halide” such as a metallic salt represented by the formula: $(M2)_xPO_yF_z$, where $0 < x \leq 3$, $0 < y \leq 4$, and $0 < z \leq 6$. *Id.*, 5:35–

61. In my opinion, the generalized cathode material that Fujiki discloses is accurately illustrated by the diagram below, which I have prepared with the assistance of Counsel as a visual aid for understanding.



54. Like Shim and Lee, Fujiki also fails to disclose PEAMs with crack portions or portions that include crystal defects. Although Fujiki mentions the “deterioration of a positive electrode active material” (Ex. 1008, 3:34–39) and the role of M2 in “stabilizing the crystal structure of the compound oxide particles at the time of charging” (*id.*, 5:32–35), none of these are equivalent to disclosing a crack portion of a PEAM particle or a portion that includes crystal defects.

VI. PRIOR ART ANALYSIS

A. Ground 1 is Deficient for Failing to Show that Shim Alone or in Combination with Lee Renders Obvious that “the Representative Element and the Fluorine are Present in a Crack Portion [or a Portion Which Includes Crystal Defects] Observed from a TEM Image”

55. In my opinion, a POSITA would not have found it obvious that “the representative element and the fluorine are present in a crack portion [or a portion which includes crystal defects] observed from a TEM image” based on the disclosure of Shim alone or in combination with Lee.

56. First, with respect to independent claim 1, Shim alone or in combination with Lee does not render obvious *fluorine* present in a crack portion observed from a TEM image. Nor does Shim alone or in combination with Lee render obvious *the representative element* (e.g., magnesium) being present in a crack portion observed from a TEM image. Moreover, with respect to independent claim 5, Shim alone or in combination with Lee also fails to render obvious the representative element and fluorine present *in a portion which includes crystal defects* observed from a TEM image.

1. The Petition Fails to Show that Shim Alone or in Combination with Lee Renders Obvious Fluorine Present in a Crack Portion Observed from a TEM Image (Claim 1)

a. Shim and Lee are Deficient

57. I understand that the Petition asserts that since Shim alone or in combination with Lee renders obvious a fluorine-containing coating, a POSITA would have “understood” that fluorine would likewise be present in crack portions. Pet., 55. I disagree.

58. In my opinion, Shim and Lee do not render obvious an applicable fluorine-containing coating in the first place. And even if they did, a POSITA would not have found it obvious for the fluorine from such a coating to be present in a crack portion. Petitioner’s discussion about elements or coating materials potentially becoming ingrained in a crack portion or a portion which includes crystal defects is not consistent with what the ’828 Patent describes when it mentions the representative element and the fluorine being present in said portions of the PEAM particle. Nevertheless, even under Petitioner’s proposed theory, Shim and Lee *still* do not render obvious fluorine present in a crack portion observed from a TEM image.

59. As I noted in Sections V.A and V.B above, Shim and Lee do not disclose crack portions whatsoever, and they certainly do not disclose or render obvious fluorine present in said crack portions. Shim instead focuses on other types of “degradation mechanisms,” such as “the dissolution of transition metal ions into the electrolytes,” or merely provides general descriptions that lack any reference to crack portions and crystal defects. Ex. 1005, 1. But not all degradation mechanisms for PEAMs include the formation of cracks. For example, in the context of surface

coatings for PEAMs, a common degradation mechanism that is addressed is the oxidative decomposition of the electrolyte, which leads to generation of acidic impurities on the PEAM particle and the dissolution of transition metals from the surface of the PEAM particles. This oxidative decomposition of the electrolyte may not include the formation of cracks whatsoever. Thus, since there are many modes of degradation associated with PEAM particles and since cracks are only a specific type of defect, Shim's general description of degradation and non-crack defects does not equate to the presence of crack portions in a PEAM particle.

60. Moreover, nothing in Shim identifies the structure shown in its Fig. 2(b) as being a "crack portion" (*see supra* ¶ 49); this is based on mere speculation by Petitioner and its declarant. In my opinion, a POSITA would not look at the alleged "crack portion" identified by Petitioner and be able to ascertain with any amount of certainty that Fig. 2(b) of Shim is a crack portion. In fact, based on my experience, the relevant region identified by Petitioner in Fig. 2(b) could correspond to a number of alternatives such as contaminants introduced during manufacturing of the PEAM or manufacturing impurities due to poor mixing of the starting materials. Lee also fails to remedy the deficiencies of Shim as it similarly never discloses crack portions and only vaguely describes the "dissolution of transition metals." Ex. 1006, 4.

61. In my opinion, the Petition does not identify *even a single prior art reference* that discloses or suggests the presence of fluorine in a crack portion. Therefore, I do

not believe a POSITA would have found independent claim 1 or independent claim 5 to be obvious given that key claim features are entirely absent from the teachings of the relied upon alleged prior art.

b. The Petition's Improper Reliance on Fey Fails to Cure the Deficiencies in Shim and Lee

62. It is my understanding that Petitioner attempts to rely on Fey (Ex. 1016, 4)—a reference that is not included in either Grounds 1 or 4—to address the deficiencies of Shim and Lee. *See, e.g.*, Pet., 25, 51, 54–55. In my opinion, however, a POSITA reading Fey in conjunction with Shim and Lee still would not find it obvious that fluorine would be present in crack portions of a PEAM particle.

63. Fey is a research paper that describes experiments performed on a cathode material comprising LiCoO_2 coated with MgAl_2O_4 —a mixed spinel oxide that does not include fluorine whatsoever—to examine the cathode's characteristics and cycling behavior. Ex. 1016, 1. In one portion of the study, cyclic voltammograms are produced and compared for (a) bare LiCoO_2 and (b) MgAl_2O_4 -coated LiCoO_2 . *Id.*, 4 and Fig. 5. By observing peaks in a cyclic voltammogram, one can identify the phase transitions of a material during cycling. As shown in Fig. 5 of Fey, the peaks “seem suppressed beyond the second sweep of the coated sample,” corresponding to suppressed phase transitions. *Id.*, 4.

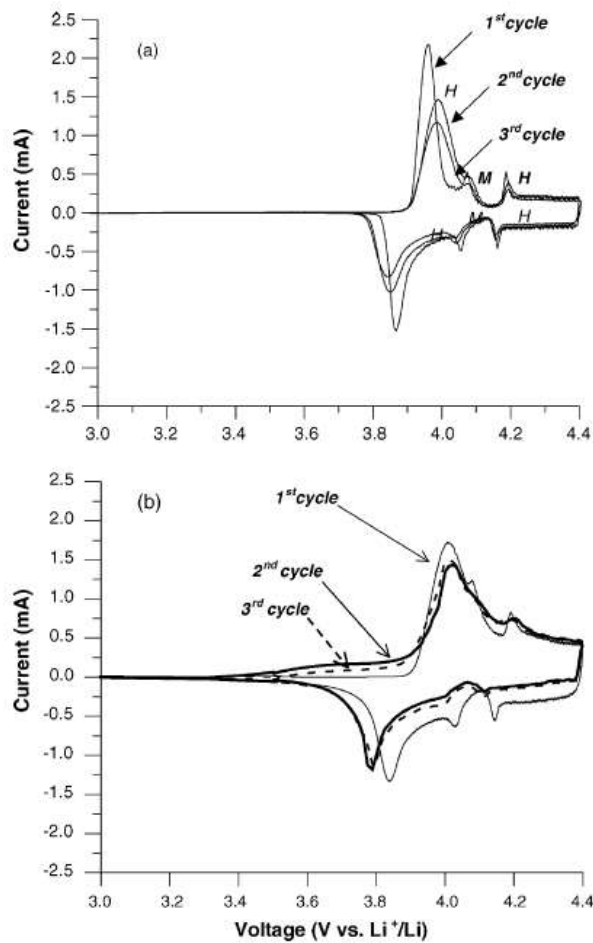


Fig. 5. Cyclic voltammograms of (a) bare LiCoO₂ and (b) 1.0 wt.% spinel-coated LiCoO₂.

Ex. 1016, Fig. 5

64. Fey's cyclic voltammograms do not provide any direct empirical evidence that MgAl₂O₄ coating material is present in a crack portion of the cathode material. However, in an effort to explain how the suppression of phase transitions occurs, the investigators in Fey speculate that an applied cycling process "could have" enabled particles of the MgAl₂O₄ coating material to "become ingrained in the crevices and cracks on the cathode surface." *Id.*, 4. Petitioner then relies on this statement

throughout the Petition to apparently support the more general notion that a POSITA would understand, in view of Fey's disclosure, that *any* coating material of a PEAM particle would be present in a crack portion of the PEAM particle observed from a TEM image.

65. In my opinion, Petitioner mischaracterizes what would be understood by a POSITA in view of Fey. Fey never discusses *fluorine* being present in a crack portion of a PEAM particle, nor does Fey address whether a fluorine-containing coating would behave similarly to a MgAl_2O_4 coating. Indeed, nothing in Fey even demonstrates that its own *MgAl₂O₄ coating* is present in a crack portion; Fey only provides a *hypothetical mechanism* that could explain the empirical results of Fey's cyclic voltammetric experiment. *Id.* (“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.”). Petitioner and Petitioner's expert appear to assume, without any justification, that a POSITA would “understand” that all coating layers would behave similarly—regardless of differences in their chemistries, coating processes, experimental conditions, etc. For example, the Petition relies on conclusory testimony from Dr. Hayes that “through the same mechanism [that Fey's MgAl_2O_4 coating might end up ingrained in cracks of the cathode material] . . . fluoride in the coating layer [of Shim's particle or

Shim's particle modified according to the disclosure of Lee] would also become present in the 'crack portion' of the PEAM particle" shown in Fig. 2(b) of Shim. Pet., 55; Ex. 1003, ¶¶ 164–65. In my opinion, this is an oversimplification of the technical field of battery materials.

66. In my opinion, a POSITA grasping the complexities and unpredictable nature of battery chemistry would have needed to know much more information before reasonably expecting that a fluorine-containing coating material would reasonably behave like Fey's MgAl_2O_4 coating material. Therefore, it would not have been obvious in view of Shim, Lee, and Fey that fluorine would be present in crack portions of a PEAM particle observed from a TEM image. The Petition's unreliable and oversimplified analysis is evident for several reasons.

67. **First**, the Petition fails to address complexities in the formation of fractures in PEAM particles that would affect whether a particular coating material becomes ingrained in crack portions. The composition of PEAM particles can substantially influence the formation of cracks in a cathode surface. *See* Ex. 2003, 13 (describing how the composition of electrode materials can affect the fracture of battery materials, for example, influencing the abundance and severity of cracks in the particle). The argument in the Petition that states that "Shim's PEAM degradation caused by cycling . . . refers to cracks" (Pet., 51 (citing Ex. 1005, 1)) is too simplistic, and I do not agree with it. Even if one were to consider the Petitioner's analysis, it

fails to account for the fact that the susceptibility of PEAMs to cracking is dependent on a range of factors including “[p]article size, material composition, pre-existing defects, state-of-charge heterogeneity, cycling time, cutoff voltage, [and] temperature.” Ex. 2003, 19 (annotated Table 1, reproduced below). As cracks become more or less abundant and/or severe in a PEAM particle, it is evident that the likelihood that a coating material would become ingrained in a crack portion would also change. Therefore, unlike Petitioner and Dr. Hayes, a POSITA would not automatically assume from Fey’s conjecture regarding MgAl_2O_4 possibly becoming ingrained in crevices and cracks of Fey’s cathode that fluorine-containing PEAM particles would behave similarly in Shim and Lee. Petitioner’s failure to recognize or address any of these differences amounts to a significant mischaracterization of what would be understood or found obvious by a POSITA.

Table 1. Comparisons of the fracture mechanism, fracture effects, and factors affecting the fracture for anodes, cathodes, and SSEs.					
	Anodes			Cathodes	SSEs
	Alloying	Intercalation (graphite)	Conversion	Intercalation	NASICONs, garnets, sulfides
Fracture mechanism	Two-phase-coherency stress, anisotropic reaction kinetics	Stress induced by volume change, gas formation, cointercalation of solvated ion	Structure reorganization and volume change	Concentration gradient stress and two-phase-coherency stress	Stress induced by growth of interphase or volume expansion of active material
Fracture effect	Specific area of active material increases, impeded diffusion of ions and electrons	Exfoliation, impedance increase, capacity fade	Structure pulverization, decayed capacity, inferior cycling stability.	Resistance increase, capacity fading	Porosity increases, impedance increases, overpotential increase
Factors affecting fracture	Size, morphology, crystallinity, C-rate	Mechanical properties, Solvent type	Morphology, material composition, structure	Particle size, material composition, pre-existing defects, state-of-charge heterogeneity, cycling time, cutoff voltage, temperature	Mechanical properties, crystallinity, defects

Ex. 2003, Table 1 (annotated)

68. **Second**, a POSITA would have known that different coating materials can have meaningfully different properties that would impact their ability to become ingrained in crack portions. *See e.g.*, Ex. 2004, 2–5 (describing many coating materials for cathode materials, including their different properties, different coating processes, and mechanisms of action); Ex. 2005, Table 2 (showing that even coatings made of the same elements in different ratios such as $\text{Li}_4\text{Ti}_5\text{O}_{12}$ and $\text{Li}_2\text{Ti}_2\text{O}_5$ can have different properties); Ex. 1005, 8 (showing, once again, that coatings made of the same elements in different ratios such as Li_2TiO_3 and $\text{Li}_4\text{Ti}_5\text{O}_{12}$ can have different properties, with $\text{Li}_4\text{Ti}_5\text{O}_{12}$ showing “much better electrochemical

performances as a coating material” due to “its excellent stability during charging/discharging”); Ex. 2006, Table 1 (showing different properties between fluoride coatings such as AlF_3 and oxide coatings such as Al_2O_3); Ex. 2007, 1–2 (demonstrating different properties for metal oxide and metal fluoride coatings and noting that “comparison of different coating chemistries on LCO across literature is not quite straight forward due to variations in parent material preparation method, coating method, coating thickness, differences in particle size, differences in rates tested, etc.”). Given these many known differences between coatings of different chemical compositions, a POSITA would not expect a fluorine-containing coating material to behave similarly to coatings having different chemistries such as Fey’s MgAl_2O_4 coating. Indeed, by nature of their differences in even basic chemical properties such as electronegativity profiles, a POSITA would expect these coatings to react and move differently in identical settings.

69. **Third**, the Petition overlooks how differences in coating manufacturing processes can also result in coatings with different properties that would impact their ability to become ingrained in crack portions in a PEAM particle. *See, e.g.*, Ex. 2008, 5–6 (comparing solid state, co-precipitation, sol-gel, hydrothermal, combustion, and spray-drying methods of synthesizing electrode materials and noting that “[d]ifferent synthesis methods have important effects on the morphology and structure properties of final products.”); Ex. 2009, 1 (proving that “different coating protocols have

critical influences on the atomic and chemical structures of surface alumina coatings as well as the electrochemical performance of the coated cathodes”); Ex. 2010, 1 (demonstrating that even cathodes coated with similar MgO coatings and prepared using similar pulsed laser deposition techniques yielded different morphology effects depending on the temperature at which coating occurs). Even at a basic level, different coating methods that result in various coating thicknesses can affect the ability of such coatings to become ingrained in cracks and crevices in the way that Petitioner alleges. Thus, given the significant influence that coating manufacturing processes can have on coating properties, a POSITA would not reasonably expect Shim’s sol-gel-formed coating (Ex. 1005, 2) or Lee’s spin-coating-formed coating (Ex. 1006, 1) to behave similarly in allegedly becoming ingrained in cracks/crevices to Fey’s “semi-alkoxide sol-gel”-formed coating despite having many manufacturing differences such as heating temperatures and times (Ex. 1016, 1–2).

c. The Petition Fails to Demonstrate Inherency, or Even that the Fluorine Present in a Crack Portion Would be Predictable in any PEAM Particle Based on Shim and Lee

70. To the extent that Petitioner appears to argue that a POSITA would have understood that, in view of Fey, a coating material such as the coatings of Shim and Lee’s particles would *inherently* or *necessarily* be present in a crack portion of a PEAM particle observed from a TEM image, I disagree with this characterization as

well. Fey does not say that particles of the coating material are always or *necessarily* present in the crevices and cracks of a cathode material. Nor is the statement in Fey a general statement that *any coating material*, deposited using *any process* must necessarily be present in the crevices and cracks. On the contrary, as I argued above (*see supra* Section VI.A.1.b), a POSITA would have understood that the fracture behavior in PEAMs is complex and can be affected by a range of factors including “[p]article size, material composition, pre-existing defects, state-of-charge heterogeneity, cycling time, cutoff voltage, [and] temperature.” Ex. 2003, 19. Accordingly, a POSITA would not have understood Fey’s speculation about fracture behavior in a non-fluorine-containing MgAl_2O_4 -coated PEAM particle (subject to very specific manufacturing processes and experimental conditions) to *necessarily* generalize to fluorine-containing PEAM particles and result in the presence of fluorine in a crack portion.

71. Indeed, a POSITA would not have even found it predictable that a fluorine-containing coating material would end up in a crack portion of a PEAM particle for similar reasons as I discuss above in Section VI.A.1.b.

2. The Petition Fails to Show that Shim Alone or in Combination with Lee Renders Obvious a Representative Element Present in a Crack Portion Observed from a TEM Image (Claim 1)

72. I understand that the Petition identifies magnesium as an example of the claimed “representative element” and offers two theories for why Shim discloses or renders obvious that the magnesium would be present in a crack portion of Shim’s particle.

73. Under the *first theory*, Petitioner refers to Shim’s alleged disclosure of magnesium diffusing into a coating of the PEAM particle and argues that a POSITA would have understood that “after having diffused into the coating,” magnesium would “become ingrained into the crack portions of Shim’s [PEAM] particle.” Pet., 53–54. Relatedly, Petitioner argues that if Shim’s particle were combined with Lee’s MgF₂ coating, then the magnesium in this coating would also be found in the crack portion. *Id.*, 55.

74. I disagree for similar reasons that I provided for why a POSITA would not have found it obvious that fluorine would be present in a crack portion (*see supra* Section VI.A.1). For example, neither Shim nor Lee disclose crack portions whatsoever, and therefore they cannot possibly disclose the representative element (e.g., magnesium) being present in said crack portions. Again, Shim and Lee both focus solely on other types of “degradation mechanisms” and provide only general

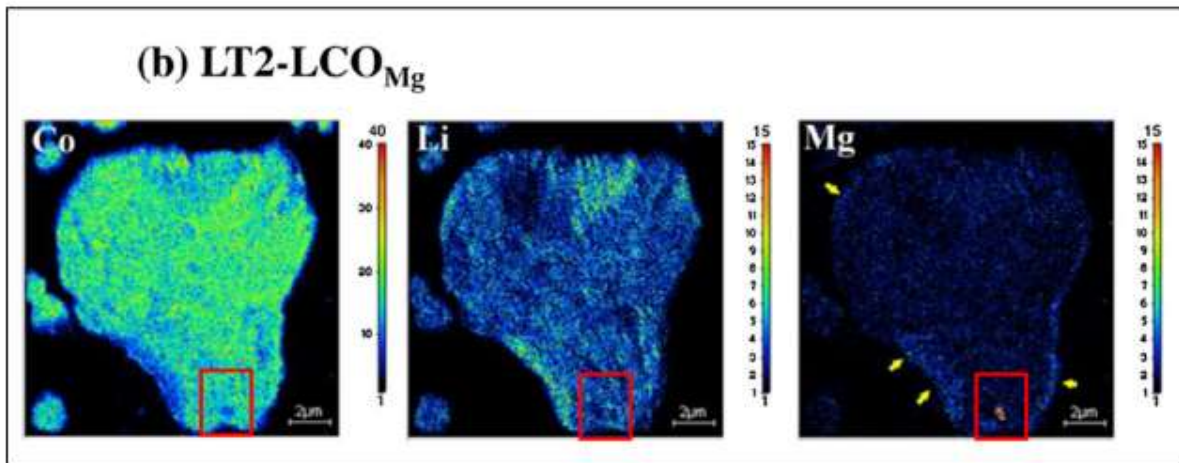
descriptions that lack any references to crack portions or portions which include crystal defects. *See* Ex. 1005, 1; Ex. 1006, 4.

75. I understand that the Petition identifies Fig. 2(b) of Shim as allegedly showing magnesium ingrained in a crack portion. Pet., 54–55. In Petitioner’s annotated version of the figure, reproduced below, Petitioner refers to the portion of Shim’s PEAM particle indicated by the red box as a “crack portion” and notes the presence of magnesium in this portion of the PEAM particle. *Id.* However, Shim never identifies the relevant region as a crack portion, and in fact, does not mention the presence of any cracks in the PEAM particle whatsoever. Without such disclosure, it is my opinion that a POSITA looking at Fig. 2(b) of Shim would not be able say with any reasonable level of confidence that the region identified by the Petitioner is a crack portion. In fact, a POSITA would recognize that the annotated region could just as well be a manufacturing impurity caused by poor mixing of the starting materials, a contaminant introduced during the manufacturing process, or some other feature.

76. I understand that Petitioner argues that the identified region of Shim’s particle represents coating material ingrained in a crack portion as a result of the process conjectured in Fey. But I find this implausible. If the high concentration of Mg found in the identified region of Shim originated from the alleged coating material, then one would expect the region to have a similar concentration of Mg as the alleged

coating material on the PEAM particle's surface. But the concentration of Mg in the identified region is *many multiples higher* than the concentration of Mg at the particle surface, and Petitioner offers no explanation for this discrepancy. In addition, Fey hypothesizes that the embedding of coating material occurs "with the application of a[n] [electrochemical] load or upon thermal cycling," (Ex. 1016, 4), while Shim does not disclose subjecting its PEAM particles to cycling whatsoever. Thus, since Shim presumably generated the nano SIMS image of Fig. 2(b) using manufactured particles with no cycling history, this would have left no opportunity for the speculated process proposed by Fey to even occur.

77. Moreover, I understand that Fig. 2(b) of Shim was imaged using nano SIMS (a technique used to measure elemental and isotopic compositions of a sample rather than structural features) and thus, at the very least, these images do not represent "a crack portion observed from a TEM image." Thus, in my opinion, a POSITA would not have found "the representative element . . . present in a crack portion observed from a TEM image" to be obvious in view of Shim alone or in combination with Lee.



Ex. 1005, Fig. 2(b) (annotated by Petitioner)

78. In my opinion, the Petitioner's reliance on Fey is also insufficient to cure the deficiencies in Shim and Lee. As I noted above in Section VI.A.1.b, Petitioner does not provide any evidence to support why a POSITA would expect the diffused magnesium in Shim or the MgF₂ coating in Lee to behave similarly to other coating materials such as Fey's MgAl₂O₄ coating by becoming ingrained or lodged in a crack portion. Thus, as I argued in Section VI.A.1.b, the Petition ignores the complexities of how fractures form in PEAM particles; it ignores the fact that coatings with differing chemical compositions can have different properties; and it ignores the fact that different coating manufacturing processes can yield coatings with different properties—all of which can affect the likelihood that a particular coating material becomes ingrained in a crack portion. Petitioner's failure to recognize these nuances

is a significant oversimplification of the technical field and therefore an inaccurate representation of what would be understood by a POSITA.

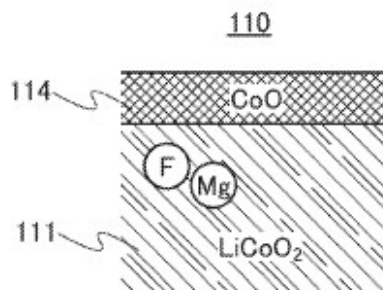
79. In my opinion, the Petition additionally fails to demonstrate that any element in the coating material of a PEAM would *inherently* be present in a crack portion (e.g., by inherently becoming ingrained in crevices and cracks on the cathode surface). Just as with fluorine in Section VI.A.1.c, Fey's limited disclosure is far from sufficient to establish that magnesium would be necessarily present in the alleged crack portions of Shim's particle. Fey does not say that particles of the coating material are always or *necessarily* present in the crevices and cracks of a cathode material. Nor does Fey provide a general statement that *any coating material* (including non-MgAl₂O₄ coatings), deposited using *any process* must necessarily be present in the crevices and cracks of a PEAM particle. Instead, a POSITA would understand that coatings with different chemistries frequently result in different fracture behavior that would affect the likelihood that coating materials become ingrained in a crack portion. *See* Ex. 2003, 13. Indeed, the fracture behavior in PEAM particles is complex and can be affected by several factors. *Id.*, 19 (annotated Table 1). Therefore, a POSITA would not understand that Fey's conjecture regarding fracture behavior in a MgAl₂O₄-coated PEAM particle (subject to very specific manufacturing processes and experimental conditions) would necessarily generalize to PEAM particles with other magnesium-based coatings

(e.g., Shim's Mg diffused dopant or Lee's MgF₂ coating), resulting in the presence of magnesium in a crack portion.

80. As I understand it, under Petitioner's *second theory* for why Shim discloses or renders obvious that the magnesium is present in a crack portion, Petitioner argues that since Shim discloses doping the bulk of the PEAM particle with magnesium, any crack portions of Shim's particle would necessarily contain magnesium. Pet., 53. Petitioner argues that since Shim discloses doping the bulk of the PEAM particle with magnesium, magnesium must therefore be "present throughout the entire particle," indicating that "crack portions of Shim's PEAM particle . . . necessarily contain magnesium." (Ex. 1003, ¶¶ 160–61; Pet., 53.) However, I emphasize again that Shim does not even *mention* crack portions in its PEAM particle, and Shim certainly does not disclose crack portions *observed from a TEM image*.

81. Moreover, the Petition does not explain how the doping of the bulk of a PEAM particle would render its presence in a crack portion observed from a TEM image obvious in the context of the '828 Patent. The '828 Patent describes that a particle 110 can include magnesium and fluorine throughout an internal region 111. Ex. 1001, 22:9–16, Fig. 5A (reproduced below).

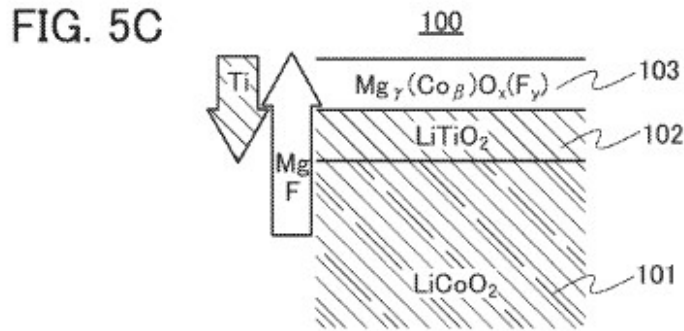
FIG. 5A



Ex. 1001, Fig. 5A

However, the '828 Patent does **not** consider this distribution of magnesium and fluorine throughout the particle 110 to constitute the presence of magnesium or fluorine in a crack portion. Instead, the '828 Patent discloses that the representative element (in this example, magnesium) is “in the vicinity of crystal defects” (*id.*, 24:11–23) or “in the crack portion” (*id.*, 24:24–27) ***after the magnesium (and fluorine) is segregated through heating*** (*id.*). See also *id.*, 22:9–54; *id.*, FIG. 5C (reproduced below) (showing how the third region 103 is formed on a surface region of the particle 110 through the segregation of the magnesium and fluorine by heating at 800°C for two hours); *id.*, 9:47–60 (stating that “the third region 103 may be present in a portion which includes crystal defects or a crack portion in the positive electrode active material 100”). Accordingly, Petitioner’s theory that the mere doping of magnesium in the bulk of the PEAM particle would make it present in a crack portion is wrong and inconsistent with the disclosure of the '828 Patent. A POSITA would not have understood the mere doping of the bulk of the PEAM

particle with magnesium as rendering it present in a crack portion observed from a TEM image.



Ex. 1001, Fig. 5C

3. The Petition Fails to Show that Shim Alone or in Combination with Lee Renders Obvious a Representative Element and Fluorine Present in a Portion Which Includes Crystal Defects Observed from a TEM Image (Claim 5)

82. I understand that while independent claim 1 of the '828 Patent recites that the representative element and the fluorine are present in a “crack portion,” independent claim 5 recites that the representative element and the fluorine are present in a “portion which includes crystal defects.”

83. To address the “portion which includes crystal defects” recited in independent claim 5, I understand that Petitioner argues (i) that “cracks are a type of crystal defect,” (ii) that a crack portion “necessarily includes crystal defects,” and (iii) that Shim’s disclosure of oxygen loss from the lattice structure and reactions between the lattice and the electrolytes are examples of crystal defects. Pet., 57–59. Petitioner

does not provide any separate reasons that the claimed “representative element” or “fluorine” would be present in a portion which include crystal defects *as distinct* from being present in a crack portion.

84. For the purpose of this analysis, Counsel has instructed that I adopt and apply (without conceding agreement to) the interpretations of “crack portion” and “a portion which includes crystal defects,” which Petitioner applies in the Petition. I have done so. Nevertheless, even assuming for the purpose of this analysis—as Petitioner asserts—that cracks and crystal defects can be conflated, Ground 1 is deficient with respect to independent claim 5 for at least the same reasons that Shim alone or in combination with Lee does not render obvious that the representative element and the fluorine are present in a crack portion (as recited in claim 1). *See supra* Sections VI.A.1 and VI.A.2.

B. Ground 4 is Deficient for Failing to Show that Fujiki Renders Obvious that “the Representative Element and the Fluorine are Present in a Crack Portion [or a Portion Which Includes Crystal Defects] Observed from a TEM Image”

85. In my opinion, Ground 4 suffers many of the same deficiencies as those discussed above in relation to Ground 1. First, for example, and with respect to independent claim 1, the Petition fails to show that Fujiki renders obvious *fluorine* present in a crack portion observed from a TEM image. Second, with respect to claim 1, the Petition fails to show that Fujiki renders obvious *the representative element*

(e.g., magnesium) present in a crack portion observed from a TEM image. Third, with respect to independent claim 5, the Petition fails to show that Fujiki renders obvious the representative element and fluorine present *in a portion which includes crystal defects* observed from a TEM image.

**1. The Petition Fails to Show that Fujiki Renders Obvious
Fluorine Present in a Crack Portion Observed from a TEM
Image (Claim 1)**

a. Fujiki is Deficient

86. I understand that the Petition asserts that since Fujiki renders obvious a fluorine-containing coating, a POSITA would have understood that fluorine would likewise be found in crack portions. Pet., 76. I disagree.

87. In my opinion, and as noted above in Section V.C, Fujiki never discusses a crack or a crack portion—let alone fluorine being present in a crack portion—and the Petition’s attempts to rectify these deficiencies are insufficient for the same reasons discussed above in relation to Ground 1. A POSITA simply would not have found independent claim 1 or independent claim 5 to be obvious given that key claim features are entirely absent from the teachings of the relied upon alleged prior art.

**b. The Petition’s Improper Reliance on Fey Fails
to Cure the Deficiencies in Fujiki**

88. It is my understanding that just as in Ground 1, Petitioner attempts to rely on Fey (Ex. 1016, 4)—a reference that is not included in either Grounds 1 or 4—to

address the deficiencies of Fujiki. *See* Pet., 76. But, in my opinion, a POSITA reading Fey in conjunction with Fujiki still would not find it obvious that fluorine would be present in crack portions of a PEAM particle.

89. As I described above, Fey is a research paper that describes experiments performed on a cathode material comprising LiCoO_2 coated with MgAl_2O_4 —a mixed spinel oxide that doesn’t include fluorine whatsoever—to examine the cathode’s characteristics and cycling behavior. Ex. 1016, 1. In one portion of the study, cyclic voltammograms are produced and compared for (a) bare LiCoO_2 and (b) MgAl_2O_4 -coated LiCoO_2 . *Id.*, 4 and Fig. 5. By observing peaks in a cyclic voltammogram, one can identify the phase transitions of a material during cycling. As shown in Fig. 5 of Fey, the peaks “seem suppressed beyond the second sweep of the coated sample,” corresponding to suppressed phase transitions. *Id.*, 4.

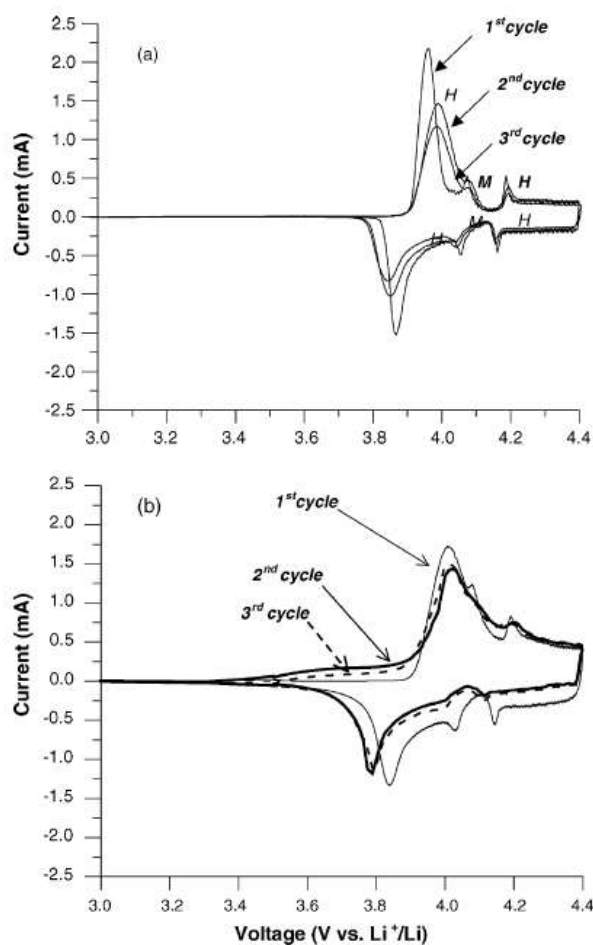


Fig. 5. Cyclic voltammograms of (a) bare LiCoO₂ and (b) 1.0 wt.% spinel-coated LiCoO₂.

Ex. 1016, Fig. 5

90. Fey's cyclic voltammograms do not provide any direct empirical evidence that MgAl₂O₄ coating material is present in a crack portion of the cathode material. However, in an effort to explain how the suppression of phase transitions occurs, the investigators in Fey speculate that an applied cycling process "could have" enabled particles of the MgAl₂O₄ coating material to "become ingrained in the crevices and cracks on the cathode surface." *Id.*, 4. Petitioner then relies on this statement

throughout the Petition to apparently support the more general notion that a POSITA would understand, in view of Fey's disclosure, that *any* coating material of a PEAM particle would be present in a crack portion of the PEAM particle observed from a TEM image.

91. In my opinion, Petitioner mischaracterizes what would be understood by a POSITA in view of Fey. Fey never discusses *fluorine* being present in a crack portion of a PEAM particle, nor does Fey address whether a fluorine-containing coating would behave similarly to a MgAl_2O_4 coating. Indeed, nothing in Fey even demonstrates that its own *MgAl₂O₄ coating* is present in a crack portion; Fey only provides a *hypothetical mechanism* that could explain the empirical results of Fey's cyclic voltammetric experiment. *Id.* (“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.”). Despite the gaps in Fey, the Petition nonetheless relies on testimony from Dr. Hayes that extrapolates from the comments made in Fey and the alleged crack portion shown in Fig. 2(b) of Shim to assert that “fluorine in Fujiki's coating would be present in the crack portions because coating materials were known to become lodged inside such cracks.” Pet., 76; Ex. 1003, ¶ 238. Petitioner and Petitioner's expert thus appear to assume, without any justification, that a POSITA would “understand” that all

coating layers would behave similarly—regardless of differences in their chemistries, coating processes, experimental conditions, etc. In my opinion, this is an oversimplification of the technical field of battery materials.

92. In my opinion, a POSITA grasping the complexities and unpredictable nature of battery chemistry would have needed to know much more information before reasonably expecting that a fluorine-containing coating material would reasonably behave like Fey's MgAl_2O_4 coating material. Therefore, it would not have been obvious in view of Fujiki and Fey that fluorine would be present in crack portions of a PEAM particle observed from a TEM image. The Petition's unreliable and oversimplified analysis is evident for several reasons.

93. **First**, the Petition fails to address complexities in the formation of fractures in PEAM particles that would affect whether a particular coating material becomes ingrained in crack portions. The composition of PEAM particles can substantially influence the formation of cracks in a cathode surface. *See* Ex. 2003, 13 (describing how the composition of electrode materials can affect the fracture of battery materials, for example, influencing the abundance and severity of cracks in the particle). The argument in the Petition that states that "PEAM degradation caused by cycling . . . refers to cracks" (Pet., 51 (citing Ex. 1005, 1)) is too simplistic, and I do not agree with it. Even if one were to consider the Petitioner's analysis, it fails to account for the fact that the susceptibility of PEAMs to cracking is dependent on

a range of factors including “[p]article size, material composition, pre-existing defects, state-of-charge heterogeneity, cycling time, cutoff voltage, [and] temperature.” Ex. 2003, 19 (annotated Table 1). As cracks become more or less abundant and/or severe in a PEAM particle, it is evident that the likelihood that a coating material would become ingrained in a crack portion would also change. Therefore, unlike Petitioner and Dr. Hayes, a POSITA would not automatically assume from Fey’s conjecture regarding MgAl_2O_4 possibly becoming ingrained in crevices and cracks of Fey’s cathode that fluorine-containing PEAM particles would behave similarly in Fujiki. Petitioner’s failure to recognize or address any of these differences amounts to a significant mischaracterization of what would be understood or found obvious by a POSITA.

94. ***Second***, a POSITA would have known that different coating materials can have meaningfully different properties that would impact their ability to become ingrained in crack portions. *See e.g.*, Ex. 2004, 2–5 (describing many coating materials for cathode materials, including their different properties, different coating processes, and mechanisms of action); Ex. 2005, Table 2 (showing that even coatings made of the same elements in different ratios such as $\text{Li}_4\text{Ti}_5\text{O}_{12}$ and $\text{Li}_2\text{Ti}_2\text{O}_5$ can have different properties); Ex. 1005, 8 (showing, once again, that coatings made of the same elements in different ratios such as Li_2TiO_3 and $\text{Li}_4\text{Ti}_5\text{O}_{12}$ can have different properties, with $\text{Li}_4\text{Ti}_5\text{O}_{12}$ showing “much better electrochemical

performances as a coating material” due to “its excellent stability during charging/discharging”); Ex. 2006, Table 1 (showing different properties between fluoride coatings such as AlF_3 and oxide coatings such as Al_2O_3); Ex. 2007, 1–2 (demonstrating different properties for metal oxide and metal fluoride coatings and noting that “comparison of different coating chemistries on LCO across literature is not quite straight forward due to variations in parent material preparation method, coating method, coating thickness, differences in particle size, differences in rates tested, etc.”). Given these many known differences between coatings of different chemical compositions, a POSITA would not expect a fluorine-containing coating material to behave similarly to coatings having different chemistries such as Fey’s MgAl_2O_4 coating. Indeed, by nature of their differences in even basic chemical properties such as electron affinity, electronegativity profile, as well as modes of bonding (e.g., oxygen is a divalent bonding element while halides such as fluorine are monovalent bonding elements), a POSITA would expect these coatings to react and move differently in identical settings.

c. The Petition Fails to Demonstrate Inherency, or Even that the Fluorine Present in a Crack Portion Would be Predictable in any PEAM Particle Based on Fujiki

95. To the extent that Petitioner appears to argue that a POSITA would have understood that, in view of Fey, a coating material such as the coatings of Fujiki’s

particles would inherently or necessarily be present in a crack portion of a PEAM particle observed from a TEM image, I disagree with this characterization as well.

96. For similar reasons as discussed above (*see supra* Section VI.B.1.b), Fey's limited disclosure is far from sufficient to establish that fluorine would be necessarily present in crack portions of Fujiki's particle (which Petitioner assumes exist). Fey does not say that particles of the coating material are always or *necessarily* present in the crevices and cracks of a cathode material. Nor is the statement in Fey a general statement that *any coating material*, deposited using *any process* must necessarily be present in the crevices and cracks. On the contrary, as I argued above, a POSITA would have understood that the fracture behavior in PEAMs is complex and can be affected by a range of factors including "[p]article size, material composition, pre-existing defects, state-of-charge heterogeneity, cycling time, cutoff voltage, [and] temperature." Ex. 2003, 19. Accordingly, a POSITA would not have understood Fey's speculation about fracture behavior in a non-fluorine-containing MgAl_2O_4 -coated PEAM particle (subject to very specific manufacturing processes and experimental conditions) to *necessarily* generalize to fluorine-containing PEAM particles and result in the presence of fluorine in a crack portion.

97. Indeed, a POSITA would not have even found it predictable that a fluorine-containing coating material would end up in a crack portion of a PEAM particle for similar reasons as I discuss above in Section VI.B.1.b.

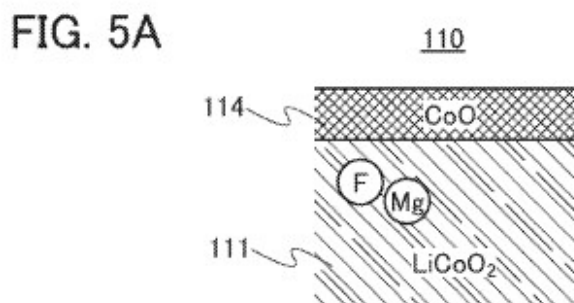
2. The Petition Fails to Show that Fujiki Renders Obvious a Representative Element Present in a Crack Portion Observed from a TEM Image (Claim 1)

98. I understand that, with respect to the claimed “representative element,” Petitioner uses magnesium as an example of the representative element and points out that Fujiki discloses that “metallic element M1” can be magnesium and is “present throughout the particle—from the surface to the interior.” Pet., 75. Petitioner then argues that based on the distribution of magnesium throughout Fujiki’s PEAM particle, a POSITA would understand that crack portions of Fujiki’s PEAM particle would contain magnesium. *Id.*, 76.

99. In my opinion, however, Fujiki does not mention crack portions in its PEAM particle whatsoever (*see supra* Section V.C), and Fujiki certainly does not disclose crack portions observed from a TEM image.

100. It is also my opinion that, contrary to Petitioner’s arguments, a POSITA would not find that the mere diffusion of magnesium throughout Fujiki’s PEAM particle would—in the context of the ’828 Patent—render obvious the presence of magnesium in a crack portion observed from a TEM image. Similarly to Fujiki, the

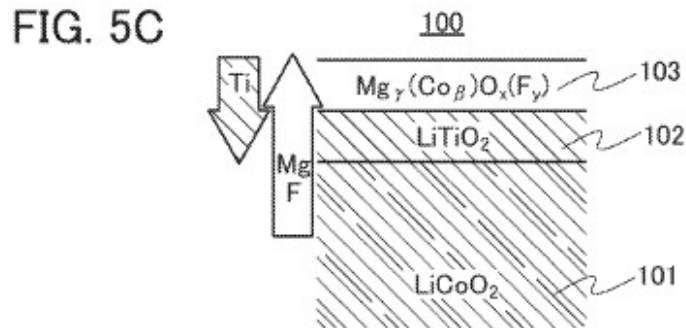
'828 Patent describes that a particle 110 can include magnesium and fluorine throughout an internal region 111. Ex. 1001, 22:9–16, Fig. 5A (reproduced below).



Ex. 1001, Fig. 5A

However, the '828 Patent does *not* consider this distribution of magnesium and fluorine throughout the particle 110 to constitute the presence of magnesium or fluorine in a crack portion. Rather, the '828 Patent discloses that the representative element (in this example, magnesium) is “in the vicinity of crystal defects” (*id.*, 24:11–23) or “in the crack portion” (*id.*, 24:24–27) ***after the magnesium (and fluorine) is segregated through heating*** (*id.*). See also *id.*, 22:9–54; *id.*, FIG. 5C (reproduced below) (showing how the third region 103 is formed on a surface region of the particle 110 through the segregation of the magnesium and fluorine by heating at 800°C for two hours); *id.*, 9:47–60 (stating that “the third region 103 may be present in a portion which includes crystal defects or a crack portion in the positive electrode active material 100”). Accordingly, Petitioner’s theory that Fujiki’s disclosed distribution of magnesium throughout the particle would make it present

in a crack portion is inconsistent with the disclosure of the '828 Patent. A POSITA would not have understood this distribution of magnesium as rendering magnesium present in a crack portion observed from a TEM image.



Ex. 1001, Fig. 5C

3. The Petition Fails to Show that Fujiki Renders Obvious a Representative Element and Fluorine Present in a Portion Which Includes Crystal Defects Observed from a TEM Image (Claim 5)

101. As discussed above in Section VI.A.3, I understand that while independent claim 1 of the '828 Patent recites that the representative element and the fluorine are present in a “crack portion,” independent claim 5 recites that the representative element and the fluorine are present in a “portion which includes crystal defects.”

102. To address the “portion which includes crystal defects” recited in independent claim 5, I understand that Petitioner argues (i) that a crack portion is simply “an example of a crystal defect” and (ii) that since Fujiki renders obvious a crack portion

observed from a TEM image, it likewise renders obvious a crystal defect observed from a TEM image. Pet., 78–79. I understand that Petitioner also argues that since Fujiki discloses that metallic element M2 has the function of stabilizing the crystal structure of the compound oxide particles at the time of charging, Fujiki “recognized that defects occur in the crystal structure of LiCoO_2 .” *Id.*, 79. However, just as in Ground 1, Petitioner does not provide any separate reasons that the claimed “representative element” or “fluorine” would be present in a portion which include crystal defects *as distinct* from being present in a crack portion.

103. For the purpose of this analysis, Counsel has instructed that I adopt and apply (without conceding agreement to) the interpretations of “crack portion” and “a portion which includes crystal defects,” which Petitioner applies in the Petition. I have done so. Nevertheless, even assuming for the purpose of this analysis—as Petitioner asserts—that cracks and crystal defects can be conflated, Ground 4 is deficient with respect to independent claim 5 for at least the same reasons that Fujiki does not render obvious that the representative element and the fluorine are present in a crack portion (as recited in claim 1). *See supra* Sections VI.B.1 and VI.B.2.

VII. CONCLUSION

104. I reserve the right to supplement my opinions to address any information obtained, or positions taken, based on any new information introduced throughout this proceeding.

105. I declare under penalty of perjury that the foregoing is true and accurate to the best of my ability.

A handwritten signature in black ink, appearing to be 'B. J.', written in a cursive style.

Date: 9/15/2023