

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent of: Ochiai et al.

U.S. Patent No.: 11,043,660

Attorney Docket No.: 12732-2367IP1

Issue Date: June 22, 2021

Appl. Serial No.: 16/923,494

Filing Date: July 8, 2020

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.

Date: 12/19/2023

By: /Brett L. Lucht/
Brett L. Lucht, Ph.D.

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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 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 15,000 times, and I have an h index of 68. 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. 11,043,660 (“the ’660 Patent”) (Ex. 1001) and alleged prior art references relating to its subject matter. I have reviewed the ’660 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. 11,043,660 (Ex. 1003);
- *Curriculum Vitae* of Dr. Troy Hayes (Ex. 1004);
- U.S. Patent App. Pub. No. US 2014/0212759 (“Blangero”) (Ex. 1005);
- U.S. Patent App. Pub. No. US 2016/0149215 (“Shim”) (Ex. 1006);
- Yamamoto et al., *Stabilization of the Electronic Structure at the Cathode/Electrolyte Interface via MgO Ultra-thin Layer during Lithium-ions Insertion/Extraction*, 82 *Electrochemistry*, 891-896 (2014) (“Yamamoto”) (Ex. 1007);
- U.S. Patent No. 10,283,770 (“Cho”) (Ex. 1008);
- Kim et al., *A New Coating Method for Alleviating Surface Degradation of $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ Cathode Material: Nanoscale Surface Treatment of Primary Particles*, 15 *Nano Letters*, 2111-2119 (“Kim”) (Ex. 1009);

- Glinski, Marek et al., *Catalytic Activity of High-Surface-Area Amorphous MgO Obtained from Upsalite*, Catalysts, MDPI (2021) (Ex. 2003);
- Zhang et al., *Oxygen Loss in Layered Oxide Cathodes for Li-Ion Batteries: Mechanisms, Effects, and Mitigation*, 122 ACS Chemical Reviews, 5641-5681 (2021) (Ex. 2004);
- He et al., *The intrinsic behavior of lithium fluoride in solid electrolyte interphases on lithium*, 117 PNAS, 73-79 (Jan. 7, 2020) (Ex. 2005);
- Xu et al., *Lithium Transport Through Lithium-Ion Battery Cathode Coatings*, J. Mater. Chem. A. 3, 17248–72 (2015) (Ex. 2006);
- Chauque et al., *Lithium titanate as anode material for lithium ion batteries: Synthesis, posttreatment and its electrochemical response*, J. of Electroanalytical Chem., 799 (2017) 142-155 (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);
- Zhu et al., *Epitaxial growth of an atom-thin layer on a $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ cathode for stable Li-ion battery cycling*, 13 Nat. Commun., 1565 (2022) (Ex. 2009);

- Pagani et al., *Epitaxial Thin Films as a Model System for Li-Ion Conductivity in $\text{Li}_4\text{Ti}_5\text{O}_{12}$* , 10, ACS Appl. Mater. Interfaces, 44494-44500 (2018) (Ex. 2010);
- WO 2010/139404 (“Ellenwood”) (Ex. 2011);
- Lettieri et al., *Critical issues in the heteroepitaxial growth of alkaline-earth oxides on silicon*, 20 J. Vac. Sci. Technol. A, 1332-1340 (2002) (Ex. 2012);
- Shim et al., *Effects of MgO Coating on the Structural and Electrochemical Characteristics of LiCoO_2 as Cathode Materials for Lithium Ion Battery*, 26 Chem. Mater., 2537-2543 (2014) (“Shim 3”) (Ex. 2013);
- U.S. Patent No. 6,514,640 (Ex. 2014);
- U.S. Patent No. 9,871,246 (Ex. 2015);
- Guo et al., *Ultra-low voltage control of magnetic properties in amorphous MgO*, 111 App. Phys. Lett. 192402 (2017) (Ex. 2017);
- Shim et al., *Characterization of Spinel $\text{Li}_x\text{Co}_2\text{O}_4$ -Coated LiCoO_2 Prepared with Post-Thermal Treatment as a Cathode Material for Lithium Ion Batteries*, 27 Chem. Mater. 27, 3273-3279 (2015) (Ex. 2019);
- Ohring et al., *Chapter 8: Epitaxy*, in “Materials Science of Thin Films, Deposition and Structure,” 2nd Edition, 419-494 (2002) (Ex. 2020);

- Pohler et al., *Seed layer stimulated growth of crystalline high Al containing (Al,Cr)₂O₃ coatings deposited by cathodic arc evaporation*, Thin Solid Films 550, 95-104 (2014) (Ex. 2021).

9. I have also reviewed various supporting references and other documentation 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 '660 Patent at the time of the earliest possible priority date of the '660 Patent. The '660 Patent was filed on July 8, 2020 and claims priority to application No. 15/638,449, filed on June 30, 2017 ("the Critical Date"). Counsel has informed me that the Critical Date represents the earliest possible priority date to which the challenged claims of '660 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 '660 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 '660 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.A-II.D do not teach or suggest “wherein the second region comprises a rock-salt crystal structure,” as recited in independent claims 1 and 6;
- The prior art references relied on in Grounds I.A-II.D do not teach or suggest “wherein a crystal orientation of at least a second part of the first region and a crystal orientation of at least a part of the second region are substantially aligned with each other,” as recited in independent claim 1, or “wherein a crystal orientation of at least a part of the first region and a crystal orientation

of at least a part of the second region are substantially aligned with each other,” as recited in independent claim 6;

- The prior art references relied on in Grounds I.A-II.D do not teach or suggest “wherein the second region extends from a surface of the positive electrode active material particle to an inside of the positive electrode active material particle where magnesium detected by a line analysis of energy dispersive X-ray spectroscopy (EDX) in a depth direction is 1/5 of a maximum peak of the magnesium in the line analysis,” as recited in independent claim 6; and
- The prior art references relied on in Grounds II.A-II.D do not teach or suggest “wherein the positive electrode active material particle comprises titanium, magnesium, fluorine, and oxygen in the second region,” as recited in independent claims 1 and 6.

III. LEGAL STANDARDS

A. Legal Standards for Obviousness

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 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.

16. 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 claimed invention and the prior art are such that the claimed invention as a whole would have been obvious before the effective filing date of the claimed invention to a person having ordinary skill in the art to which the claimed invention 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.

17. 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.

18. 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."

19. 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."

20. 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.

21. 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.

22. 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.

23. 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.

24. 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.

25. 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.

26. 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.

27. 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.

28. 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).

B. Level Of Ordinary Skill in The Art

29. 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 ’660 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., 20. 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.

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

IV. OVERVIEW OF THE '660 PATENT

31. 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.

32. As the '660 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, 2:15-19. At the time of the invention of the '660 Patent, a known issue with LIBs was the reduction in capacity due to repeated charge and discharge cycles. *Id.*, 2:20-31. 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 and breaking of PEAM particles after repeated charge and discharge cycles. Such cracking and breaking of PEAM particles “make[] a new surface of the positive electrode active material exposed” and cause the surface to be “in contact with an electrolyte solution to cause a decomposition reaction,” which leads to degrading of the “charge and discharge characteristics” of the battery. *Id.*, 4:22-28.

33. 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 ’660 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, 4:32-36.

34. The ’660 Patent describes and claims PEAM particles with coatings that were a substantial and unexpected improvement compared to prior coating materials and techniques. The inventors proposed novel protective layers that could be used in PEAM particles to mitigate deleterious effects of crack portions and crystal defects in the PEAM particle. Ex. 1001, 24:43-55.

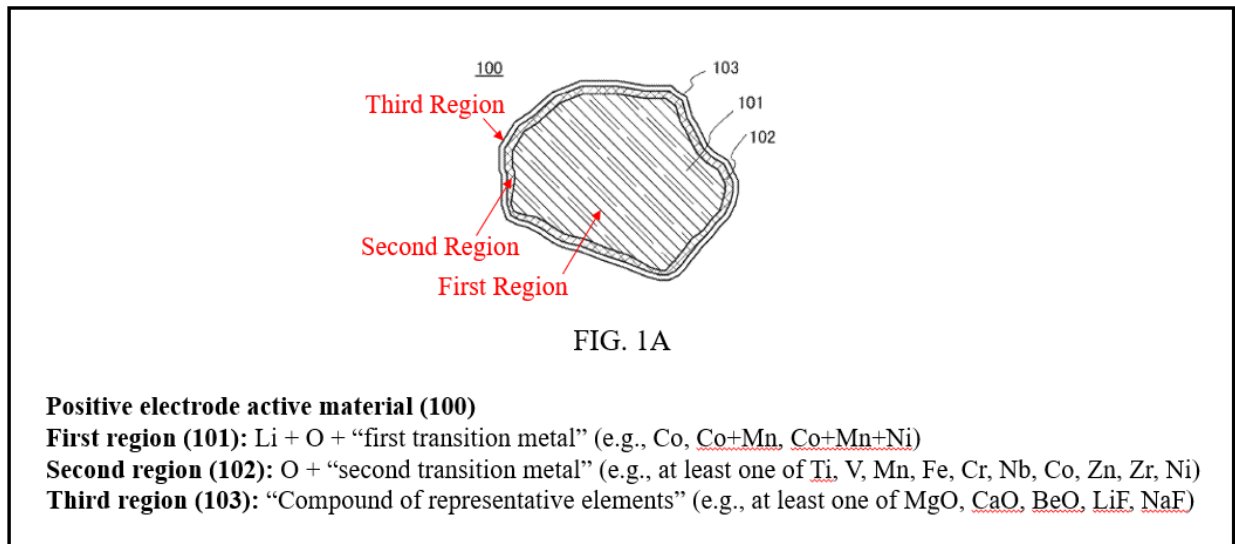
35. As an example, the ’660 Patent discloses a PEAM particle 100 having a first region 101, a second region 102, and a third region 103. *See id.*, 10:12-13:57. The first region 101 contains “a composite oxide of lithium and a first transition metal,” wherein the first transition metal can be cobalt only (Co); cobalt and manganese (Mn); or cobalt, manganese, and nickel (Ni). *Id.*, 10:24-26, 10:29-31. The first region 101 “preferably has a layered rock-salt crystal structure.” *Id.*, 10:27-28. The second region 102 contains “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.*, 11:6-16. The second

region 102 “preferably has a rock-salt crystal structure,” and “preferably has a crystal structure which is the same as that of the third region 103[.]” *Id.*, 11:22-23, 11:59-61. The third region 103 contains “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.*, 12:18-25. The third region 103—which can include fluorine and a representative element such as magnesium—is provided as an example of a protective layer of the PEAM particle 100. *See id.*, 12:28-32 (“[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.*, 12:34-37 (“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 id.*, Figs. 5C, 6D (showing that the third region 103 includes fluorine and magnesium). The third region 103 “preferably has a rock-salt crystal structure,” like the second region. *Id.*, 12:61-63.

36. The ’660 Patent explains that “a state where structures of two-dimensional interfaces have similarity is referred to as ‘epitaxy,’” and “a state where three-dimensional structures have similarity or orientations are crystallographically the same is referred to as ‘topotaxy.’” Ex. 1001, 9:34-41. Topotaxy is observed when “orientations of crystals in two regions...are substantially aligned with each

other.” *Id.*, 9:41-44. The ’660 Patent describes that the second region is “preferably formed by heteroepitaxial growth from the first region 101,” and the third region is “preferably formed by heteroepitaxial growth from the second region 102.” *Id.*, 13:59-62. The ’660 Patent explains that “[a] region formed by heteroepitaxial growth becomes topotaxy which has crystal orientations substantially three-dimensionally aligned with those of a region serving as a base,” meaning that “the first region 101, the second region 102, and the third region 103 can be topotaxy.” *Id.*, 13:62-67.

37. The different regions of the PEAM are summarized in annotated FIG. 1A, reproduced below.

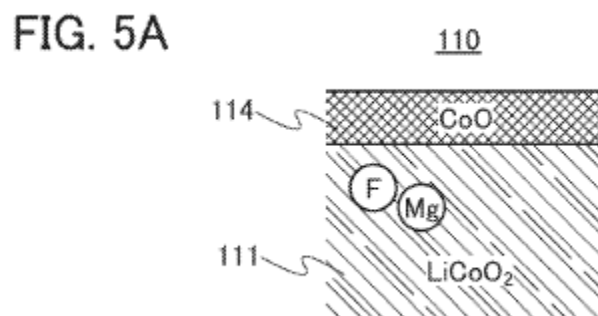


Ex. 1001, Fig. 1A (annotated)

38. I note that the formation, and hence presence, of a protective layer, such as the third region 103, yields protections in these areas where conventional coatings

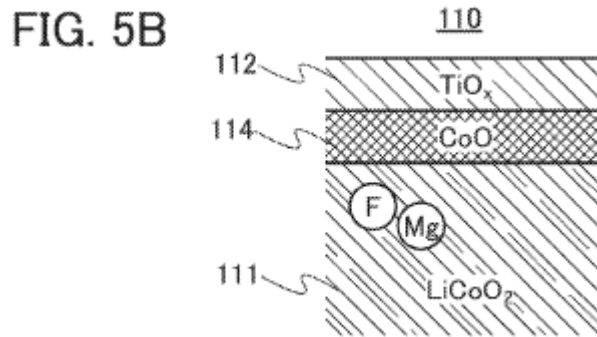
did not. For example, Figs. 5A–5C and 6A–6D of the '660 Patent illustrate a segregation model of elements contained in a PEAM particle, showing how during a manufacturing process that involves heating (e.g., at 500°C to 1200°C), representative elements such as magnesium (Mg) and fluorine (F) can unexpectedly segregate towards a superficial portion of the PEAM particle to form the third region 103. Ex. 1001, 10:49-53, 23:54-24:24, 27:20-28:37.

39. In more detail, and starting, for example, with Fig. 5A of the '660 Patent, a particle 110 can initially include a cobalt oxide layer 114 in contact with a region 111 containing lithium cobaltate, magnesium, and fluorine. *Id.*, 22:51-55, 23:3-5.



Ex. 1001, Fig. 5A

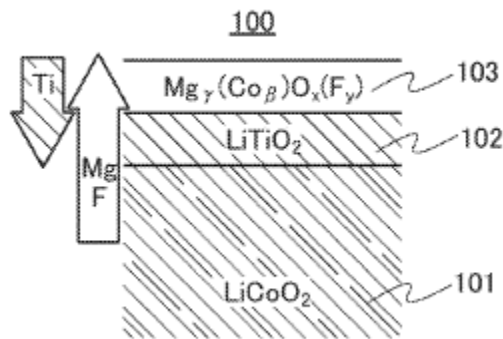
40. The particle 110 is then described as being coated with a material containing titanium (e.g., a titanium oxide) using a sol-gel method to produce the state of the particle 110 shown in Fig. 5B. *Id.*, 22:56-61.



Ex. 1001, Fig. 5B

41. 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.*, 22:62-23:5. According to the '660 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. *Id.*; *see also id.*, 24:30-55. This heating process can form the second region 102 and the third region 103 of the PEAM 100. *Id.*, 24:62-25:3. 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.*, 12:34-37.

FIG. 5C



Ex. 1001, Fig. 5C

42. The '660 Patent further demonstrates the utility of this approach by demonstrating that a PEAM particle formed through this process showed “excellent initial charge and discharge capacity, rate characteristics, and cycle characteristics.” *Id.*, 65:48-52.

V. OVERVIEW OF THE CITED REFERENCES

A. Blangero

43. I have been informed by Counsel and understand that Petitioner relies on Blangero in Grounds I, II.B and II.D of the Petition to argue that the invention of the '660 Patent would have been obvious to a POSITA. Blangero is a published patent application directed to a “cathode material having a high packing density, a high rate performance, improved discharge capacity and showing high stability during extended cycling at high charge voltage.” Ex. 1005, [0059]. Blangero states that a need existed in the art for “high capacity $LiCoO_2$ based cathodes which can be cycled in a stable manner in real cells at higher voltages.” *Id.*, [0005]. Blangero

identified prior art approaches including coating methods, such as with “LiF protective coatings[.]” *Id.*, [0006]. But Blangero noted a problem with these coatings, namely that “coated dense LiCoO_2 often has a lower reversible capacity, so that a part of the gain of energy density by charging to higher voltage is annulled by lower intrinsic capacity.” *Id.*

44. Blangero approached this problem by developing a “lithium metal oxide powder” that can be used “as a cathode material in a rechargeable battery,” consisting of (i) “a core material consisting of the elements Li, a metal M and oxygen,” where the metal M “has the formula $\text{M}=\text{Co}_{1-a}\text{M}'_a$,” and M’ is “either one or more metals of the group consisting of Al, Ga and B,” and (ii) “a surface layer consisting of a mixture of the elements of the core material and inorganic N oxide particles, wherein N is either one or more metals of the group consisting of Mg, Ti, Fe, Cu, Ca, Ba, Y, Sn, Sb, Na, Zn, Zr and Si.” *Id.*, [0009]; *see also id.*, [0059]. The “core material has hexagonal layered crystal structure described as an ordered rock salt-type crystal structure with space group R-3m.” *Id.*, [0060]. The “existence of a defect-free core is an inherent feature of the cathode material of the actual invention.” *Id.*, [0061]; *see also id.*, [0085] (“The inventors have made two surprising observations, which are believed to be essential aspects of the invention”), [0086] (noting “surprising” observation that “[t]he core of the particles is lithium stoichiometric and substantially free from defects.”).

45. I note that Blangero identifies that in one embodiment, the powder has the “general formula $\text{LiCo}_{1-a-b}\text{M}'_a\text{M}''_b\text{O}_2$,” where “M’ compris[es] either one or more elements from the group Al, Mg, Ti, Fe, Cu, Ca, Ba, Y, B, Sn, Sb, Na, Ga, Zn” and “M” compris[es] either one or more elements from the group F, P, S, Zr.” *Id.*, [0011]. Blangero notes that the “elements F, P, and S may be present on the surface under the form of LiF, Li_3PO_4 and Li_2SO_4 .” *Id.*

46. Blangero goes on to describe a method for manufacturing the claimed lithium metal oxide powder, involving (i) “providing a first mixture consisting of a first M-comprising and a first Li-comprising precursor powder,” (ii) “sintering the first mixture in an oxygen comprising atmosphere at a temperature T_1 of at least 600°C , thereby obtaining a Li-enriched lithium metal oxide compound, the metal being M,” (iii) “providing a second M-comprising precursor powder,” (iv) “mixing the Li-enriched lithium metal oxide compound and the second M-comprising precursor powder, thereby obtaining a second mixture,” and (v) “sintering the second mixture in an oxygen comprising atmosphere at a temperature T_2 of at least 600°C .” *Id.*, [0016]-[0020]; *see also id.*, [0021]-[0026], [0027]-[0032]. In other words, during manufacturing, the powder in Blangero undergoes two separate mixing and sintering (i.e. heating) steps.

47. Blangero notes that the invention’s surface layer “has an inhomogeneous composition compared to the core with a compositional gradient of

the different M, M', Li and O elements.” *Id.*, [0062]. Blangero further notes that the surface is “enriched of elements N such as Mg, Ti, Fe, Cu, Ca, Ba, Y, Sn, Sb, Na, Zn, Zr and Si and, in one embodiment, is formed by segregation from the core and accumulation of these metal dopants at the surface of the particles,” whereas “[i]n the core, these dopants are substantially absent.” *Id.* In a key paragraph, Blangero notes that the “[a]uthors could not indisputably establish the chemical nature of the oxides formed at the surface and therefore speculate that, for example in the case of Mg, Si and Ti doping, possible forms, but not limited to, are LiMO_2 , MgO , CoO , $\text{Co}_{1-\phi}\text{Mg}_\phi\text{O}\dots$, Co_3O_4 , $\text{Mg}_\delta\text{Co}_{3-\delta}\text{O}_4\dots$, TiO_2 , Li_2TiO_3 , SiO_2 , $\text{Li}_\epsilon\text{Si}_\lambda\text{O}_\pi[\dots]$ ” *Id.*

48. Blangero then describes a comparison between a cathode produced using a prior art process and a cathode produced using Blangero’s disclosed process. Blangero obtains a “state of the art titanium and magnesium doped LiCoO_2 (noted LCO-1)” using a “standard high temperature solid state synthesis (=the first firing step) by mixing the precursor with Li_2CO_3 .” *Id.*, [0109]. I note that LCO-1 “is also referred to as the lithium doped cobalt oxide ‘parent’ of Example 1 and Example 2.” *Id.* To prepare Example 1 of Blangero’s invention, “95 mol % of LCO-1 and 5 mol % of 0.25 mol% Ti and 0.5 mol % Mg doped $\text{Co}(\text{OH})_2$ ” were mixed, then “placed in an alumina crucible and heated (=the second firing step) at 925°C for 12 hours under constant air flow.” *Id.*, [0110]. The “chemical composition of the surfaces of

the particles of LCO-1 and Ex1” was investigated using X-Ray Photoelectron Spectroscopy (XPS) Analysis. *Id.*, [0129]; *see also id.*, Fig. 4a and 4b. The results are shown in Table 3, which I have reproduced below:

TABLE 3		
elemental composition of the surface of the grains of LCO-1 and Ex1.		
at. %	LCO-1	Ex1
Li	21.7	15.3
Co	8.7	12.0
O	41.9	37.5
C	23.1	26.5
S	1.3	0.8
Ca	0.6	0.1
F	0.9	0.4
Ti	0.9	1.1
Na	0.3	0.3
Si	Below detection limit	1.1
P	0.4	0.6
Mg	Below detection limit	4.4

Id., [0129]. Notably, I see that Example 1 showed increased amounts of cobalt (Co), titanium (Ti), and magnesium (Mg), and decreasing amounts of lithium (Li), oxygen (O), and fluorine (F) on the surface as compared to the prior art LCO-1 composition. The dependent claims demonstrate that F, P, and S (present as LiF, Li₃PO₄, and Li₂SO₄) are apparent contaminants that are intended to be limited. *See id.*, cl. 37 (“The lithium metal oxide powder of claim 33, wherein the surface layer further comprises less than 2000 ppm of one or more of LiF, Li₃PO₄ and Li₂SO₄”); *see also id.*, cl. 62. I note that Blangero never explains where the fluorine that is detected in Table 3 originates, or indicates that its presence is either intentional or desirable.

49. Blangero went on to use XPS as a tool to analyze the “evolution of Mg and Ti as function of particle depth” for Example 1 “by means of XPS depth profiling

as shown on FIG. 8.” *Id.*, [0135]. Blangero’s depth profiling showed that the “concentration of Mg and Ti decreases rapidly over the first 50 nm of the particles,” but the amounts “do not decrease down to zero[.]” *Id.* Importantly, I note that Blangero does not disclose the alignment of the crystal orientation of the core and surface materials, or whether the core and surface material orientations are aligned with one another.

B. Shim

50. I have been informed by Counsel and understand that Petitioner relies on Shim as a secondary reference in Ground I of the Petition to argue that the invention of the ’660 Patent would have been obvious to a POSITA. Shim is a published patent application directed to a “positive active material for a rechargeable lithium battery having excellent cycle-life characteristics.” Ex. 1006, [0008]; *see also id.*, Abstract. Specifically, Shim teaches that the positive active material “includes a lithium intercalation compound; and a lithium titanium oxide...on the surface of the lithium intercalation compound” with the formula “ $\text{Li}_{4-x}\text{M}_x\text{Ti}_y\text{O}_{12-z}$.” *Id.*, [0011]; *see also id.*, [0053]. In this formula, “M is an element selected from Mg, Al, Ga, La, Tb, Gd, Ce, Pr, Nd, Sm, Ba, Sr, Ca, and combinations thereof.” *Id.*, [0016]; *see also id.*, [0056]. Alternately, the “lithium intercalation compound may be an M-doped compound, where M is selected from Mg, Al, Ti, La, Tb, Gd, Ce, Pr, Nd, Sm, Ba, Sr, Ca, and combinations thereof.” *Id.*, [0019]. I note that Shim teaches

that the “lithium titanium oxide may be positioned on the surface of the lithium intercalation compound in a form of an island coating,” where Shim defines “island coating” as “a discontinuous coating formed of spherical, semi-spherical, non-spherical, or amorphous shape particles.” *Id.*, [0058]. Shim alleges that the “resulting positive active material exhibits improved ion conductivity and electric conductivity,” and “thus, excellent rate capability and cycle-life characteristics during the charge and discharge of the battery may be obtained.” *Id.*, [0059].

51. Shim describes that its positive active material is prepared by “mixing a titanium compound-containing liquid and a lithium intercalation compound to prepare a mixture; drying the resulting mixture to obtain a dried product; and heat-treating (e.g., by firing) the dried product.” *Id.*, [0067]. The “titanium compound” “may be titanium butoxide, titanium isopropoxide, titanium dioxide, or a combination thereof.” *Id.*, [0069]. Shim explains that the concentration of the titanium compound in the titanium compound-containing liquid should be “of about 1.5 wt% to about 3.0 wt%, based on the total amount of the liquid,” in order to successfully form a compound “on the surface of the lithium intercalation compound.” *Id.*, [0070]-[0071]. If the concentration is out of this range, Shim notes that “undesired impurities such as, for example, M-doped Li_2TiO_3 and/or the like may be formed.” *Id.* Likewise, Shim teaches that “a mixing ratio of the titanium compound and the lithium intercalation compound may be a weight ratio ranging

from about 0.0092:1 to about 0.073:1,” leads to an improvement in “capacity characteristics,” whereas using ratios outside this range can lead to a decrease in capacity. *Id.*, [0074].

52. Shim further discloses that “a starting material that generally remains as an impurity during the manufacturing of a lithium intercalation compound (for example, Li_2CO_3 remaining during the manufacturing of LiCoO_2), reacts with a titanium compound to form a lithium titanium oxide, such that a doping element M (initially doped on the lithium intercalation compound) is then doped on the lithium titanium oxide.” *Id.*, [0077].

53. Shim then describes a series of Examples of Shim’s invention along with Comparative Examples that appear to be from the prior art. Shim discloses several scanning transmission electron microscope (STEM) images showing cross-sections of the active material. I note that Petitioner relies on two of these STEM images in Figures 4B and 5B to demonstrate the alignment of the crystal structure between the coating regions and the lithium intercalation regions. *See, e.g.*, Pet., 35-36 (reproducing and annotating Ex. 1006, Fig. 4B, 5B). These images, however, are taken of Comparative Examples 2 and 6, respectively, and are **not** depictions of Shim’s invention that Petitioner otherwise purports to rely on. *See* Ex. 1006, [0134]-[0135]. Indeed, I note that Comparative Examples 2 and 6 use a titanium butoxide ethanol solution with a concentration of 0.46 wt% and 1.35 wt%, respectively, both

below the 1.5 wt % to 3.0 wt% range that Shim teaches can be used successfully to form the titanium compound on the surface of the lithium intercalation compound. *See id.*, [0070]-[0071].

C. Yamamoto

54. I have been informed by Counsel and understand that Petitioner relies on Yamamoto as a secondary reference in Grounds I.B, II.C, and II.D of the Petition to argue that the invention of the '660 Patent would have been obvious to a POSITA. Yamamoto is a research article that describes coating a LiCoO_2 thin-film model electrode with magnesium oxide (MgO) to “improve[] the durability of the cathode during high-potential cycling.” Ex. 1007, 1. I note that Yamamoto tests two MgO coatings: one deposited onto the LiCoO_2 films for 30 seconds at room temperature (RT-MgO-LCO), and one deposited onto the LiCoO_2 films for 60 seconds at high temperature, 700°C (HT-MgO-LCO). *Id.* Yamamoto characterized the thin films using various techniques, including as one example energy dispersive X-ray spectroscopy (EDX). *Id.*, 1-2. Yamamoto discloses that the EDX scan “detected a Mg containing layer of approximately 5 nm in thickness outside Co containing layer for the RT-MgO-LCO,” as shown in Figure 1(b), while the HT-MgO-LCO instead “has a Mg layer within the LiCoO_2 surface,” (data not shown). *Id.*, 2.

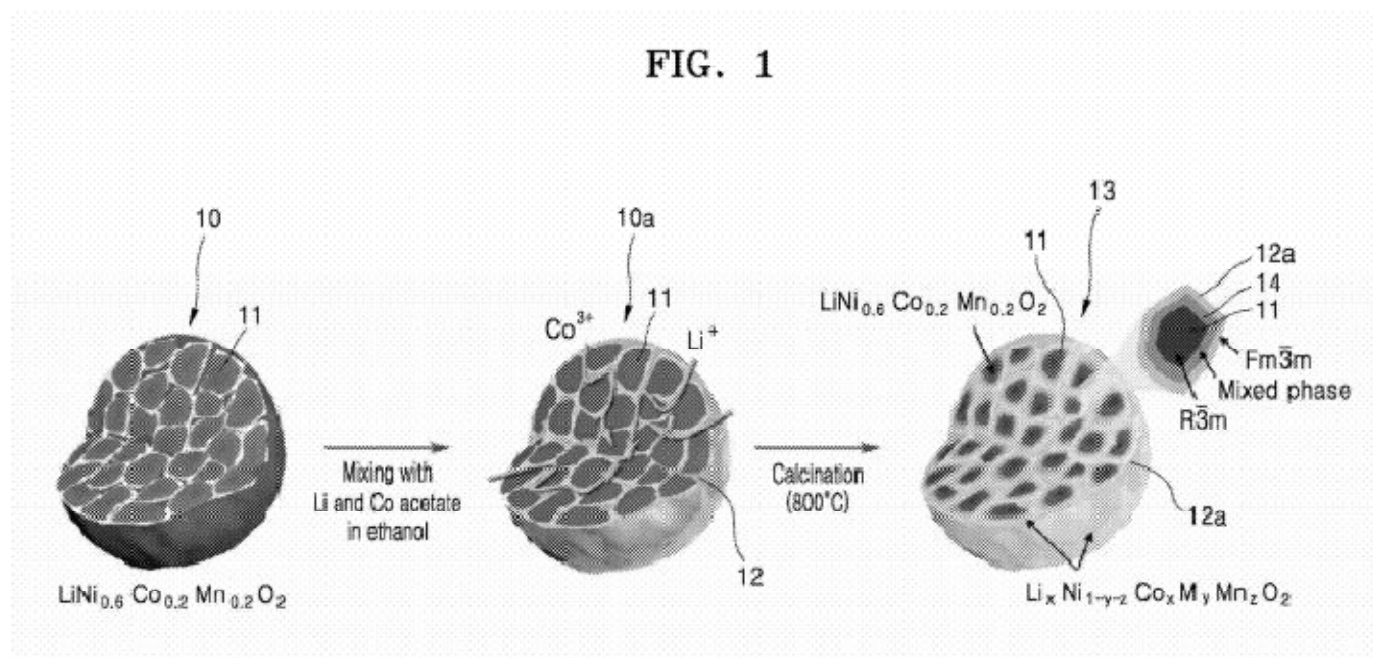
55. Yamamoto goes on to conclude that the “temperature of MgO coating influences the surface structure of LiCoO_2 .” *Id.*, 3. Specifically, Yamamoto

describes that in “the RT-MgO-LCO, the MgO layer only covers LiCoO₂ surface, leading to unfavorable increase in the interfacial resistance,” whereas in the “HT-MgO-LCO, the solid solution of Mg stabilizes the interfacial structure upon cycling at high potentials.” *Id.*, 4.

D. Cho

56. I have been informed by Counsel and understand that Petitioner relies on Cho as a reference in Ground II of the Petition to argue that the invention of the '660 Patent would have been obvious to a POSITA. Cho is a patent directed to a “composite cathode active material, including a nickel-based lithium transition metal oxide secondary particle[.]” Ex. 1008, Abstract. Cho describes that the “nickel-based lithium transition metal oxide secondary particle includ[es] a coating layer containing lithium and cobalt on a surface of a primary particle of the secondary particle.” *Id.*

57. I have reproduced Figure 1 below, which is an exemplary illustration of Cho’s “nickel-based lithium transition metal oxide secondary particle 10” showing “an agglomerate of primary particles 11.” *Id.*, 6:42-46. I note that exemplary nickel-based lithium transition metal oxide secondary particles **10** include LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂ (NCM622), LiNi_{0.5}Co_{0.3}Mn_{0.2}O₂, or LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂. Ex. 1008, 6:46-49.



Ex. 1008, Fig. 1

58. Cho discloses that its “cathode may include at least one technical feature, such as composition and particle diameter, that is different from the composite cathode active material in addition to the composite cathode active material,” such as “cathode active material used in the art.” Ex. 1008, 9:16-20. Exemplary cathode active material used in the art includes “a lithium cobalt oxide, a lithium nickel cobalt manganese oxide, a lithium nickel cobalt aluminum oxide, a lithium iron phosphate, or a lithium manganese oxide.” *Id.*, 9:21-26.

E. Kim

59. I have been informed by Counsel and understand that Petitioner relies on Kim as a secondary reference in Ground II of the Petition to argue that the

invention of the '660 Patent would have been obvious to a POSITA. Kim is a research article that describes a new coating method for alleviating surface degradation of $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ cathode material. Ex. 1009, 1. I note that Kim appears to be an article underlying the Cho patent and specifically contains several figures that appear to be identical to those found in Cho. *E.g., compare* Ex. 1008, Figs. 1, 4A, 4B, 7A, *with* Ex. 1009, 2-3 (Figs. 1a, 1b, 1c, 2d).

VI. PRIOR ART ANALYSIS

A. Ground I is Deficient for Failing to Show that Blangero Alone or in Combination with Shim or Yamamoto Renders Obvious Independent Claims 1 and 6

60. In my opinion, Ground 1 is deficient for at least three reasons. First, with respect to both independent claims 1 and 6, in my opinion, Petitioner's argument that Blangero discloses a "rock-salt crystal structure" is speculation that is inconsistent with Blangero's own disclosures. Second, I do not believe that Petitioner has shown that Blangero and Shim, once they are combined, have the relative crystal orientations claimed in independent claims 1 and 6. Third, with respect to claim 6, in my opinion Petitioner has not demonstrated that the distribution of particles in Blangero would in fact be represented by Yamamoto's EDX line analysis in order to meet the claimed magnesium distribution in the second region.

1. The Petition Fails to Show that Blangero Alone or in Combination with Shim Discloses a “Second Region that Comprises a Rock-Salt Crystal Structure” (Elements 1.4, 6.4)

61. I understand that Petitioner argues in turn that three separate disclosures from Blangero allegedly teach a second region that comprises a rock-salt crystal structure for Ground I.A and I.B—specifically, that each of (i) magnesium oxide (MgO), (ii) lithium fluoride (LiF), or (iii) “TiO₂, that may also be lithiated” disclose this type of crystal structure. Pet., 27-30. But, in my opinion, Petitioner has not shown that any of these three compounds is present in the second region, let alone that one is present and has a rock-salt crystal structure.

i. Petitioner has not demonstrated that MgO is present in Blangero or necessarily has a rock-salt crystal structure

62. I understand that Petitioner argues “Blangero discloses that its PEAM material includes a surface layer comprising MgO due to ‘segregation from the core’” and “[a] POSITA would have known that at least the MgO portion of Blangero’s coating would have a rock-salt crystal (*i.e.* NaCl) structure.” Pet., 27-28. I disagree.

63. Petitioner’s argument is based only on speculation, as, in my opinion, it would not have been obvious to a POSITA that MgO is present in Blangero’s coating based on Blangero’s disclosures. Instead, I note that Blangero quite

explicitly states that the surface layer is inhomogeneous and of indeterminate composition:

The surface layer has an *inhomogeneous composition* compared to the core with a *compositional gradient* of the different M, M', Li and O elements.... Authors *could not indisputably establish the chemical nature of the oxides formed at the surface* and therefore *speculate* that, for example in the case of Mg, Si and Ti doping, *possible forms*, but not limited to, are LiMO_2 , **MgO**, CoO , $\text{Co}_{1-\phi}\text{Mg}_\phi\text{O}$ with $\phi \leq 1$, Co_3O_4 , $\text{Mg}_\delta\text{Co}_{3-\delta}\text{O}_4$ with $\delta \leq 1$, TiO_2 , **Li₂TiO₃**, SiO_2 , $\text{Li}_\varepsilon\text{Si}_\lambda\text{O}_\pi$ with $2 \leq \varepsilon \leq 8$, $1 \leq \lambda \leq 2$ and $3 \leq \pi \leq 7$ [.]

Ex. 1005, [0062].¹

64. Providing no explanation for its selection, Petitioner plucks MgO from a laundry list of M' or M'' metal elements and N dopants that Blangero identifies as possibilities, but admits could not be “indisputably establish[ed].” *Id.* For example, Blangero discloses that “[t]he surface layer has an inhomogeneous composition compared to the core with a compositional gradient of the different M, M', Li and O elements” and that “[t]he surface is enriched of elements N such as Mg, Ti, Fe, Cu, Ca, Ba, Y, Sn, Sb, Na, Zn, Zr and Si and...formed by segregation from the core and accumulation of these metal dopants at the surface of the particles” in which “elements F, P and S may be present on the surface.” *Id.*; *see also id.*, [0011] (“M' comprising either one or more elements from the group Al, Mg, Ti, Fe, Cu, Ca, Ba,

¹ All emphasis added unless otherwise noted.

Y, B, Sn, Sb, Na, Ga, Zn; M” comprising either one or more elements from the group F, P, S, Zr.”). In picking out MgO from these lists, in my opinion, Petitioner misrepresents and overstates Blangero’s disclosure of MgO. For example, I see that Petitioner summarizes paragraph 59 as simply “describing Mg and Ti oxides at a ‘surface layer,’” (Pet., 27), when what paragraph 59 actually says is “one or more metals of the group consisting of Mg, Ti, Fe, Cu, Ca, Ba, Y, Sn, Sb, Na, Zn, Zr and Si,” (Ex. 1005, [0059]). I see no explanation for why Petitioner chose magnesium as opposed to the other metals that Blangero discloses.

65. Additionally, in my opinion Petitioner makes an unsupported assumption when it assumes that it would have been obvious to a POSITA that MgO, to the extent present at all, would **only** form a rock-salt crystal structure. But to the contrary, MgO is known to a POSITA to be available in amorphous forms. *See e.g.*, Ex. 2003, 1; Ex. 2017, 2. Accordingly, in my view Petitioner has failed to explain why Blangero’s “speculat[ive]” MgO, assuming it is present on the surface at all, would then necessarily also be present in a rock-salt crystal structure. Ex. 1005, [0062]. To the contrary, in my opinion, Blangero’s disclosure that the surface layer “has an inhomogeneous composition” would lead a POSITA to infer that the structure was **not** an ordered rock-salt crystal structure, but rather amorphous instead.

66. The most Petitioner can allege (incorrectly, in my opinion) is that MgO, if present in Blangero, would necessarily form a rock-salt crystal structure. But that assumption is unsupported and, in my opinion, unsupportable. Petitioner has offered only an assumption, not grounded in Petitioner's prior art, that MgO is present in rock-salt crystal structure, when the nature of its formation could also have led to a disordered or amorphous crystal structure.

ii. Petitioner has not demonstrated that LiF is present in Blangero or has a rock-salt crystal structure

67. I note that Petitioner next makes the argument that "Blangero's disclosure of lithium fluoride (LiF) 'on the surface' of the particle discloses this limitation." Pet., 28. But, in my opinion, Petitioner's argument ignores other of Blangero's disclosures, including a discussion of prior art where Blangero noted that coated LiCoO_2 "often has a lower reversible capacity," including when "LiF protective coatings" were used, and sought to *improve* over such prior art coatings. *See* Ex. 1005, [0006]-[0008]. Indeed, one of Petitioner's background references confirms that "LiF is an undesired product of the electrolyte degradation...due to its low permeability to Li-ions and electrons" which can "deteriorate the performance of the cathode." Ex. 1021, 2; *see also* Ex. 2004, 5-6. Therefore, in my opinion a POSITA simply would not have been motivated to select a LiF coating as a desirable coating to apply to Blangero's surface layer.

68. In my opinion, Blangero's disclosure that "[t]he elements F, P and S *may be present* on the surface under the form of LiF, Li₃PO₄ and Li₂SO₄" is not sufficient to show the presence of this element. Ex. 1005, [0011]. I note that Petitioner argues "Blangero makes clear that such LiF is in the 'surface layer' in addition to oxides of magnesium and/or titanium," (Pet., 29), suggesting that Petitioner believes that LiF is present in Blangero's coating as well as oxides of magnesium and/or titanium. But as I discussed above, Petitioner's position is based only on speculation, as Blangero explicitly states that its "[a]uthors *could not indisputably establish the chemical nature of the oxides formed at the surface*["] Ex. 1005, [0062]. While Blangero indicates that "F...*may* be present on the surface under the form of LiF," in my opinion, Blangero fails to conclusively demonstrate that LiF *is* in fact present on the PEAM surface. *Id.*, [0011]. Indeed, while I note that fluorine is identified by X-Ray Photoelectron Spectroscopy (XPS) in Table 3, I also note that the corresponding spectrum in Figure 5(b) does *not* identify LiF, but rather identifies Li₂CO₃ and LiCoO₂. *Id.*, Fig. 5(b).

69. Furthermore, I note that Petitioner again assumes (and again without evidence) that LiF, to the extent present at all in Blangero's surface, has a rock-salt crystal structure. But LiF (just as I explained with MgO) was known to also be available in amorphous forms. *See e.g.*, Ex. 2005, 4-5 (identifying "an amorphous outer layer" of LiF "which likely consists of organic/polymeric species from

electrolyte decomposition”); Ex. 2004, 5-6 (“The Li_2CO_3 together with LiF can form an *amorphous* SEI layer coated on the particle surface...which reduces the ionic conductivity of the cathode surface and undermines the connectivity between neighboring cathode particles.”). Indeed, here, I understand that Petitioner argues that LiF would result from “electrolyte decomposition,” through the “hydrolysis of LiPF_6 ,” an electrolyte salt. Pet., 12 (citing Ex. 1021, 1). But in my opinion, this method of deposition could make it even more likely that LiF would be deposited in an amorphous form, rather than with an ordered rock-salt crystal structure. *Id.* Accordingly, in my opinion, Petitioner has not shown that a POSITA would have understood that LiF is present, let alone that LiF is present *and* has a rock-salt crystal structure.

70. The most Petitioner can allege (again, incorrectly, in my opinion) is that LiF, if it is present in Blangero, would necessarily form a rock-salt crystal structure. But, just as with MgO, that assumption is unsupported. Petitioner has offered only an assumption, not grounded in Petitioner’s prior art, that LiF is present in rock-salt crystal structure, when the nature of its formation through decomposition of the electrolyte formulation could also have led to a disordered or amorphous crystal structure.

iii. Petitioner has not demonstrated that lithium titanium oxide is present in Blangero or has a rock-salt crystal structure

71. I note that Petitioner then argues that “Blangero renders this limitation at least obvious in view of its surface layer further comprising ‘TiO₂, that may also be lithiated,’ or lithium titanate.” Pet., 29 (citing Ex. 1005, [0094], [0059], [0062]).

72. Petitioner’s position is again based on speculation, as, in my opinion, it would not have been obvious to a POSITA that lithium titanium oxide is in fact present in Blangero’s coating based on its disclosure. I see no examples in Blangero that show lithium titanium oxide present in the surface layer. Instead, seemingly based on hindsight, Petitioner alleges that lithium titanium oxide is present based on an expansive list of possible combinations derived from laundry lists of possible metal elements and N dopants that might be present on the surface, but that Blangero itself states could not be established with certainty. Blangero states that “[a]uthors *could not indisputably establish the chemical nature of the oxides formed at the surface* and therefore *speculate* that, for example in the case of Mg, Si and Ti doping, *possible forms*, but not limited to, are LiMO₂, MgO, CoO, Co_{1-φ}Mg_φO with $\varphi \leq 1$, Co₃O₄, Mg_δCo_{3-δ}O₄ with $\delta \leq 1$, TiO₂, Li₂TiO₃, SiO₂, Li_εSi_λO_π with $2 \leq \varepsilon \leq 8$, $1 \leq \lambda \leq 2$ and $3 \leq \pi \leq 7$ [.]” Ex. 1005, [0062]. Therefore, in my opinion, the disclosure of “TiO₂” or “Li₂TiO₃” as possibilities is not enough to show that lithium titanium oxide is in fact present in Blangero’s surface layer.

73. I note that Petitioner also again erroneously assumes (without evidence) that the form of any lithium titanium oxide present on a cathode surface will be a rock-salt crystal structure. But on the contrary, lithium titanium oxide can also be amorphous in form. *See e.g.*, Ex. 2007, 4. I note that Petitioner has failed to explain why the “speculate[d]” forms of lithium titanium oxide, if even present in Blangero, would necessarily be present in a rock-salt crystal structure. Furthermore, since Blangero’s list of “speculate[d]” “possible forms” includes compounds that lack a rock-salt crystal structure, such as Co_3O_4 , in my opinion it would not have been obvious to a POSITA that lithium titanium oxide would necessarily be required to have a rock-salt crystal structure if it was found in Blangero’s surface layer.

74. Additionally, contrary to Petitioner’s assertion, (*see* Pet., 29-30), it is my view that a POSITA would not have been motivated to change the crystal structure disclosed in Blangero based on the description in Wang’s 2012 study (Ex. 1029), because Blangero and Wang have different substrate materials. I note that Petitioner points to Fig. 4(c) and 4(d) in Wang to allege that there is a motivation to combine LiTiO_2 coating, which can sometimes have a rock-salt crystal structure, with the PEAM in Blangero because LiTiO_2 allegedly has better characteristics than the Li_2TiO_3 species disclosed in Blangero. But Wang teaches the use of LNCM (e.g., $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$), which I note is itself coated with LiTiO_2 . *See* Ex. 1029, 4 (“...the coated LNCM microspheres are described as surface- LiTiO_2 -rich

microspheres, with $\text{Li}[\text{Ni}_x\text{Mn}_y\text{Ti}_{1-x-y}]\text{O}_2$ domains in the surface layer. This unique coating layer indicates a strong interaction between LiTiO_2 and the host structure”). From this disclosure, in my opinion, a POSITA would have understood that Wang’s disclosure describes characteristics caused by a **combination** of a coating material made of LiTiO_2 and LNCM with a specific host structure. Because the LiCoO_2 substrate in Blangero is a different substrate from the LNCM substrate in Wang, it would not have been obvious to a POSITA that the effect shown in Wang would have also been obtained when LiTiO_2 was used with a different substrate—i.e., when applied to LiCoO_2 .

75. Finally, as I discussed above, the most Petitioner can allege (incorrectly, in my opinion), is that lithium titanium oxide, if it is present in Blangero, would necessarily form a rock-salt crystal structure. But, just as with MgO and LiF , that assumption is unsupported. Petitioner has offered only an assumption, not grounded in Petitioner’s prior art, that lithium titanium oxide is (i) present and (ii) has a rock-salt crystal structure.

2. The Petition Fails to Show that Blangero Alone or in Combination with Shim Renders Obvious the Claimed Relative Crystal Orientation (Elements 1.7 and 6.7)

76. In my view, Petitioner provides no evidence from Blangero that shows “wherein a crystal orientation of at least a second part of the first region and a crystal orientation of at least a part of the second region are substantially aligned with each

other.” Ex. 1001, cl. 1; *see also* cl. 6. While Petitioner purports to rely upon principles of epitaxy to disclose this limitation, I note that Blangero itself is silent with regard to epitaxy being applicable in any of its compositions, and a POSITA would not have found Blangero’s conditions to facilitate epitaxial growth for several reasons I discuss further below.

77. Because Blangero is silent as to alignment of the crystal structure of its surface and core, Petitioner relies in large part upon the crystal structure alignment disclosed in another reference, Shim, without explaining how or why Shim and Blangero would be combined, or why a POSITA would have expected the same crystal structure or alignment when Shim and Blangero use markedly different processes. Additionally, I note that Petitioner appears to completely ignore the fact that Shim also teaches a *spinel* structured compound for use in its coating that has improved properties.

i. A POSITA would not have understood Blangero to disclose the claimed crystal alignment

78. Petitioner alleges that “a POSITA would have understood that Blangero’s first region (comprising LiCoO_2) and second region (comprising Li_2TiO_3 and MgO in part) are in substantial alignment due to the principles of epitaxy,” but this allegation, in my opinion, is not supported by the evidence. Pet., 33. As a preliminary matter, as I explain in detail above, Petitioner’s assumption that either

Li_2TiO_3 or MgO is present in the second (i.e., surface) region is unsupported by Blangero's own disclosures. Next, I note that Blangero is silent as to either epitaxy or topotaxy. Epitaxy refers to a two-dimensional type of crystal growth or material deposition in which new crystalline layers are formed with one or more well-defined orientations with respect to the crystalline seed layer. *See* Ex. 2009, 2. Three-dimensional growth is more correctly referred to as topotaxy. *See* Ex. 1001, 9:34-44. A POSITA would have understood that not all growth on a given surface is epitaxial/topotaxial, and that epitaxial/topotaxial growth requires specific conditions that facilitate that type of growth. *See, e.g.,* Ex. 2010, 2, 6 (epitaxial growth achieved on MgO single crystal films but not on MgO polycrystalline films); EX-2020, 5 ("differences in film and substrate chemistry and coefficient of thermal expansion can strongly influence the electronic properties and perfection of the interface"); EX-2021, 7 (discussing "main preconditions" needed to stimulate epitaxial growth). An amorphous surface layer would not lend itself to epitaxial/topotaxial growth, and would be unlikely to have alignment with the "first region," or core. *See, e.g.,* Ex. 2012, 3 (formation of "amorphous" material "will in a worst case, eliminate epitaxy."); EX-2020, 7 ("crystalline metals deposited on amorphous substrates" yields "resulting films [that] are quite defective.").

79. Although I see that Petitioner points to another Shim reference (Ex. 1032, "Shim 2015") to explain that Li_2TiO_3 can be epitaxially grown on the surface

of LiCoO_2 , Petitioner completely ignores the different fabrication methods used in Blangero and Shim. It would **not** have been obvious to a POSITA that Li_2TiO_3 could (or would) have been grown epitaxially/topotaxially on the surface of LiCoO_2 in Blangero as taught by Shim 2015, particularly if the surface of Blangero was at least partially amorphous. Furthermore, Petitioner failed to address why a POSITA would have **necessarily** expected similar results despite significant differences found in the fabrication methods used by Blangero and Shim, as I discuss in more detail below.

ii. The Petition does not explain how a POSITA would have combined Shim and Blangero to maintain Shim's crystal structure alignment

80. I understand that Petitioner alleges that “a POSITA would have been motivated to use Shim's Li_2TiO_3 coating layer in Blangero's particle, as this coating layer would be in substantial alignment with Blangero's LiCoO_2 layer.” Pet., 36-37. In my opinion, this allegation is unsupported by the record evidence. Nowhere in the Petition does the Petitioner clearly articulate *how* a POSITA would “use” Shim's coating in Blangero's process. For instance, if I assume that Petitioner intends that the “use” of Shim's coating means that the coating would be **added** to Blangero's existing surface layer, then I see nothing in either Shim or in Blangero that teaches or even suggests that Shim's coating would somehow be in substantial alignment with Blangero's disclosed surface layer. Ex. 1005, [0062]. Indeed, in my view this

type of combination would be a hybrid composition that is neither contemplated by nor supported by the data provided by Shim or Blangero. Shim and Blangero are directed to two different types of coating processes, and neither contemplates being combined with other, different, coating techniques. Additionally, I note that even if a combination of coating techniques was somehow contemplated, I see nothing in either Shim or Blangero that suggests a POSITA would have a reasonable expectation of success in combining the two methods and achieving alignment between portions of the first and second regions. Indeed, in my opinion, if Shim's coating was *added* to Blangero, I see no explanation in the Petition as to what would constitute the "first" region or the "second" region in the hybrid composition containing both Blangero and Shim's coatings.

81. Alternately, if I assume instead that Petitioner meant to propose *substituting* Blangero's surface layer with Shim's Li_2TiO_3 coating layer, in my opinion such a modification would not have been obvious to a POSITA because Blangero's method of fabrication is starkly different from Shim's. I note that in Example 1, Blangero states that LCO-1 was prepared "according to the process explained in WO2010-139404." Ex. 1005, [0109] (citing Ex. 2011 ("Ellenwood")). I have reviewed Ellenwood, and I note that Ellenwood states that prior to its invention, there were "two major preparation routes to prepare...monolithic LiCoO_2 ," one of which involved using "a source of cobalt (like Co_3O_4) with

relatively small particle[s] is mixed with a source of lithium (like Li_2CO_3) and fired at sufficient high temperature with a sufficient excess of lithium.” Ex. 2011, 2:15-17. Ellenwood explains that this method “basically results in a failure.” *Id.*, 2:23. Ellenwood then goes on to explain the method of its invention, which involves a “precursor compound consisting of either one or more of a non-sintered agglomerated cobalt oxide, hydroxide and oxy-hydroxide powder” with a specific particle size. *Id.*, 3:30-4:2. I note that this is consistent with Blangero’s identification of $\text{Co}(\text{OH})_2$ as its desired precursor. Ex. 1005, [0109].

82. Shim, on the other hand, discloses that it uses Co_3O_4 as its cobalt source. *See* Ex. 1006, [119] (“ Li_2CO_3 , Co_3O_4 and Mg were mixed.”); *id.*, [0129] (Example 1 preparing PEAM “according to the same (or substantially the same) method as in Comparative Example 2.”). In other words, Shim uses the cobalt source that Ellenwood—the source of Blangero’s preparation method—teaches “basically results in a failure.” Ex. 2011, 2:15-23. Thus, in my opinion, a POSITA familiar with Blangero and Ellenwood would simply not have been motivated to use Shim’s method. Further, in my view, even if using Co_3O_4 does not result in total “failure,” it is likely that, at a minimum, a POSITA would have expected the resulting features of the lithium cobalt oxide to be different between Blangero’s and Shim’s processes.

83. Moreover, I note that several additional differences between Blangero and Shim’s coating processes would also have been expected to affect the results a

POSITA would have attained. As just one additional example, in Blangero, the second step, much like the first step, uses solid state synthesis, meaning solid precursors are added and heated. Ex. 1005, [0111]. In contrast, in Shim, the surface coating is generated via a solution phase reaction of titanium butoxide. Ex. 1006, [0129]. Numerous other differences exist between Blangero and Shim's processes as well. *Compare* Ex. 1005, [0109] ("A state of the art titanium and magnesium doped LiCoO_2 (noted LCO-1) is obtained by means of a standard high temperature solid state synthesis (=the first firing step) by mixing the precursor with Li_2CO_3 "), [0110] ("The homogeneous mixture is placed in an alumina crucible and heated (=the second firing step) at 925°C . for 12 hours under constant air flow. After cooling, the resulting powder (Ex1) is sieved and characterized."), *with* Ex. 1006, [0119] (mixture of Li_2CO_3 , Co_3O_4 and MgO was "heated up to 1000°C at a rate of $5^\circ\text{C}/\text{min}$ for 6 hours and then naturally cooled down"), [0120] ("adding titanium butoxide to ethanol and agitating the mixture at room temperature for about 2 hours"), [0121] (agitating equal masses of $\text{LiCo}_{0.95}\text{Mg}_{0.05}\text{O}_2$ with titanium butoxide ethanol solution for 2 hours, then drying in an 80°C oven for 24 hours), [0122] (heating dried powder to " 950°C at $5^\circ\text{C}/\text{min}$ in a furnace under an air atmosphere and then heat-treating at 950°C for 5 hours"), [0130] (modification for Example 2); *see also* Ex. 1032, 2-3. Therefore, not only are there differences in the synthesis of the primary particles, but there are also differences in the way the coatings are

created that a POSITA would understand lead to significant differences in the coating structure.

84. Additionally, as I noted above, epitaxial/topotaxial growth is highly process dependent. Ex. 2010, 2, 6 (epitaxial growth achieved on MgO single crystal films but not on MgO polycrystalline films); EX-2020, 5 (“differences in film and substrate chemistry and coefficient of thermal expansion can strongly influence the electronic properties and perfection of the interface”); EX-2021, 7 (discussing “main preconditions” needed to stimulate epitaxial growth); Ex. 2012, 3 (noting considerations for achieving epitaxy). Epitaxial and topotaxial growth generally depends on the surface conditions, but in my view, Petitioner has not established what the surface conditions of Blangero or Shim would be, what the features of their combination be, let alone demonstrated that they would be similar or would be expected to support epitaxial/topotaxial growth. Moreover, in my opinion, a POSITA would not have expected epitaxial/topotaxial growth, or the same type of crystal orientation based on the fabrication differences between Blangero and Shim that I discuss above. I note that Petitioner does not addresses the differences in the methods of fabrication (or how a POSITA would have combined them, if at all), and, thus, in my opinion, has not shown that the claimed crystal alignment limitation would have been obvious over the prior art.

85. Thus, in my opinion, Petitioner has not demonstrated that it would have been obvious to a POSITA that epitaxial growth of Shim's Li_2TiO_3 on the surface of Blangero's LiCoO_2 would have necessarily occurred based on the teachings of Shim due to the different fabrication methods of Blangero and Shim.

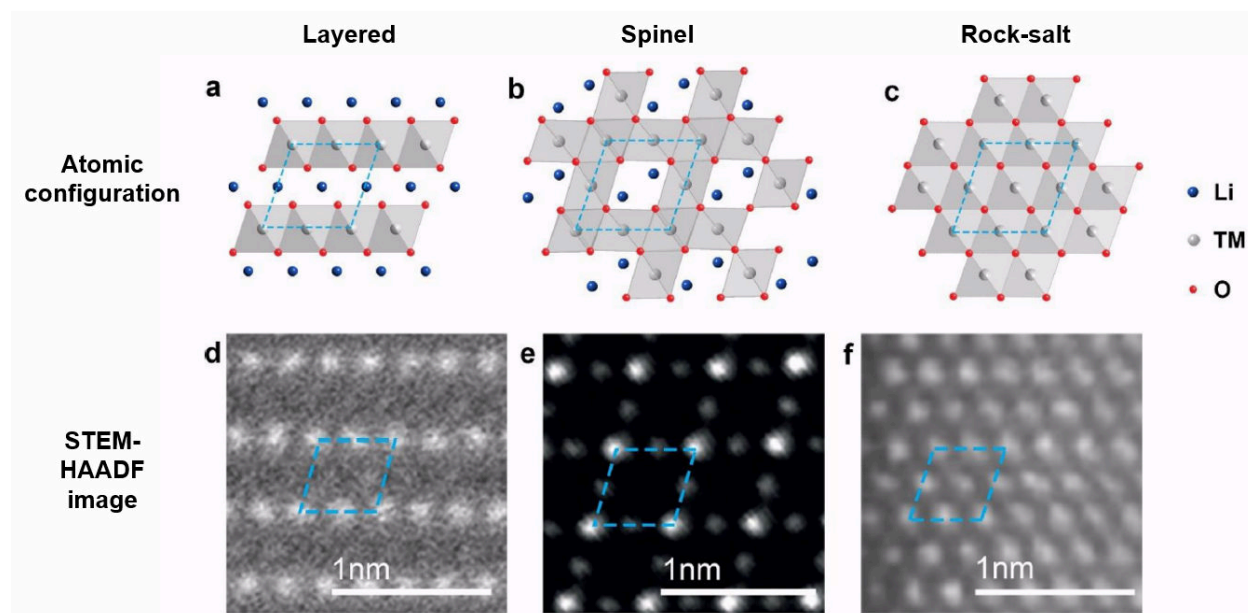
iii. Shim teaches the use of a spinel crystal structure in the lithium titanium oxide coating

86. I understand that the Petition alleges that "Shim discloses a coating (*i.e.*, second region) containing Li_2TiO_3 and magnesium (similar to Blangero's second region) with a crystal structure in substantial alignment with that of Shim's LiCoO_2 (*i.e.*, first region)," reproducing Figures 4B and 5B from Shim. Pet., 34-36.

87. As an initial matter, I note that these two figures appear to show the results from using *prior art* coating methods, not Shim's invention that the Petition otherwise relies upon. Rather, Figures 4B and 5B are the results of Comparative Examples 2 and 6. *See* Ex. 1006, [0035], [0038].

88. More importantly, I note that to the extent that Figures 4B and 5B disclose either a layered rock-salt crystal structure or a rock-salt crystal structure, Shim does not suggest using those crystal structures in its coating. To the contrary, Shim instead teaches that the lithium titanium oxide "may be a compound having a *spinel* structure[.]" Ex. 1006, [0061]; *see also id.*, [0017] ("The lithium titanium oxide may have a spinel structure."). A spinel structure is distinct from the layered

rock-salt crystal structure and the rock-salt crystal structure required by the claims of the '660 Patent, as I show in the annotated figure below:



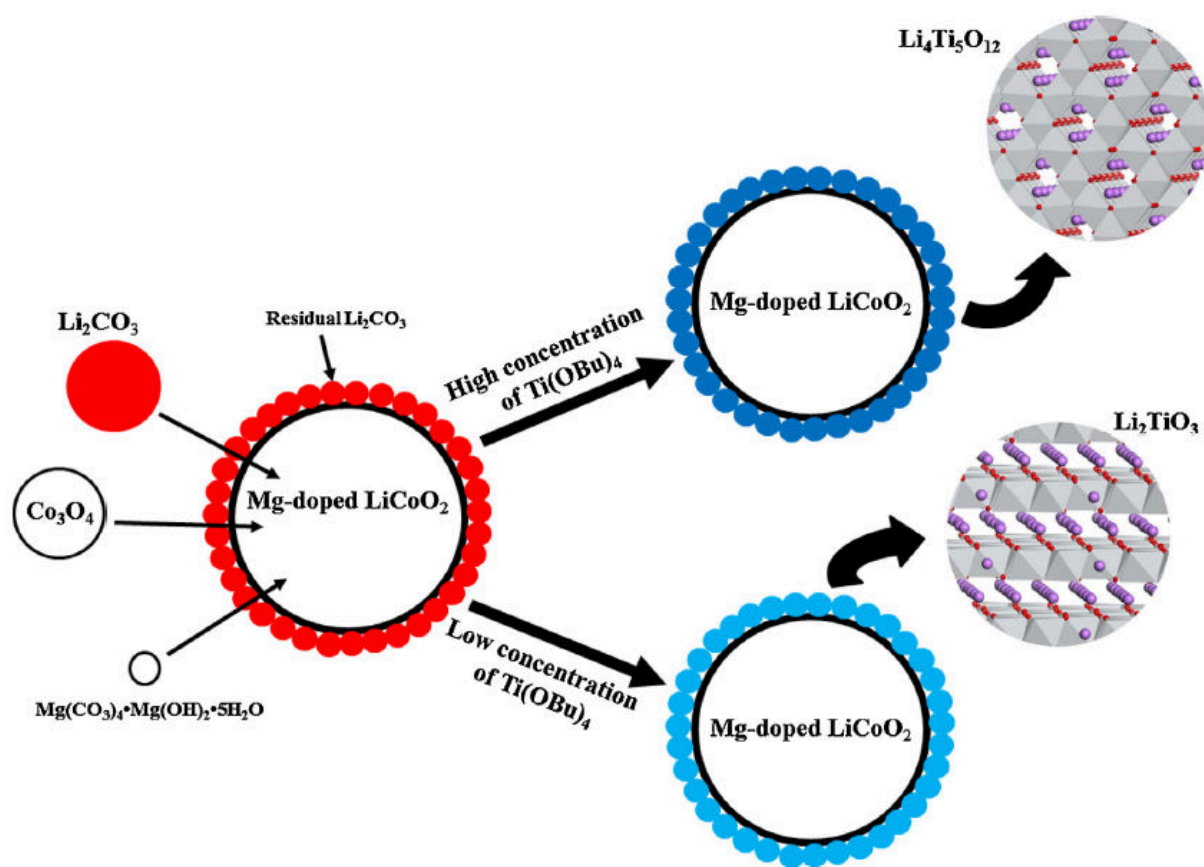
Adapted from Ex. 2004, 6²; *see also* Ex. 1032, 2 (Scheme 1). Critically, I note that Shim teaches that “unlike the compound having a layered structure,” a compound with a spinel structure “can remain stable (without a structural transition) during charge and discharge.” Ex. 1006, [0061]. Thus, in my opinion, a POSITA would understand Shim to teach that a spinel structure is not only a possible crystal

² STEM-HAADF, or scanning transmission electron microscope high-angle annular dark field imaging, is an imaging technique used to identify atomic structure.

structure, but, further, that the spinel structure is in fact the preferred crystal structure for a coating.

89. I note that Shim's preference for spinal crystal structures is evidenced by additional articles published by Shim,³ which confirm the benefits of using a coating material with a spinel structure. Petitioner even cites as one of its own exhibits a 2015 paper by Shim in *Electrochimica Acta* (Ex. 1032, "Shim 2015") that "revealed that two completely different lithium titanate phases (monoclinic Li_2TiO_3 and spinel $\text{Li}_4\text{Ti}_5\text{O}_{12}$) can be obtained as the coating materials depending on the concentration of a titanium source." Ex. 1032, 1. When low concentrations of $\text{Ti}(\text{OBu})_4$ are used, a layered structure (i.e. a layered rock-salt crystal structure) of Li_2TiO_3 is formed. *Id.*, 2. On the other hand, when high concentrations of $\text{Ti}(\text{OBu})_4$ are used, a spinel structure of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ is formed. *Id.* Shim demonstrates this phenomenon in Scheme 1, which I have reproduced below:

³ Both Shim 2015 and Shim 3 were disclosed to the Examiner during prosecution of the application that issued as the '660 Patent. *See* Ex. 1002, 263-265, 308-310.



Scheme 1. Synthesis and Coating Process of Lithium Titanate-Coated LiCoO₂.

Ex. 1032, 2 (Scheme 1).

I note that of the two coating materials, Shim 2015 explains that the spinel structured compound is superior, “secur[ing] better electrochemical performances than the Li₂TiO₃ one...due to its ‘zero-strain’ characteristic.” *Id.*; see also *id.*, 5 (“Therefore, it is considered that the coating layer of LT2-LCO_{Mg} (spinel Li₄Ti₅O₁₂) provides better performances for LiCoO₂ cathode than that of LT0-LCO_{Mg} (monoclinic Li₂TiO₃)”); *id.*, 8 (“Due to its excellent stability during charging/discharging, the spinel Li₄Ti₅O₁₂, which was obtained from the high concentration of Ti source, showed much better electrochemical performances as a coating material than the

monoclinic Li_2TiO_3 ”). Shim 2015 explains that the “discrepancy in effectiveness of the coatings” comes from the “structural or phase stability during cycling[.]” *Id.*, 5. Another article by Shim, (Ex. 2019 “Shim 3”) notes that “spinel materials are attractive in terms of their high Li^+ ion conductivity and good thermal stability” and they relieve “the difficulties associated with a Li-rich cathode, such as the operation voltage delay and thermal instability[.]” Ex. 2019, 1. Shim 3 speculates that the “spinel phase” can “function[] as a protection layer in order to prevent electrolytes from reacting with the LiCoO_2 materials,” “significantly enhanc[ing]” stability. *Id.*, 4; *see also id.*, 6-7 (noting “effective coating layer exhibits a dual role” by “prevent[ing] side reactions as a protection layer and function[ing] as conductive agent, thus improving the electrical behavior.”). It appears that Petitioner completely ignores the disclosures of a spinel structure in Shim. As such, in my opinion, the Petition fails to explain why a POSITA considering the Shim references would select a layered rock-salt crystal structure instead of the spinel structure that Shim teaches has superior and desirable properties.

90. Therefore, in my view, Petitioner has not shown that the combination of Blangero and Shim discloses or otherwise renders obvious the claimed crystal structure alignment.

3. The Petition Fails to Show that Blangero Alone or in Combination with Shim and/or Yamamoto Renders Obvious the Claimed Magnesium Distribution (Element 6.6)

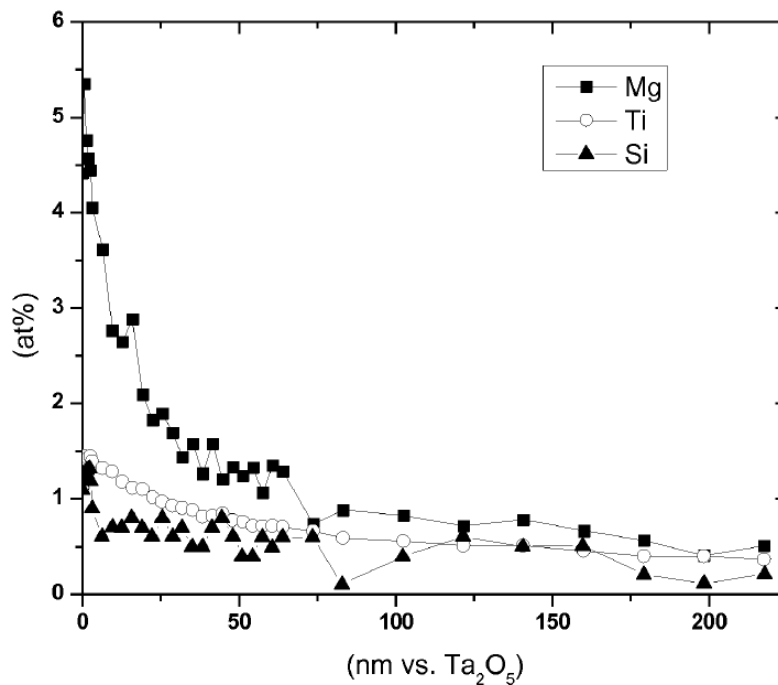
91. I understand that Petitioner asserts that “Blangero renders [Element 6.6] obvious either alone or in combination with Shim and/or Yamamoto.” Pet., 51. In my opinion, Petitioner’s assertion overlooks key differences between Blangero and Yamamoto⁴ that prevent Yamamoto’s disclosure from applying to Blangero’s PEAM.

92. First, I understand that it is undisputed that Blangero does not disclose measurements using energy dispersive X-ray (EDX) spectroscopy, as required by Element 6.6. *See* Ex. 1001, cl. 6. Instead, Blangero discloses the use of X-ray Photoelectron Spectroscopy (XPS), which I note is a different technique than EDX. In my opinion, the results of Blangero’s XPS and Yamamoto’s EDX spectroscopy highlight the fact that Blangero and Yamamoto disclose different coating methods that result in different distributions of magnesium on the cathode surface. I note that Yamamoto teaches a method for depositing MgO on “LiCoO₂ thin-film electrodes prepared via pulsed laser deposition[.]” Ex. 1007, 1. Blangero, on the other hand, does not use pulsed laser deposition, but instead uses the Ellenwood solid state

⁴ I note that Petitioner does not cite any specific disclosure in Shim (Ex. 1006) for Element 6.6, despite including Shim in the combination.

synthesis method that I describe above. Ex. 1005, [0109] (citing Ex. 2011). Additionally, in my opinion, Yamamoto's MgO coating does not necessarily apply to Blangero because, as discussed at length above, Blangero's "[a]uthors ***could not indisputably establish the chemical nature of the oxides formed at the surface and...possible forms...***not limited to...***MgO***[" Ex. 1005, [0062]. Therefore, the same flawed assumption that, in my view, is fatal to Petitioner's obviousness analysis of a rock-salt layer (Elements 1.4 and 6.4) and the alignment of the first and second regions (Elements 1.7 and 6.7) is equally fatal to Element 6.6.

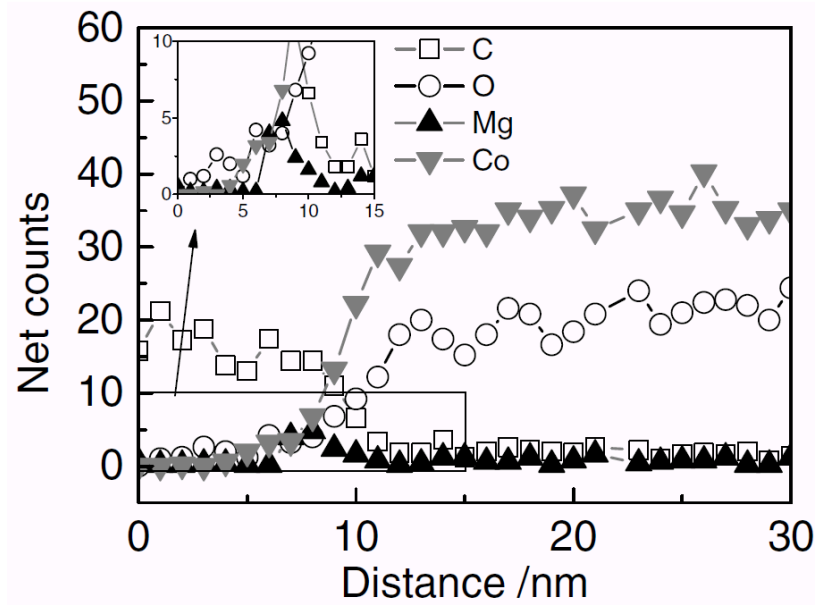
93. In my opinion, the respective data of Blangero and Yamamoto further demonstrate that the two coating methods yield different results. Figure 8 of Blangero demonstrates that, using Blangero's coating method, XPS determines that magnesium is present in substantial quantities (>1 at%) as far as 70 nm into the surface:



Ex. 1005, Fig. 8.

I note that Petitioner acknowledges this result. Pet., 46-47 (Blangero’s XPS spectrum “shows that the concentration of magnesium peaks at the surface (0 nm depth) of a coated LiCoO₂ particle” is ~5.4 atomic %, and that its concentration decreases to ~1 atomic % “over the first 50 nm of the particle.”).

94. Yamamoto’s EDX spectrum, on the other hand, demonstrates that Yamamoto’s coating technique resulted in magnesium being found in a narrow band just near the surface:



Ex. 1007, Fig. 1(b).

See also id., 2 (“The EDX scan detected a Mg containing layer of approximately 5 nm in thickness.”). Thus, in my opinion, Yamamoto and Blangero result in quite different distributions of magnesium into a surface. Petitioner’s allegation that “Blangero corroborates Yamamoto’s EDX line scan analysis” is therefore flawed. Pet., 46.

95. For this additional reason, in my opinion Petitioner has not shown that its Ground I.B prior art renders obvious claim 6.

B. Ground II is Deficient for Failing to Show that Cho Alone or in Combination with Kim Renders Obvious Independent Claims 1 and 6

96. In my opinion, Ground 2 is also deficient for at least four reasons. First, with respect to both independent claims 1 and 6, Petitioner’s argument that Cho or Kim disclose a PEAM containing titanium, magnesium, fluorine, and oxygen is

unsupported by their own disclosures, which never disclose fluorine present in the PEAM, and only disclose magnesium and titanium in a long laundry list of potential metals to use in the PEAM. Instead, Cho and Kim teach a POSITA to use nickel and manganese. Second, because Cho and Kim do not disclose the claimed elements, in my opinion they do not support a finding that when the claimed elements are present, they are present in either the claimed crystal structure or with the alignment of the crystal structure between the first and second regions claimed in independent claims 1 and 6. Third, with respect to claim 6, in my opinion the EDX spectra disclosed by Cho and Kim fail to show the claimed distribution of magnesium (as no magnesium is present), and further suggest that even if magnesium were present, it would not be present in the claimed distribution. Rather than decreasing to 1/5 of peak magnesium, Cho and Kim's data indicates that the compound would have relatively constant distribution within the PEAM.

1. Petition Fails to Show that Cho Alone or in Combination with Kim Renders Obvious “[a] positive electrode active material particle comprises titanium, magnesium, fluorine, and oxygen in the second region” (Elements 1.6 and 6.5)

i. Petitioner has not demonstrated that fluorine is present in Cho or Kim

97. At the outset, I note that none of the examples in Cho and Kim contemplates the inclusion of fluorine, as required by elements 1.6 and 6.5, in a

second region (e.g., the coating layer) of the particle. Petitioner does not argue otherwise.

98. Instead, I understand that Petitioner argues that Cho “teaches that the second region comprises fluorine, especially in view of a LiPF_6 electrolyte,” and therefore “[a] POSITA would have understood that such reactions with LiPF_6 would lead to the formation of a fluorine-coating layer on the cathode surface.” Pet., 68. Additionally, I understand that Petitioner argues that because Kim “explains that ‘a new solid electrolyte interface (SEI) layer, comprising *decomposition products of the electrolyte*, is formed along the active surface of the primary particles (grains) developed by the microcracks’ of the cathode material,” “when LiPF_6 is used in the electrolyte (as both Kim and Cho teach), Kim teaches that fluorine-containing layer is formed on the particle.” Pet., 68-69 (citing to Ex. 1009, 2–3).

99. Contrary to Petitioner’s arguments, however, Cho and Kim fail to disclose any particle having a second region (e.g., coating layer) containing fluorine. Although Petitioner alleges that a SEI layer containing fluorine is formed on the particle when decomposition occurs, in my opinion this position is contrary to the purpose of Cho and Kim, which are directed to *preventing* the formation of a SEI by using a coating, e.g., a Li/Co solution, to treat the outer surface of the particles. See e.g., Ex. 1009, 2 (“The *SEI layers on the surface of the grains inhibit electron and lithium transport* through the grain–electrolyte interface, thus diminishing the

electrical performance. *To resolve the structural and thermal instability* of the Ni-rich cathode materials, *many researchers have implemented surface modification such as surface coating*) (footnote omitted) (emphasize added), *id.* (“Herein, we propose...*surface treatment* of primary particles of...cathode material using a mixed ethanol solution with dissolved lithium and cobalt precursors. This new approach resulted in *improved structural and thermal stability*....”); Ex. 1008, 4:15-20 (“When the primary particles contact the electrolyte, a resistance layer, such as a *solid electrolyte interphase (SEI) layer, may be formed* on a surface of the primary particles, and the *electrochemical characteristics* of the cathode active material may *deteriorate*.”). In my view, neither Cho nor Kim discloses or even suggests that fluorine is present in the second region of the particle because the protective surface coatings that these references teach are designed to *prevent* such degradation, and, in turn, *prevent* fluorine from being deposited on the particle surface. I note that it was well known in the art as of the Critical Date that fluorine deposition was undesirable, and even harmful, to a cathode surface. *See* Ex. 1021, 2; *see also* Ex. 2004, 5-6. For at least this reason, in my opinion a POSITA would have understood that the presence of fluorine was undesirable and would have sought to avoid fluorine deposition.

100. I understand that Petitioner argues that Cho discloses a second region containing fluorine because Cho’s “coating layer may include [] a product obtained

by a sol-gel reaction of compositions including a *lithium salt*,” but then Petitioner appears to conflate this disclosure with Cho’s earlier discussion of LiPF_6 use in electrolyte solutions. Pet., 68 (emphasis in original). In my opinion, Petitioner’s arguments here are also flawed.

101. Contrary to Petitioner’s arguments, in my opinion, when a POSITA reads Cho as a whole, it is clear that Cho teaches certain salts associated with forming its coating layer, such as lithium salts and the cobalt salts that contain “each a nitrate, a sulfate, a chloride, or an acetate” and “each of which contains lithium and cobalt.” Ex. 1008, 6:23-28; *see also id.*, 6:50-55 (“The...particle **10** may be mixed with an ethanol solution including a lithium salt, such as lithium acetate, and a cobalt salt”). In contrast, Cho’s later discussion of LiPF_6 is directed to uses of lithium salts in electrolyte solutions—*not* the formation of Cho’s coating layer. *Id.*, 12:12-34 (“[A]n electrolyte may be prepared.... The organic electrolytic solution may be prepared by dissolving a lithium salt in an organic solvent.... The lithium salt may be any one of various lithium salts used in the art. Examples of the lithium salt include LiPF_6”). Accordingly, in my opinion, Petitioner has failed to show that Cho discloses or suggests the inclusion of fluorine in a second region.

102. Ultimately, I note that Petitioner has provided no actual evidence that fluorine is deposited, let alone that it forms the claimed crystal structure and

alignment. Petitioner has offered, again, only assumptions that are unsupported by Petitioner's prior art.

ii. Petitioner has not demonstrated that Mg or Ti are present in Cho and Kim

103. I understand that Petitioner alleges that Cho teaches elements 1.6 and 6.5 because “Cho teaches that its ‘coating layer may include a coating element compound, such as an *oxide* of a coating element,’ including Mg and Ti, ‘or a mixture thereof.’” Pet., 68.

104. In my opinion, Petitioner's arguments on this point are deficient. As I mentioned earlier, Cho is the patent underlying the Kim article, and both Cho and Kim show experimental data collected from LiNiCoMnO₂-based cathodes that include a coating that, notably, contains no magnesium or titanium, let alone fluorine. In fact, Kim does not disclose magnesium or titanium at all. Yet I see that Petitioner maps Cho's “coating layer 12a” to the claimed second region, despite the fact that none of the examples in Cho or Kim show a coating layer that contains Ti, Mg or F. See Ex. 1008, 14:59-60 (“a coating layer included a Li[Ni_{0.55}Co_{0.25}Mn_{0.2}]O₂ particle”), 15:12-14 (“coated Li[Ni_{0.55}Co_{0.25}Mn_{0.2}]O₂ particle”); *id.*, 15:23-24, 15:50-52; *see also* Ex. 1009, 2 (describes “surface treatment...using a mixed ethanol solution with dissolved lithium and cobalt precursors.”). Therefore, in my opinion a POSITA would not

have found it obvious to have arrived at the claimed “second region” composition based on the teachings of Cho and Kim.

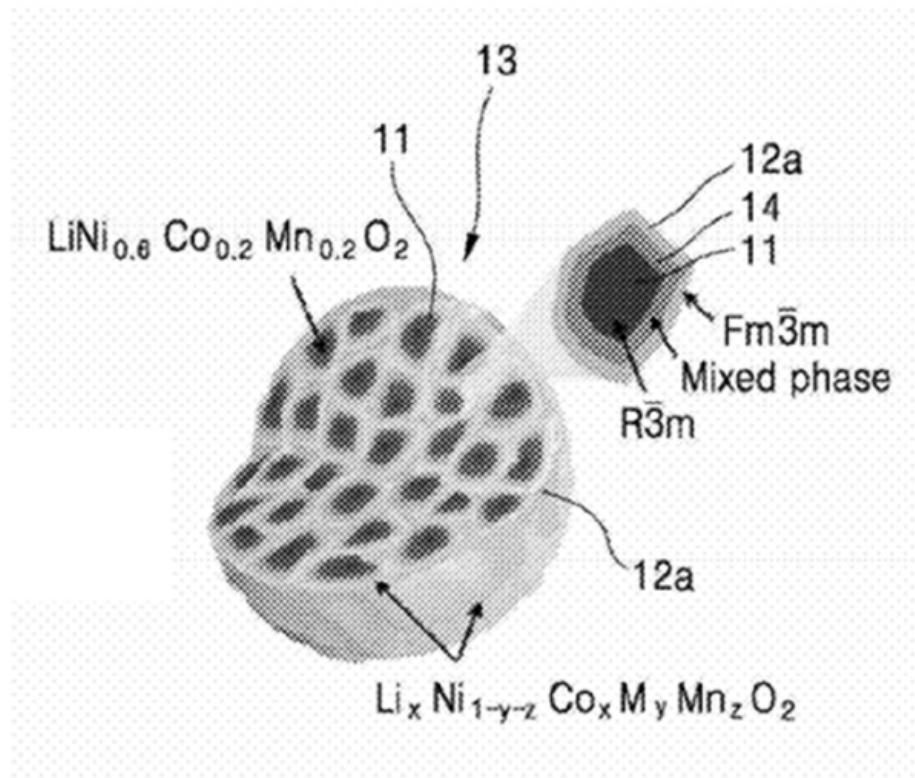
105. To arrive at the ’660 Patent’s claim limitation through Cho and Kim, Petitioner appears to rely on hindsight to pick out Ti and Mg from an expansive list of metals and combinations of metals provided in Cho’s disclosure—lists that I note are found nowhere in Kim. Cho discloses that its “coating layer may include a compound represented by Formula 2:”



where “M is a transition metal” can include at least 17 individual metals such as “one or more of Mg, Ca, Sr, Ba, Ti, Zr, Nb, Mo, W, Zn, Al, Si, Mn, Cr, Fe, V, or a rare earth element.” Ex. 1008, 7:55-60, 7:38-39. While I note that Ti and Mg are listed in Cho as potential transition metals, the number of possible combinations resulting from 17 potential transition metals in a formulation would end up being vast—resulting in 3.6×10^{14} possible combinations (e.g., $\text{Li}_a\text{Ni}_{1-x-y-z}\text{Co}_x\text{MgMn}_z\text{O}_2$, $\text{Li}_a\text{Ni}_{1-x-y-z}\text{Co}_x\text{CaMn}_z\text{O}_2$, $\text{Li}_a\text{Ni}_{1-x-y-z}\text{Co}_x\text{AlMn}_z\text{O}_2$, $\text{Li}_a\text{Ni}_{1-x-y-z}\text{Co}_x\text{MgCaMn}_z\text{O}_2$, etc...)—even without further accounting for other potential main elements (e.g., Li, Ni, Co, Mn, O₂). I note that Petitioner provides no rationale for why a POSITA would choose either magnesium or titanium, or why a POSITA would choose to use them together, particularly when all of Cho and Kim’s examples consistently use nickel and manganese instead.

2. Petition Fails to Show that Cho Alone or in Combination with Kim Renders Obvious that “[a] first region comprises a layered rock-salt crystal structure” and “[a] second region comprises a rock-salt crystal structure” (Elements 1.3-1.4 and 6.3-6.4)

106. I understand that Petitioner alleges that Cho discloses a “first region [that] comprises a layered rock-salt crystal structure” and “second region [that] comprises a rock-salt crystal structure” because “Cho’s first region has ‘a layered structure (a space structure of R-3m),’” and “Cho’s second region has ‘coating layer 12a’ with ‘rock-salt layer structure (a space group of Fm-3m),’” as depicted in Figure 1 (reproduced below). Pet., 63-67; Ex.1008, 7:7–14.



Ex. 1008, Fig. 1 (excerpted).

In my opinion, Petitioner's arguments have several deficiencies.

107. First, in my opinion, Petitioner's sole focus on Cho's Figure 1 is misleading, because it suggests to me that Cho's only disclosure is of a layered rock-salt crystal structure (i.e., space group R-3m) and a rock-salt crystal structure (i.e., space group Fm-3m). But that is not Cho's only disclosure. An inspection of Cho's Figure 1 shows that this example is specific to Ni-based active compositions, and more specifically, what are referred to as NCM compositions (i.e., LiMO_2 , with M = Ni, Co, and Mn). See Ex. 1008, 7:7-10 ("As shown in FIG. 1...a **coating layer 12a contains NCM** having a rock-salt layer structure (a space group of Fm3m) on a surface of the primary particle"); see also 6:53-65 ("The coating layer **12a** may include a compound represented by Formula 2 below: $\text{Li}_a\text{Ni}_{1-x-y-z}\text{Co}_x\text{M}_y\text{Mn}_z\text{O}_2$...[where] $0.9 < a < 1.5$, M is a transition metal, $0 < x \leq 0.3$, $0 \leq y \leq 0.1$, $0 < z \leq 0.3$."). But it was well-known that many of the cathode active materials disclosed by Cho, such as lithium iron phosphate (LiFePO_4), do **not** have a layered rock-salt crystal structure. Ex. 1008, 9:7-58; Ex. 2014, 13:55-56 ("Electrode materials with the olivine structure LiFePO_4 (triphylite)"); Ex. 2015, 1:60-63 (" LiFePO_4 [has] been most widely studied among olivine-structured compounds.").

108. While it might have been understood that Cho's Figure 1 and its examples are specific to crystal structures associated with Ni-based composite

cathode active material, in my opinion it would not have been obvious to a POSITA that those same crystal structures would be applicable to *other* “cathode active material[s]” known in the art. Ex. 1008, 9:16-20 (“The cathode may include...in addition to the composite cathode active material...a cathode active material used in the art.”), *id.*, 9:21-22 (“Examples of the cathode active material used in the art may include one or more of a lithium cobalt oxide...”). Accordingly, it is my opinion that a POSITA would not have understood nor found obvious that the known cathode active materials disclosed by Cho, such as lithium cobaltate, exhibit the same crystal structures as those associated with Cho’s specific Ni-based active materials.

109. Second, I note that the composition formulations depicted in Figure 1 are missing critical elements from the ’660 Patent’s claim limitations that a POSITA would understand were likely to affect the crystal structure. For example, Cho’s Figure 1 does not include fluorine, as required by claims 1 and 6 and as I discuss in more detail above. Moreover, I see the Petitioner points to nothing in Cho, in any of its example or disclosure, that suggests that the claimed second region, which requires titanium, magnesium, fluorine, and oxygen all together, would have the same crystal structure as shown in Cho’s limited depiction.

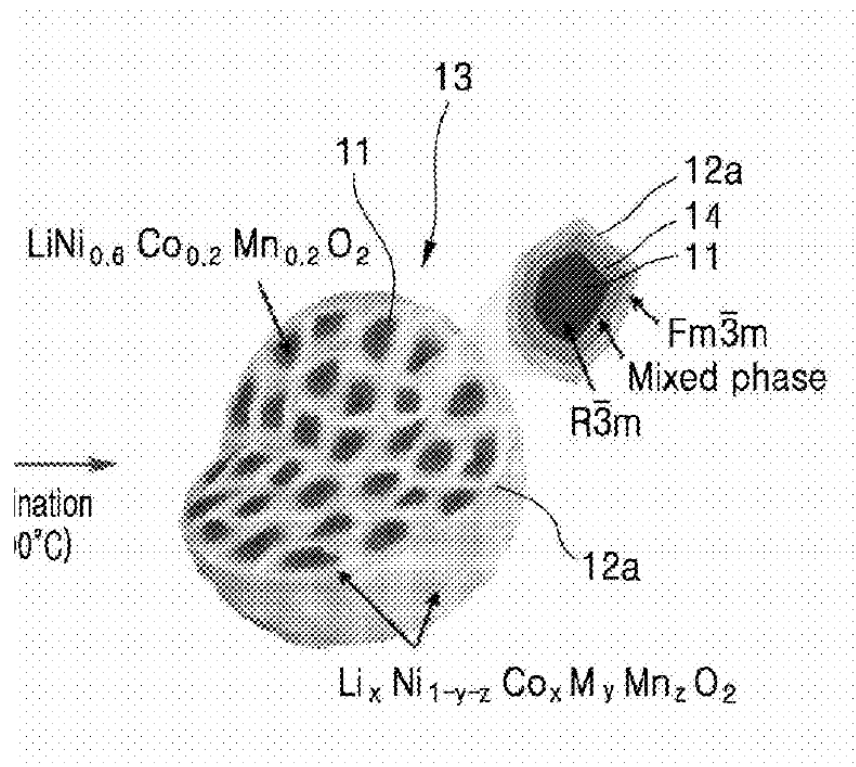
110. As one example, regarding Petitioner’s argument that the “second region comprises fluorine...in view of a LiPF_6 electrolyte” because “[a] POSITA would have understood that such reactions of LiPF_6 would lead to the formation of

a fluorine-coating layer on the cathode surface,” I note that nothing in Cho nor Kim teaches or suggests that fluorine is present in the second region, or that it originates from a decomposition product of LiPF_6 , let alone that it has the claimed rock-salt crystal structure. Pet., 68. In my opinion, the Petition entirely fails to address whether or how fluorine deposited through a decomposition process would have been expected to result in a rock-salt crystal structure. As discussed above, to the extent Petitioner argues that fluorine is deposited on the surface in the form of LiF , it is my view that a POSITA would have understood that LiF can also be present in amorphous form, and therefore is not limited to only rock-salt crystal structure. See Ex. 2005, 4-5; Ex. 2004, 5-6.

3. Petition Fails to Show that Cho Alone or in Combination with Kim Renders Obvious the Claimed Relative Crystal Structure (Elements 1.7 and 6.7)

111. I understand that Petitioner alleges that Cho teaches elements 1.7 and 6.7 because “Cho teaches a ‘cation-mixing layer’ that is also part of the second region, since it comprises the coating and ‘ha[s] a rock-salt phase’” (Pet., 66 (citing Ex. 1008, 5:48–52)), and “Cho’s Figure 7C demonstrates substantial alignment between the rock-salt crystal of the second region (cation mixing layer) and the layered rock-salt crystal of the first region” (Pet., 69). However, in my opinion, Petitioner’s arguments are flawed for several reasons.

112. Contrary to Petitioner's assertions, I note that Cho's "cation-mixing layer" is **not** part of Cho's coating layer. Instead, it seems to me that Petitioner is conflating Cho and Kim's disclosure of the "cation-mixing layer" and the "coating layer." To be clear, Cho expressly discloses that "the coating layer [is] formed on a surface of the primary particle," and that the primary particle itself—not the coating layer—can have a cation-mixing layer. Ex. 1008, 5:51-53 ("A surface phase **of the primary particle** may be a cation-mixing layer having a rock-salt phase"). Additionally, I note that Cho explains that "[a]n intermediate region 14 having a mixed phase may be formed **between** the primary particle 11 and the coating layer 12a." *Id.*, 7:12-14. But again, the "intermediate region 14" is distinguished as being a separate region from Cho's coating layer. *See id.*, Fig. 1, Claim 11 ("a mixed phase is between the primary particle and the coating layer."). I note that even Petitioner acknowledges that the "mixed phase" is formed **between** Cho's first and second regions. *See Pet.*, 63 ("As shown above, Cho teaches a first region with space group R-3m and second region 12a with space group Fm-3m. Moreover, **Cho teaches a 'mixed phase' formed between the two regions.**") (footnote and internal citations omitted).



Ex. 1008, Fig. 1 (excerpted).

113. But even if Cho’s “intermediate layer” were somehow considered to be part of Cho’s second region, i.e., the coating layer (which in my opinion it is not), Petitioner’s allegation that Cho and Kim render obvious elements 1.7 and 6.7 still fails. Pet., 69-70. Kim discloses that the crystal orientation of the first region (i.e., surface of the primary particles) and the second region (e.g., coating layer) associated with Ni-rich cathodes of the Cho-Kim combination would be *dissimilar* because the surface layer is composed of “primary particles (grains) [that] are aggregated” and “have different crystallographic orientation and slip planes.” Ex. 1009, 1; *see also id.* (“The Ni-rich cathode materials...are composed of round-shape

secondary particles in which many micrometer-sized *primary particles (grains) are aggregated*” and “all these primary particles have *different crystallographic orientation and slip planes*, anisotropic lattice volume expansion or contraction between the grains during lithium intercalation or deintercalation”). In my opinion, it would not have been obvious to a POSITA that any coating layer (or even an intermediate layer) would be in the same crystal orientation with an underlying second region made up of an aggregated underlying substrate having a plurality of “different crystallographic orientation and slip planes.”

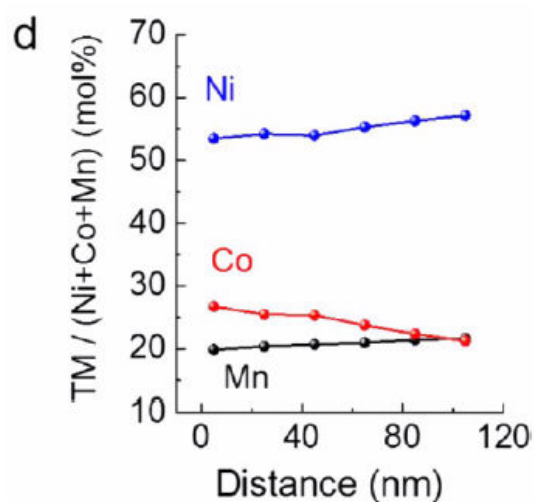
114. Lastly, I see that Petitioner also points to Figures 8B, but it is unclear to me why, since in my view this figure appears to be irrelevant. Figure 8B shows “Comparative Example 1,” which I note are *uncoated* NCM particles instead of NCM particles that include a coating layer. I understand that Petitioner mapped the coated NCM particles as containing the “second region” in the claims. *See* Pet., 63 (“Cho teaches a...second region 12a with space group Fm-3m.”) (citing to Ex. 1008, Fig. 1).

4. Petition Fails to Show that Cho Alone or in Combination with Kim and/or Yamamoto Renders Obvious the Claimed Magnesium Distribution (Element 6.6)

115. I understand that Element 6.6 requires Petitioner to show that some combination of Cho, Kim, and Yamamoto discloses “wherein the second region extends from a surface of the positive electrode active material particle to an inside

of the positive electrode active material particle where magnesium detected by a line analysis of energy dispersive X-ray spectroscopy (EDX) in a depth direction is $\frac{1}{5}$ of a maximum peak of the magnesium in the line analysis[.]” Ex. 1001, cl. 6. I understand that Petitioner argues that “Cho renders this element obvious.” Pet., 82. But as I discuss above, Cho does not disclose a *single* example using magnesium; instead, both Cho and Kim (i.e., the scientific article underlying Cho), *only* include data using nickel and manganese. See Ex. 1008, 14:59-60, 15:12-14, 15:23-24, 15:50-16:5; Ex. 1009, 2. I note that Kim is completely silent as to magnesium, and the only place magnesium can be found at all is in Cho, in a list of over seventeen different potential transition metals. Ex. 1008, 7:55-60, 7:38-39. Therefore, in my opinion, it follows that neither Cho nor Kim can disclose the required distribution of magnesium.

116. Additionally, to the extent magnesium is somehow incorporated into Cho or Kim’s particles, in my opinion the evidence in Cho and Kim indicates that the concentration of magnesium would more likely be *constant* across the particle. In Figure 1 of Kim (similar to Figures 5B and 6B in Cho), reproduced below, EDX spectra show roughly constant mass amounts of the three transition metals, Ni, Co, and Mn, as deep as 100 nm into Kim’s particle:



Ex. 1009, 2 (Fig. 1) (excerpted).

See also Ex. 1008, Fig. 5B, Fig. 6B. If we were to assume, as Petitioner argues, that magnesium is substituted for either nickel or manganese, then in my opinion the above figure suggests the distribution would be relatively constant. I note that Petitioner points to no evidence in Cho or Kim to suggest that, if magnesium were present, then at any point in the particle it would reach 1/5 of the maximum peak as required by the claims. Therefore, in my view, Petitioner's argument that "the concentration of magnesium in a PEAM particle is a result-effective variable whose optimization could have been discovered by routine optimization" is incorrect. Pet., 83. The concentration and distribution of magnesium will ultimately depend on the process that is used to create the PEAM particle.

117. I understand that Petitioner then relies upon Yamamoto's disclosure for its EDX line analysis. But Petitioner provides no explanation for how a POSITA

would combine Yamamoto with Cho and Kim, when they disclose different coating materials and different coating techniques. On the one hand, I note that Yamamoto only discloses using MgO as a coating material. Cho and Kim, on the other hand, disclose a coating that includes Li, Ni, Co, Mn, and O. If Petitioner's position is true (it is not, for the reasons I discuss above), then Cho and Kim disclose an even more complex coating layer that also includes Mg, Ti, and F. But nowhere does Cho or Kim disclose MgO as a coating. Therefore, it is not possible to draw conclusions about what the resulting composition would be when Yamamoto and Cho's coatings are combined.

118. Similarly, I note that Yamamoto and Cho disclose different coating processes. Cho and Kim use a "mixed ethanol solution with dissolved lithium and cobalt precursors." Ex. 1009, 2; Ex. 1008, 14:23-56. Yamamoto, on the other hand, uses "pulsed laser deposition" to apply extremely thin coatings of MgO. Ex. 1007, 1-2 ("The EDX scan detected a Mg containing layer of approximately 5 nm in thickness"). Petitioner provides no explanation for how these two techniques could possibly be combined, or why a POSITA would be motivated to combine them in the first place. Petitioner provides a cursory argument that their "complementary teachings" "would have involved routine implementation with no technical risks." Pet. 79, but in my opinion that is simply boilerplate language that provides no information about how a POSITA would actually have implemented the alleged

combination. In my view, Petitioner has not shown that Cho and Yamamoto should (or even could) be combined.

119. Additionally, Petitioner's attempts to rely on Yamamoto's EDX line analysis results fall short. I note that Petitioner provides no rationale for why a POSITA would look to Yamamoto for EDX results when Cho itself discloses EDX results for its particles. Moreover, as I noted above, Yamamoto's EDX line analysis is specific to particles with a thin layer of MgO applied using pulsed laser deposition, and Petitioner provides no explanation for how or why Yamamoto's EDX line analysis would apply to Cho, particularly when Cho's own EDX line analysis is shown to be markedly different.

120. Finally, in my opinion, Petitioner's argument that "a POSITA would have been motivated to combine Cho and Yamamoto for the reasons explained in Section X.B.1," (Pet., 83), falls flat because Section X.B.1 discusses a different set of prior art—i.e., a combination of Blangero, Shim and Yamamoto—and therefore is simply not applicable to the Ground II.C combination of Cho, Kim, and Yamamoto.

VII. CONCLUSION

121. 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.

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