

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

AMPEREX TECHNOLOGY LTD.
Petitioner,

v.

SEMICONDUCTOR ENERGY LABORATORY CO., LTD.
Patent Owner

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

Issued: June 22, 2021

Filed: July 8, 2020

Inventor: Teruaki Ochiai et al.

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

Case IPR2023-01349

DECLARATION OF TROY A. HAYES, PH.D., P.E.
IN SUPPORT OF PETITION FOR
***INTER PARTES* REVIEW OF U.S. PATENT NO. 11,043,660**

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1001	U.S. Patent No. 11,043,660 (“’660 Patent”)
1002	File History of U.S. Patent No. 11,043,660 (“’660 FH”)
1003	Declaration of Dr. Troy Hayes in Support of <i>Inter Partes Review</i> of U.S. Patent No. 11,043,660
1004	<i>Curriculum Vitae</i> of Dr. Troy Hayes
1005	U.S. Patent Pub. No. 2014/0212759 A1 (“Blangero”)
1006	U.S. Patent Pub. No. 2016/0149215 A1 (“Shim”)
1007	Yamamoto et al., <i>Stabilization of the Electronic Structure at the Cathode/Electrolyte Interface via MgO Ultra-thin Layer During Lithium-ions Insertion/Extraction</i> , 82 <i>Electrochemistry</i> , 891–96 (2014) (“Yamamoto”)
1008	U.S. Patent No. 10,283,770 (“Cho”)
1009	Kim et al., <i>A New Coating Method for Alleviating Surface Degradation of $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{MnO}_2$ Cathode Material: Nanoscale Surface Treatment of Primary Particles</i> , 15 <i>Nano Lett.</i> , 2111–19 (2015) (“Kim”)
1010	Shim et al., <i>Effects of MgO Coating on the Structural and Electrochemical Characteristics of LiCoO_2 as Cathode Materials for Lithium Ion Battery</i> , 26 <i>Chem. Mater.</i> 2537–43 (2014)
1011	Yu et al., <i>The Investigation of Ti-Modified LiCoO_2 Materials for Lithium Ion Battery</i> , 262 <i>Journal of Power Sources</i> , 136–39 (2014) (“Yu”)
1012	Lee & Park, <i>Interface Characterization of MgF_2-Coated LiCoO_2 Thin Films</i> , 230 <i>Solid State Ionics</i> , 86–91 (2013) (“Lee”)

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1013	Ashcroft & Mermin, SOLID STATE PHYSICS, Cengage Learning, 1st ed. (1976)
1014	Int'l Patent No. WO 2013/048048
1015	U.S. Patent No. 9,048,495 B2
1016	CN 102610806 (translated)
1017	U.S. Patent Application 2007/0224506 A1
1018	Dedryvère et al., <i>Surface Film Formation on Electrodes in a LiCoO₂/Graphite Cell: A Step by Step XPS Study</i> , 174 Journal of Power Sources, 462–68 (2007)
1019	Solano, <i>Development of Artificial Surface Layers for Thin Film Cathode Materials</i> , Material Chemistry, Université de Bordeaux (2015)
1020	Aurbach et al., <i>On the Capacity Fading of LiCoO₂ Intercalation Electrodes: The Effect of Cycling, Storage, Temperature, and Surface Film Forming Additives</i> , 47 Electrochimica Acta, 4291–306 (2002)
1021	Aykol et al., <i>Thermodynamic Aspects of Cathode Coatings for Lithium-Ion Batteries</i> , Advanced Energy Materials, 1400690 (2014)
1022	Kitagawa et al., <i>Application of Fluorine-containing Solvents in High Voltage Operation</i> , 78 Electrochemistry, 345–48 (2010)
1023	Sun et al., <i>Role of AlF₃ Coating on LiCoO₂ Particles During Cycling to Cutoff Voltage Above 4.5 V</i> , 156 Journal of the Electrochemical Society, A1005–10 (2009)
1024	Aboulaich et al., <i>High Voltage Stability and Enhanced Electrochemical Performances of LiCoO₂ by CeF₃ Coating</i> , IEEE (2015)

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1025	M.C. Rao, <i>Structure and Surface Morphology of LiCoO₂ Thin Film Cathodes Prepared by Pulsed Laser Deposition</i> , 2 J. Intense Pulsed Lasers & Applications in Advanced Physics, 35–38 (2012)
1026	Lu et al., <i>Probing the Origin of Enhanced Stability of “AlPO₄” Nanoparticle Coated LiCoO₂ During Cycling to High Voltages: Combined XRD and XPS Studies</i> , 21 Chem. Mater., 4408–24 (2009)
1027	JP2012-018914 (“Patent Document 1”) (translated)
1028	JP2015-201432 (“Patent Document 2”) (translated)
1029	Wang et al., <i>Infiltrative Coating of LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ Microspheres with Layer-Structured LiTiO₂: Toward Superior Cycling Performances for Li-ion Batteries</i> , J. Mater. Chem. A (2012)
1030	Xu et al., <i>Lithium Transport Through Lithium-ion Battery Cathode Coatings</i> , 3 J. Mater. Chem. A, 17248–72 (2015)
1031	Bai et al., <i>Performance Improvement of LiCoO₂ by Molten Salt Surface Modification</i> , 167 J. Power Sources, 504–09 (2007)
1032	Shim et al., <i>Synergistic Effects Of Coating and Doping for Lithium Ion Battery Cathode Materials: Synthesis And Characterization Of Lithium Titanate-Coated LiCoO₂ with Mg Doping</i> , Electrochimica Acta 186, 201–08 (2015)
1033	Mizushima et al., <i>A New Cathode Material for Batteries of High Energy Density</i> , Mat. Res. Bull., 783–89 (1980)
1034	Yang et al., <i>Hybrid Microwave Synthesis and Characterization of the Compounds in the Li-Ti-O System</i> , 175 J. Power Sources 1, 575–80 (2008)

Exhibit	Description
1035	Li et al., <i>Lithium Titanate Epitaxial Coating on Spinel Lithium Manganese Oxide Surface for Improving the Performance of Lithium Storage Capability</i> , 6 ACS Appl. Mater. Interfaces 21, 18742–50 (2014)
1036	Eom et al., <i>M₃ (PO₄)₂-Nanoparticle-Coated LiCoO₂ vs LiCo_{0.96}M_{0.04}O₂ (M = Mg and Zn) on Electrochemical and Storage Characteristics</i> , 155 Journal of the Electrochemical Society 3, A201–05 (2008)
1037	Kleykamp, <i>Phase Equilibria in the Li-Ti-O System and Physical Properties of Li₂TiO₃</i> , 61–62 Fusion Engineering and Design, 361–66 (2002)
1038	Bai et al., <i>Performance Improvement of LiCoO₂ by MgF₂ Surface Modification and Mechanism Exploration</i> , 134 Electrochimica Acta, 347–54 (2014)
1039	Katsui et al., <i>Epitaxial Growth of (104)- and (018)-Oriented LiCoO₂ films on MgO Single Crystals Prepared by Chemical Vapor Deposition</i> , 218 Surface and Coatings Technology, 57–61 (2013)
1040	Kumar, A. and Kumar, J., <i>On the Synthesis and Optical Absorption Studies of Nano-Sized Magnesium Oxide Powder</i> , 69 Journal of Physics and Chemistry of Solids 11, 2764–72 (2008)
1041	Declaration of Jennifer Babbitt in Support of <i>Inter Partes</i> Review of U.S. Patent No. 11,043,660
1042	Federal Court Management Statistics (June 2023)
1043	Time to Trial, Virginia Eastern District (September 2, 2018–September 2, 2023), <i>available at</i> https://search.docketnavigator.com/ (last accessed September 2, 2023)

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

I. ASSIGNMENT

1. I have been retained as an expert witness on behalf of Amperex Technology Ltd. (“Amperex” or “Petitioner”) to offer technical opinions in connection with the above-captioned Petition for Inter Partes Review (“IPR”) of U.S. Patent No. 11,043,660 (“the ’660 Patent”) (Ex. 1001).

2. I have been asked to provide my independent analysis of the ’660 Patent in light of the prior art patents and publications cited below.

3. I have been asked to provide my opinions regarding whether claims 1 through 10 of the ’660 Patent (the “Challenged Claims”) are invalid as being anticipated and/or obvious to a person having ordinary skill in the art (a POSITA) at the time of the alleged invention.

4. In preparing my Declaration, I reviewed the ’660 Patent, the file history of the patent (Ex. 1002), prior art references, technical references, and other publications from the time of the alleged invention, which are discussed herein.

5. For the purposes of my Declaration, I have been asked to assume that the priority date of the alleged invention recited in the ’660 Patent is July 5, 2016 (hereinafter the “Priority Date”).

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

compensation based on my time actually spent analyzing the '660 Patent, the prior art patents and publications cited below, and issues related thereto, and I will not receive any added compensation based on the outcome of any IPR or other proceeding involving the '660 Patent. I have no financial interest in Amperex.

II. BACKGROUND AND QUALIFICATIONS

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

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

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

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

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

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

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

evaluations also included extensive discussions with battery factory personnel related to battery design and manufacturing before and after the factory visits to drive corrective action in order to improve the battery design and manufacturing quality. These discussions and evaluations include but are not limited to the positive and negative electrode material chemistry and composition, electrolyte composition and additives, as well as the separator design. My experience at the factory level began in 2005 and includes evaluations of factories and the associated existing and forward-looking battery technologies of manufacturers in China, Taiwan, Korea, Japan, Indonesia, the United States, France, Germany, and Switzerland. I developed Exponent's lithium-ion battery factory evaluation program and, through discussions with my teams, meetings with clients, and reviews of reports, have been directly involved in evaluations of many additional factories and associated battery designs. Through projects working with original equipment manufacturers (OEMs) and battery cell and pack manufacturers starting in 2005, I am also very familiar with the trends and motivations in the battery industry, including trends related to battery separator design as well as electrolyte chemistry and positive and negative battery electrode materials. My projects involved discussions with OEMs and battery manufacturers related to improvements in battery design, including separator characteristics, electrolyte composition and additives, as well as positive and

negative electrode chemistry and design. This includes work with many consumer electronics OEMs and their associated battery manufacturing partners.

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

14. I have presented at more than two dozen conferences, seminars, webinars, training courses, or workshops on various topics pertaining to battery design, manufacturing, and failure. Exemplary topics I have presented on include “How Do You Choose a Battery Manufacturer for Your Device? Part 2: Cell and

Pack Manufacturing Audits”; “Catastrophic Thermal Battery Failures in Vaping Devices and in Pockets: Why Do They Happen?”; “Considerations When Choosing Batteries Made in China”; “Detecting Failures and Manufacturing Defects in Li-ion Batteries”; “Manufacturing, Testing and Recycling Safe EV Li-ion Batteries”; “Analyzing Electronic Product Battery Failures”; “Key Manufacturing Practices to Achieve High Quality Cells”; “Design and Manufacturing of Safe Lithium Ion Batteries for EVs”; “Identifying a Non-safe Li-ion Battery in the Production Cycle to Avoid Safety Recalls and Maintaining Your Brand’s Trust”; “Methodologies of Identifying Root Causes of Failures in Lithium Ion Battery Packs”; and “Use of CT Scanning for Defect Detection in Lithium Ion Batteries.”

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

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

III. MATERIALS AND OTHER INFORMATION CONSIDERED

17. In forming the opinions expressed in this Declaration, I relied upon my education and many years of experience in the relevant field of the art and have considered the viewpoint of a person having ordinary skill in the art (a POSITA) as of the Priority Date of the '660 Patent.

18. I have considered the materials referenced in this Declaration, including the '660 Patent, its file history, and other documents listed in the Exhibit List to the '660 Petition, reproduced above. In particular, I have considered the prior art references listed below.

19. U.S. Patent Application Number 2014/0212759 A1 ("**Blangero**") (Ex. 1005) was filed on May 29, 2012 and published on July 31, 2014 and is prior art under AIA 35 U.S.C. § 102(a)(2).

20. U.S. Patent Application Number 2016/0149215 A1 ("**Shim**") (Ex. 1006) was filed on November 20, 2015, and published on May 26, 2016 and is prior art under AIA 35 U.S.C. § 102(a)(2).

21. Yamamoto et al., *Stabilization of the Electronic Structure at the Cathode/Electrolyte Interface via MgO Ultra-thin Layer During Lithium-ions Insertion/Extraction* ("**Yamamoto**") (Ex. 1007) was published on October 5, 2014 and is prior art under AIA 35 U.S.C. § 102(a)(1).

22. U.S. Patent No. 10,283,770 (“**Cho**”) (Ex. 1008) is a United States Patent effectively filed December 17, 2015, and is prior art under AIA 35 U.S.C. §§ 102(a)(2) and 102(d).

23. Kim et al., *A New Coating Method for Alleviating Surface Degradation of LiNi_{0.6}Co_{0.2}MnO₂ Cathode Material: Nanoscale Surface Treatment of Primary Particles* (“**Kim**”) (Ex. 1009) was published on February 10, 2015, and is prior art under AIA 35 U.S.C. § 102(a)(1).

24. These prior art were not considered by the Examiner during prosecution and predate the ’660 Patent, which claims priority to a foreign application filed on July 5, 2016. Blangero and Yamamoto, and Blangero were previously before the Examiner during prosecution, but were not discussed or relied upon in an office action. See generally Ex. 1002.

IV. UNDERSTANDING OF PATENT LAW

25. I understand that prior art to the ’660 Patent includes patents and printed publications in the relevant art that predate the Priority Date of the ’660 Patent.

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

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

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

29. I understand that certain factors—often called “secondary considerations”—may support or rebut an assertion of obviousness of a claim. I understand that such secondary considerations include, among other things, commercial success of the alleged invention, skepticism of those having ordinary skill in the art at the time of the alleged invention, unexpected results of the alleged invention, any long-felt but unsolved need in the art that was satisfied by the alleged invention, the failure of others to make the alleged invention, praise of the alleged invention by those having ordinary skill in the art, and copying of the alleged invention by others in the field. I further understand that there must be a nexus—a connection—between any such secondary considerations and the alleged invention.

I also understand that contemporaneous and independent invention by others is a secondary consideration tending to show obviousness.

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

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

- Combining prior art elements according to known methods to yield predictable results;
- Simple substitution of one known element for another to obtain predictable results;

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

V. SUMMARY OF OPINIONS

32. It is my opinion that Claims 1–5 of the ’660 Patent are rendered obvious by Blangero alone or in combination with Shim.

33. It further my opinion that Claims 6–10 of the ’660 Patent are rendered obvious by Blangero alone or in combination with Shim and/or Yamamoto.

34. It is further my opinion that Claims 1–3 of the ’660 Patent are rendered obvious by Cho, either alone or in combination with Kim.

35. It is further my opinion that Claims 4 and 5 of the ’660 Patent are rendered obvious by Cho, either alone or in combination with Kim and/or in combination with Blangero.

36. It is further my opinion that 6–8 of the '660 Patent are rendered obvious by Cho, either alone or in combination with Kim and/or in combination with Yamamoto.

37. It is further my opinion that Claims 9 and 10 of the '660 Patent are rendered obvious by Cho, either alone or in combination with Kim and/or Yamamoto and/or Blangero.

VI. OVERVIEW OF TECHNOLOGY AND OF THE '660 PATENT

A. Introduction to the Technology

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

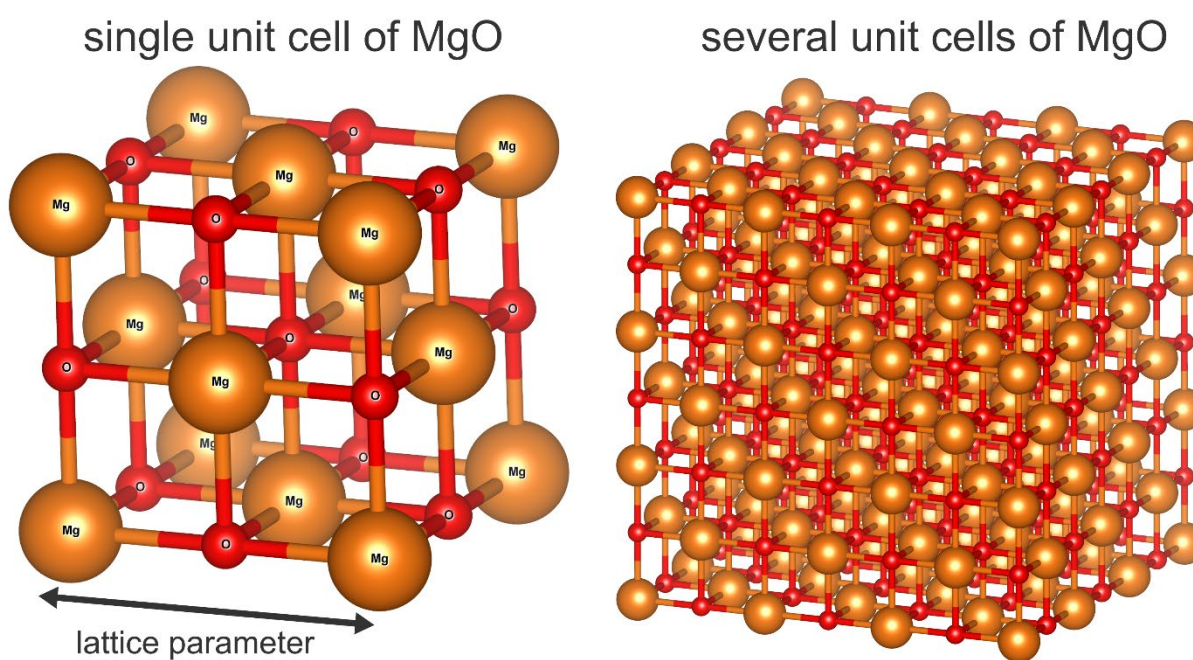
39. To help provide context to the '660 Patent, I provide a brief review of relevant lithium-ion battery development as well as the underlying materials science concepts and characterization techniques.

1. Crystal Structures Generally

40. Crystalline solids consist of atoms arranged in a three-dimensional repeating structure. While individual crystals commonly contain many trillions of atoms, crystal structures can be simplified by identifying the repeating building block—or “unit cell”—which is repeated in three dimensions. The length of the unit cell is known as the lattice parameter, and an idealized crystal can be constructed by translating the unit cell along its three principal axes. The crystal structure can also be described by its symmetries, commonly referred to as its space group, expressed in a specific space group notation.

41. Using the rock salt crystal structure as an example, the unit cell is a cube with cations at the corner and face center sites, and anions at the edge centers and body center. Equivalently, the cations and anions both form face-centered-cubic (“FCC”) crystal structures offset one half lattice parameter along a principal axis. Rock salt therefore consists of two intertwined FCC structures with the anions in the interstitial sites of the cations (spaces between cation positions) and the cations in the interstitial sites of the anions (spaces between anion positions). The binary rock salt structure belongs to the space group $Fm\bar{3}m$. The F in the space group indicates

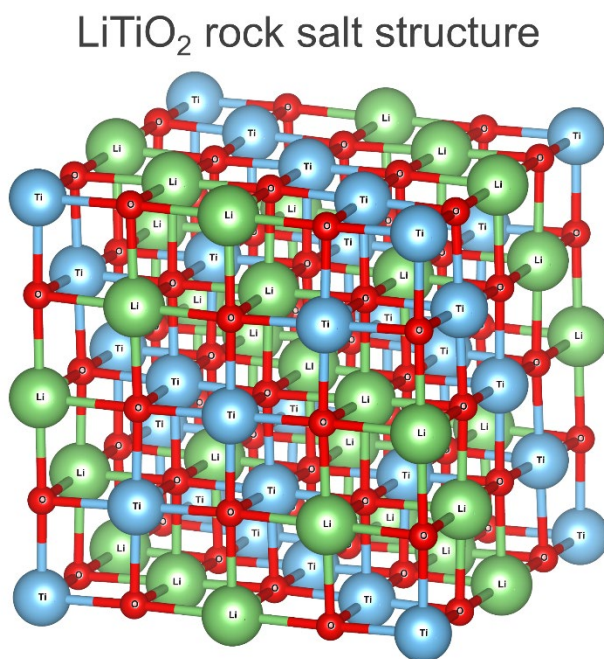
that the structure is FCC, the $\bar{3}$ (read as “three bar”) describes the four 3-fold rotoinversion symmetry axes, and the two m ’s describe mirror planes. The unit cell of magnesium oxide (MgO)—one of many compounds known to form the rock salt structure—is shown on the left of the following figure. The right image demonstrates the periodic repetition of unit cells to construct a crystal structure.



2. Rock Salt and Layered Rock Salt Structures

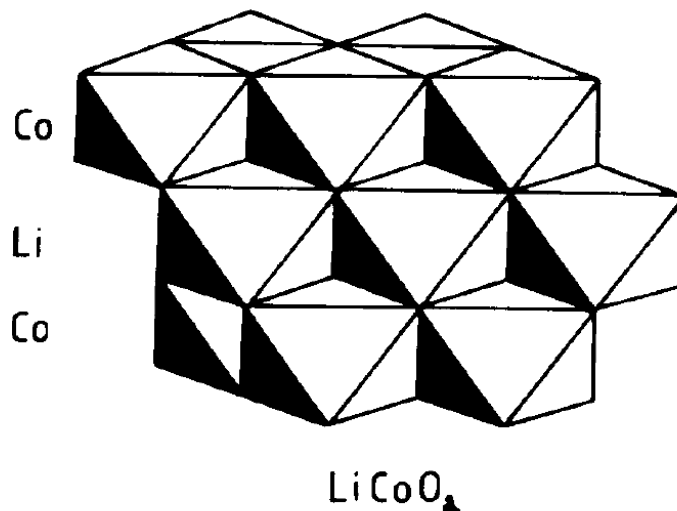
42. The rock salt crystal structure is common among ionic compounds. Salts such as sodium chloride (NaCl), lithium fluoride (LiF), and cesium fluoride (CsF), and metal oxides such as magnesium oxide (MgO) and calcium oxide (CaO) are examples of substances that adopt the rock salt structure. These are all specifically binary rock salts with one anionic element and one cationic element. However, rock-salt-like structures can also form with multiple cationic elements

such as rock-salt-like lithium titanate (LiTiO_2), where lithium and titanium atoms alternate in the cation sites of the rock salt structure, as shown below. The two cations of LiTiO_2 lower the symmetry of the structure to the space group $Fd\bar{3}m$.



43. A similar rock-salt-like structure that has received a great deal of attention is the layered rock salt structure of LiCoO_2 , or “lithium cobalt oxide” (“LCO”). In 1980, the late Nobel Prize winner, John B. Goodenough, and collaborators reported that LiCoO_2 can reversibly extract and insert lithium ions as “A New Cathode Material for Batteries of High Energy Density.” With this report, the authors introduced a new family of cathode materials. They described these “layered rocksalt” or “ordered-rocksalt” structures as containing “close-packed

oxygen arrays” where the “ Li^+ and M^{3+} ions [where M is cobalt or another transition-metal] occupy alternate (111) layers of the rocksalt structure.” Ex. 1033, 2. Figure 1 from this paper illustrating the layered rock salt structure of LiCoO_2 is shown below.

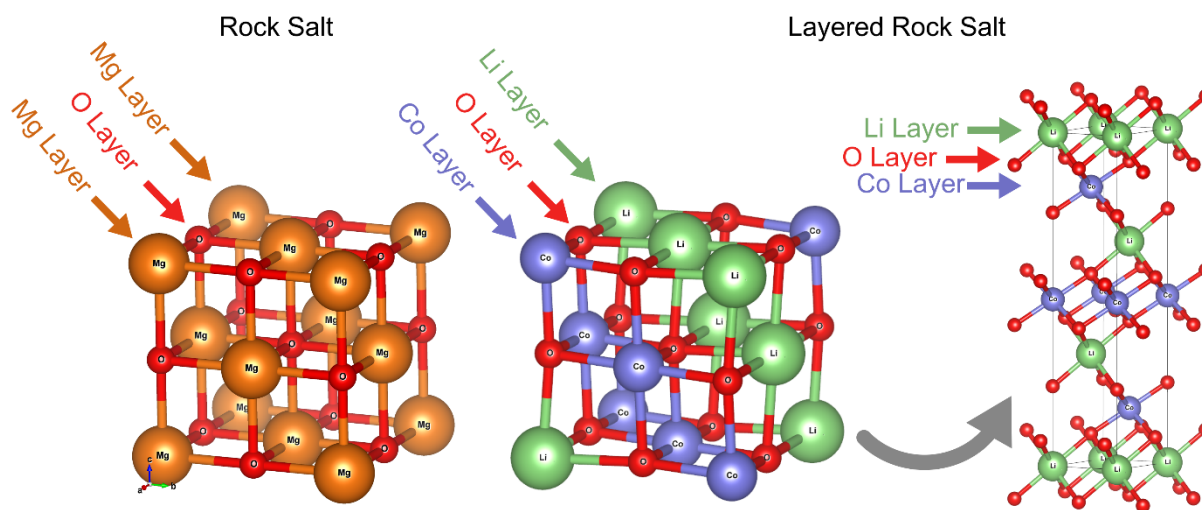


Id., Fig. 1.

44. Since this first paper introducing LiCoO_2 as a cathode material, many other layered rock salt structures have been developed for lithium-ion batteries, including analogues where cobalt is partially substituted with other elements such as manganese, nickel, and aluminum. The most common analogues are $\text{LiNi}_x\text{Co}_y\text{Al}_z\text{O}_2$ abbreviated as “NCA” and $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$ abbreviated as “NMC” or “NCM,” where $x+y+z = 1$.

45. As shown below, LiCoO_2 can be depicted as a rock-salt-like structure with alternating diagonal layers of lithium and cobalt atoms or from a rotated

perspective as a layered material with rhombohedral or trigonal symmetry. Note that, due to the alternating lithium and cobalt layers that break three of the four 3-fold rotoinversion symmetry axes, LiCoO_2 does not have the same symmetry as binary rock salts (e.g., MgO). Rather, LiCoO_2 belongs to the space group $R\bar{3}m$, where R indicates “rhombohedral” which is equivalent to “trigonal.”



3. Interfaces and Crystal Alignment

46. An interface is the “internal surface” between two materials in contact with one another, such as the interface between a painted metal surface and the paint on that surface or, similarly, the interface between a cathode particle and its coating. When two crystalline materials have sufficiently similar crystal structures and

similar lattice parameters¹, crystallographic alignment of the two structures is energetically favorable. If an interface between such materials is exposed to high enough temperatures to enable substantial atomic mobility—either during the coating process or afterwards during annealing—it was well known that the coating would become crystallographically aligned to the underlying material (such as a cathode particle) to minimize interfacial energy.

47. To characterize the crystallographic alignment of two materials at an interface, techniques sensitive to crystallographic alignment are necessary. Transmission electron microscopy (TEM) is therefore commonly used to investigate the crystallographic alignment between particles and their coatings.

4. Transmission Electron Microscopy

¹ More inclusive conditions can be used to predict when two materials are energetically preferred to be crystallographically aligned at an interface. This includes using multiples (or fractions) of unit cells, comparing the symmetries of the anion sublattices, measuring anion separation distances, comparing the (approximate) periodicity of atoms in specified planes, and using analogies to well-studied materials.

48. TEM is a characterization technique used to study materials at length scales from around 0.00001 m (10 μm) to the atomic scale, less than 0.0000000001 m ($< 1 \text{ \AA}$). TEM was first demonstrated in the 1930s, and has been regarded as an important tool for studying the basic structure of materials and interfaces for many decades, including long before July 5, 2016, which I understand to be the assumed priority date of the '660 Patent.

49. A TEM operates by directing a beam of electrons at a thin sample. A fraction of these electrons is transmitted through the sample and reach the detector, whereas the rest are scattered in other directions or absorbed by the sample. The electrons may illuminate the full area to be imaged and then be collected on a screen or a camera, similar to a traditional light microscope, or they may be focused into a fine probe which is then scanned over the sample with the electrons collected in a series of detectors. The scanning mode of operation is referred to as scanning transmission electron microscopy (STEM).

50. When the electron beam interacts with the sample, several different physical phenomena can occur. In a typical imaging scenario, the most likely outcome for any given electron is that it will be transmitted without interacting, in which case it will follow a straight trajectory through the sample along the axis of the instrument. Images formed with the transmitted electrons are termed “brightfield” images. In contrast, it is possible through the use of apertures or

detector placement to only collect electrons which have been scattered away from the transmitted electron beam. These images are broadly termed “darkfield” images. One particular type of darkfield image is collected in a STEM by using an annular detector (basically a ring detector) placed such that it collects a cone of electrons scattered at a high angle away from the transmitted beam. These images are termed high-angle annular darkfield (HAADF) images.

51. The contrast of an image—*i.e.*, the difference between lighter and darker areas—is determined by which sorts of electrons are collected. In brightfield images, areas that are thicker or composed of heavier elements will be dark because the electrons are less likely to be transmitted through those areas. HAADF is a popular method because the chance of an electron to be scattered into very high angles depends primarily on the atomic number of the atoms the electron beam interacts with, and elements further down the periodic table produce brighter contrast. It is possible with HAADF images to distinguish between different elements, and areas with heavier elements look brighter.

a. Electron Diffraction

52. Electron diffraction, often referred to as selective-area electron diffraction (SAED) when used in TEM, is a technique that exploits the wavelike properties of electrons to extract atomic structural information about the material

under study. When used in conjunction with TEM, it allows researchers to study the arrangement of atoms in materials with very high spatial resolution.

53. In electron diffraction, a beam of electrons strikes the sample. The electrons interact with the atoms in the sample, causing them to scatter in different directions. When striking different planes of atoms within a periodic lattice, some scattering angles are made much more likely than others due to the process of diffraction. The scattering angles which are enhanced by diffraction are described by Bragg's Law, or:

$$n\lambda = 2d\sin\theta$$

where n is the diffraction order, which is any positive integer, λ is the wavelength of the electrons, d is the spacing between atomic layers, and θ is the angle between the incoming electron beam and the atomic layers. Electron scattering into angles other than θ from that set of atomic layers will be reduced by destructive interference, whereas scattering into θ will be increased by constructive interference. A three-dimensional crystal will have many planes which all produce Bragg scattering. Placing a detector after the lens system in a TEM will result in a pattern of bright spots called a diffraction pattern. Each spot in the pattern results from a set of planes for which the Bragg condition is met, and each spot can then be related to one set of crystal planes in the sample. There is an inverse relationship in the Bragg equation between the spacing d and the angle θ , where the largest unique repeated spacings

in the structure are scattered through the smallest angles. The innermost spots in the diffraction pattern are thus representative of the main dimensions in the basic repeat unit of the structure. The positions of the diffraction spots in the pattern can thus be analyzed to help determine the crystal symmetry, unit cell dimensions, and the orientation of the crystal lattice.

54. Given the very high resolution of the TEM, it can also be used to form images of the atomic arrangement called “lattice images.” Lattice images exhibit repeated patterns which have the same periodicity and orientation as the crystal structure of the sample. Lattice images can be processed digitally with a method called a “fast Fourier transform” (FFT) which, like a diffraction pattern, produces a pattern of spots where each spot corresponds to a different periodic spacing in the image. In other words, FFT analysis of a TEM image helps reveal the repeating patterns of the atoms. The resulting FFT pattern can be interpreted in the same way as an electron diffraction pattern to help determine the crystal symmetry, unit cell dimensions, and the orientation of the crystal lattice.

b. TEM as a Tool to Investigate Crystal Alignment

55. TEM may be used in two ways to understand the crystal alignment of two materials that share an interface. The first method uses real-space (direct) visualization of the atomic structure. This method requires lattice images. The second technique uses reciprocal-space imaging. The reciprocal space data may be

collected as the FFT patterns of lattice images, or as SAED patterns. Since both real-space and reciprocal-space visualizations are only two-dimensional projections of three-dimensional structures, careful interpretation is necessary to make reliable conclusions from the data.

c. Energy-Dispersive X-Ray Spectroscopy

56. Energy-dispersive X-ray spectroscopy (“EDS” or “EDX”), or equivalently, energy-dispersive X-ray analysis (EDXA or EDAX) is a technique that analyzes the X-rays produced during electron illumination to determine the elemental composition of a defined region within a sample. EDS is a complementary tool to electron microscopy and can be conducted in both the scanning electron microscope (SEM) and transmission electron microscope (TEM).

57. The working principle of EDS involves the interaction of the electron beam with core electrons of constituent elements within the sample. Specifically, an incident electron can cause a core electron to be ejected from the inner shell, leaving a vacant hole. An electron from an outer shell of the atom then fills its place, releasing energy in the form of an X-ray as it transitions from a higher-energy state of the outer shell to the hole of the inner shell. The energies of the electrons in an atom are characteristic of each element in the periodic table, so these X-rays are known as characteristic X-rays. By collecting a population of X-rays excited from

the sample and analyzing the intensity of X-rays at various energies, the elemental composition of a sample can be determined.

5. X-ray Photoelectron Spectroscopy

58. X-ray photoelectron spectroscopy (“XPS”) is a characterization technique that provides elemental information from approximately the outermost 1–10 nm of a sample surface. The working principle is based on the photoelectric effect, in which samples exposed to electromagnetic radiation emit electrons. If the incoming radiation is of a narrow, suitable, and well-defined energy, the resulting electrons emitted have kinetic energies that are related to their binding energies. X-rays are the electromagnetic radiation in the right energy range to eject the electrons from an atom. As discussed above in the context of EDS, the distribution of energies of the electrons around an atom are characteristic of each element. This phenomenon allows for elemental identification according to the following (simplified) expression:

$$E_B = h\nu - KE$$

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

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

60. Due to its high surface sensitivity, XPS is often used to investigate the surface chemistry of battery materials. Notably, XPS can provide insight into the elemental composition of surface coatings on electrode particles and the composition of the solid electrolyte interphase (“SEI”) or the cathode electrolyte interphase (“CEI”). Furthermore, due to the inherently shallow photoelectron escape depth, XPS analysis is commonly understood to be representative of a sample surface only, in contrast to other chemical and spectroscopic techniques that can probe elemental composition further into the bulk regions of a sample (such as EDS when used with SEM). The formation of chemical bonds also influences the binding energy of emitted photoelectrons, so XPS analysis can identify the presence and types of chemical bonds on a sample surface, provided that those bonds cause large enough energy changes to the binding energies to be spectrally resolved.

6. Lithium-Ion Battery Components and Operation

61. A lithium-ion battery has the basic components of a negative electrode (*i.e.*, anode) and a positive electrode (*i.e.*, cathode), a separator between the

electrodes, and an electrolyte. The positive electrode typically consists mainly of layered lithium metal oxide microparticles capable of reversible lithium-ion extraction/insertion (deintercalation/intercalation), commonly lithium cobalt oxide (LiCoO_2) (also referred to as LCO), in batteries for consumer electronics, or an analogous material, coated on an aluminum foil current collector. The negative electrode typically consists mainly of graphite microparticles capable of reversible lithium-ion insertion/extraction coated on a copper foil current collector. Both electrode coatings also typically contain conductive additives (*e.g.*, carbon black) and binder material (*e.g.*, polyvinylidene fluoride (PVDF) for the positive electrode and a mix of carboxymethyl cellulose (CMC) and styrene-butadiene rubber (SBR) for the negative electrode). Note the use of conductive additives is important and common practice, because the active materials, especially the positive electrode active materials (PEAMs), lack good electronic conductivity on their own (when in the form of a network of particles), and the charge/discharge process ideally involves electrons getting to and from all the active material. The separator is typically a thin polymer membrane consisting of porous polyethylene and/or polypropylene that maintains electrical isolation between the electrodes but, when paired with the electrolyte, allows transport of lithium ions between the electrodes during charge/discharge. The separator is commonly paired with an inorganic heat resistant layer to reduce the shrinkage of polymer membrane during high temperature, thereby

enabling the cell to endure higher temperature without experiencing thermal runaway. Electrolytes are typically liquids that wet the separator and serve as the medium through which lithium ions are transported from one electrode to the other. They are commonly mixtures of organic solvents (e.g., carbonates) and a dissolved lithium salt, typically lithium hexafluorophosphate (LiPF_6). Various additives may also be added to the electrolyte to enhance the cell stability, safety, and performance.

62. When a lithium-ion battery is charged, lithium ions are extracted from the positive electrode material, diffuse and migrate through electrolyte and separator, and are inserted into the negative electrode material. In an analogous manner, during discharge, lithium ions are extracted from the negative electrode, diffuse and migrate through electrolyte and separator, and are inserted into the positive electrode.

63. Electrode active materials are typically described in their discharged state. For example, LiCoO_2 is the discharged state of a common PEAM. Upon charging, LiCoO_2 is converted to $\text{Li}_{1-x}\text{CoO}_2$, where x is approximately 0.5 in the fully charged state. The crystal structure of LiCoO_2 and related layered rock salts is described in Section VI.A.2 above.

64. Manufacturing of lithium-ion pouch cells—for use in consumer electronics, for example—typically involves these steps (at a minimum): (1) mixing of electrode powder active materials, binders, and solvent (involves both wet and dry mixing steps); (2) coating the electrode layers onto foil current collectors; (3)

drying the electrodes; (4) calendaring the electrodes; (5) cutting/slitting/notching the electrodes; (6) tab cutting and welding to the electrodes; (7) winding/stacking of the electrodes with the separator layer; (8) hot pressing of cell winding/stack; (9) packaging the electrode assembly in a pouch; (10) cell drying; (11) injecting electrolyte; (12) undergoing formation and aging; (13) degassing and final sealing; and (14) aging.

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

66. The SEI and CEI are critical to the longevity of the battery since they help shield the bulk electrode materials from further interactions with the electrolyte, which effectively limits further electrolyte decomposition reactions. This self-passivating effect is due to the catalytic nature of the bare electrode active material

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

67. Importantly, if a fluorine-containing salt is used in the electrolyte, such as LiPF_6 —as is industry standard (*see* Ex. 1021, 1), the corresponding SEI and CEI layers are known to contain fluorine in various compounds such as lithium fluoride, fluorophosphates, and lithiated fluorophosphates. Of such species, lithium fluoride is a notable example of an electrochemically inactive compound under nominal lithium-ion battery operating conditions. *See id.* Although the initial formation reaction of lithium fluoride (during CEI or SEI formation) is believed to originate from LiPF_6 hydrolysis or the consumption of a small fraction of lithium from the positive electrode active material, the resulting lithium fluoride layer limits further electrolyte decomposition reactions from occurring as described in previous paragraphs. *See, e.g.,* Ex. 1018 (showing formation of a LiF layer on cathode surface due to LiPF_6 hydrolysis), Ex. 1019, 86 (describing fluorine “on the surface” of cathode particles due to “formation of LiF derivatives”); Ex. 1020, 12 (describing

that XPS measurements on “LiCoO₂ powder stored in LiPF₆/alkyl-carbonate solutions always shows LiF peaks”); Ex. 1021, 1 (“Solid LiF . . . precipitates on the electrode particles[.]”).

7. Degradation in Common Commercial Lithium-Ion Battery Chemistries

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

69. During charge and discharge, the positive and negative electrode active materials typically experience volumetric changes as lithium is inserted (“intercalated”) and extracted (“deintercalated”). As a result of the repeated stress related to lithium intercalation and deintercalation, the positive and negative electrode active material particles can become damaged over time.

70. The PEAM, which is typically LiCoO₂ in batteries for consumer electronics, is well-known to form cracks and crevices near the surface of the particles during cycling. The initial few cycles of a lithium-ion battery are performed during the formation process of manufacturing. With further cycling, a greater population of particles have been observed with cracks, which is consistent with

fatigue expected for repeated volume changes. When cracks are generated during cycling, new active material surface area is exposed to the electrolyte. These regions are then passivated through further reactions with the electrolyte to form a new interphase layer at the crack location. This process leads to the consumption of electrolyte and the build-up of deposits on the positive electrode material, degrading the performance of the cell. Furthermore, as the number of cracked particles increases, there is a greater likelihood of poor electronic contact among the particles. This phenomenon consequently leads to an increase in cell impedance.

71. LiCoO_2 and other layered rock salt PEAMs are also known to degrade through the irreversible rearrangement of atoms near particle surfaces. Under appropriate conditions, the transition metal can migrate to and occupy a lithium site within the lattice. In LiCoO_2 , this has been shown to result in the formation of Co_3O_4 in a spinel structure and CoO in a rock salt structure. Ex 1036. These processes are generally exacerbated by high-voltage operation and exposure to elevated temperatures.

72. Most commercial lithium-ion battery electrolyte chemistries utilize a LiPF_6 salt dissolved in a mixture of organic carbonate-based solvents and are susceptible to breakdown. Beyond the decomposition processes induced by electrochemical cycling that result in the growth of interfacial layers on the positive and negative electrodes, common electrolytes utilizing LiPF_6 are also known to react

with trace amounts of water in the electrolyte to form hydrofluoric acid (HF). The hydrolysis reaction of LiPF₆ was proposed as:



Ex. 1021, 6. It was widely understood that HF was detrimental to cathode stability and battery performance. Since PEAMs are often lithium transition metal oxides (e.g., LiCoO₂), they are highly susceptible to etching by acidic species, including HF. This degradation pathway results in the loss of active material and transition metal dissolution into the electrolyte. Ex. 1008, 3–4 (“It is well known that acid (such as HCl and *HF*) can be generated from the reaction between water and salts (such as LiClO₄ and *LiPF*₆) in the electrolyte. . . . The acid-containing electrolyte attacks the surface of the cathode during cycling.”) (emphases added); *see also* Ex. 1020, 2 (“a LiPF₆ solution always contains HF contamination.”); Ex. 1023, 1 (describing “capacity loss of LiCoO₂” “mainly due to the interfacial impedance growth between the cathode and electrolyte, which results from side reactions involving LiPF₆”).

8. Strategies to Improve LiCoO₂ Cathode Materials

73. Over the last several decades and across nearly all industry sectors that utilize lithium-ion batteries, four critical goals have guided battery engineering efforts: (1) maximize energy density, (2) maximize power performance, (3) maximize cycle life, and (4) minimize safety concerns.

74. Although the theoretical capacity of LiCoO_2 is as high as ~ 274 mAh/g, the maximum attainable practical capacity is far lower (approximately 137 mAh/g when cycled in the range of 3.0–4.2 V in a battery). To increase the cell-level energy density, a primary option is to increase the upper cut off voltage, which involves the extraction of more lithium ions from the positive electrode material (*e.g.*, where more than 50% of the lithium has been extracted from the positive electrode). While this avenue provides an immediate increase in capacity, it subjects the electrodes to new/accelerated degradation mechanisms that can compromise the long-term health of the cell. For example, charging LiCoO_2 above ~ 4.2 V vs. Li/Li^+ can cause a phase change in the LCO particles, resulting in a change in the unit cell dimensions, the generation of internal stress within the bulk particle, and an increase the likelihood of PEAM particle cracking, which can lead to loss of electrical contact between particles and increased electrolyte consumption due to the exposure of fresh surfaces. Cell operation at elevated voltages is also known to increase the susceptibility to cobalt dissolution from the PEAM and can drive additional electrolyte decomposition. As a result, cells used at elevated voltages can suffer from rapid increases in impedance (impacting rate capability) and capacity loss.

75. Doping has been a widely explored strategy to improve the electrochemical performance of LiCoO_2 . Cobalt can be partially substituted with other inactive elements, such as magnesium, aluminum, titanium, and others, to

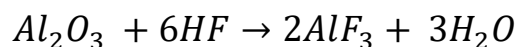
improve both high-voltage and long-term cycling stability. *See* Ex. 1010, 1 (“[V]arious dopants . . . have been proposed to improve the electrochemical performances. In particular, the Mg-doped LiCoO₂ has been intensively investigated because of its enhanced electronic conductivity and thermal or cycling stability.”); Ex. 1011, 1 (“[P]artial doping of cobalt with metal ions leads to extended cyclability and increase in capacity of lithium ion cells by enhancing structural stability.”); Ex. 1008, 1 (“An inexpensive element, Mg has been used as a doping material to enhance the electrochemical properties of cathodes.”). To synthesize such materials, dopant precursors can be introduced simultaneously with cobalt and lithium precursors and subsequently calcined.

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

77. Following these early works, researchers went on to explore a wide range of different coatings, including a host of different oxides, phosphates, and

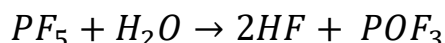
fluorides. *See, e.g.,* Exs. 1006, 1010, 1008, 1007, 1008, 1009. Using different synthesis routes and precursors, researchers have been able to develop different coating morphologies, including rough/patchy coatings, thin uniform coatings, and thicker “core-shell” coatings.

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



The scavenging reaction results in the conversion of some of the metal oxide coating (in this case Al_2O_3) on the surface of the particle to a metal fluoride (e.g., AlF_3). Ex. 1021, 2 (“[A] fluoride layer can form on top of an HF-scavenging oxide coating as a product of the HF-scavenging reaction.”); Ex. 1038, 1 (discussing “conversion” of metal oxides to metal fluorides, and stating that “metal fluorides are most likely to be the ultimate products which make genuine contribution in repeated electrochemical cycling”). In this manner, the fluoride becomes another barrier that prevents the active material from subsequent reactions with HF. Although this reaction reduces the amount of HF present in a cell, as shown above, it results in the

formation of water. This water can further hydrolyze the LiPF_6 electrolyte salt, generating more HF:



Ex. 1021, 1; Ex. 1036, 4 (describing H_2O from cathodes “leading to a continuous increase of HF content”).

79. To this end, it was further understood in the art that metal fluoride coatings could be used directly (as opposed to being formed through scavenging reactions). Studies found that fluorine-containing coatings “serve[] as a protective layer, preventing direct contact with the reactive electrolyte.” Ex. 1008, 4; *see also id.*, 5 (“ MgF_2 coating layer seemed to react with acidic electrolyte and incur damage on cycling instead of the electrode material.”), 6 (“[T]he MgF_2 coating layer was effective as a protective layer to prevent the dissolution of the transition metal . . . which may lead to enhanced electrochemical properties.”); Ex. 1024, 2 (“In recent years, fluorine in the form of metal fluorides such as AlF_3 , CaF_2 , MgF_2 , LaF_3 , CeF_3 , FeF_3 is considered to be one of the most promising coating materials to improve electrochemical performance of cathode, due to its high stability. This could be attributed to the reduction of the structural change of active material from HF attack by a thin coating layer preventing the direct contact between delithiated cathode and liquid electrolyte.”); Ex. 1030, 2 (finding that metal fluoride coatings

improved the thermal stability of LiCoO₂ electrodes); Ex. 1027, 1 (finding that metal fluoride coatings protected LiCoO₂ from hydrofluoric acid attack).

80. Indeed, well before the '660 Patent, it was known to those of skill in the art that magnesium, titanium, and fluorine (in particular) could be used in the coating layer—and would be beneficial in coatings on positive electrode active materials. *See* Ex. 1014, [0011]–[0013] (disclosing TiF, MgF₂, TiF₃, and TiF₄ coatings); Ex. 1015, 2:66–3:15 (disclosing various coatings, including TiF, MgF₂, TiF₃, and TiF₄); Ex. 1016, 1:24–31 (disclosing one element from the group “Ti, Zr and Hf” used with one element from the group “F or P”); Ex. 1017, [0010] (disclosing surface oxides of “titanium, silicon (Si), magnesium, and zirconium”). Further, some studies indicated multiple coating layers could be layered on top of the bulk particle. *See* Ex. 1017, [0077] (“[T]he surface layer is arranged on the coating layer”).

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

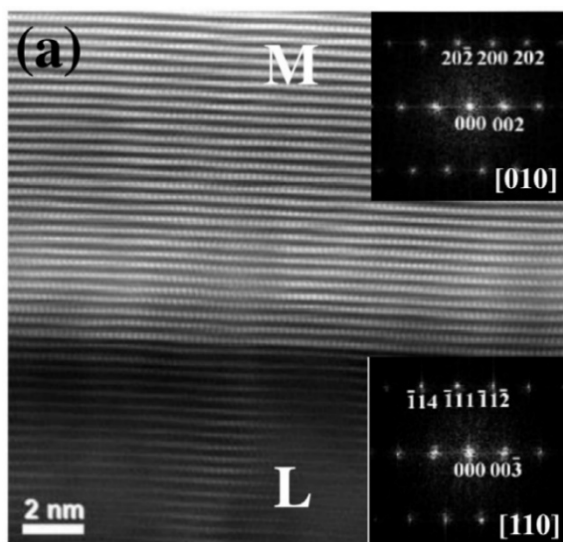


Ex. 1021, 1; *see also* Ex. 1019, 86 (describing fluorine present “on the surface” of the cathode particle “caused by the formation of LiF derivatives”); Ex. 1021 at 1 (“Solid LiF . . . precipitates on the electrode particles[.]”); Ex. 1020, 12 (describing that XPS measurements on “LiCoO₂ powder stored in LiPF₆/alkyl-carbonate solutions always shows LiF peaks”). Some studies found this formation of a fluorine-containing layer to be beneficial. *See* Ex. 1022, 3 (concluding that a fluorine-containing solvent “provides an electrochemically stable surface film on a LiCoO₂ cathode and ***significantly improves the cycle performance*** under a high-voltage condition”) (emphasis added).

82. POSITAs would have additionally known about tradeoffs in using such coatings. For example, while a thicker, uniform coating can provide better protection against irreversible reactions with the electrolyte, improving the cycle life, it can also hinder lithium ion and electron transport, penalizing the high-rate performance. To minimize the tradeoffs, researchers have focused on developing coating materials that provide better lithium ion and electronic conductivity, as explained by Shim et al. in 2015. *See* Ex. 1032. The authors describe using different lithium titanium oxides—also called “lithium titanates”—as coating materials for Mg-doped LiCoO₂ because they are known lithium-ion conductors. Building on their previous work focused on MgO-coated LiCoO₂ (Ex. 1010), Shim et al. found that Mg-doping of the lithium titanate coating significantly improved its electrical conductivity, allowing

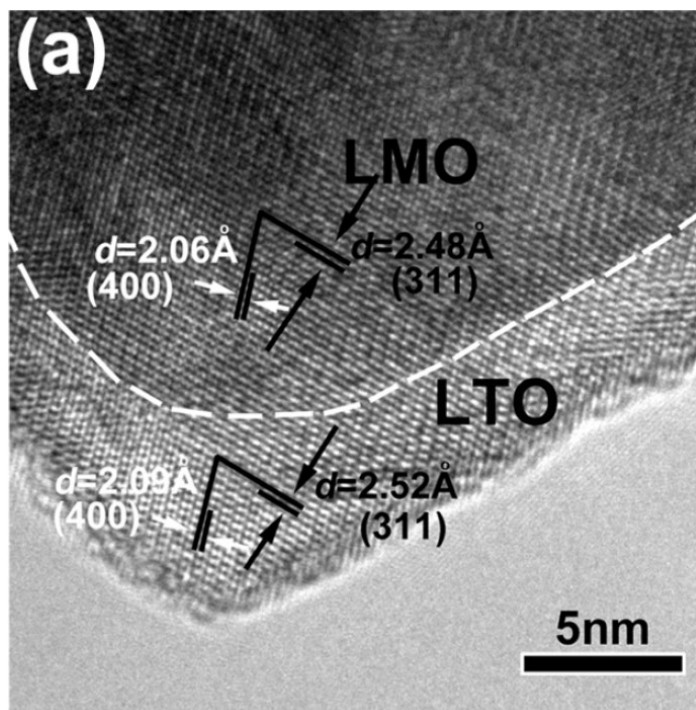
the authors to overcome previous performance issues faced by lithium titanate coatings.

83. In addition to developing coatings with improved conductivity, researchers have also highlighted the importance of creating good crystallographic matches between the coating layer and the bulk. Evidence of good crystallographic alignment was demonstrated in the TEM images presented by Shim 2015.



Ex. 1032, Fig. 3(a).

84. In the case of their monoclinic lithium titanate coating (shown in the image above), the authors note: “the Li_2TiO_3 crystal was epitaxially grown on the surface of LiCoO_2 as a coating layer.” *Id.*, 4. The benefits of this approach were explained in a paper that Shim et al. referenced from the previous year by Li et al. Ex. 1035. This group also created an epitaxial coating of lithium titanate on positive electrode active material (LiMn_2O_4 in this case), as shown in the image below.



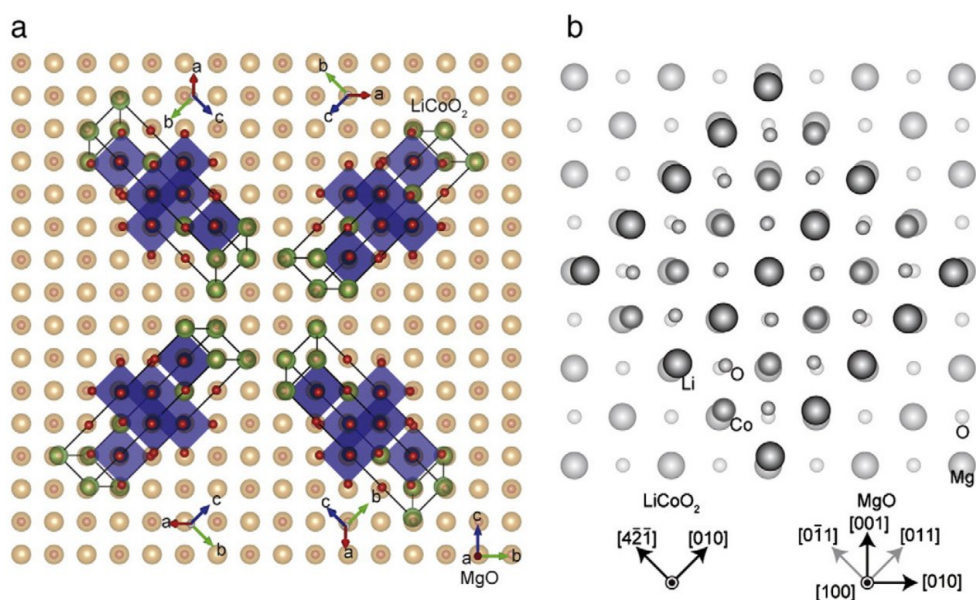
Id., Fig. 3(a).

85. Li et al. explaining the motivation for their work as follows:

“Epitaxial coating, as a distinctive coating strategy, is mainly applied in the semiconductor field, which is direct growth of a thin film on the surface of the host material with the same lattice orientation. It can lead to a dense and tight film to effectively suppress undesirable side reactions and the formation of voids between the host materials and the coating layer during cycling. Besides, there is the same lattice orientation and small lattice mismatch between the coating layer and the host material eliminating the apparent interface of coating layer and host material and decreasing the stacking faults to form an efficient tunnel for Li^+ -ion diffusion. Consequently, epitaxial coating may be an ideal strategy for improving the electrochemical performance of cathodes.” *Id.*, 1.

In this manner, Li et al. teach that epitaxial coatings (that involve a crystallographic alignment of the coating layer with the bulk) improve both lithium-ion transport and cycling stability, two of the most important functions of a coating layer.

86. The crystallographic alignment between rock salt and layered rock salt had also been explored. For example, Katsui et al. demonstrated the epitaxial growth of LiCoO_2 films on three different orientations of bulk MgO single crystals. Ex. 1039. In each case, the same crystallographic alignment was achieved, one where the cubic-closest-packed anion sublattice of LiCoO_2 was aligned with that of MgO , as shown in the figure below, demonstrating the crystallographic compatibility of these two structures.



Ex. 1039, Figs. 3(a)-(b).

9. Lithium Titanate (LTO)

87. Lithium titanate has attracted considerable research attention due to its potential use as a substitute for graphite as the negative electrode active materials in lithium-ion batteries. In comparison to graphite, lithium titanate can provide

increased cycling stability, improved lithium ion transport properties, and a reduced risk for lithium plating (a potential problematic occurrence in lithium-ion battery operation). In addition, as discussed in the previous section, lithium titanate has also been investigated as a coating material for PEAMs (*e.g.*, Ex. 1029, 1032) due to its stability and lithium-ion conductivity.

88. There are various lithium titanate compounds that consist of stoichiometric amounts of lithium, titanium, and oxygen. While $\text{Li}_4\text{Ti}_5\text{O}_{12}$ —which assumes the cubic spinel crystal structure—is a common phase considered for negative electrode use in battery applications, it has been well known in the field that other phases can be synthesized or formed during lithiation (during charging of the battery) or through degradation reactions with the electrolyte.

89. Other reported structures include: $\text{Li}_2\text{Ti}_3\text{O}_7$ which assumes an orthorhombic structure, Li_2TiO_3 which can assume cubic rock salt structures (α or γ phases) or a monoclinically distorted rock-salt-like structure (β phase), and LiTiO_2 which also assumes the cubic rock salt structure discussed in Section VI.A.2. Exs. 1034, 1037.

10. Magnesium Oxide (MgO)

90. As discussed above, MgO is an ionic compound known to crystallize in the rock salt structure with the $Fm\bar{3}m$ space group. As shown below, in 1976, Ashcroft and Mermin taught that MgO has the “sodium chloride” structure.

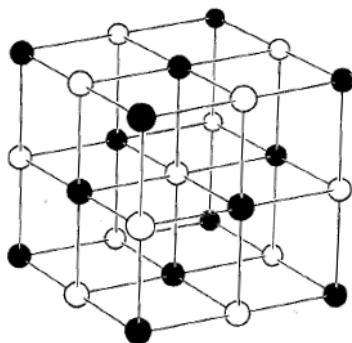


Figure 4.24

The sodium chloride structure. One type of ion is represented by black balls, the other type by white. The black and white balls form interpenetrating fcc lattices.

Table 4.5

SOME COMPOUNDS WITH THE SODIUM CHLORIDE STRUCTURE

CRYSTAL	a (Å)	CRYSTAL	a (Å)	CRYSTAL	a (Å)
LiF	4.02	RbF	5.64	CaS	5.69
LiCl	5.13	RbCl	6.58	CaSe	5.91
LiBr	5.50	RbBr	6.85	CaTe	6.34
LiI	6.00	RbI	7.34	SrO	5.16
NaF	4.62	CsF	6.01	SrS	6.02
NaCl	5.64	AgF	4.92	SrSe	6.23
NaBr	5.97	AgCl	5.55	SrTe	6.47
NaI	6.47	AgBr	5.77	BaO	5.52
KF	5.35	MgO	4.21	BaS	6.39
KCl	6.29	MgS	5.20	BaSe	6.60
KBr	6.60	MgSe	5.45	BaTe	6.99
KI	7.07	CaO	4.81		

Ex. 1013, 80.

91. MgO is understood to crystallize at moderate synthesis temperatures due to the relatively large size of the Mg^{2+} cation and the resulting preference for octahedral bonding. For instance, X-ray diffraction patterns demonstrating MgO crystallization after sintering at 500 °C and above are shown below by Kumar and Kumar:

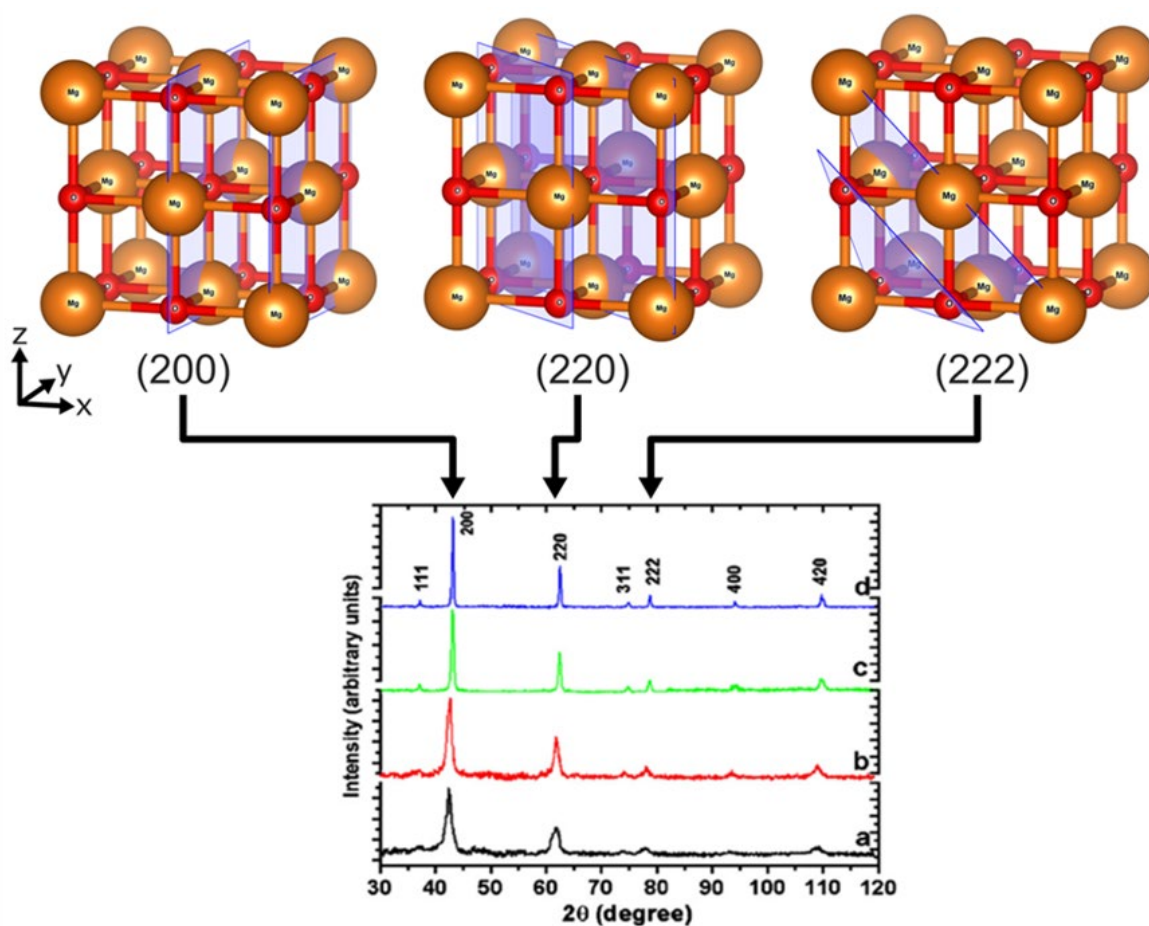


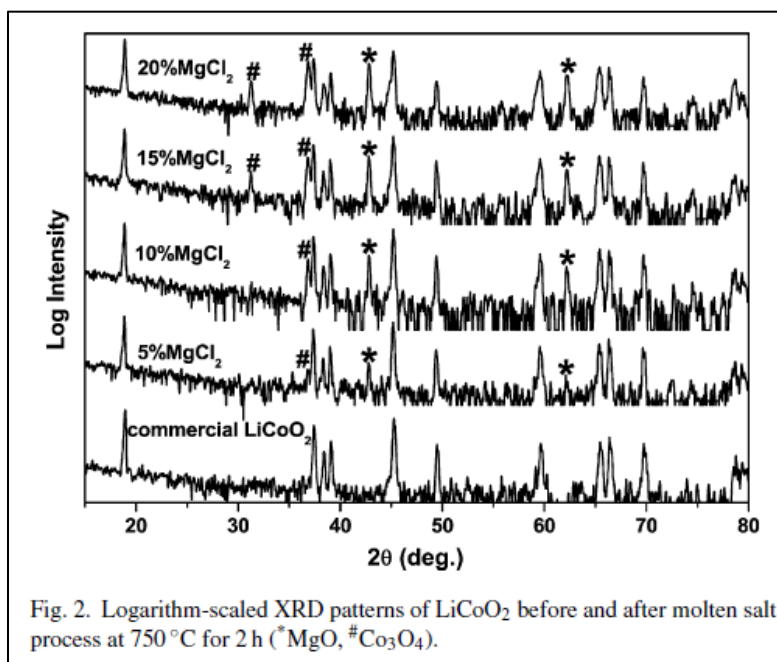
Fig. 4. X-ray diffraction patterns of dried sol-gel product after decomposition at (a) 500, (b) 600, (c) 800, and (d) 1000 °C for 2 h each. These indicate formation of f.c.c. (NaCl-type) phase of magnesium oxide.

Ex. 1040, Fig. 4 (crystal structure images added). Particularly notable are the diffraction peaks labeled with the corresponding crystal planes (Miller indices in (hkl) notation) where, for example, (200) corresponds to the set of planes that includes the unit-cell faces of the two intertwined FCC lattices that make up the rock salt structure. As Kumar and Kumar indicate with respect to Figure 4, “[t]hese indicate formation of f.c.c. (NaCl-type) phase of magnesium oxide.” *Id.*

92. Additionally, Xu et al. explained that only one phase of MgO—“a rocksalt cubic structure of NaCl (space group $Fm-3m$)”—is “the stable phase at room

temperature and pressure conditions,” meaning crystalline MgO will be in the rock-salt phase under normal conditions. Ex. 1030, 16.

93. Similarly, in 2007 Bai et al. used MgO as a coating to improve the performance of lithium-ion cathodes. Ex. 1031, 1. Figure 2 shows the X-ray diffraction, or XRD patterns, of LiCoO_2 after being treated with MgO. As shown below, peaks marked with asterisks (*) correspond to MgO, which a POSITA would have understood to be consistent with the (200) and (220) peaks characteristic of rock salt MgO:



Id., Fig. 2.

94. MgO is a material that has been extensively investigated as a coating material for LiCoO_2 . *See, e.g.*, Ex. 1010; Ex. 1007, Ex. 1031. Researchers have been interested in MgO as a coating because, in addition to providing a physical barrier

between the PEAM and the electrolyte (like other metal oxides), there is a reported benefit attributed to magnesium diffusion into the near surface of the LiCoO_2 to form a solid-solution phase. This diffusion process is understood to increase the stability of LiCoO_2 through a “pillaring effect” where Mg ions occupy the Li layer near the surface of the LiCoO_2 and help prevent the collapse of the layered structure at elevated voltages (when more than 50% of the Li has been removed). *See, e.g.*, Ex. 1010; Ex. 1007, Ex. 1031.

B. Overview of the '660 Patent

95. I understand that the '660 Patent issued on June 22, 2021 from U.S. Application No. 16/923,494, filed on July 8, 2020. For purposes of this declaration, I have been asked to assume that the '660 Patent is entitled to a Priority Date of July 5, 2016.

1. Claims

96. The '660 Patent has 10 claims, including independent claims 1 and 6. The '660 Patent has 8 dependent claims.

2. Summary of the Specification

97. The '660 Patent address a battery degradation problem that was well known to a POSITA—“[w]hen charge and discharge cycles are repeated, deformation of the particles of the positive electrode active materials, such as cracking or breaking, might occur.” *Id.*, 4:20–22. To address this deformation,

cracking, and breaking issue, the '660 Patent proposes a “coating film” over a PEAM’s surface to suppress the “decrease in capacity due to charge and discharge cycles.” *Id.*, 4:5–8.

98. Before prosecution—as admitted by the '660 Patent—lithium-ion batteries were “actively developed,” ('660 Patent, 1:48–50) and these coating films were known. *See id.*, 1:66–2:2 (“Thus, improvement of a positive electrode active material has been studied to increase the cycle performance and the capacity of the lithium ion secondary battery (Patent Document 1 and Patent Document 2).”). For example, (as stated in admitted background art) it was known that “[t]he formation of the *surface layer* of the positive electrode active material . . . can reduce the energy barrier in insertion and removal of lithium at the positive electrode active material surface.” Ex. 1027, Abstract (emphasis added). PEAMs with “a *surface layer* consisting of a layered structure of lithium cobalt oxide comprising element M (M is Mg and/or Al), and at least a layered structure of lithium cobalt oxide solidified by Ti” were also known. Ex. 1028, Abstract (emphasis added). As further admitted by the '660 Patent, its claimed coating materials were known. Ex. 1001, 22:35–38 (“It is generally known that, at the time of synthesizing particles of the composite oxide containing lithium, cobalt, magnesium, and fluorine . . .”).

99. Considering the robust lithium-ion battery prior art, the '660 Patent’s purported invention is using regions within a PEAM. *Id.*, 2:42–48. For example, the

'660 Patent states that a “first region contains an oxide of lithium and a first transition metal and has a layered rock-salt crystal structure,” a “second region contains a non-stoichiometric compound containing an oxide of a second transition metal and the non-stoichiometric compound has a rock-salt crystal structure,” and “third region contains a compound of representative elements and the compound of representative elements has a rock-salt crystal structure.” *Id.*, 2:62–3:3. There may be overlap in the latter two regions (*id.*, 2:49–51), and “it is preferable that crystal orientations of the first region and the second region be partly aligned with each other and crystal orientations of the second region and the third region be partly aligned with each other.” *Id.*, 3:11–16.

3. Summary of the Prosecution History

100. The application which lead to the '660 Patent was filed with 24 claims. On September 21, 2020, the application received a Non-Final Rejection, where the examiner rejected all pending claims under 35 U.S.C. §§ 102 and 103 because certain claims were anticipated by Liu (CN 102610806 B) and obvious over Liu in view of Gao (US 2016/0104880 A1). Ex. 1002, 104–88 (Sep. 21, 2020 Non-Final Rejection). In response, the applicant amended certain claims to add, among others, a “positive electrode,” having a “first region compris[ing] a layered rock-salt crystal structure” and a “second region compris[ing] a crystal structure which is different

from the layered rock-salt crystal structure.” *Id.*, 219–24 (Dec. 21, 2020 Amendment and Reply).

101. On January 11, 2021—despite the applicants’ amendments adding PEAM regions—the applicant received a Final Rejection rejecting all claims under 35 U.S.C. § 112. *Id.* (Jan. 11, 2021 Final Rejection). Among other defects, the examiner found that the specification “[d]id not reasonably provide enablement for a second region comprising a crystal structure which is different from a layered rock-salt crystal structure.” *Id.*, 1311 (Jan. 11, 2021 Final Rejection). The examiner stated that “[t]he limitation ‘wherein the second region comprises a crystal structure which is different from the layered rock-salt crystal structure’ encompasses a second region with any crystal structure.” *Id.*, 1311-12; *see also id.*, 1313-15 (“It is unclear with what 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.”).

102. On January 29, 2021 during an Examiner Interview, the applicant proposed “canceling claims 7, 9–12, 19, and 21–25,” “amending claims 1 and 13 . . . to ‘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,’” and “amending claim 13 . . . to ‘wherein the second region covers at least a first part of the first region.’” *Id.*, 1345 (Feb. 4, 2021 Examiner

Interview Summary Record 1). The applicant made these changes on February 10, 2021.

VII. LEVEL OF ORDINARY SKILL IN THE ART

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

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

105. In determining the characteristics of a hypothetical person of ordinary skill in the art of the '660 Patent at the time of the claimed invention, I considered several things, including the type of problems encountered in this field, and the rapidity with which innovations were made. I also considered the sophistication of the technology involved, and the educational background and experience of those actively working in the field, and the level of education that would be necessary to understand the '660 Patent.

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

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

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

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

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

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

VIII. CLAIM CONSTRUCTION

112. I understand that terms in the challenged claims should be given their plain and ordinary meaning, considering the specification.

IX. DETAILED INVALIDITY ANALYSIS

A. Background on Prior Art References

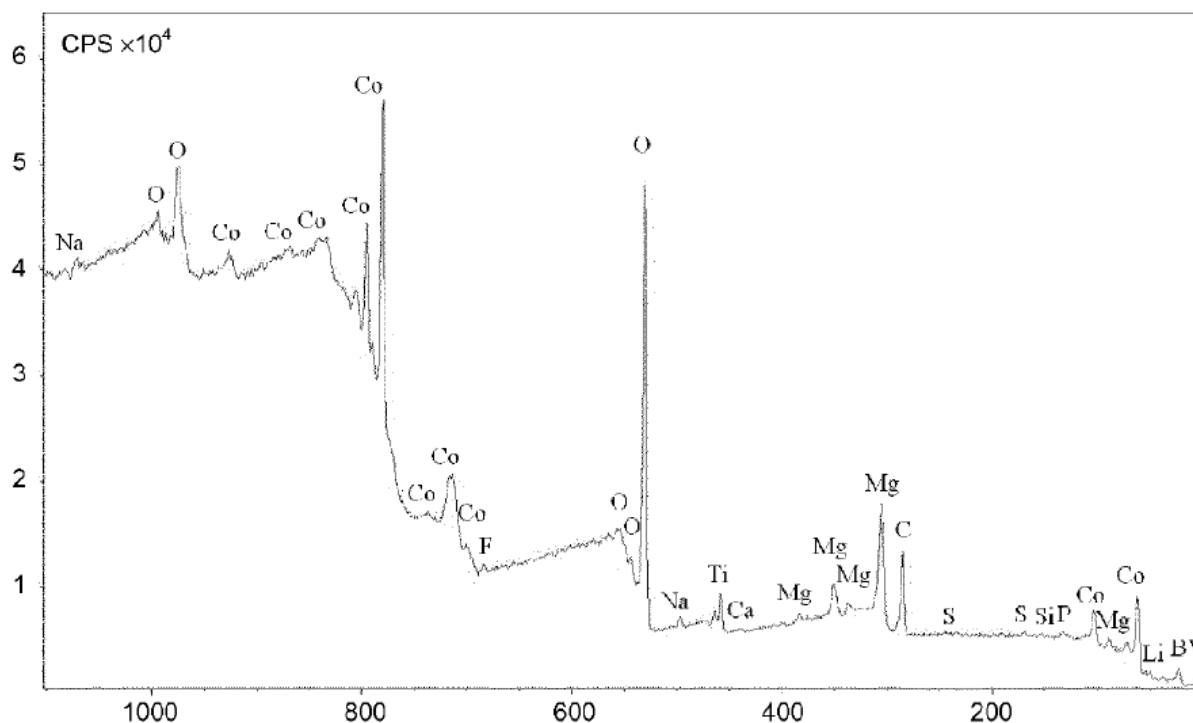
1. Overview of Blangero (Ex. 1005)

113. US Pat. Pub. No. 2014/0212759 A1 (“Blangero”) is a patent application filed on May 20, 2012 and is directed to the development of lithium metal oxide particles coated with a mixture of the elements of the core material and one or more metal oxides for as a PEAM in a lithium-ion battery. Specifically, Blangero teaches

“[a] lithium metal oxide powder for use as a cathode material in a rechargeable battery, consisting of a core layer and a surface layer, the core having a layered crystal structure consisting of the elements Li, a metal M and oxygen.” Ex. 1005, Abstract.

114. In particular, Blangero describes improving the stability of LiCoO_2 cathodes by introducing a coating, or surface layer. *Id.*, ¶¶ [0004], [0005]–[0007]. Specifically, Blangero explains “[b]y careful selection of the compounds, such as Mg, Zr, Si and Ti, the process allows to achieve an in-situ coating of the final power with a layer that is enriched in oxidized compounds, like MgO and TiO_2 , that may also be lithiated.” *Id.* [0094]. Blangero explains that “[t]he surface layer,” is “preferably between 20 nm and 100 nm thick.” *Id.*, ¶ [0066].

115. “Example 1” had a core of “titanium and magnesium doped LiCoO_2 ” and a surface comprising “the elements of the core material (Li, Co) and inorganic metal oxides comprising Mg and Ti.” *Id.*, ¶¶ [0094], [0108]–[0110]. Blangero’s XPS analysis of the surface of the Example 1 particle expressly assigned peaks corresponding to cobalt, oxygen, magnesium, titanium, and fluorine:

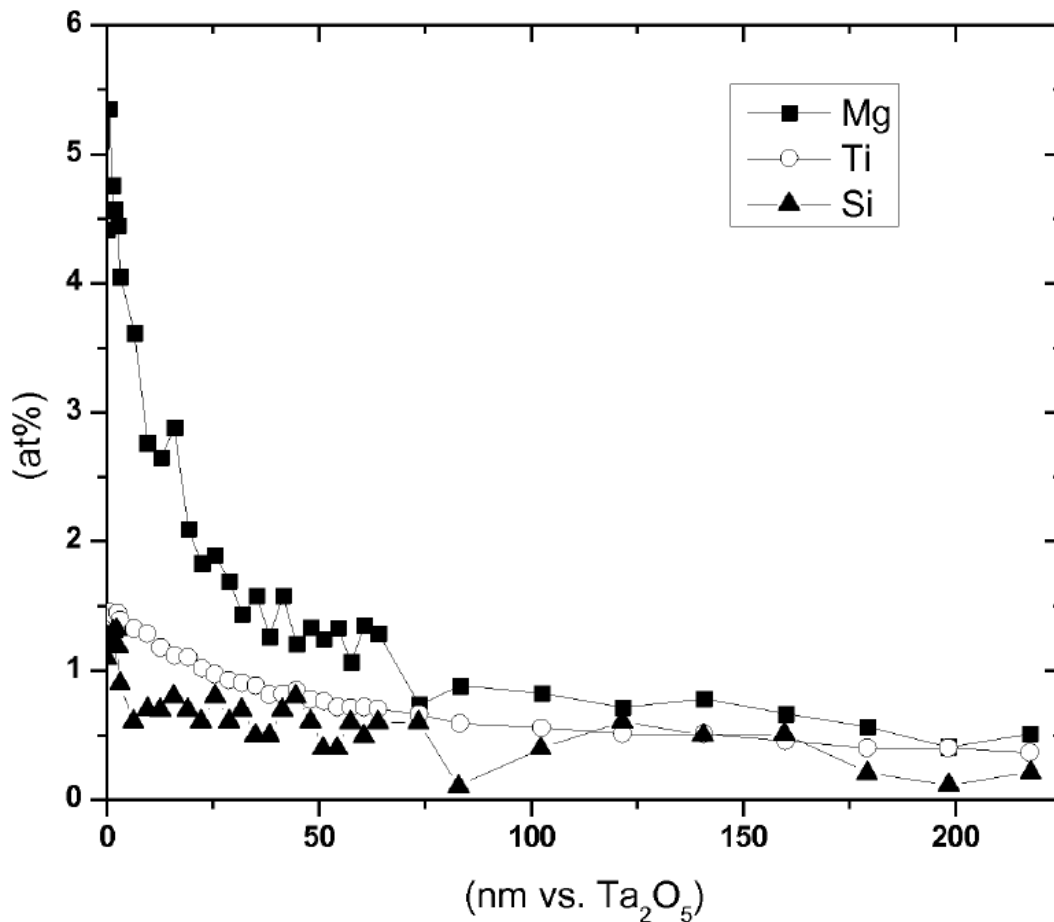


Id., Fig. 4(b), ¶ [0129]. Table 3 similarly confirms that Example 1 has detectable levels of cobalt, oxygen, magnesium, titanium, and fluorine on the particle's surface:

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], Table 3 (stitched). The claims further confirm that the disclosed PEAM has a surface layer that contains Mg, Ti, O and F. *Id.*, cls. 33, 37, 53, 62. Blangero further discloses that “the concentration of M' and M'' at the surface of the lithium metal oxide powder is at least 5 times the concentration of M' and M'' in the bulk of the powder” where M' includes magnesium. *Id.*, ¶ [0011].

116. Blangero also examined Mg and Ti as a function of particle depth through XPS depth profiling. Figure 8 shows that “[t]he concentration of Mg and Ti decreases rapidly over the first 50 nm of the particles.” *Id.* ¶ [0035]. The measurements “suggest an in-situ coating mechanism” where Mg and Ti “are accumulated at the surface of the LiCoO₂ particle in oxidized form.” *Id.* As shown below, the concentration of Mg peaks at the surface, at a depth of 0 nm, at approximately 5.4 atomic % and decreases to approximately 1 atomic % “over the first 50 nm of the particle[.]”



Id., Fig. 8, ¶ [0135].

2. Overview of Shim (Ex. 1006)

117. US Pat. Pub. No. 2016/0149215 A1 (“Shim”) is a patent application from Samsung SDI filed on November 20, 2015 and is directed to the development of a positive electrode for application in a high-performance lithium-ion battery. Specifically, Shim teaches the use of a doped lithium titanium oxide on the surface of a “lithium intercalation compound” as the positive electrode.

118. Shim discloses various compositions for lithium intercalation compounds. For example, Shim states that “[t]he lithium intercalation compound may be an M-doped compound (where M is Mg, Al, Ti, La, Tb, Gd, Ce, Pr, Nd, Sm, Ba, Sr., Ca, or a combination thereof).” *Id.*, ¶ [0063]. Shim teaches several “chemical formulae” that can represent the lithium intercalation compound. *See id.*, ¶ [0065]. Moreover, Shim provides several examples with specific lithium intercalation compounds, including LiCoO_2 . *Id.*, ¶ [0125].

119. Shim discloses examples of PEAM coatings. For example, Shim states that the coating can be “represented by Chemical Formula 1 on the surface of the lithium intercalation compound.



. . . [where], $0 < x \leq 3$, $1 \leq y \leq 5$, $-0.3 \leq z \leq 0.3$, and M is an element selected from Mg, Al, Ga, La, Tb, Gd, Ce, Pr, Nd, Sm, Ba, Sr, Ca, and combinations thereof.” *Id.*, ¶¶ [0011]–[0016]. Figures 4B, 5B, and 6B show a coating on the surface of the lithium intercalation compound:

FIG. 4B

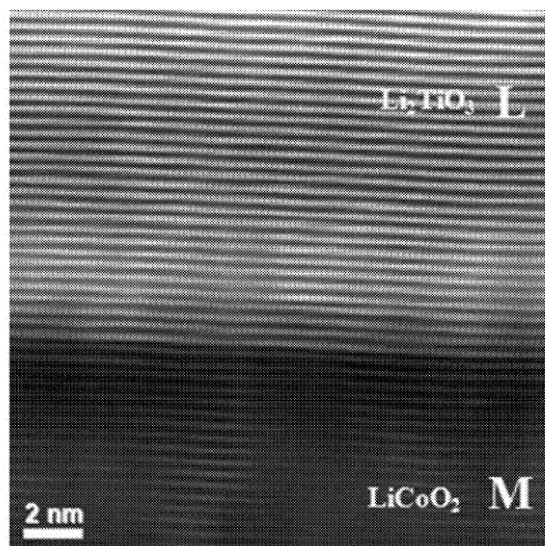


FIG. 5B

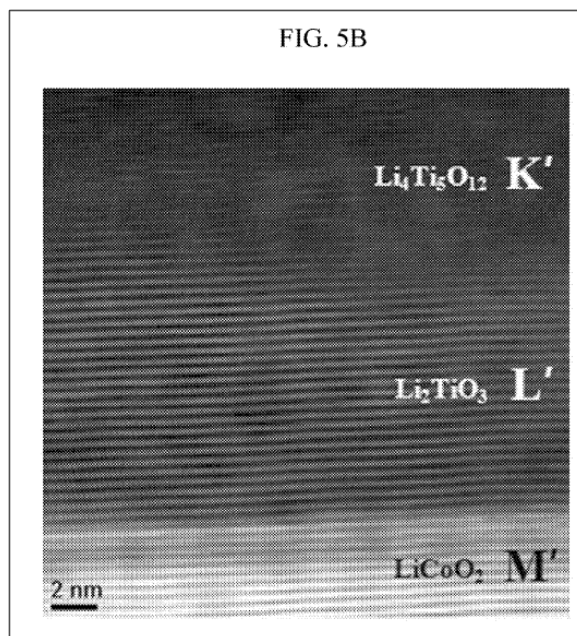
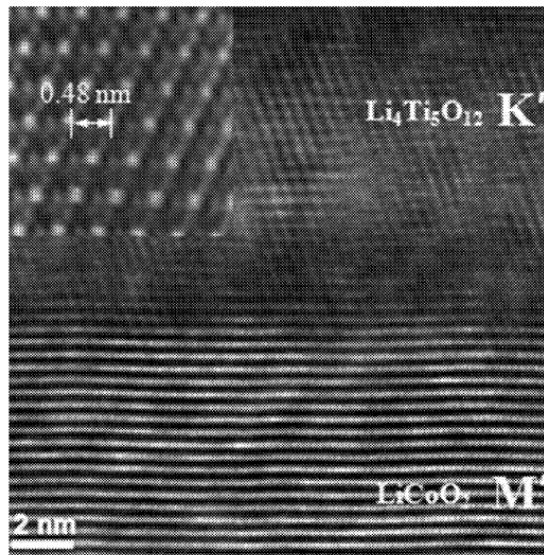


FIG. 6B



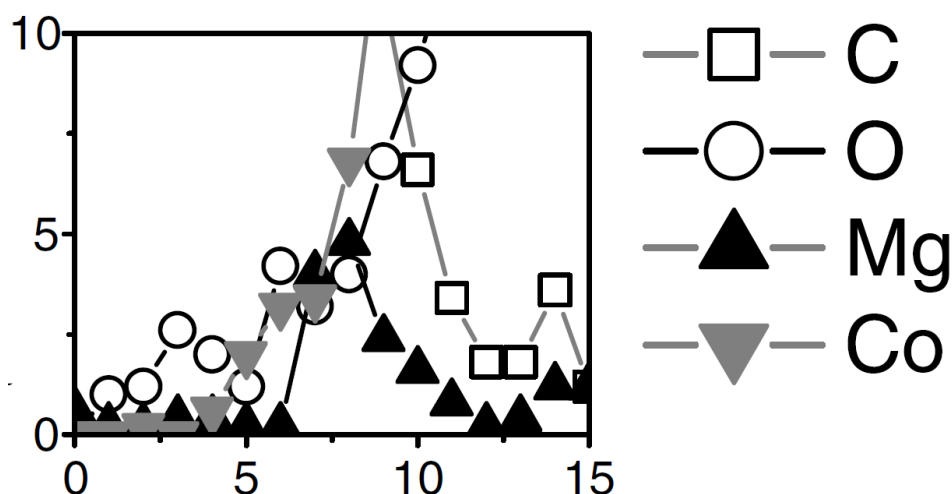
Id., Figs. 4B, 5B, and 6B. In the above images, LiCoO_2 is the lithium intercalation compound and lithium titanate is the coating. The angles between the bright and dark lines in the coating regions and the lithium intercalation regions shows that the crystal structures are substantially aligned.

120. Shim describes the evaluation of different LiCoO_2 PEAMs using SEM/EDS to characterize the morphology and elemental distribution near the surface. “As shown in FIG. 2B [EDX analysis of Example 1], Ti and Mg were simultaneously positioned on the surface of the active material. This result shows that Mg, a doping element in the LiCo-MgO -was diffused into the coating layer during the heat treatment.” *Id.*, [0131]. Shim also describes evaluating these different PEAMs using TEM with FFT image analysis to analyze the crystal structure.

3. Overview of Yamamoto (Ex. 1007)

121. 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*, 891–96 (“Yamamoto”) (Ex. 1007) was published on October 5, 2014. Yamamoto investigates the degradation mechanism and coating effects at the cathode/electrolyte interface using MgO-coated LiCoO₂ thin-film electrodes. *Id.*, 1. The investigation found “[t]he MgO coating improves the durability of the cathode during high-potential cycling.” *Id.* Further, “the MgO coating suppresses drastic distortions of local structure at the LiCoO₂ surface.” *Id.*

122. Specifically, Yamamoto undertook EDX line scan analysis to characterize the amount of magnesium through a depth of the particle. As shown below, the concentration of magnesium (y-axis, net counts) varies as a function of depth (x-axis, nanometers):



Id., Fig. 1 (excerpted).

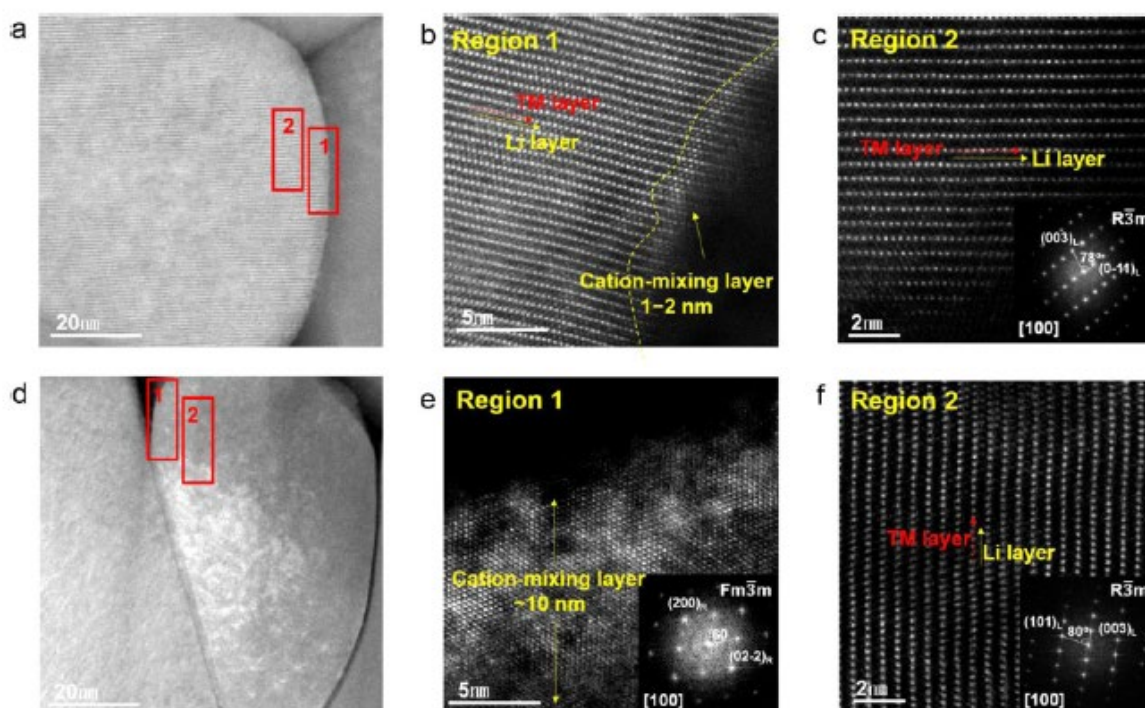
4. Overview of Cho (Ex. 1008)

123. U.S. Patent No. 10,283,770 (“Cho”) is a patent application from authors at Samsung SDI filed on December 17, 2015 and is directed to the development of a composite cathode active material for application in a high-performance lithium-ion battery. Specifically, Cho teaches a PEAM particle that includes a coating layer containing lithium and cobalt on the surface of a primary particle. Ex. 1008, 2:5–10. Cho further teaches that the primary particle can possess a layered structure, while the coating layer can possess a rock-salt structure. Specifically, Cho indicates that “the cathode active material may be . . . LiCoO_2 ” (*id.*, 10:8–9) while the “exterior” of the particle may have a rock-salt structure—“an exterior of the primary particle at region 1 had a structure having a disordered phase and that the crystal structure having a rock-salt structure (an isometric system, a space group $\text{Fm}[-3]\text{m}$). Region 2 was formed of a mixed phase. The coating layer formed on a surface of the primary particle may have a structure that does not have a boundary between the primary particle and the layer.” *Id.* 17:58–67.

5. Overview of Kim (Ex. 1009)

124. Kim is a peer-reviewed research article pertaining to the surface modification of PEAM particles and is directed towards improving the structural and thermal stability of the PEAM. Two of the authors of Kim overlap with those of Cho, as do several of the images used in both references. *See* Exs. 1008, 1009.

125. Like Cho, Kim discloses PEAM primary particles as consisting of two regions. The inner region, which comprises most of the particle, possesses the nominal layered phase, while the outer region is labeled the “cation-mixing” phase. *Id.*, Abstract, 2. Importantly, and as shown below, Kim uses transmission electron microscopy and fast Fourier transform (FFT) analysis to determine that these two regions possess the $R\bar{3}m$ and the $Fm\bar{3}m$ space groups, respectively:



Id., Fig. 2 (annotations in original).

126. Kim also tested the impact of the surface coating on cycling performance and thermal stability of the PEAM. Electrochemical cells were fabricated using surface-modified PEAMs and compared to analogous cells fabricated using unmodified PEAMs. The PEAM with surface treatment showed

improvement in cycling stability with no loss in high-rate cycling performance. Through postmortem cross-sectional SEM and TEM analysis, Kim observed less intergranular cracking and less severe phase changes at the primary particle surface for the PEAM with surface treatment. Furthermore, Kim also found that the surface modification improved the thermal stability of the PEAMs. The surface-modified PEAMs exhibited a higher oxygen evolution temperature when subjected to calorimetric testing. Kim attributes these benefits to the presence of the surface cation-mixing layer (possessing an $Fm\bar{3}m$ space group), which acts as a “pillar layer” and provides additional structural stability when more lithium is extracted from the PEAM at high voltages. *Id.*, 8.

B. Ground I.A: Claims 1–5 Are Rendered Obvious by Blangero Alone or in Combination with Shim

127. In my opinion, Shim in combination with Blangero renders obvious a lithium-ion secondary battery described by the challenged claims.

1. Motivation to Combine Blangero with Shim

128. In my opinion, a POSITA would have been motivated to combine Blangero with Shim. Specifically, Blangero and Shim are in the same technical field and are directed to improving PEAMs for enhanced battery performance. Both Blangero and Shim describe the development of substantially similar doped LiCoO_2 cathode particles with metal oxide surface coatings that contain titanium and

magnesium. Ex. 1006, ¶ [0011] (“[A] positive active material for a rechargeable lithium battery includes a lithium intercalation compound; and *a lithium titanium oxide* . . . on the surface of the lithium intercalation compound.”) (emphasis added) [0053], [0056], [0134]; Ex. 1005, ¶¶ [0004], [0005], [0094], [0108]–[0110] (describing particles based on LiCoO_2 doped and coated with Mg and Ti). Moreover, Blangero and Shim disclose manufacturing PEAM particles with similar manufacturing techniques that include a two-step firing (sintering) process that results in the formation of a LCO bulk in the center of the PEAM and a surface coating. Ex. 1005, ¶¶ [0108]–[0110]; Ex. 1006, ¶¶ [0119]–[0123], [0129].

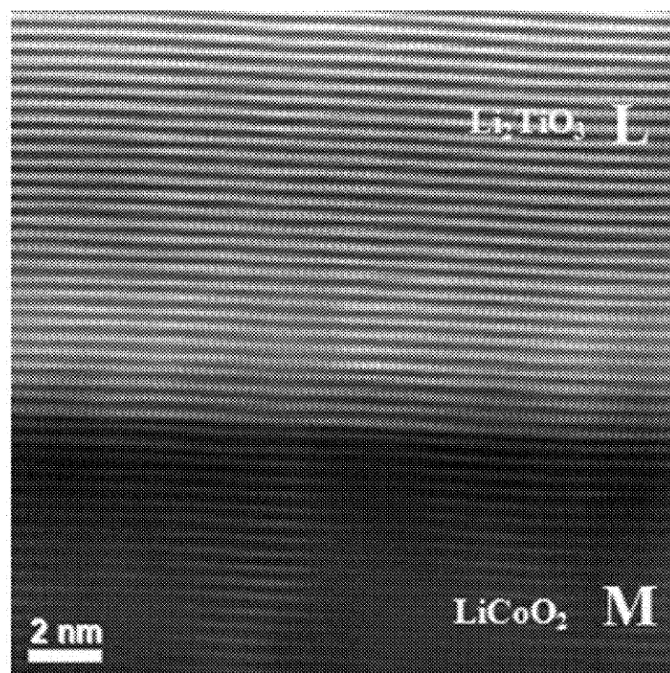
129. Indeed, both Blangero and Shim describe the same goal of using magnesium and titanium dopants and coatings in the development of PEAM particles as an approach to achieve a lithium-ion battery with improved electrochemical performance. *See* Ex. 1005, ¶¶ [0004], [0005]–[0007] (describing coating LiCoO_2 to improve stability while maintaining capacity); Ex. 1006, ¶¶ [0057] (improving “electrical characteristics at the surface of the active material” using coating containing Mg and Ti), [0059] (describing “excellent rate capability and cycle-life characteristics” due to doping lithium titanate coating), [0060]. Both references teach doping LiCoO_2 with various dopants, including “Mg, Al, Ti,” and others. Ex. 1005, ¶¶ [0004], [0062], [0087], [0109] (describing “state of the art Mg and Ti doped LiCoO_2 ”); (Ex. 1006, ¶¶ [0019], [0025], [0063]). In my opinion, a

POSITA would have had a reasonable expectation of success in combining the teachings Blangero and Shim. It was well known that transition metals including magnesium and titanium could be added as dopants to the bulk of a PEAM particle. It was further known in the art that the use of such metals in a coating layer improved PEAM performance. The combination of the teachings in Blangero and Shim would have required only routine experimentation and would require applying known manufacturing techniques from these references to build an improved PEAM particle for use in a secondary lithium ion battery.

130. Blangero also teaches that the use of titanium and magnesium precursors during the two-part synthesis process results in an “in-situ” coating of a surface layer that “is formed during the second sintering by spontaneous segregation of the dopants from the core.” Ex. 1005, ¶ [0090]. This surface layer includes “enriched in oxidized compounds, like MgO and TiO₂, that may also be lithiated.” Ex. 1005, ¶¶ [0090], [0094]. Blangero teaches that this surface layer is beneficial because the “inert coating layer provides additional safety when the powders are in contact with the electrolyte of the battery.” *Id.*, ¶¶ [0093]–[0094]. Blangero further discloses that the coating may include Li₂TiO₃. *Id.*, ¶ [0134]. This addresses the problem where uncoated PEAMs react with the electrolyte, causing electrolyte decomposition and, over time, results in a loss of capacity and an increase in the battery’s impedance. *Id.* [0093]–[0094]. Shim similarly teaches using magnesium

and titanium as dopants and as a coating on the surface of a lithium intercalation compound in a PEAM. Specifically, Shim teaches a “lithium titanium oxide” “positioned on the surface of the lithium intercalation compound in a form of an *island coating*.” Ex. 1006, ¶ [0058]. Shim’s island coating, which is preferably doped with magnesium, exhibits “excellent rate capability and cycle-life characteristics during the charge and discharge of the battery.” *Id.*, ¶ [0059]. Further, Shim discloses the inclusion of magnesium as a dopant in the bulk of the LiCoO_2 , and in the disclosed Li_2TiO_3 coating layer and in the enumerated example and comparative examples. *Id.* ¶¶ [0117]–[0149]. I have reproduced a TEM image of a cross section of the surface of Shim’s PEAM according to Shim’s Example 2 below (Shim notes that Mg is present in both layers but not included in the figure annotations):

FIG. 4B



Id. Fig. 4B. As shown in the figure above, Li_2TiO_3 is present on the surface of LiCoO_2 lithium intercalation compound. *Id.*, Fig. 4b. While it is not labeled in the above image, the bulk and coating layers also include magnesium. *Id.*, ¶ [0134].

131. Both Blangero and Shim disclose considerably similar two-firing-step solid state synthesis processes. For example, in both Example 1 disclosed by Blangero and Example 1 disclosed by Shim, initial precursors are mixed and then fired (sintered) to create a doped- LiCoO_2 base material, and then additional precursors are mixed in and a second firing step (925 °C for 12 hours in Blangero and 950 °C for 5 hours in Shim) is performed to help form the surface coating. *See* Ex. 1005, ¶ [0110] (describing a second firing step at 925 °C); Ex. 1006, ¶ [0118]

(describing a second firing step at 950 °C). In fact, both Blangero and Shim describe the diffusion of the dopants from the bulk to the surface during the second firing step as important in the creation of their coatings.

132. For these reasons, in my opinion a POSITA would have understood that Blangero and Shim teach highly similar, specific technological features. A POSITA would have understood the teachings of Blangero's doped LCO particles with surface coatings to be compatible with other doped LCO particles with surface coatings, including the particles disclosed by Shim. Thus a POSITA would have been motivated to combine the teachings of Blangero with Shim.

2. Independent Claim 1

a. [Preamble]: A lithium-ion secondary battery comprising:

133. In my opinion, to the extent the preamble is limiting, Blangero discloses it. Blangero is directed to a "lithium metal oxide powder for use as a cathode material in a rechargeable battery." Ex. 1005, Abstract. Blangero explains "[d]ue to their high energy density rechargeable lithium and lithium-ion batteries can be used in a variety of portable electronic applications" and that "[c]ommerciably available lithium-batteries typically consist of graphite-based anode and LiCoO₂-based cathode materials" *Id.* ¶ [0002]. Blangero identified a need for "LiCoO₂-based materials with increased specific capacity." *Id.* Accordingly, Blangero explains "[i]t is an object of the present invention to define a cathode material having a high

packing density, high rate performance, improved discharge capacity and showing high stability during extended cycling at high charge voltage for high end secondary battery applications. *Id.* ¶ [0008].

- b. [Element 1.1]: a positive electrode, the positive electrode comprising a positive electrode active material particle comprising lithium cobaltate,

134. In my opinion, Blangero discloses this limitation. A POSITA would have known that lithium cobaltate generally refers to LiCoO_2 , also called lithium cobalt oxide or LCO, and was commonly used as the intercalation compound in the bulk of a positive electrode active material, or PEAM, in lithium-ion batteries. The '660 specification confirms this, referring to lithium cobaltate as LiCoO_2 . Ex. 1001, 22:31–33 (“A region 111 of the drawings contains lithium, cobalt, magnesium, and fluorine, and **lithium cobaltate (LiCoO_2)** is a main component of the region 111.”).

135. Blangero describes the well-documented use of “ LiCoO_2 -based cathode materials” in the art, including their regular use in consumer electronics, studies on the size of PEAM particles and packing density, Mg and Ti doping of LiCoO_2 particles, and the use of various coatings or chemical modifications. Ex. 1005, ¶ [0002], [0004]–[0007]. Blangero discloses using $\text{LiCo}_{1-a-b}\text{M}'_a\text{M}''_b\text{O}_2$, where $0 < a \leq 0.05$ and $0 < b \leq 0.02$, M' can be one or more elements including magnesium and titanium, and M'' can be fluorine. *Id.*, ¶ [0011]. In one embodiment, “Li-enriched lithium metal oxide compound is LiCoO_2 with dense monolithic particles of at least

5 and preferably at least 10 to 20 micrometers” and Blangero notes that “[m]any commercial prior art LiCoO_2 particles already have this desired morphology.” *Id.* ¶ [0072]. Blangero further describes measuring PEAMs prepared using “state of the art Ti and Mg-doped lithium cobalt oxide” which it refers to as LCO-1, LCO-3, and LCO-4, that are “prepared according to a conventional method.” *Id.* ¶ [0087]. Enumerated Examples 1–4 use “Li-stoichiometry controlled LiCoO_2 -based cathode materials.” *Id.* ¶¶ [0108]–[0118]. Example 5 uses state-of the art LiCoO_2 -based materials with lithium overstoichiometry. *Id.* ¶¶ [0119]–[0122].

- c. [Element 1.2]: wherein the positive electrode active material particle comprises a first region and a second region,

136. In my opinion, Blangero discloses this limitation and discloses a positive electrode active material particle that comprises a first region and a second region. Blangero discloses a coating or “surface layer” (second region) over the surface of “ LiCoO_2 materials” (first region). *See id.*, ¶¶ [0009]–[0011], [0027], [0059], [0062], [0066], [0094]. The use of coatings were well known in the art, which Blangero confirms noting “[t]o achieve high voltage stability, LiCoO_2 materials are usually coated (for example with Al_2O_3) or otherwise chemically modified (e.g. by providing a fluorinated surface” and for this reason treated LiCoO_2 is used in commercial batteries. *Id.*, ¶¶ [0006]–[0007]. Blangero provides a “summary” of the disclosed invention which is a “lithium metal oxide powder for

use as a cathode material in a rechargeable battery” consisting of two major components: (1) “a core material consisting of the elements Li, a metal M and oxygen,” where M is at least 95% cobalt; and (2) “a surface layer consisting of a mixture of elements of the core material and inorganic N oxide particles,” where N is one or more of a group consisting of magnesium and titanium. *Id.*, ¶ [0009]. In one embodiment Blangero’s PEAM has a “core,” as described in ¶ [0010] and elements “present on the surface” as described in ¶ [0011]. Blangero describes another embodiment with a “method for manufacturing a lithium metal oxide powder consisting of a core material and a surface layer, the core material having a layered crystal structure consisting of the elements Li, a metal M and oxygen.” *Id.*, ¶ [0027]; *see also id.*, ¶¶ [0059] (describing a PEAM with “a core material consisting of the elements LI, the metal M and oxygen . . . and an electron-insulating surface layer consisting of a mixture of the elements of the core materials or inorganic N-based oxides or combinations thereof” where N may include metals consisting of Mg and Ti), [0062] (describing MgO, TiO₂, and/or Li₂TiO₃ in the surface layer), [0066] (indicating “[t]he surface layer is typically between 20 nm to 200 nm, and is preferably between 20 nm and 100 nm thick”), [0094] (explaining “a cathode material protected by an insulating surface layers will solve this problem”—referring to deterioration of battery performance due to unwanted electrolyte decomposition

on the surface of the PEAM—“by physically separating tetravalent cobalt ions from the electrolyte and eventually preventing further electrolyte reduction.”).

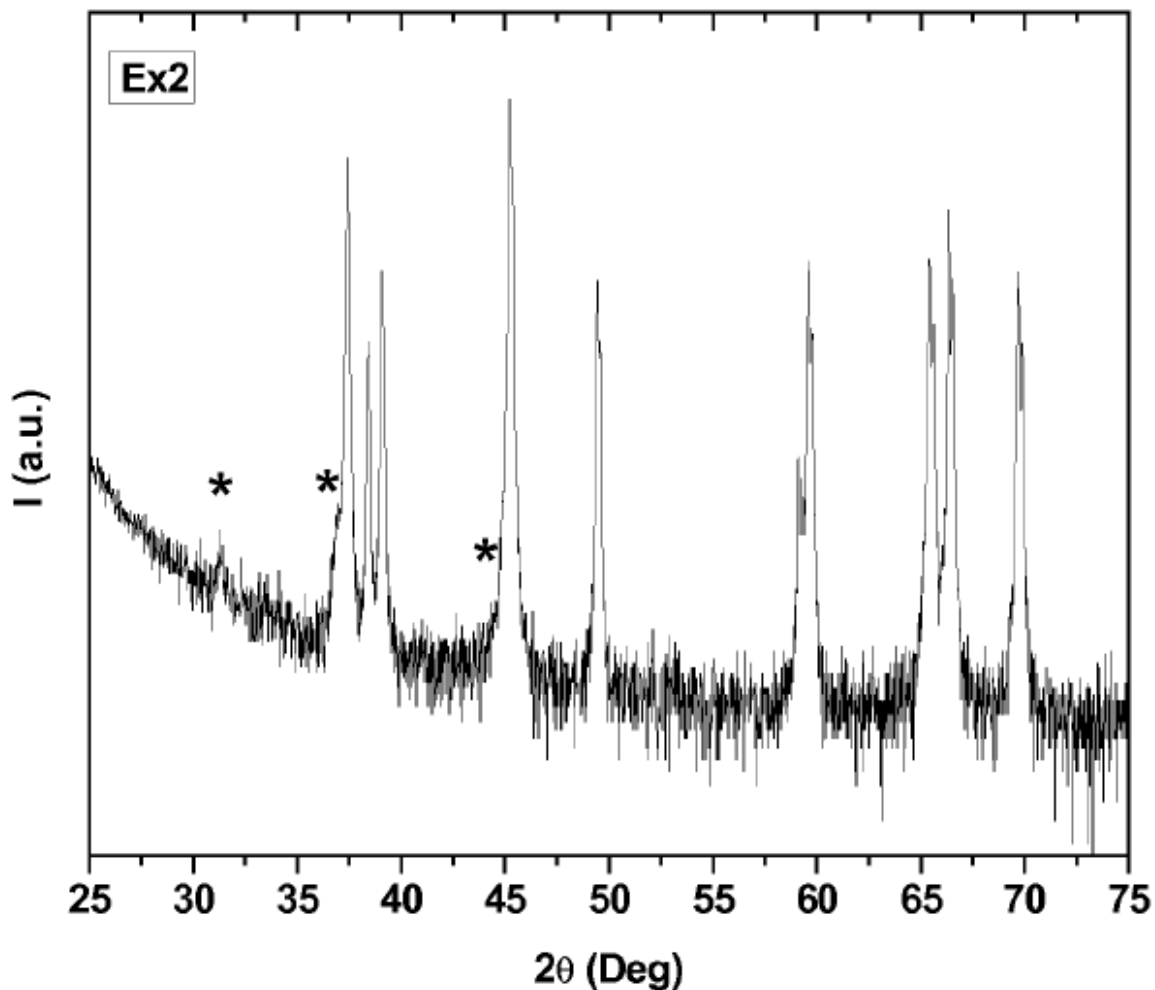
137. Blangero observes that when “dopants N are present, the surface is formed during the second sintering by spontaneous segregation of the dopants from the core.” *Id.*, ¶ [0090]. This forms an “in-situ” coating where “N dopants such as Mg and Ti are expelled and accumulated at the surface of the particles.” *Id.* The end result is the formation of “an in-situ coating of the final [LiCoO₂] powder with a layer that is enriched in oxidized compounds, like MgO and TiO₂, that may also be lithiated.” *Id.*, ¶¶ [0090], [0093], [0135]. Blangero explains that “[t]his inert coating layer provides additional safety when the powders are in contact with the electrolyte of the battery.” *Id.*, ¶ [0094].

- d. [Element 1.3]: wherein the first region comprises a layered rock-salt crystal structure,

138. In my opinion, Blangero discloses a first region, wherein the first region comprises a layered rock-salt crystal structure. Layered rock-salt crystal structures were well understood by a POSITA. In addition to this understanding, the specification of the '660 Patent defines “layered rock-salt crystal structure” as having a “space group of . . . R-3m,” as opposed to “space group Fm-3m of a general rock-salt crystal.” Ex. 1001, 8:67–9:3. In my opinion, a POSITA would have known that a LiCoO₂ PEAM would have a layered rock-salt structure (R-3m). *See* Ex. 1006,

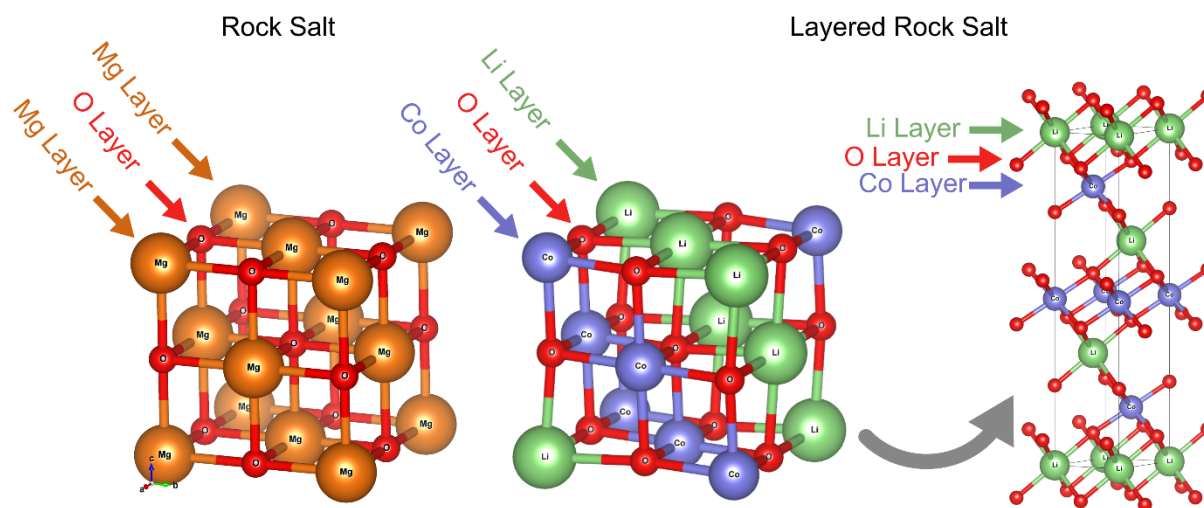
Figs. 4C, 5C, 6C (fast Fourier transform images showing R-3m structure of LiCoO₂); Ex. 1010, 3 (confirming the “hexagonal structure of LiCoO₂ (R-3m)”); Ex. 1025, 1; Ex. 1026, 4–5.

139. Blangero confirms that the LiCoO₂ first region of its PEAM has layered rock-salt crystal structure with a space group of R-3m: “[t]he core has a *layered* structure—it is known that lithium cobalt[a]te consists of layers of lithium that lie between slabs or layers of octahedra formed by cobalt and oxygen atoms[.]” Ex. 1005, ¶ [0010]; *see also id.*, ¶ [0027] (“In another embodiment, the method for manufacturing a lithium metal oxide powder consisting of a core material and a surface layer, the core material having a **layered crystal structure** consisting of the elements Li, a metal M and oxygen....”). Blangero examined one embodiment, Example 2, via X-ray diffraction (XRD) and determined “the main peaks” of the XRD pattern of are “attributed to the layered LiCoO₂-based oxide (*R-3m space group*)”:

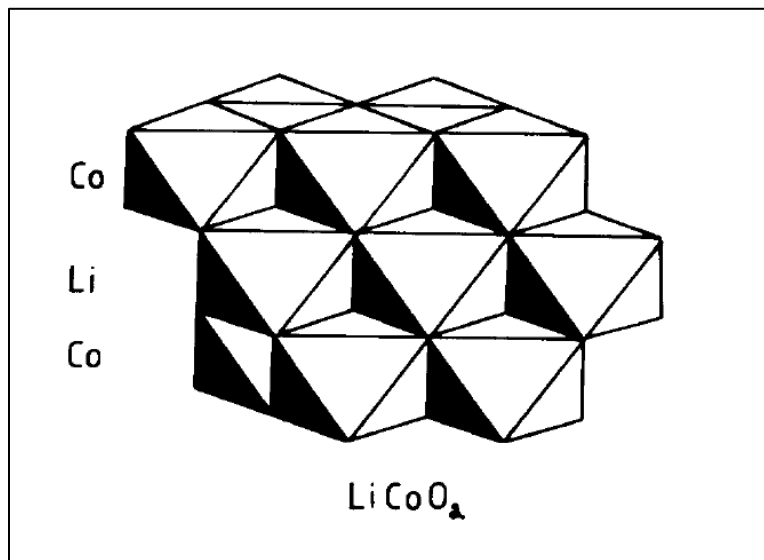


Id., Fig. 3, ¶ [0126]; *see also id.*, ¶¶ [0027] (“[T]he core material having a **layered crystal structure** consisting of the elements Li, a metal M and oxygen”), [0060], [0124], [0149]. A POSITA would have understood that the disclosed R-3m space group of the “layered LiCoO_2 -based oxide” is a layered rock-salt, as claimed by the ’660 patent.

140. This disclosure in Blangero confirms that LiCoO_2 was generally known to exist in a layered rock-salt structure (R-3m). Lu (a 2009 study) describes indexing their X-ray diffraction results of LiCoO_2 material (including coated and uncoated, pristine and cycled samples) “to a hexagonal unit cell with rhombohedral symmetry having space group R-3m.” Ex. 1026, 4–5. Similarly, Rao (a 2012 study) noted that “[t]he structure of layered LiCoO_2 has Rhombohedral symmetry and belongs to the space group $R\bar{3}m$, and is ideally suited to accommodate large changes in Li concentration. This crystal structure consists of close-packed oxygen ion layers separated by alternating layers of Li and Co ions.” Ex. 1025, 1. Rock-salt and layered rock-salt structures are shown below (using MgO and LiCoO_2 as respective examples):



Additionally, Mizushima et al. showed a similar layered rock-salt structure in 1980:



Ex. 1033, Fig. 1, 2 (“[I]t was then natural to consider the transition-metal lithium oxides LiMO_2 crystallizing in the layered structure illustrated in Fig. 1. The Li^+ and M^{3+} ions occupy alternate (111) layers of the rocksalt structure[.]”).

e. [Element 1.4]: wherein the second region comprises a rock-salt crystal structure,

141. In my opinion, Blangero renders obvious a second region wherein the second region comprises a rock-salt crystal structure. Blangero discloses forming a surface layer on a PEAM particle comprising MgO . *See* Ex. 1005, ¶¶ [0059] (describing “an electron-insulating surface layer consisting of a mixture of the elements of the core materials or inorganic N-based oxides” where N may be Mg), [0062] (describing a PEAM including “ MgO ” on the surface as a result of “segregation from the core and accumulation of [] metal dopants at the surface of the particles”), [0094] (explaining this is beneficial to battery performance by

“physically separating tetravalent cobalt ions from the electrolyte and eventually preventing further degradation” through the “careful selection of the compounds” including Mg and Ti which achieves “an in-situ coating of the final powder with a layer that is enriched in oxidized compounds, like MgO and TiO₂”).

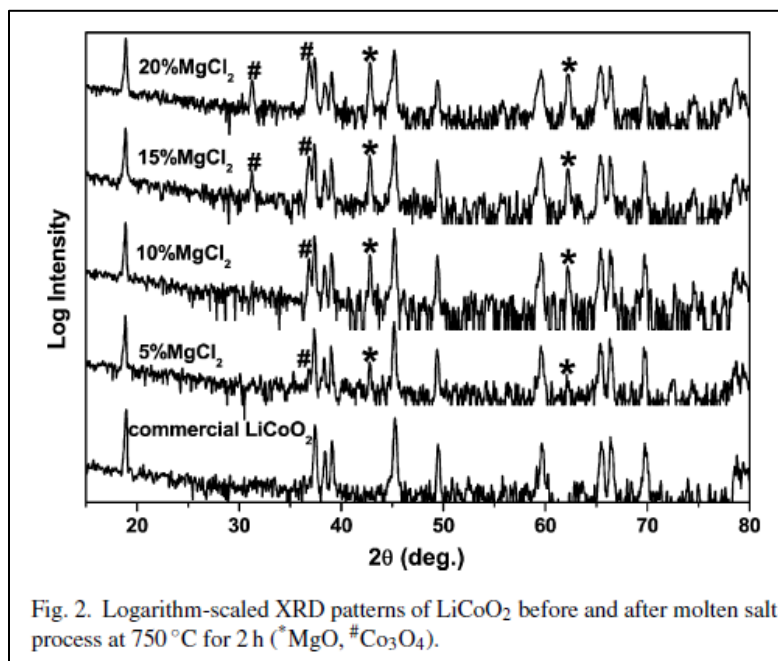
142. In my opinion, A POSITA would have known that at least the MgO portion of Blangero’s coating would have had a rock-salt crystal structure, as described in Section VI.A.10 above. A textbook from 1976 confirms that MgO has long been known to assume this crystal structure:

Table 4.5
SOME COMPOUNDS WITH THE SODIUM CHLORIDE STRUCTURE

CRYSTAL	<i>a</i> (Å)	CRYSTAL	<i>a</i> (Å)	CRYSTAL	<i>a</i> (Å)
LiF	4.02	RbF	5.64	CaS	5.69
LiCl	5.13	RbCl	6.58	CaSe	5.91
LiBr	5.50	RbBr	6.85	CaTe	6.34
LiI	6.00	RbI	7.34	SrO	5.16
NaF	4.62	CsF	6.01	SrS	6.02
NaCl	5.64	AgF	4.92	SrSe	6.23
NaBr	5.97	AgCl	5.55	SrTe	6.47
NaI	6.47	AgBr	5.77	BaO	5.52
KF	5.35	MgO	4.21	BaS	6.39
KCl	6.29	MgS	5.20	BaSe	6.60
KBr	6.60	MgSe	5.45	BaTe	6.99
KI	7.07	CaO	4.81		

Ex. 1013, 80 (annotated). It was well understood in the art that MgO is stable in a rocksalt structure under normal conditions. For example, Xu noted that the rock-salt

crystal structure for MgO was “the stable phase at room temperature and pressure conditions,” meaning crystalline MgO will be in the rock-salt phase under normal conditions. Ex. 1030. The crystalline nature of the MgO would also have been obvious to a POSITA given the synthesis methods disclosed in Blangero. Specifically, the second firing step involving 925 °C for 12 hours. *See, e.g.*, Ex. 1005 ¶ [0110]. As discussed in Section VI.A.10, MgO is well known to crystallize even at substantially lower temperatures (e.g., 500 °C) (and will do so more readily at 925 °C). Ex. 1030. Bai 2007 discloses a solid state synthesis involving firing at 750 °C for 2 hours, a lower temperature and shorter duration than Blangero, and still reported evidence of the formation rock salt MgO (longer sintering times at higher temperatures in Blangero would be more conducive to the crystallization of MgO in the rock salt structure). Figure 2 shows the X-ray diffraction (XRD) patterns, of the resulting MgO-coated LiCoO₂—as shown below, peaks marked with asterisks (*) correspond with MgO, which a POSITA would have understood to be consistent with the 200 and 220 peaks characteristic of rock salt MgO (see further discussion on MgO in Section VI.A.10):



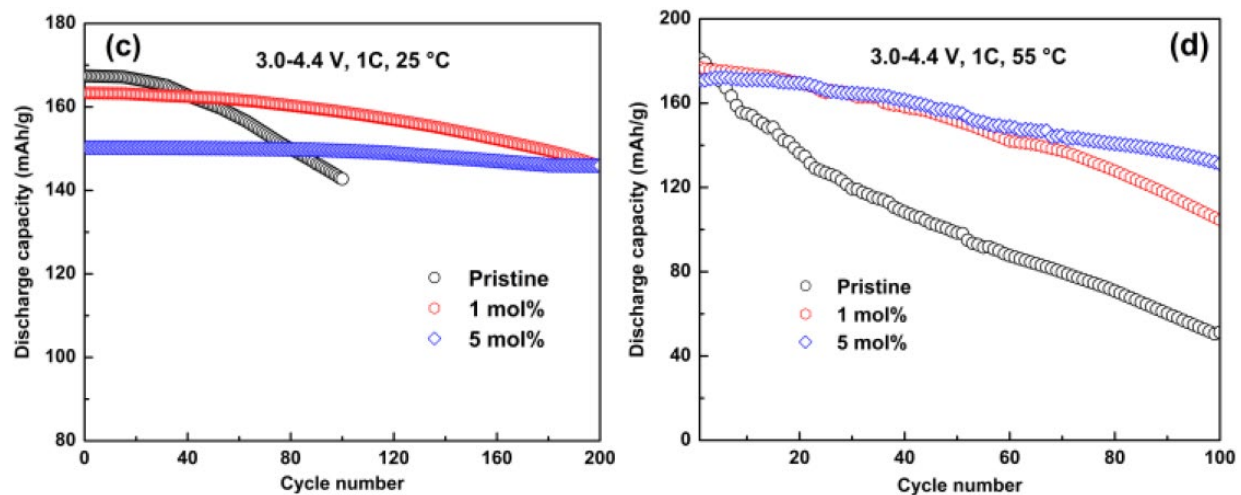
Id., Fig. 2. A POSITA would have understood that, from this figure and disclosure, the fact that XRD analysis revealed discernible peaks associated with MgO means that the MgO is crystalline and in the rock salt structure. *See id.*, 3 (“Besides hexagonal LiCoO_2 , diffraction peaks of MgO and Co_3O_4 are observed[.]”).

143. Separately, and for the same reasons as described above, in my opinion Blangero’s disclosure of lithium fluoride “on the surface under the form of LiF ” discloses this limitation. Ex. 1005, ¶ [0011]. Blangero also recognizes that in the art “materials are usually coated” and that “ LiF protective coatings” were known. *Id.*, ¶ [0006]. As shown in the chart above from Ashcroft & Mermin, *SOLID STATE PHYSICS*, Cengage Learning, 1st ed. (1976), LiF also has rock salt structure. Ex. 1013, 80. Blangero repeatedly references surface LiF in the specification and claims. Ex. 1005, ¶ [0011], cls. 33, 37, 53, 62. In my opinion, a POSITA would have

understood from these disclosures that Blangero discloses at least a region of the second region comprising a rock-salt structure.

144. In the alternative, is it my opinion that Blangero's disclosure of lithiated titanium oxide on the surface of the particle renders this limitation obvious. Blangero's surface layer further comprises "TiO₂, that may also be lithiated." Ex. 1005, ¶ [0094]. Blangero explains that "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" can be achieved by using "an electron-insulating surface layer" consisting of a mixture of metal oxides, including Ti. *Id.*, ¶ [0059]; *see also id.*, ¶ [0062] (describing a surface enriched with metal oxides including Li₂TiO₃). Moreover, it was well known at the time that lithium titanate could assume a rock-salt structure. *See* Section VI.A.9. Indeed, "[r]ock salt-type LiTiO₂," which exhibited "superior structural stability . . . as compared to other Li-Ti-O oxides." Ex. 1029, 1, 4; Ex. 1034; Ex. 1037.

145. In my opinion a POSITA would have had a reasonable likelihood of success in using rock-salt lithium titanate as a coating to improve lithium-ion battery electrochemical characteristics. Below, I have reproduced the figures from Wang's 2012 study that shows that the cycling performance of PEAMs was improved by using rock salt LiTiO₂ coatings of 1 and 5 mol % compared to a "pristine" PEAM that did not include such coating:



Id., Fig. 4(c)–(d). As shown above, PEAMs that did not have the rock salt LiTiO_2 coatings experienced significant degradation in their discharge capacity after approximately 100 cycles. By contrast, PEAMs with rock salt LiTiO_2 coatings had much less degradation at 100 cycles and even retained better performance at 200 cycles than the pristine PEAM did at 100 cycles. This same study from Wang noted that “*rock salt-type LiTiO_2 can be used as the suitable candidate of coating materials* toward developing higher energy density electrode materials together with longer cycle life for LIBs,” *Id.*, 5 (emphasis added). Wang further found that rock salt LiTiO_2 coatings “*exhibited inspiring high-voltage cycling capability* as well as *thermal stability* than the pristine counterpart” and that “[s]uch impressive cycling performances are attributed to the *robust structure feature of LiTiO_2* protective layer and the *strengthened interfacial stability* between [the] coating layer” and the bulk material, or lithium intercalation compound. *Id.* (emphasis added). Therefore,

it is my opinion that a POSITA would have known that the lithium titanate coating in Blangero's PEAM could have been a rock-salt form of lithium titanate.

146. For these reasons, it is my opinion that Blangero discloses a PEAM comprising a second region wherein the second region comprises a rock-salt crystal structure. Blangero discloses the presence of MgO as well as the presence of LiF on the surface of the particle and both of these compounds were known to adopt a rock-salt structure and each of which independently satisfies the claim limitation. Further, Blangero in view of the knowledge of a POSITA further renders this limitation obvious through its disclosure of lithium titanate. It was known in the art that lithium titanate commonly exists in rock salt phases, and a rock salt phase of lithium titanate had been demonstrated improve PEAM performance as a coating layer. Thus a POSITA would have been motivated to use a rock-salt lithium titanate coating with Blangero's PEAM.

- f. [Element 1.5]: wherein the second region covers at least a first part of the first region,

147. In my opinion, Blangero discloses this limitation. As explained above in Section IX.B.2.c, Blangero's PEAM consists of "a core material" (first region) and "a surface layer" (second region). Ex. 1005, ¶ [0009]; *see also id.*, ¶¶ [0010], [0011], [0027], [0059], [0062], [0066], [0094]. Blangero explains that "[w]hen dopants N are present, the surface is formed during the second sintering by

spontaneous segregation of the dopants from the core” forming an “in-situ” coating. *Id.*, [0090]. During this segregation process, dopants “are expelled” from the core material “and are accumulated at the surface of the particles,” thus forming a surface layer. *Id.*, ¶ [0090]. Blangero explains that “a cathode material protected by an insulating surface layer will solve” the problem of decomposition reactions between the electrolyte and PEAM surface “by physically separating tetravalent cobalt ions from the electrolyte and eventually preventing further electrolyte reduction.” *Id.*, ¶¶ [0093]–[0094]. Thus the “surface layer” provides a physical barrier between the core and the electrolyte solution, and necessarily covers at least a part of the first region. *see also id.* ¶¶ [0093]. [0135]. Blangero confirmed at least a first and second region in the particle by means of XPS depth profiling, which showed “[t]he concentration of Mg and Ti decreases rapidly over the first 50 nm of the particles” thus proving an “in-situ segregation mechanism of Mg and Ti.” *Id.*, ¶ [0135].

- g. [Element 1.6]: wherein the positive electrode active material particle comprises titanium, magnesium, fluorine, and oxygen in the second region, and

148. In my opinion, Blangero discloses this limitation. As explained above in Section IX.B.2.c, Blangero’s PEAM comprises “a surface layer consisting of a mixture of elements of the core material and inorganic N *oxide* particles,” where N is one or more of a group consisting of magnesium and titanium. *Id.*, ¶ [0009]; *see also id.*, ¶¶ [0010], [0011], [0027], [0059], [0062] (describing MgO, TiO₂, and/or

Li₂TiO₃ in the surface layer), [0066], [0094]. Specifically, the core may contain dopant metal M, which may be a variety of metals including magnesium and titanium, that are concentrated on the particles surface. *Id.*, ¶¶ [0010] – [0011]. In another embodiment, “the surface layer” consisted of various metals and “inorganic N-based oxides” where N may be Mg or Ti. *Id.*, ¶¶ [0011], [0027], [0059]. In one embodiment, “[t]he surface layer is an inhomogeneous composition compared to the core with a compositional gradient of the different M, M’, and Li and O elements” and “[t]he surface is enriched of elements N” which may be Mg or Ti. *Id.* ¶ [0062]. The resulting surface layer is “typically between 20 to 200 nm, and preferably between 20 and 100 nm thick.” *Id.*, ¶ [0066]. To form the coating layer, Blangero indicates that “careful selection of the compounds such as Mg . . . and Ti, the process allows to achieve an in-situ coating of the final powder with a layer that is enriched in oxidized compounds, like MgO and TiO₂.” *Id.*, ¶ [0094].

149. In my opinion, Blangero further discloses presence of fluorine in the surface layer. Specifically, Blangero teaches “[t]he element[] F . . . may be present on the surface under the form of LiF[.]” *Id.* ¶ [0011]. Blangero confirms that the presence of LiF in the “surface layer” in addition to metal oxides on the surface of the particle, including oxides of magnesium and/or titanium. Ex. 1005, ¶ [0011], cls. 33, 37, 53, 62.

titanium:

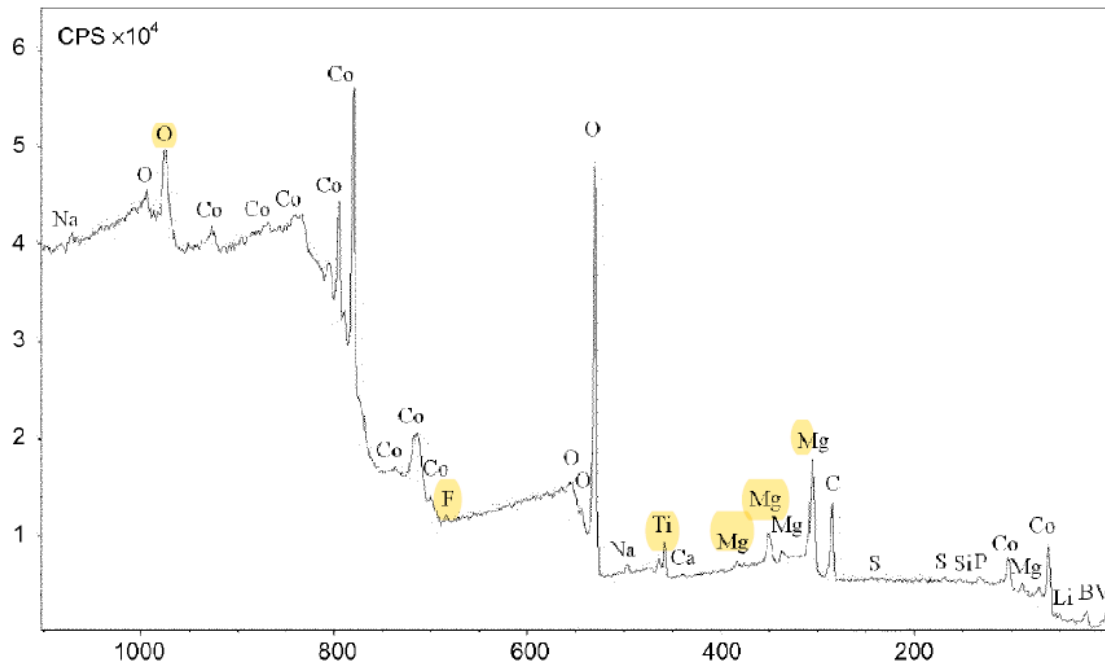


Figure 4(b)

magnesium:

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], Table 3; *see also id.*, [0094], [0096], [0108]–[0110]. As shown above, both Blangero’s XPS analysis and Table 3 confirm that cobalt, oxygen, magnesium, titanium, and fluorine are present on the surface of the particle. Thus, it is my opinion that Blangero discloses titanium, magnesium, fluorine, and oxygen in the second region.

- h. [Element 1.7]: 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.

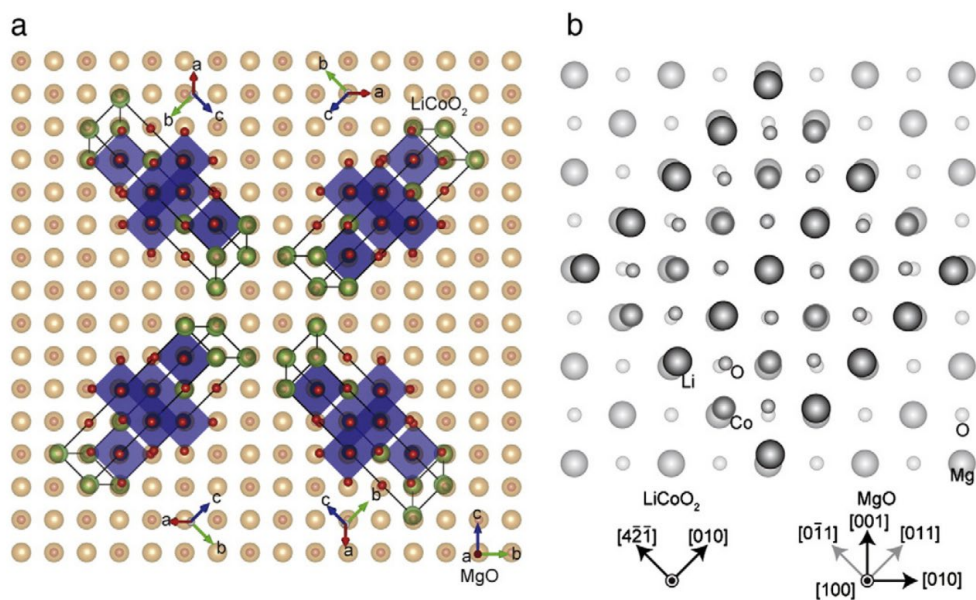
151. In my opinion, Blangero renders this limitation obvious either alone, with the knowledge of a POSITA, or in view of Shim. The ’660 Patent explains that “crystal orientations” are “substantially aligned with each other” “where the directions of the cubic closest packed structures formed of anions are aligned with

each other.” Ex. 1001, at 9:7–10; *see also id.* (describing viewing such alignment with common techniques, including TEM, STEM, HAADF-STEM, ABF-STEM, “and the like”). The ’660 Patent also explains that in such TEM images, the two claimed regions are “substantially aligned” when “an angle between the repetition of bright lines and dark lines in the layered rock-salt crystal [*i.e.*, first region] and the repetition of bright lines and dark lines in the rock-salt crystal [*i.e.*, second region] is less than or equal to 5°[.]” *Id.*, 9:22–28.

152. The phenomenon of epitaxial growth was well understood in the art. For instance, a 2015 paper by the same authors of Shim discloses that Li_2TiO_3 , one of the LTO compounds Blangero discloses, was “epitaxially grown on the surface of LiCoO_2 as a coating layer,” and that the “[001] direction of LiCoO_2 (*c*-axis) was ***completely parallel*** to the [001] direction of Li_2TiO_3 .” Ex. 1032, 4. Shim 2015 discloses the use of a two-firing-step solid state synthesis where the second firing is performed at 950 °C for 6 hours, similar to the second firing step in Blangero, 925 °C for 12 hours. Ex. 1006, ¶ [0118] (describing a second firing step at 950 °C); Ex. 1005, ¶ [0110] (describing a second firing step at 925 °C). In the light of the similar synthesis methods, a POSITA would expect the Li_2TiO_3 disclosed on the surface of the PEAM in Blangero to form in a considerably similar manner to the Li_2TiO_3 formed in Shim 2015, including its phase and alignment with the bulk.

153. Other studies have observed that “epitaxial coating may be an ideal strategy for improving the electrochemical performance of cathodes” due to the “the same lattice orientation and small lattice mismatch between the coating layer and the host material” (*i.e.*, lithium intercalation compound). Ex. 1035, 1, Fig. 3(a) (showing substantial alignment between an LMO cathode material and LTO layer).

154. In my opinion, a POSITA would also expect the MgO disclosed by Blangero to form with substantial alignment with the LiCoO_2 core given the established crystallographic compatibility of these two structures and the high-temperature, long-duration second firing step disclosed by Blangero. As discussed in Section VI.A.7, Katsui et al. demonstrated the epitaxial growth of LiCoO_2 films on three different orientations of bulk MgO single crystals. Ex. 1039. In each case, the same crystallographic alignment was achieved, one where the cubic-closest-packed anion sublattice of LiCoO_2 was aligned with that of MgO, as shown in the figure below, demonstrating the crystallographic compatibility of these two structures.



Ex. 1039, Figs. 3(a)-(b). As shown in the above schematic, LiCoO_2 was deposited on MgO and the two layers exhibited the same crystallographic alignment. The extent of lattice mismatches between the LiCoO_2 and MgO layers was “small” which “suggested the hetero-epitaxial growth of LiCoO_2 films on the MgO single-crystal substrates.” *Id.*, at 2. In my opinion, a POSITA would have understood that “hetero-epitaxial growth” refers to epitaxial growth between two different materials.

155. In the alternative, in my opinion the combination of Blangero and Shim renders this limitation obvious. Shim discloses using a coating (second region) that contains oxides of magnesium, and titanium, similar to Blangero’s coating, as discussed above in Section IX.B.1. Below, I have reproduced annotated Figures 4B, 5B, and 6B which are STEM images of PEAM cross sections that depict the coating, or second region, and lithium intercalation compound LiCoO_2 , first region. To

determine the angle between the two regions I superimposed a vertical line on top of each image. I then measured the angle between the vertical line and a plane of the crystal structure in the STEM image (for both the first and second regions). Once I obtained the angle of each region from a vertical line, I found the difference between the two measured angles. Below, I reproduced these annotations and have included the measured angle between the lines on the annotated figures:

FIG. 4B

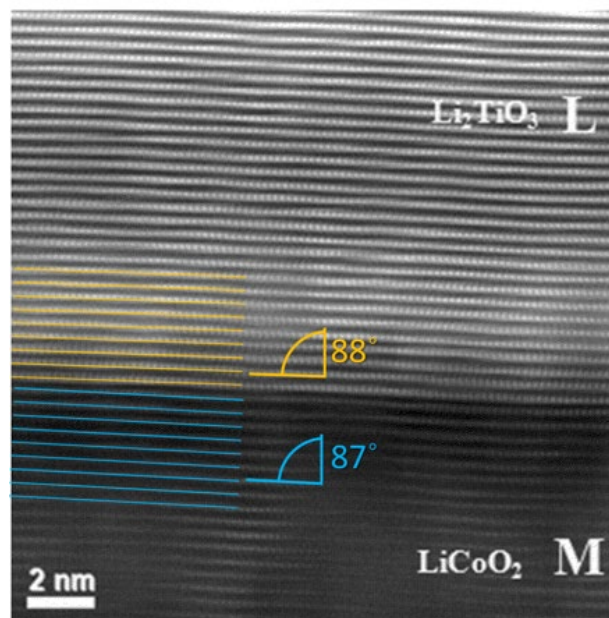
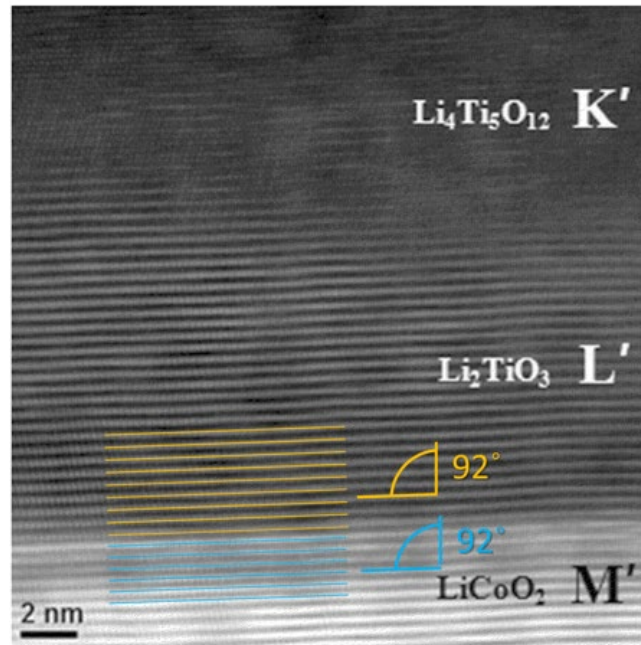


FIG. 5B



Ex. 1006, Figs. 4B, 5B. As shown above in annotated Figures 4B and 5B, the angles between the bright and dark lines in the coating regions and the LiCoO₂ regions roughly horizontal and measure 1° and 0° respectively, with an error margin of approximately +/-2° degrees or less. In taking these measurements, I confirmed that even the least aligned layers in the images shown above were well within an angle of 5° from each other. Because these angles are less than 5°, Shim's first region and second region are substantially aligned under the '660 Patent's definition.

156. In my opinion, a POSITA would have understood that Shim's teachings of alignment between crystal orientations of the first and second regions would apply equally to Blangero's first and second regions for the reasons described above in Section IX.B.1. In particular, both references use highly similar lithium intercalation

compounds—LiCoO₂ doped with magnesium and titanium—and substantially similar coatings to form a PEAM particle. For example, as shown in Figures 4B and 5B above, Shim observed several compounds and confirmed that the PEAM particle included LiCoO₂ and Li₂TiO₃ and that both compounds further included magnesium. Ex. 1006, ¶¶ [0134]–[0135]. Blangero similarly teaches a PEAM with an enhanced surface layer surface layer “consisting of a mixture of the elements of the core material” (*i.e.*, lithium), and “inorganic N oxide particles,” including oxides of “Mg, Ti.” Ex. 1005, ¶ [0009]; *see also id.*, ¶ [0094] (describing MgO and lithiated TiO₂ in the surface layer). Blangero further indicates that the titanium may be present in the coating layer as Li₂TiO₃. *Id.*, ¶ [0134]. In my opinion, a POSITA would have understood that Blangero and Shim’s similar core compounds (first region) and coating compounds (second region) indicate that Shim’s teachings of crystal orientation alignment apply to Blangero.

3. Dependent Claim 2

- a. [Element 2.0]: The lithium-ion secondary battery according to claim 1,

157. In my opinion, Blangero, alone or with Shim, renders claim 1 obvious.

See supra, § IX.B.2.

- b. [Element 2.1]: wherein the titanium, the magnesium, the fluorine, and the oxygen are provided in a range from the surface of the positive electrode active material particle to a depth of 5 nm.

158. Blangero discloses titanium, magnesium, fluorine, and oxygen provided in a range from the surface of the PEAM particle to a depth of 5 nm. As I explained above, Blangero discloses a surface layer containing titanium, magnesium, fluorine, and oxygen. *See supra*, §§ 136, IX.B.2.g. Blangero’s “surface layer is typically between 20 nm to 200 nm, and preferably between 20 nm and 100 nm thick.” Ex. 1005, ¶ [0066]; *see also id.*, cl. 36 (“wherein the thickness of the surface layer is less than 100 nm”). Thus, in my opinion a POSITA would have understood that the titanium, magnesium, fluorine, and oxygen present in Blangero’s surface layer would be found within the first 5 nm from the surface of the particle.

159. Further, the ’660 Patent explains that “[t]he measurement range of the XPS is from the surface of the particle of the positive electrode active material 100 to a region at a depth of approximately 5 nm.” Ex. 1001, 19:10–12. By using XPS, “the concentration of elements . . . present at a depth approximately 5 nm from the surface can be analyzed quantitatively.” *Id.* 19:14–18. Blangero uses XPS to evaluate “the surfaces of the particles of LCO-1 and Ex1.” Ex. 1005, ¶ [0129]. Table 3 shows the quantitative results of the elements on the surface of Blangero’s PEAM particle:

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., Table 3 (stitched). In my opinion, a POSITA would have understood that the detectable levels of Li, Co, O, F, Ti, and Mg at the surface of the particle indicate that in Blangero's Example 1 all of these elements are present within 5 nm of the PEAM's surface.

4. Dependent Claim 3

- a. [Element 3.0]: The lithium-ion secondary battery according to claim 1,

160. In my opinion, Blangero, alone or with Shim, renders claim 1 obvious.

See supra, § IX.B.2.

- b. [Element 3.1]: wherein the positive electrode further comprises a conductive additive, and

161. Blangero renders this limitation obvious. First, it was well known in the art that a conductive additive was regularly used in the positive electrode of

lithium-ion batteries. *See, e.g.*, Ex. 1005; Ex. 1010. A POSITA would have known to use a conductive additive with Blangero's PEAM to produce a positive electrode, as is standard practice in lithium-ion battery manufacturing. Moreover, a POSITA also would have recognized that Blangero's PEAM particles have "low electrical conductivity" on the surface which aids in "preventing undesirable side-reactions." Ex. 1005, ¶¶ [0062], [0065], [0092], [0094]. In my opinion, a POSITA would have known that conductive additives, which were necessary to build operational positive electrodes at the time (and continue to be so today), would certainly have been necessary in Blangero's PEAM given his discussion of the low electrical conductivity of the PEAM particle surface coatings. To this end, a POSITA would have known to use a conductive additive with Blangero's particle to enable sufficient electrical conductivity within the positive electrode to allow operation of the battery with high discharge rate capability.

162. In fact, Blangero discloses achieving high-rate capability. Table 8 shows the discharge capacity, C-rate performance and average voltage at 4.4 V for Blangero's LCO-4 and Example 4. Ex. 1005, Table 8, ¶ [0158]. The C-rate shows the discharge rate of a cell containing an electrode built with the PEAM, where 1C refers to the current that would result in a 1-hour discharge based on the nominal capacity of the battery, 20C refers to 20-times the 1C current (or the current that would result in a 3-minute discharge based on the nominal capacity of the battery,

etc). As shown in Table 8 below, both LCO-4 and Example 4 (one of Blangero's embodiments) have strong C-rate performances:

TABLE 8								
discharge capacity, C-rate properties, average voltage <V> at 15 C and specific energy Es for LCO-4 and Ex4 at 4.4 V.								
Sample	DQ1	1 C (%)	5 C (%)	10 C (%)	15 C (%)	20 C (%)	<V>	Es mWh/g
LCO-4	172.9	97.3	94.6	93.2	91.9	90.1	3.76	597
Ex4	173.1	99.0	96.8	95.2	94.0	92.8	3.81	620

Id. Table 8. For example, the particle of Example 4 was able to maintain 99% of its capacity at a 1C current. It was further able to discharge 92.8% of its capacity at a 20C current. In fact, the particle of Example 4 was found to be “highly suitable for high power applications where maintaining high specific energy at high C-rate is required.” *Id.*, ¶ [0158]. To achieve such high discharge rates and maintain a high average voltage, a POSITA would have understood that the positive electrode used by Blangero comprising the PEAM ultimately possessed good electrical conductivity despite the described high-resistance of the PEAM itself.

163. In the alternative, it is my opinion that Blangero in combination with Shim renders this limitation obvious. Like Blangero, Shim discloses achieving “excellent rate capability.” Ex. 1006, ¶ [0059]. While Blangero does not describe the specific components in its positive electrode, Shim, which describes considerably similar PEAMs, discloses “the positive active material layer further includes . . . *a*

conductive material.” Ex. 1006, ¶ [0081]. “Any suitable electrically conductive material may be used as the conductive material” which “is included to provide electrode conductivity” including “a carbon-based material such as *natural graphite, artificial graphite . . . a carbon fiber, and/or the like.*” *Id.*, ¶ [0083] (emphasis added). In my opinion, a POSITA would have been motivated to combine Shim’s conductive material with Blangero’s PEAM for all the reasons explained above in Section IX.B.1.

- c. [Element 3.2]: wherein the conductive additive comprises carbon fiber, graphene or multilayer graphene.

164. In my opinion, Blangero, alone or with Shim, renders this limitation obvious. To offset the low conductivity the PEAMs disclosed in Blangero, it would have been obvious for a POSITA to experiment with different conductive additives to create a positive electrode with good conductivity. In addition to carbon black, other forms of carbon materials, such as carbon fiber, graphene, and multilayer graphene, were well known candidates for conductive additives. A POSITA would have known to use a conductive additive with Blangero’s particle which was a well-established practice in battery manufacturing at the time.

165. In the alternative, Blangero and Shim render this limitation obvious. Shim states that “[a]ny suitable electrically conductive material may be used as the conductive material” like “a carbon-based material such as *natural graphite,*

artificial graphite . . . a carbon fiber, and/or the like.” Ex. 1006 ¶ [0083] (emphasis added). Also, using “graphene” or “multilayer graphene” would have been obvious to a POSITA from Shim, since natural and artificial graphite are themselves composed of many layers of graphene. Moreover, such materials were well known in the art and were widely used prior to the ’660 Patent. For example, during prosecution of the ’660 Patent, the examiner found that a 2015 patent application published in 2016 (U.S. 2016/0104880) (“Gao”) disclosed the use of the claimed conductive additive. Ex. 1002 (Part 1), 285. In my opinion, a POSITA would have been very familiar with the use of conductive additives including the use of graphene and multilayer graphene, or graphite. A POSITA would have been motivated to use these additives for the reasons described above in Sections IX.B.1 and 157.

5. Dependent Claim 4

- a. [Element 4.0]: The lithium-ion secondary battery according to claim 1,

166. In my opinion, Blangero, alone or in combination with Shim, renders claim 1 obvious. *See supra*, § IX.B.2.

- b. [Element 4.1]: wherein a relative value of a concentration of the titanium is greater than or equal to 0.05 and less than or equal to 0.4 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of cobalt is defined as 1.

167. In my opinion, Blangero discloses this limitation. Blangero discloses a “lithium metal oxide powder for use as a cathode material in a rechargeable battery”

consisting of doped LiCoO_2 , the core of the PEAM, and a “surface layer” consisting of elements of the core material and “one or more metals” including magnesium and titanium. Ex. 1005, ¶¶ [0009], [0072], [0091], [0093], [0094], [0108]. In Example 1, Blangero describes a LiCoO_2 -based cathode material that has “an electron insulating surface comprising the elements of the core material (Li, Co) and inorganic metal oxides comprising Mg and Ti.” *Id.*, ¶ [0108]. Table 3, reproduced and annotated below, compares Example 1 against LCO-1, which is a “Ti and Mg-doped lithium cobalt oxide” particle and is the “parent” of Example 1. *Id.*, ¶¶ [0087], [0108]–[0110]. Example 1 is prepared with “95 mol % of LCO-1 and 5 mol % of 0.25 mol % Ti and 0.5 mol % Mg doped $\text{Co}(\text{OH})_2$.” ¶ [0110]. Table 3 shows “[t]he chemical composition of the surfaces of the particles of LCO-1 and Ex 1” as determined by XPS analysis:

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], Table 3 (annotations added); *see also id.* Fig. 4 (showing XPS spectra for Example 1). Table 3 shows that the surface of the particle of examples 1 has a Ti:Co ratio of 1.1/12.0. This equals a ratio of 0.0917 and falls within the claimed range of 0.05 to 0.4, disclosing this element.

168. I note that other references confirm that this ratio was known in the art. For example, Yu similarly shows a Ti:Co ratio of 0.0849 at the surface of a doped LiCoO₂ particle. Ex. 1011, 3, Table 3. Yu teaches a decreasing concentration gradient of titanium through the PEAM particle. After 4000 seconds of sputtering, Yu notes that “[t]he intensity for Ti becomes lower gradually” and eventually “hardly visible.” *Id.* By contrast, “the intensity for Co becomes obviously higher with the increasing sputtering time.” *Id.* Yu discloses measured Ti:Co concentrations of a PEAM at different sputtering times in Table 3:

Table 3 The Ti/Co atomic ratio of LiCo _{0.998} Ti _{0.002} O ₂ powders with different sputtering times.					
Sputtering time (s)	0	500	1000	2500	4000
Ti/Co (%)	8.49	3.02	2.17	1.25	0.75

Ex. 1011, 3, Table 3 (annotated). The spectral data underlying the results in Table 3 are presented in Figure 2. *Id.*, 2, Fig. 2.

169. As I annotated in the red box above, at 0 seconds of sputtering time—the surface of the sample—the ratio of titanium to cobalt is 8.49%, or 0.0849. Ex. 1011, 3, Table 3. At this surface point, when the concentration of cobalt (the first

transition metal) is defined as 1, the concentration of titanium (the second transition metal) is 0.0849, and therefore falls in the range of 0.05 to 0.4, as required by the claim.

170. A POSITA would have also known that the Ti:Co concentration is a result-effective variable and that the performance of a battery would be influenced by the titanium-cobalt ratio because titanium is electrochemically inert in the cathode. A POSITA further would have known that the amount of titanium included in a PEAM particle is easily adjusted during cathode synthesis. Moreover, a POSITA would have understood that despite enhancing battery stability and performance, titanium is not electrochemically active and therefore a POSITA could have discovered an optimal Ti:Co concentration ratio with routine experimentation. Accordingly, a POSITA would have understood it may be beneficial to increase the concentration of surface titanium, would have been motivated to experiment with it to improve cathode performance, and could have easily done so.

171. Therefore, in my opinion Blangero discloses this limitation.

6. Dependent Claim 5

- a. [Element 5.0]: The lithium-ion secondary battery according to claim 1,

172. In my opinion, Blangero, alone or in combination with Shim, renders claim 1 obvious. *See supra*, § IX.B.2.

- b. [Element 5.1]: wherein a relative value of a concentration of the magnesium is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of cobalt is defined as 1.

173. In my opinion, Blangero and Shim render this element obvious. As explained above in reference to Claim 4 (in Section IX.B.5) Blangero describes a LiCoO₂-based cathode material that has “an electron insulating surface comprising the elements of the core material (Li, Co) and inorganic metal oxides comprising Mg and Ti” in Example 1. Ex. 1005, ¶¶ [0009], [0072], [0091], [0093], [0094], [0108]. Table 3 shows “[t]he chemical composition of the surfaces of the particles of LCO-1 and Ex 1” as determined by XPS analysis:

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., ¶ [0108], Table 3; *see also id.*, Fig. 4(b). Table 3 shows that the surface of the particle of Example 1 has a Mg:Co ratio of 4.4/12.0. This equals a ratio of 0.367,

which approximates 0.4 in the context of Claim 5 where the range is defined with values expressed as one decimal place. In my opinion, Blangero also discloses a PEAM particle with a higher concentration of magnesium at the surface of the particle than disclosed by Example 1. Example 2 is comprised of “94 mol % of LCO-1 and 6 mol % of 0.25 mol % Ti and 0.5 % mol % Mg doped $\text{Co}(\text{OH})_2$.” *Id.*, ¶ [0111]. Example 1, by comparison, is comprised of “95 mol % of LCO-1 and 5 mol % of 0.25 mol % Ti and 0.5 % mol % Mg doped $\text{Co}(\text{OH})_2$.” *Id.* By increasing from 5 mol % to 6 mol %, the amount of magnesium used in manufacturing Example 2 increases by 20% compared to the amount used in Example 1, while the amount of cobalt decreases by approximately 1% (from 96 mol % to 95 mol %). Accordingly, a POSITA would have understood that the relative concentration of magnesium to cobalt derived from Table 3 is not a floor and instead some of Blangero’s samples would have higher ratios. If, for example, this increase in magnesium were to cause a corresponding 20% increase in surface magnesium and a 1% decrease in surface cobalt, the ratio of Mg:Co would increase from 0.367 to 0.444. In my opinion, this further renders the claimed range obvious.

174. In my opinion, a POSITA would have found the Mg:Co ratio within the range of 0.4 to 1.5 obvious based on the above ratio taught by Blangero. A POSITA would have known that magnesium can improve the performance of LiCoO_2 -based batteries, including “excellent rate capability and cycle-life characteristics

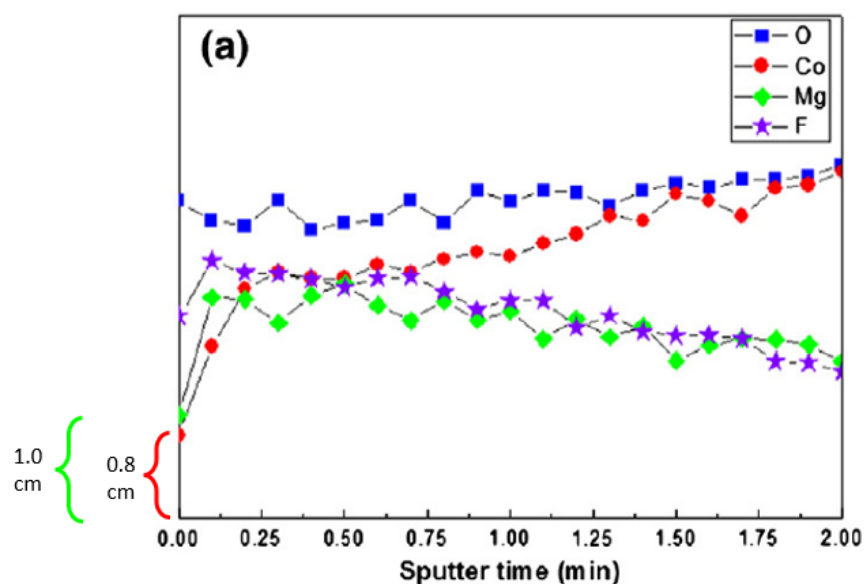
(Ex. 1006, ¶ [0059]), and protection of the cathode active material from unwanted interactions with the electrolyte. Ex. 1005, ¶¶ [0093]–[0094] (using Mg or MgO in a coating layer “provides additional safety” to the cathode material from the battery’s electrolyte, preventing “deterioration of the electrochemical performance” and “loss of capacity”).

175. A POSITA would have also known that the Mg:Co concentration is a result-effective variable and that the performance of a battery would be influenced by the magnesium-cobalt ratio because magnesium is not electrochemically active in the cathode. A POSITA further would have known that the amount of magnesium included in a PEAM particle is easily adjusted during cathode synthesis. Accordingly, a POSITA would have understood it may be beneficial to increase the concentration of surface magnesium, would have been motivated to experiment with it to improve cathode performance, and could have easily done so.

176. I further note that a POSITA would have been aware of other studies showing Mg:Co concentration ratios at the surface of particles within the claimed range. For example, Lee teaches using an MgF₂ coating with a LiCoO₂ PEAM. Ex. 1008. Lee describes an XPS-based depth analysis in which a sample is sputtered, or etched (e.g., by argon ions) while XPS measurements are conducted intermittently. As the sample is etched, subsurface regions of the sample are revealed, enabling the composition to be measured at various depths. That is, a longer etching time

correlates with a measurement deeper into the sample, while an etching time of 0 corresponds to a measurement at the surface of the sample.

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



Ex. 1008, Fig. 5(a) (y-axis annotations added). A sputtering time of 0 minutes, the Co concentration measured 0.8 centimeters on the Y-axis, while the Mg concentration measured 1.0 cm on the Y-axis. Assuming that the Y-axis begins at 0,

this would reflect a relative concentration of magnesium to cobalt (Mg:Co) of 1.25 at the surface of the particle.²

C. Ground I.B: Claims 6–10 Are Rendered Obvious by Blangero Alone or in Combination with Shim and/or Yamamoto

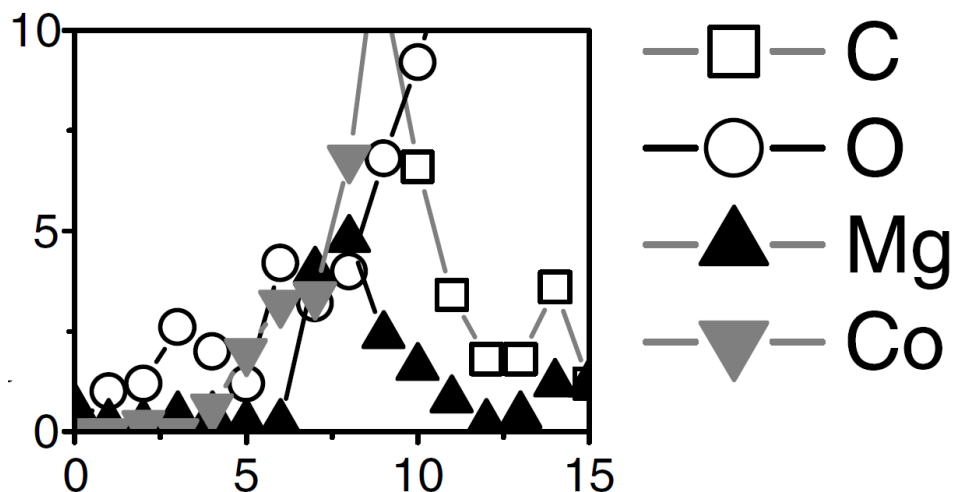
1. Motivation to Combine Blangero with Yamamoto

178. In my opinion, a POSITA would have been motivated to combine Blangero's particle with Yamamoto's MgO coating, thus rendering the limitation obvious. Both Blangero and Yamamoto describe using MgO in a coating layer in a PEAM to stabilize the layered structure of LiCoO₂. Ex. 1006 ¶ [0093]–[0094] (using oxidized compounds like MgO to stabilize the PEAM and protect the surface from reactions with the electrolyte); Ex. 1010, 5 (“In the case of the MgO-coated sample heat-treated at 810 °C, the Mg ions in the Li sites have a role of ‘pillars’ to preserve

² A POSITA would have also understood that if the Y-axis began at a value higher than 0, the Mg:Co concentration ratio would decrease or converge to a minimum ratio of 1. For example, if the Y-axis of Fig. 5(a) began at a value corresponding to 1.0 centimeters, the Mg:Co concentration ratio would be $(1.0+1.0)/(1.0+0.8) = (2.0/1.8) = 1.11$. The higher the value that the Y-axis begins at, the closer the Mg:Co concentration ratio would be to 1.0.

the layered structure, particularly, in the charged (delithiated) state, which is a main reason it has enhanced cycling ability.”).

179. Yamamoto discloses observing a PEAM particle using an EDX line scan. Ex. 1007, 2, Fig. 1(b). A POSITA would have understood from Yamamoto that a magnesium coating on the surface of a PEAM will exhibit a peak concentration near the surface that decreases radially inward toward the center of the PEAM. In the examined PEAM, there was “a Mg containing layer of approximately 5 nm in thickness.” *Id.* The line scan, reproduced below, is consistent with this description: there is no Mg detected in the first ~7 nm measured followed by a section from approximately 7 to 13 nm that contains magnesium.

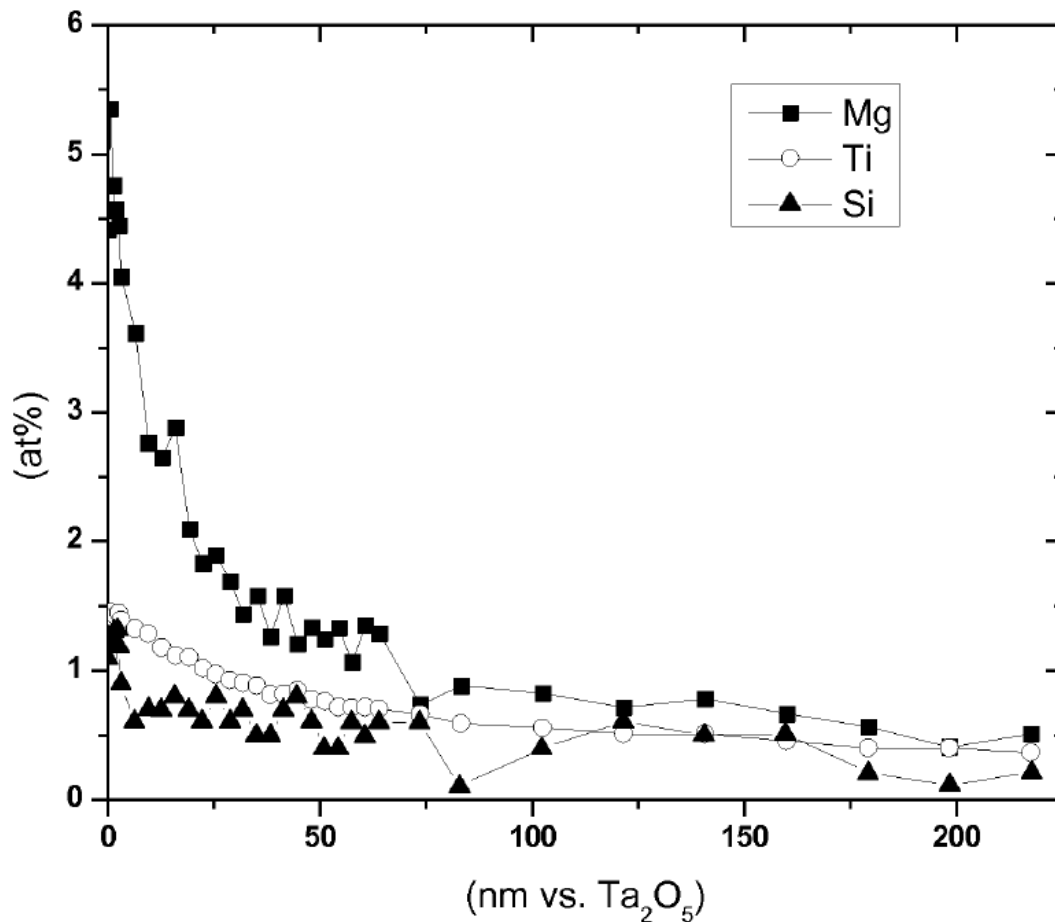


Id., Fig. 1 (excerpted).

180. A POSITA would have understood that using the EDX line scan like that used in Yamamoto shows both the location of an element within a PEAM

particle and further shows the concentration of that element, reflected by the height of the line on the graph. Shown as black upwards triangles in the graph above, magnesium concentration (which is essentially 0 for the first 6 nm) approximately peaks at a depth of 8 nm and at a value of 5 net counts, and then decreases as the line analysis continues. *Id.* The magnesium concentration reaches 1/5 of its peak (*i.e.*, 1 net count) at an approximate depth of 12 nm. *Id.*

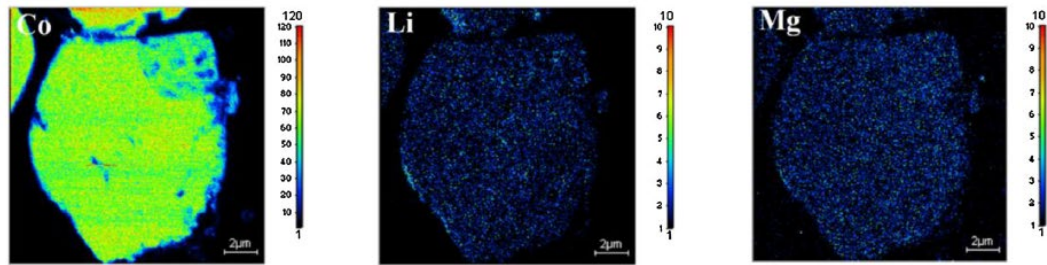
181. Applying Yamamoto's findings and well-known measurement technique to Blangero would have been a routine matter. A POSITA would have further expected to observe that the concentration of magnesium would decrease from the surface to the center of the PEAM as this was a well-documented and observed phenomenon. For example, Blangero similarly shows a PEAM with a maximum concentration of magnesium, shown as a square in the graph below, on the particle's surface with the magnesium concentration decreasing radially inward in the particle:



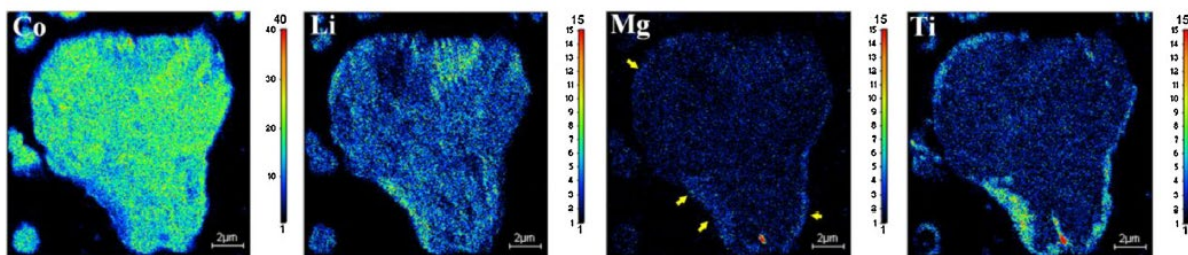
Ex. 1005, Fig. 8, ¶ [0135]. Blangero confirms that “[t]he surface layer” is “preferably between 20 nm and 100 nm thick” (*id.*, ¶ [0066]), which is the approximate depth range where magnesium concentration reaches ~1 at%. Blangero further discloses that “the concentration of M’ and M’’ at the surface of the lithium metal oxide powder is at least 5 times the concentration of M’ and M’’ in the bulk of the powder” where M’ includes magnesium. *Id.*, ¶ [0011]. This corroborates Yamamoto’s teaching that magnesium concentration decreases to 1/5 of its peak value throughout the second region.

182. It was known in the art that the concentration of magnesium in a PEAM particle, including in Mg-doped PEAM particles, would exhibit this concentration profile. Ex 1032 at 1. Blangero confirms “[t]he surface is enriched of elements N such as Mg [and] Ti . . . and, in one embodiment, is formed by segregation from the core and accumulation of these metal dopants at the surface of the particles. In the core, these dopants are substantially absent.” Ex. 1005, ¶ [0062]; *see also id.*, ¶ [0090] (“When dopants N are present, the surface is formed during the second sintering by spontaneous segregation of the dopants from the core.”). Other researchers specifically observed this segregation process in Mg-doped LiCoO₂ coated with lithium titanate. Ex. 1032, 3. The images below depict PEAMs doped with Mg²⁺ ions and then coated with lithium titanate. As a result of this coating, the Mg from the bulk of the PEAM migrates from the bulk to the coating layer. Figure 2, reproduced below, shows “the Mg²⁺ ions are homogenously distributed over the entire particle of the uncoated-LCO_{Mg} whereas the Mg map of the LT2-LCO_{Mg} particle exhibits slightly larger population at the edges of particle (yellow arrows) than in the bulk (inner area), which is in accord with the Ti map even though its intensity is much smaller.” *Id.* at 3.

(a) uncoated-LCO_{Mg}



(b) LT2-LCO_{Mg}



Id., Fig. 2.

183. In my opinion, a POSITA would have understood that even the Mg-doped PEAMs disclosed in Blangero would have a peak of magnesium at the surface that decreases inwardly until a point of 1/5 maximum peak when detected by a line analysis of EDX. Accordingly, a POSITA would have understood that Blangero's Mg-doped PEAMs would exhibit the claimed concentration 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.

184. Further, both Blangero and Yamamoto teach LiCoO₂ PEAMS with surface MgO exhibit improved durability and cycling characteristics of the battery. See Ex. 1005, ¶¶ [0032], [0062], [0092]; Ex. 1007, 1 ("MgO coating improves the

durability of the cathode during high-potential cycling”). As I explain above with respect to Blangero, the inclusion of an enriched “inert coating layer” that includes oxidized compounds like MgO “provides additional safety when the powders are in contact with the electrolyte of the battery.” Ex. 1005, ¶¶ [0094].

185. Finally, a POSITA would have recognized that Blangero and Yamamoto are both directed to the same goal for substantially similar PEAMs: improving performance of LiCoO₂-based batteries. Both Blangero and Yamamoto teach coatings containing magnesium and oxygen to attain this specific benefit. *See* Ex. 1005, ¶¶ [0032], [0062], [0071]–[0072], [0092], [0108]–[0121] (describing treating enumerated Examples and precursor LCO compounds alike with magnesium—including with reference to “state of the art titanium and magnesium doped LiCoO₂”); Ex. 1007, 1 (“The MgO coating improves the durability of the cathode during high-potential cycling. . . . [T]he MgO coating suppresses drastic distortions of local structure at the LiCoO₂ surface.”). Therefore, it is my opinion that a POSITA would have been motivated to combine Blangero with Yamamoto.

2. Independent Claim 6

a. [Preamble]: A lithium-ion secondary battery comprising

186. In my opinion, Blangero discloses the preamble. *See supra*, § IX.B.2.a.

b. [Element 6.1]: a positive electrode, the positive electrode comprising a positive electrode active material particle comprising lithium cobaltate,

187. In my opinion, Blangero discloses this limitation. *See supra*,
§ IX.B.2.b.

- c. [Element 6.2]: wherein the positive electrode active material particle comprises a first region and a second region,

188. In my opinion, Blangero discloses this limitation. *See supra*,
§IX.B.2.c.

- d. [Element 6.3]: wherein the first region comprises a layered rock-salt crystal structure,

189. In my opinion, Blangero discloses this limitation. *See supra*,
§ IX.B.2.d.

- e. [Element 6.4]: wherein the second region comprises a rock-salt crystal structure,

190. In my opinion, Blangero discloses this limitation. *See supra*,
§ IX.B.2.e.

- f. [Element 6.5]: wherein the positive electrode active material particle comprises titanium, magnesium, fluorine, and oxygen in the second region,

191. In my opinion, Blangero discloses this limitation. *See supra*,
§ IX.B.2.g.

- g. [Element 6.6]: 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

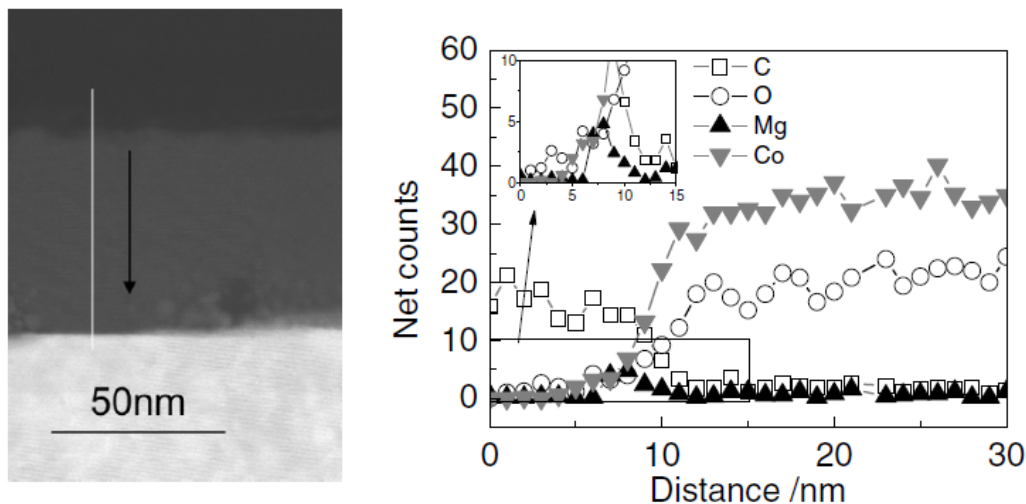
direction is $\frac{1}{5}$ of a maximum peak of the magnesium in the line analysis, and

192. In my opinion, Blangero renders this limitation obvious either alone, in combination with a POSITA's knowledge, or in combination with Yamamoto. The surface of Blangero's particle, which expressly contains magnesium (*see supra*, § IX.B.2.g; Ex. 1006, ¶¶ [0059], [0062], [0094]), is one end of the second region. *See supra*, § IX.B.2.f (citing Ex. 1006, ¶ [0009]; *see also id.*, ¶¶ [0010], [0011], [0027], [0059], [0062], [0066], [0094]. A POSITA would have found it obvious detect the magnesium in the surface layer of Blangero's PEAM using a line analysis of energy dispersive X-ray spectroscopy (EDX) which was a well known measurement technique. *See supra* § VI.A.4.c. Further, a POSITA would have found it obvious based on their knowledge as informed by Yamamoto that the second region extends from the surface of the PEAM to a point where the amount of magnesium (as measured by EDX line analysis) is $\frac{1}{5}$ of the maximum peak of magnesium in the line analysis. Similarly, Blangero confirms that "[t]he surface layer" is "preferably between 20 nm and 100 nm thick" (*id.*, ¶ [0066]), which is the approximate depth range where magnesium concentration reaches ~1 at%. Blangero further discloses that "the concentration of M' and M'' at the surface of the lithium metal oxide powder is at least 5 times the concentration of M' and M'' in the bulk of the powder" where M' includes magnesium. *Id.*, ¶ [0011]. Accordingly, the concentration of magnesium

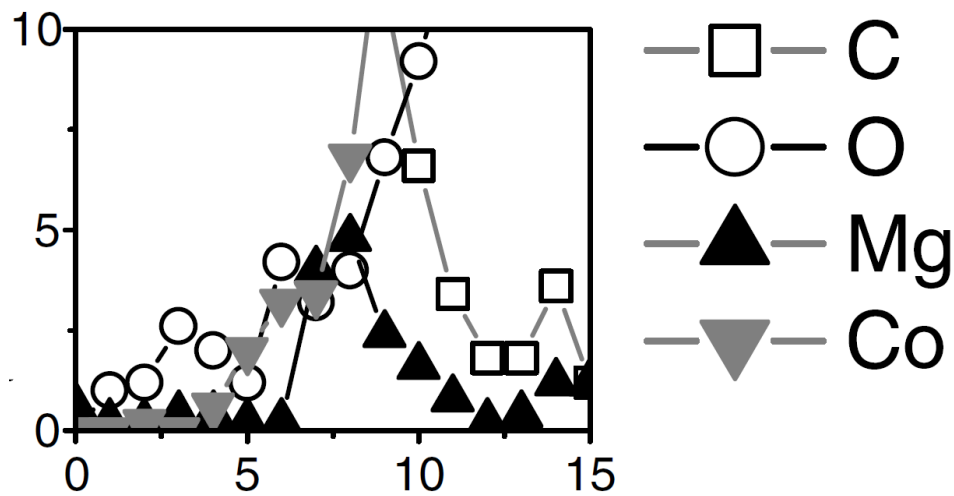
at the surface of Blangero's PEAM particle is approximately five times the concentration in the bulk.

193. Yamamoto, confirms that a POSITA could observe this phenomena if they examined Mg-coated PEAMs. Yamamoto studied MgO coatings on LiCoO₂ particles, and characterized the magnesium content in the coating region through EDX line scans, as shown below:

(b)



Ex. 1007, Fig. 1(b). As shown above (left image), Yamamoto's line scan started from outside the particle (in the dark gray area above the particle) and proceeded downwards into the light gray area. Yamamoto's line analysis is consistent with this, as there is no Mg detected in the first ~7 nm measured. For clarity, below I reproduce Yamamoto's inset line scan concerning the first 15 nm of the line scan on the x-axis (and limited to concentrations no higher than 10 net counts on the y-axis):

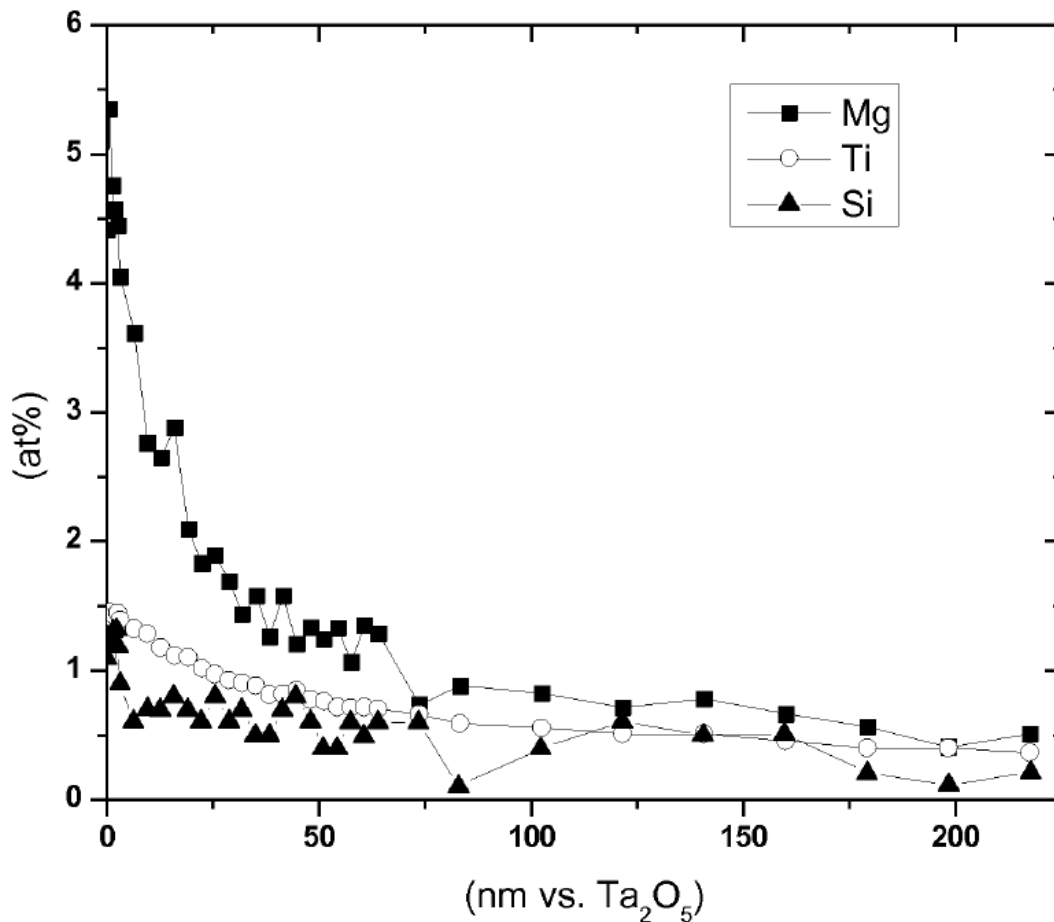


Id. (excerpted). As shown above by the black upwards triangles, magnesium concentration (which is essentially 0 for the first 7 nm) approximately peaks at a depth of 8 nm and at a value of 5 net counts, and then decreases as the line analysis continues. *Id.* The magnesium concentration reaches 1/5 of its peak (*i.e.*, 1 net count) at an approximate depth of 12 nm. *Id.*

194. Thus, a POSITA would have understood from Yamamoto that its second region (*i.e.*, containing the MgO coating) starts from the surface of the material (at a depth of ~7 nm) and extends to a depth of ~12 nm, where the magnesium concentration reaches 1/5 of the value at its peak. In fact, Yamamoto states that “[t]he EDX scan detected a Mg containing layer of approximately 5 nm in thickness outside [the] Co containing layer.” *Id.*, 2. From Yamamoto’s teachings, a POSITA would have understood that a PEAM with an MgO coating formed

according to well-known techniques like those used in Yamamoto would have its magnesium concentration reach 1/5 of its peak at the bottom of the layer.

195. Blangero corroborates Yamamoto's EDX line scan analysis and similarly shows a PEAM with a maximum concentration of magnesium on the particle's surface with the magnesium concentration decreasing radially inward in the particle. Ex. 1005, Fig. 8, ¶ [0135]. Blangero describes a PEAM with a "surface layer," or a second region, which is "preferably between 20 nm and 100 nm thick." *Id.*, ¶ [0066]. As shown below, the magnesium concentration at the surface of Blangero's particle (0 nm depth) is ~5.4 atomic %, which is the maximum concentration found in the particle. *Id.* The concentration of magnesium decreases to ~1 atomic % "over the first 50 nm of the particle[]":



Ex. 1005, Fig. 8, ¶ [0135]. This corroborates Yamamoto’s teaching that magnesium concentration decreases to 1/5 of its peak value throughout the second region.

196. Thus, in my opinion, a POSITA (who would have had knowledge of Yamamoto) would have understood that Blangero renders obvious the claimed second region and claimed magnesium concentration profile. As shown in the EDX line scan above, a POSITA would have understood a fundamental concept from Yamamoto: that “a Mg containing layer of approximately 5 nm in thickness” (*id.*) such as MgO experiences a concentration peak within that 5 nm range, and

eventually decreases to 0 farther away from the surface, and into the bulk of the particle. Accordingly, at some given depth, the concentration of magnesium must reach 1/5 of its maximum concentration—indeed, at some given depth, the concentration of magnesium will be 75% of its peak, 50% of its peak, 25% of its peak, etc.

197. Finally, a POSITA would have also known that the Mg:Co concentration is a result-effective variable and that the performance of a battery would be influenced by the magnesium-cobalt ratio because magnesium is not electrochemically active in the cathode. A POSITA further would have known that the amount of magnesium included in a PEAM particle is easily adjusted during cathode synthesis. Accordingly, a POSITA would have understood it may be beneficial to increase the concentration of surface magnesium, would have been motivated to experiment with it to improve cathode performance, and could have easily done so. To the extent Blangero does not explicitly disclose the claimed ratio, it would be obvious for a POSITA to obtain this through routine experimentation.

198. Accordingly, in my opinion this limitation is obvious over Blangero in view of a POSITA's knowledge of well-known measurement techniques like EDX line analysis as well as a POSITA's knowledge that the concentration of a coating element would decrease radially inwardly in the PEAM. In the alternative, a

POSITA would have been motivated to combine the teachings of Blangero and Yamamoto as described above in Section IX.C.1

- h. [Element 6.7]: 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.

199. In my opinion, Blangero in combination with Shim renders this limitation obvious. Like claim 1, the “second region” of claim 6 refers to a coating comprising titanium, magnesium, fluorine, and oxygen. *See supra* § IX.C.2.f (Element 6.5); Ex. 1001, cl. 6, 11:5–12:16 (describing titanium, magnesium, fluorine, and oxygen in a second region over the first region), 12:17–13:57. Thus Blangero and Shim render this limitation obvious for the reasons described above in Section IX.B.2.h.

3. Dependent Claim 7

- a. [Element 7.0]: The lithium-ion secondary battery according to claim 6,

200. In my opinion, Blangero, either alone or in combination with Shim and/or Yamamoto, renders claim 6 obvious. *See supra*, § IX.C.2.

- b. [Element 7.1]: wherein the titanium, the magnesium, the fluorine, and the oxygen are provided in a range from the surface of the positive electrode active material particle to a depth of 5 nm.

201. In my opinion, Blangero discloses this limitation. *See supra*, § IX.B.3.b.

4. Dependent Claim 8

- a. [Element 8.0]: The lithium-ion secondary battery according to claim 6,

202. In my opinion, Blangero, either alone or in combination with Shim and/or Yamamoto, renders claim 6 obvious. *See supra*, § IX.C.2.

- b. [Element 8.1]: wherein the positive electrode further comprises a conductive additive, and

203. In my opinion, Blangero and Shim render this limitation obvious. *See supra*, § IX.B.4.b.

- c. [Element 8.2]: wherein the conductive additive comprises carbon fiber, graphene or multilayer graphene.

204. In my opinion, Blangero and Shim render this limitation obvious. *See supra*, § IX.B.4.c.

5. Dependent Claim 9

- a. [Element 9.0]: The lithium-ion secondary battery according to claim 6,

205. In my opinion, Blangero, either alone or in combination with Shim and/or Yamamoto, renders claim 6 obvious. *See supra*, § IX.C.2.

- b. [Element 9.1]: wherein a relative value of a concentration of the titanium is greater than or equal to 0.05 and less than or equal to 0.4 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of cobalt is defined as 1.

206. In my opinion, Blangero discloses this limitation. *See supra*, § IX.B.5.b.

6. Dependent Claim 10

- a. [Element 10.0]: The lithium-ion secondary battery according to claim 6,

207. In my opinion, Blangero, either alone or in combination with Shim and/or Yamamoto, renders claim 6 obvious. *See supra*, § IX.C.2.

- b. [Element 10.1]: wherein a relative value of a concentration of the magnesium is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of cobalt is defined as 1.

208. In my opinion, Blangero renders this limitation obvious. *See supra*, § IX.B.6.b.

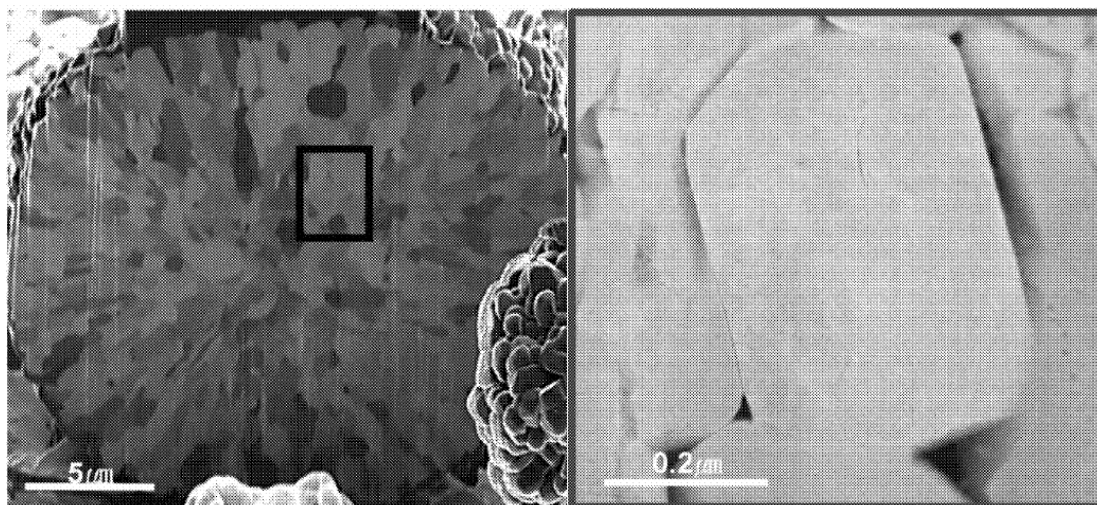
D. Ground II.A: Claims 1–3 and 6–8 Are Rendered Obvious by Cho Alone or in Combination with Kim

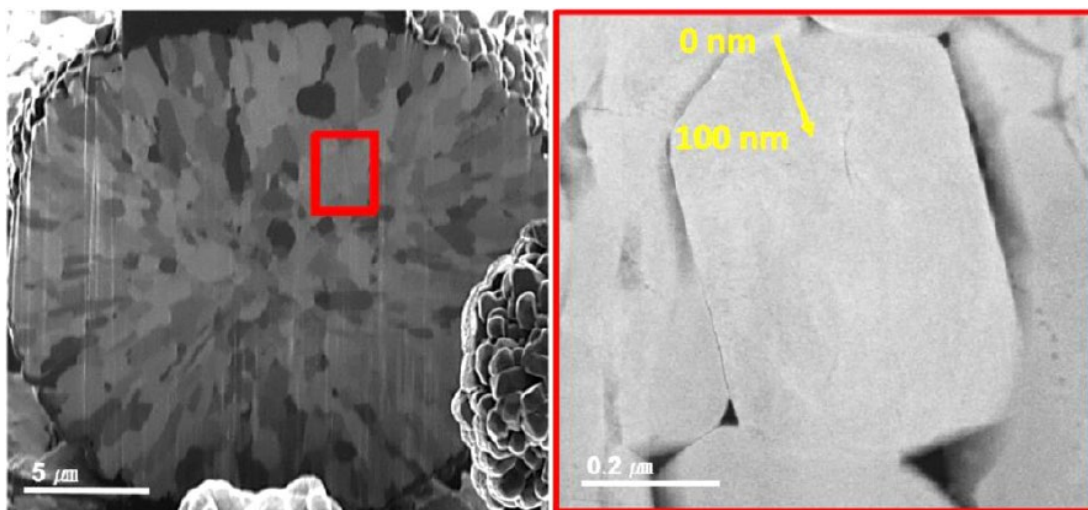
1. Motivation to Combine Cho with Kim

209. In my opinion, a POSITA would have been motivated to combine Cho and Kim for several reasons. Cho and Kim are two publications stemming from the work of the same research group—in fact, they contain many of the same images. Accordingly, they both describe the same goal of improving lithium-ion battery cyclability using a cathode active material coating. Thus, a POSITA would have read Cho and Kim together and would have understood their teachings to be complementary for several reasons.

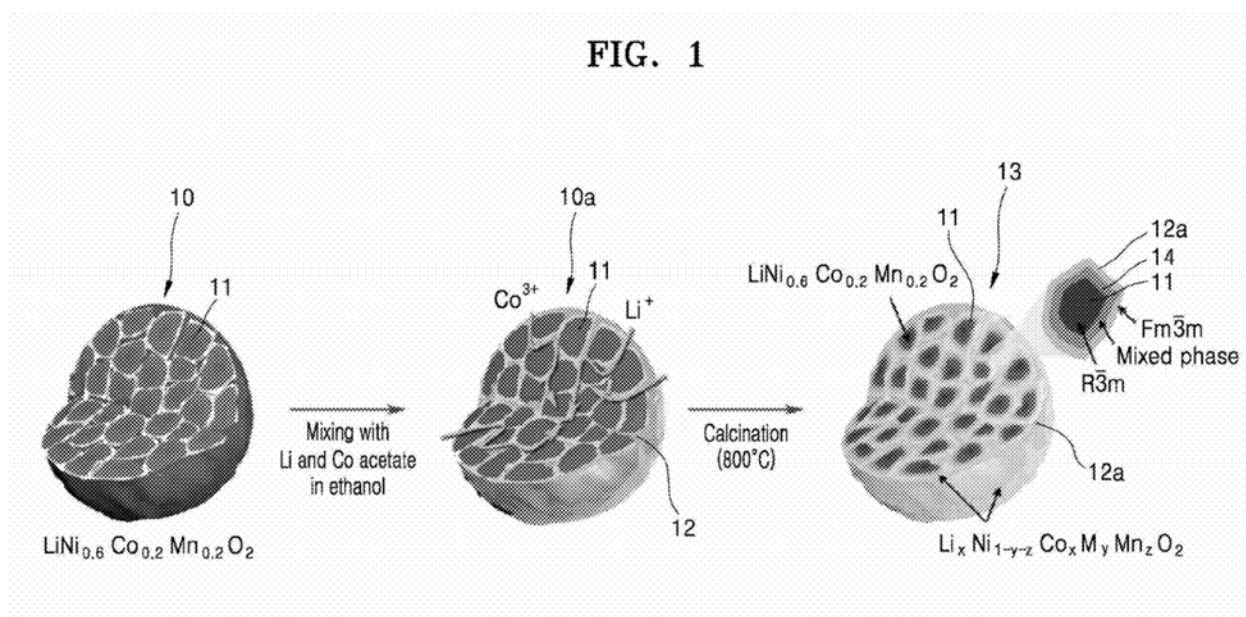
210. Cho and Kim are related references meant to be understood together. For instance, Kim's authors and Cho's inventors substantially overlap, including Hyejung Kim and Jaephil Cho. *See* Ex. 1008; Ex. 1009. Both references additionally refer to the Ulsan National Institute of Science and Technology. Moreover, Kim was published on February 10, 2015, which is around the filing date of Cho's application, December 17, 2015. *Id.* Because these two references were borne from the same research group at similar times and cover nearly identical subject matter (including several figures in common, as described below), a POSITA would have found them compatible and would have read and understood them together.

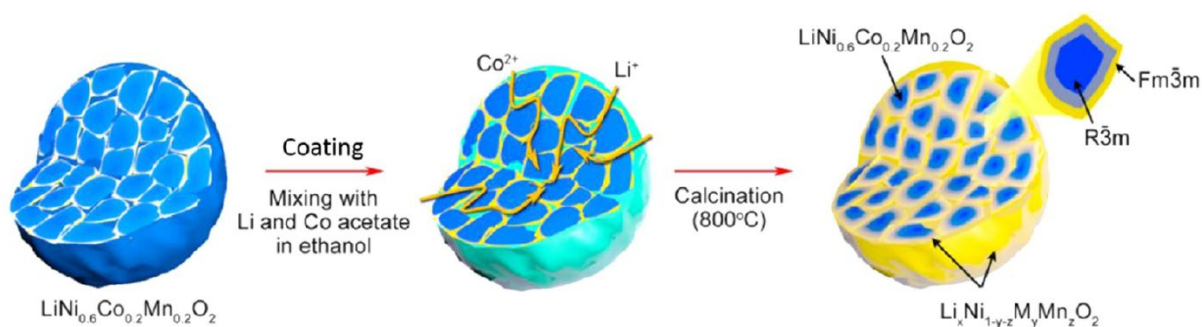
211. Additionally, both references discuss the same technical features, including lithium-ion batteries, coatings, crystalline structures, PEAMs, and battery performance, as indicated in the common images below:



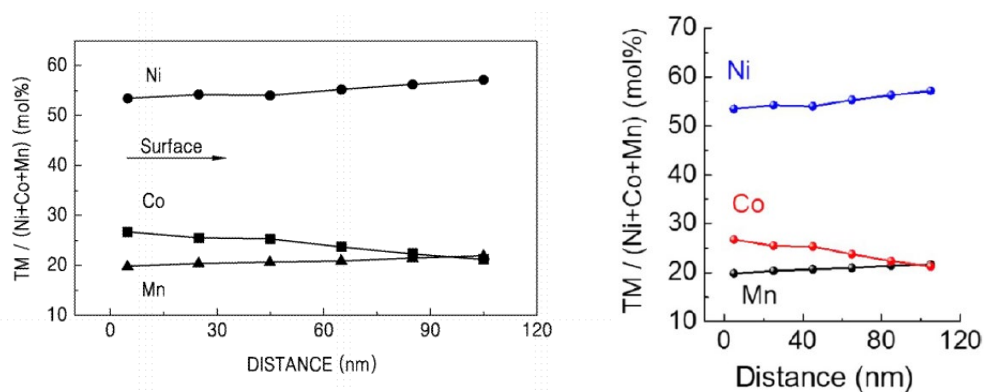


Ex. 1008, Figs. 4A–B (top); Ex. 1009, Fig. 1(b)–(c) (bottom) (all annotations in original).

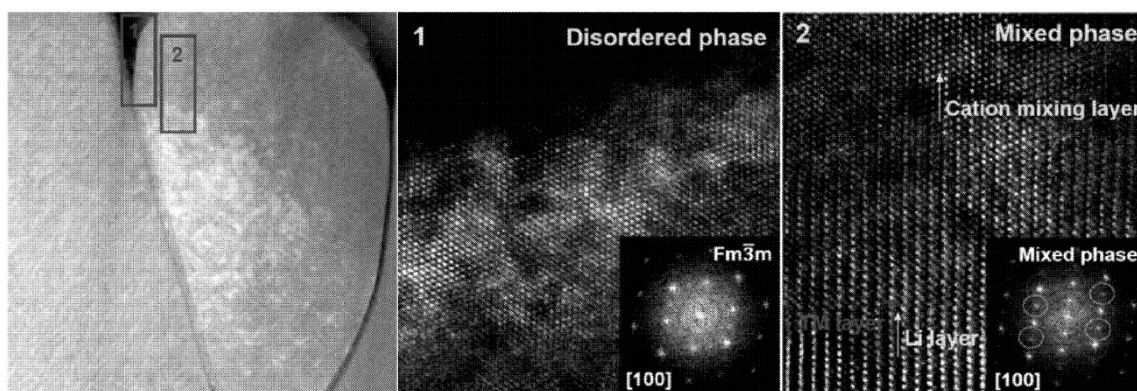


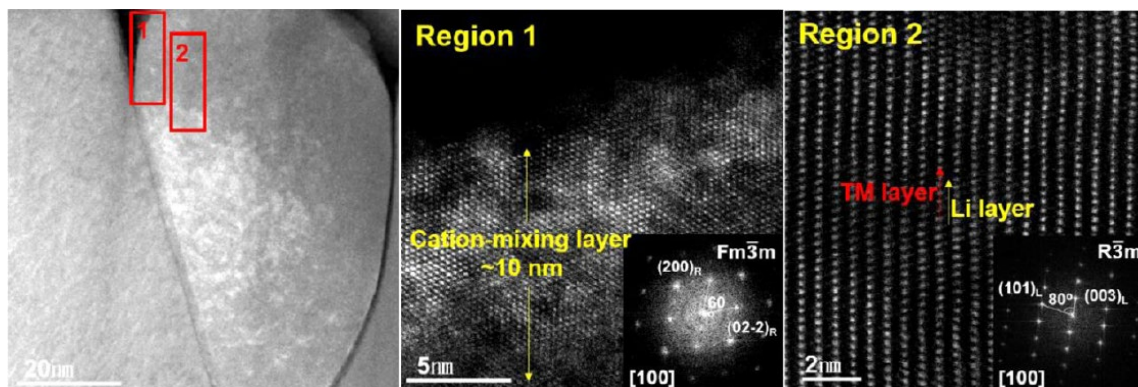


Ex. 1008, Fig. 1 (top); Ex. 1009, Fig. 1(a) (bottom) (all annotations in original).

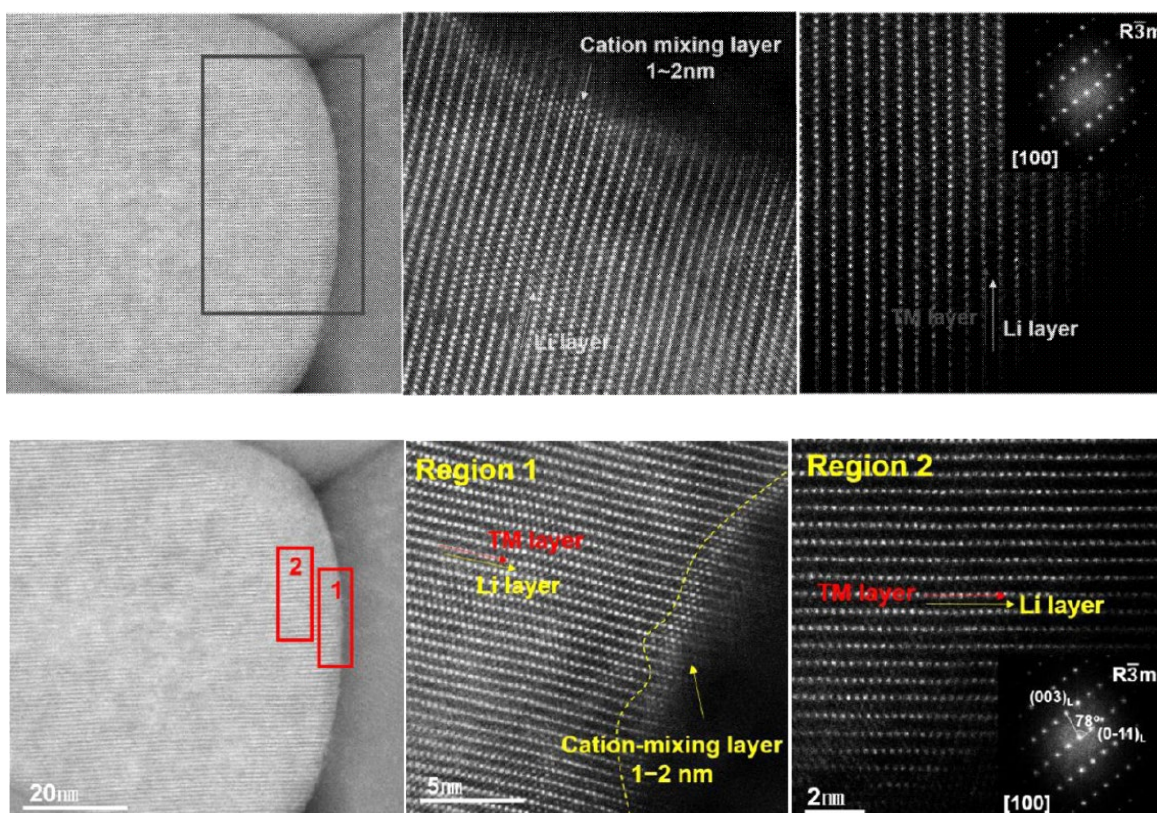


Ex. 1008, Fig. 5B (left); Ex. 1009, Fig. 1(d) (right).





Ex. 1008, Figs. 7A–C (top); Ex. 1009, Figs. 2(d)–(e) (bottom).



Ex. 1008, Figs. 8A–C (top); Ex. 1009, Figs. 2(a)–(c) (bottom) (showing similar, rotated images) (all annotations in original).

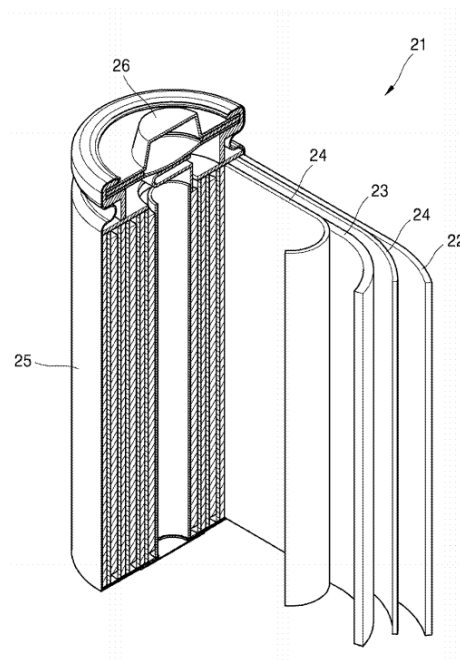
212. Accordingly, a POSITA would have recognized that Kim and Cho are two highly related references that stem from many of the same experiments from the

same research group, with the common goal of using PEAM coatings to achieve “improved structural and thermal stability under severe conditions such as high cutoff voltage (>4.4 V) and high temperature (>50 °C).” Ex. 1009, 2; Ex. 1008, 4:31–57, 6:3–7. Thus, a POSITA would be motivated to consider these references together.

2. Independent Claim 1

a. [Preamble]: A lithium-ion secondary battery comprising:

213. In my opinion, Cho discloses the preamble. Cho involves “[a] composite cathode active material” for a “lithium ion battery.” Ex. 1008, Abstract, 12:62–63. Moreover, Cho teaches improving the electrochemical performance of lithium-ion batteries. *Id.*, 4:31–57. For instance, a “schematic perspective view of a lithium battery” (*id.*, 2:31–32) is shown below in Figure 2:



Id., Fig. 2.

- b. [Element 1.1]: a positive electrode, the positive electrode comprising a positive electrode active material particle comprising lithium cobaltate,

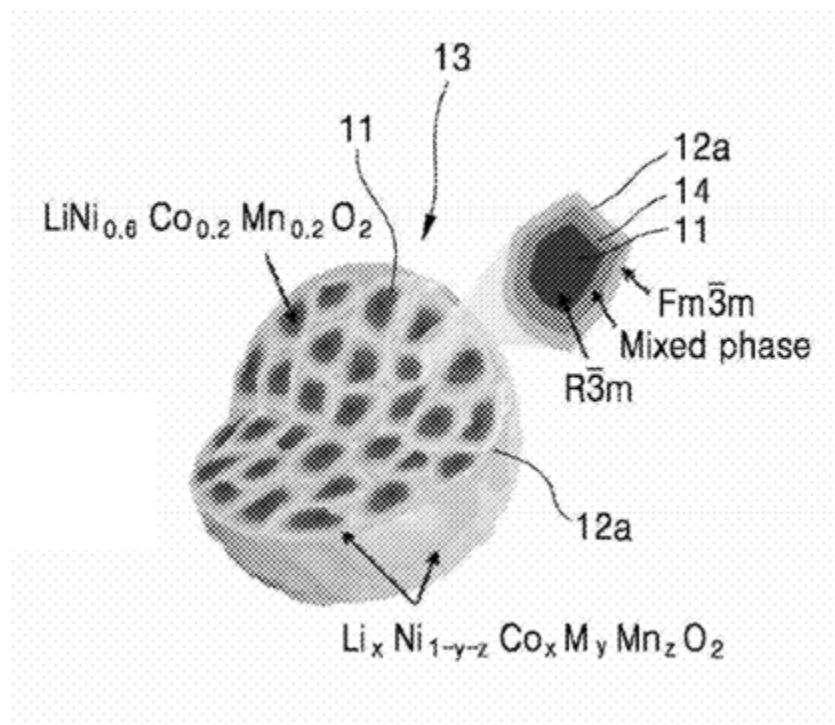
214. In my opinion, Cho discloses element 1.1. Cho teaches that “lithium cobaltate” is an “[e]xample[] of the cathode active material.” Ex. 1008, 9:21–22 (“Examples of the cathode active material used in the art may include one or more of a *lithium cobalt oxide*.”) (emphasis added); *see also id.*, 10:8–9 (“For example, the cathode active material may be . . . *LiCoO₂*[.]”) (emphasis added). As I previously explained, a POSITA would have understood that lithium cobalt oxide refers to LiCoO₂, a well-known and common PEAM used in lithium-ion batteries.

215. Additionally, Cho clarifies that “[a]ny cathode active material used in the art may be further used” in its disclosed battery (*id.*, 9:25–26)—thus, a POSITA would have understood that Cho’s disclosures are broad and not limited to any particular active material (like lithium nickel cobalt manganese oxide (NCM), as taught in some embodiments). In fact, Cho explicitly states that “the cathode active material may be a compound that is represented by one of [several] formulae,” including LiCoO₂. *See id.* 9:27–58. For example, Cho discloses a PEAM formula $Li_aA_{1-b}B_bD_2$, which is simply LiCoO₂ where A is cobalt, B can be cobalt, D is oxygen, *a* is 1, and *b* is 0—all values that Cho discloses. *See id.* In addition, given the similar crystal structures and similar challenges faced by NCM-type and LCO-

type PEAMs, it would have been logical for a POSITA to consider Cho in the context of LCO.

- c. [Element 1.2]: wherein the positive electrode active material particle comprises a first region and a second region,

216. In my opinion, Cho discloses this limitation. Cho discloses that the PEAM or first region “may have a coating layer on a surface thereof[.]” Ex. 1008, 9:59–60. For example, the “coating layer 12a may have a distinct boundary with the primary particle 11” or that the boundary may form a mixed phase that “may not be clearly distinctive” (*i.e.*, the mixed phase region below):



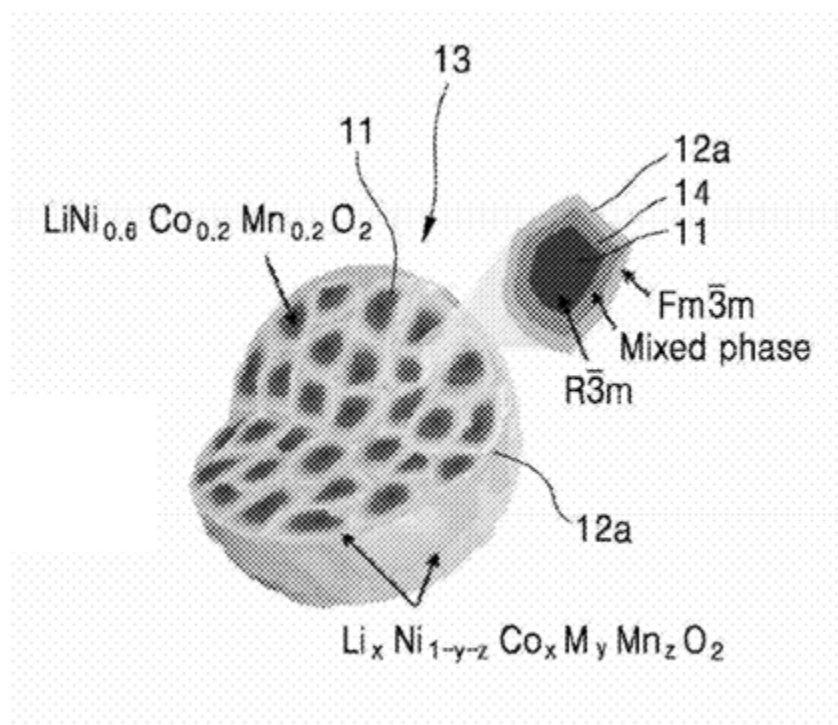
Id., Fig. 1 (excerpted), 7:15–18; *see also id.* 1:35–40, 2:21–25, 7:61–64, 14:57–61, 15:8–52, Figs. 7, 8. As shown above, Cho discloses a coating in a second region

(12a) and an active material in a first region 11. *See id.*, Fig. 1. Although Figure 1 shows a schematic of an NCM-based PEAM, as I explained above in Section IX.D.2.b, Cho expressly indicates that its invention and teachings apply to LiCoO_2 PEAMs. Ex. 1008, 10:8–9 (“[T]he cathode active material may be . . . ***LiCoO₂***[.]”).

- d. [Element 1.3]: wherein the first region comprises a layered rock-salt crystal structure,

217. In my opinion, Cho discloses this limitation. The '660 Patent defines “layered rock-salt crystal structure” as having a “space group of . . . $R\bar{3}m$,” as opposed to “space group $Fm\bar{3}m$ of a general rock-salt crystal.” Ex. 1001, 8:67–9:3.

218. Cho’s first region 11 has “a layered structure (a space structure of $R\bar{3}m$)” (Ex, 1008, 7:10-13):



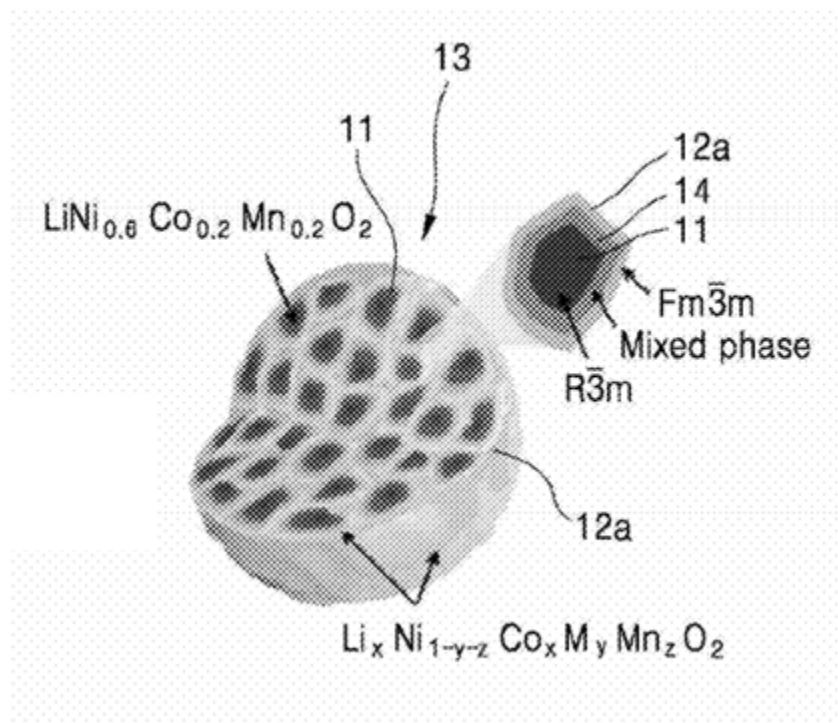
Id., Fig. 1 (excerpted). Figure 1 shows an NCM-based PEAM, but as explained above, Cho also teaches a LiCoO_2 first region with an R-3m space group. *See supra*, § IX.D.2.b; Ex. 1008, 10:8–9.

219. Further, in my opinion a POSITA would have been aware that a LiCoO_2 PEAM would have a layered rock-salt structure (R-3m). As I explain above in Section IX.B.2.d, Lu found that “[t]he crystallographic parameters of pristine and cycled, bare and coated LiCoO_2 in the rhombohedral symmetry” had “space group R-3m.” Ex. 1026, 4–5. Similarly, Rao noted that “[t]he structure of layered LiCoO_2 has Rhombohedral symmetry and belongs to the space group $R\bar{3}m$, and is ideally suited to accommodate large changes in Li concentration. This crystal structure consists of close-packed oxygen ion layers separated by alternating layers of Li and Co ions.” Ex. 1025, 1.

- e. [Element 1.4]: wherein the second region comprises a rock-salt crystal structure,

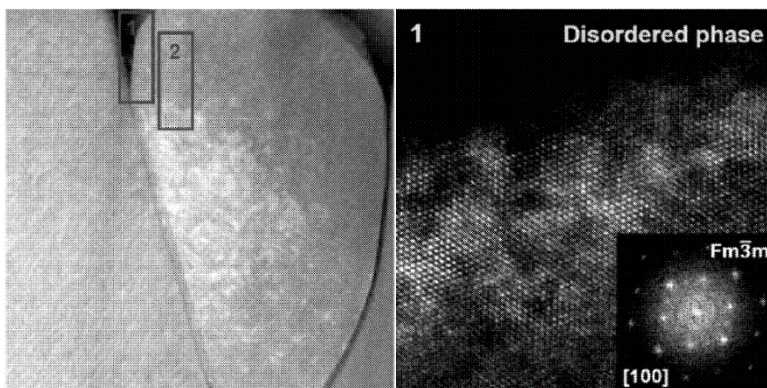
220. In my opinion, Cho discloses this limitation. The ’660 Patent states that a “general rock-salt crystal” (as opposed to a layered rock-salt crystal) has a “space group Fm-3m.” Ex. 1001, 8:67–9:3.

221. As shown below, Cho’s second region (“coating layer 12a”) has a “rock-salt layer structure (a space group of Fm-3m)”:



Ex. 1008, Fig. 1 (excerpted), 7:7–14.

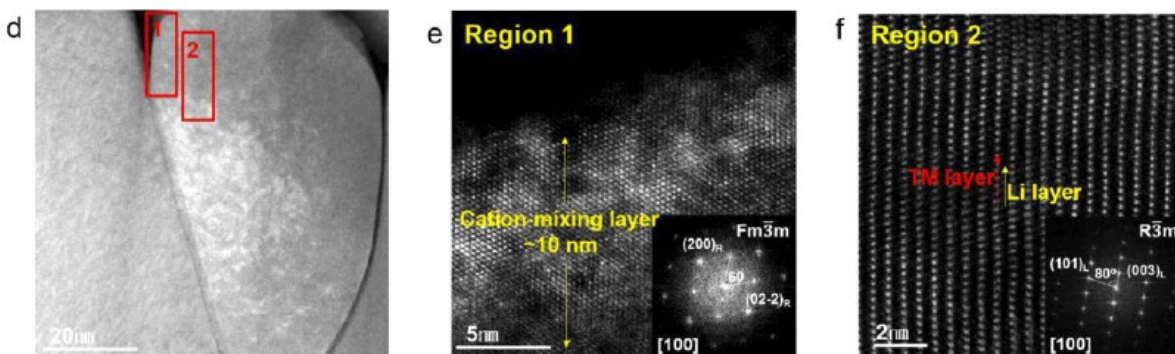
222. Cho’s figures further confirm that the coatings have a rock salt structure, as shown below:



Id., Figs. 7A (left), 7B (right). Figure 7A shows a cross-sectional image of the coated PEAM containing a “region 1” on the “exterior” of the particle, which a POSITA would have understood to be the coating. Ex. 1008, 17:44–48. Figure 7B shows an

atomic-resolution STEM image of the coating at region 1. Cho discloses a FFT patent of region 1 that is identified that is indicative of the space group $Fm\bar{3}m$. *See id.*; *see also id.*, 17:58–61 (describing “an exterior of the primary particle at region 1 . . . having **a rock-salt structure** (an isometric system, a **space group $Fm\bar{3}m$)**”) (emphasis added).

223. Cho also discloses a “cation-mixing layer” that “does not have a layer distinction or a boundary” with the PEAM. *Id.*, 5:48–52. A POSITA would have understood that this cation-mixing layer is part of the second region, since it is a coating and “ha[s] a rock-salt phase.” *Id.* As shown above in Figure 7B, Cho discloses that “[a] **surface phase** of the primary particle was a cation-mixing layer having a rock-salt phase.” *Id.*, 17:64-67. In fact, the FFT analyses disclosed in Kim (which includes the same figures), teach that the PEAM has an $R\bar{3}m$ layered rock salt structure, while the cation-mixing layer has an $Fm\bar{3}m$ rock salt structure:

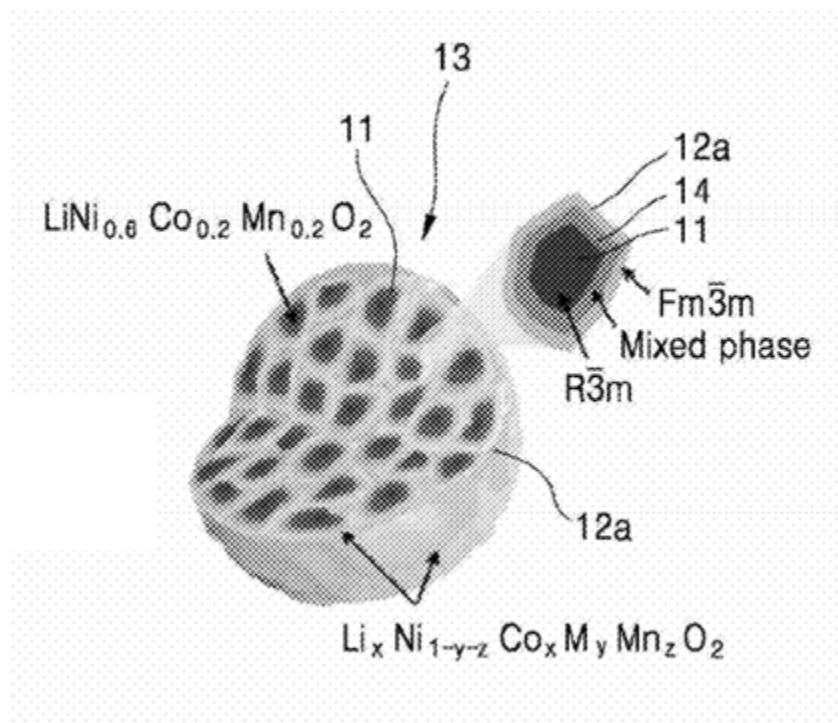


Ex. 1009, Fig. 2(d)–(f). As explained above, a POSITA would have understood that Kim and Cho present the same results, figures, experiments, and research groups, and would consider them together. *See* § IX.D.1.

224. Cho’s crystal terminology differs slightly from the ’660 Patent. For example, Cho uses the term “layered structure” when referring to the first region’s layered rock salt structure, and “rock-salt layer structure” when referring to the coating’s rock salt structure. Ex. 1008, 7:7–14. However, Cho makes clear that the first region’s structure has an R-3m space group (*i.e.*, layered rock salt), while the coating’s structure has an Fm-3m space group (*i.e.*, rock salt), consistent with the definitions of the ’660 Patent. *See id.*; Ex. 1001, 8:67–9:3, cl. 1.

- f. [Element 1.5]: wherein the second region covers at least a first part of the first region,

225. In my opinion, Cho discloses this limitation. Cho explicitly states that “[t]he coating layer may cover the primary particle(s)[.]” Ex. 1008, 5:54–55; *see id.*, 4:41–42, 7:7–12 (describing “coating layer 12a” (space group of Fm-3m) “***on a surface of the primary particle***” with “space structure of R[-]3m”) (emphasis added). Moreover, Cho discloses first region 11 (which can be LiCoO₂) covered by a coating layer 12a:



Ex. 1008, Fig. 1 (excerpted).

- g. [Element 1.6]: wherein the positive electrode active material particle comprises titanium, magnesium, fluorine, and oxygen in the second region, and

226. In my opinion, Cho discloses this limitation. Cho discloses that the coating layer or second region may be an oxide of a mixture of titanium and magnesium—in other words, a coating containing titanium, magnesium, and oxygen. For example, Cho states the “coating layer may include a coating element compound, such as an oxide of a coating element” like Mg and Ti (or other elements) “or a mixture thereof.” Ex. 1008, 9:59–6:2

227. Further, Cho teaches that such a coating layer comprises fluorine. Cho’s battery contains an LiPF_6 electrolyte (Ex. 1008, 12:13–30, 16:4–9), and Cho

explicitly states that “[t]he coating layer may include [] a product obtained by a sol-gel reaction of compositions including a *lithium salt* or a cobalt salt[.]” *Id.*, 5:29–31. A POSITA would have understood that a fluorine-coating layer (which could be a lithium salt, LiF) would be formed on the cathode surface from reactions with the LiPF₆ electrolyte, from the following reaction:

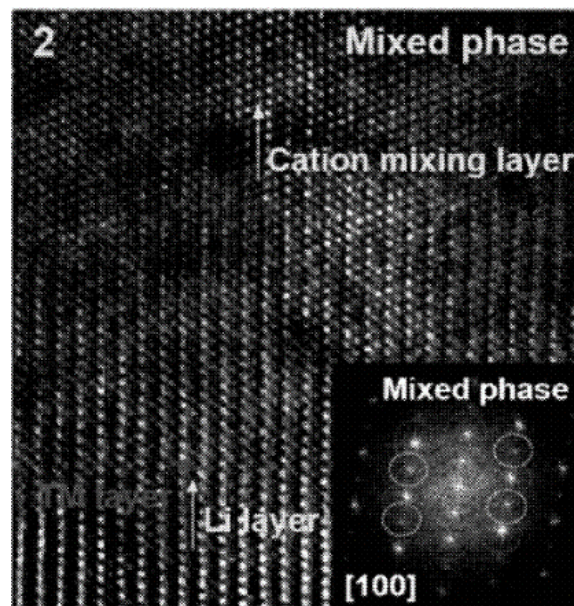


228. A POSITA would have further understood Cho’s coating would comprise fluorine in view of Kim’s disclosures. For example, Kim states that “a new solid electrolyte interface (SEI) layer, comprising *decomposition products of the electrolyte*, is formed along the active surface of the primary particles (grains) developed by the microcracks” (emphasis added) of the cathode material. Ex. 1009, 2–3. In other words, decomposition products of the LiPF₆ (including a fluorine-containing layer) will be found along the “active surface” of the cathode material. As explained above, a POSITA would have read Kim and Cho together because they present the same results, figures, experiments, and were authored by the overlapping researchers and research groups. *See* § IX.D.1.

- h. [Element 1.7]: 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.

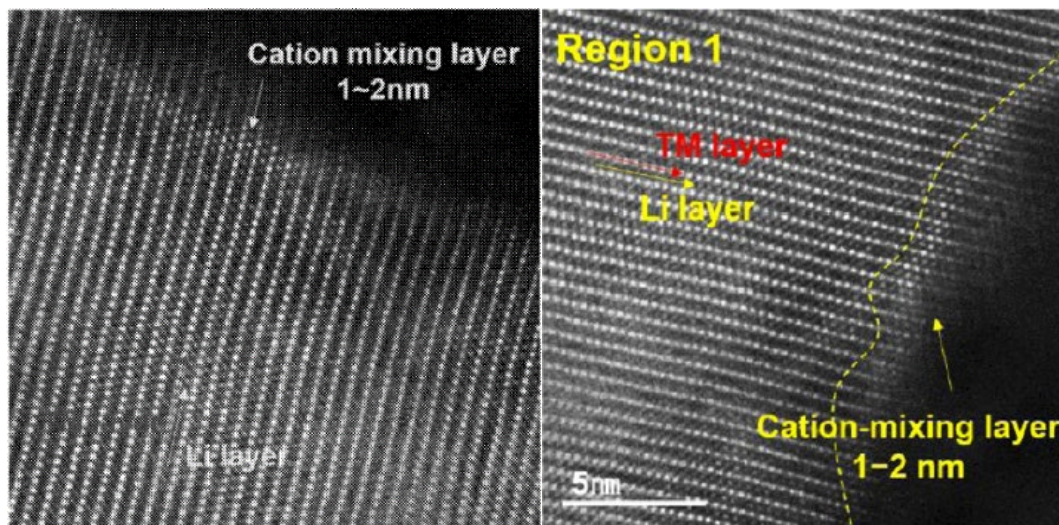
229. In my opinion, Cho discloses this limitation. As explained in the '660 Patent, substantial alignment of “crystal orientations” occurs when “an angle between the repetition of bright lines and dark lines in the layered rock-salt crystal [*i.e.*, first region] and the repetition of bright lines and dark lines in the rock-salt crystal [*i.e.*, second region] is less than or equal to 5°.” Ex. 1001, 9:22–29.

230. Cho shows substantial alignment between the first region’s layered rock-salt crystal structure and the second region’s rock-salt crystal structure:



Ex. 1008, Fig. 7C. As I explain above in Section IX.D.2.e, a POSITA would have understood that the cation-mixing layer is the second region (as it has space group Fm-3m) (Ex. 1008, Fig. 7B) and the base material (the lower region in the image) is the first region. Moreover, the angles between the bright and dark lines between the first and second region measure 0°, as the bright and dark lines are vertical.

231. Substantial alignment between Cho's first and second regions is further shown by Figure 8B, when compared to Kim's Figure 2B, which is the same image as Cho's Fig. 8B but merely rotated and annotated:



Ex. 1008, Fig. 8B (left); Ex. 1009, Fig. 2(b) (right) (annotations in original). As shown above, the bright and dark lines between “Region 1” and the “Cation-mixing layer” seamlessly extend in straight lines across the boundary between these regions. Thus, the angles between the bright and dark lines between the first and second region measure approximately 0°. As explained above, a POSITA would have read Kim and Cho together because they present the same results, figures, experiments, and were authored by the overlapping researchers and research groups. *See* § IX.D.1.

3. Dependent Claim 2

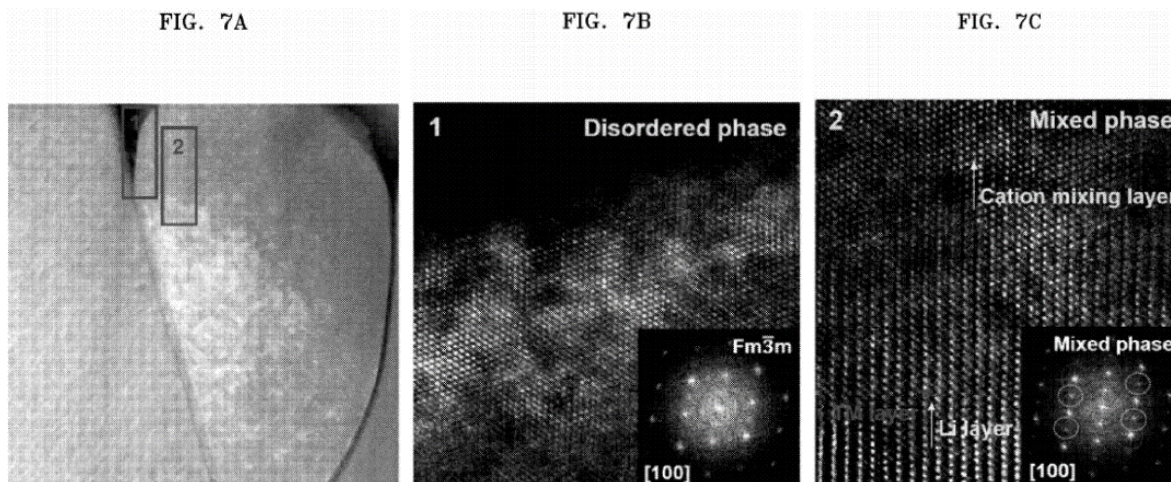
- a. [Element 2.0]: The lithium-ion secondary battery according to claim 1,

232. In my opinion, Cho renders claim 1 obvious, either alone or with Kim.

See supra, § IX.D.2.

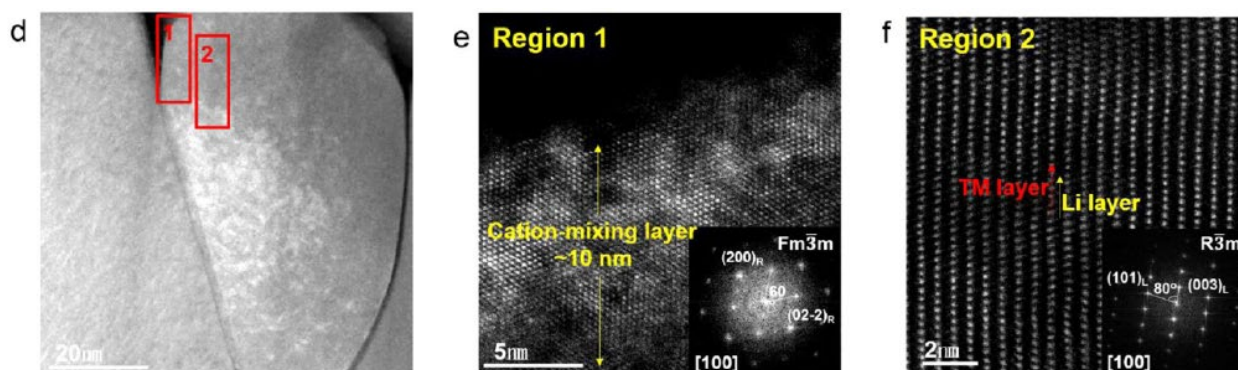
- b. [Element 2.1]: wherein the titanium, the magnesium, the fluorine, and the oxygen are provided in a range from the surface of the positive electrode active material particle to a depth of 5 nm.

233. In my opinion, Cho discloses this limitation. As I explain above in Section IX.D.2.g, Cho teaches a coating layer comprising oxygen, titanium, and magnesium. Ex. 1008, 2:5–11, 9:59–10:2, 12:13–30. The “thickness of the coating layer may be about 20 nm or less, or, for example, in a range of about 1 nm to about 20 nm.” *Id.*, 5:9–11, *see also id.*, 1:49–50 (“A thickness of the coating layer may be about 20 nm or less”), 5:11–13 (“When the thickness of the coating layer is within this range, a lithium battery may have excellent capacity and lifespan characteristics without deterioration of discharge capacity.”), cl. 3. Cho’s Figures 7A–C show TEM images of the primary particle in Cho’s Example 1, containing “a coating layer” (*id.*, 14:59):



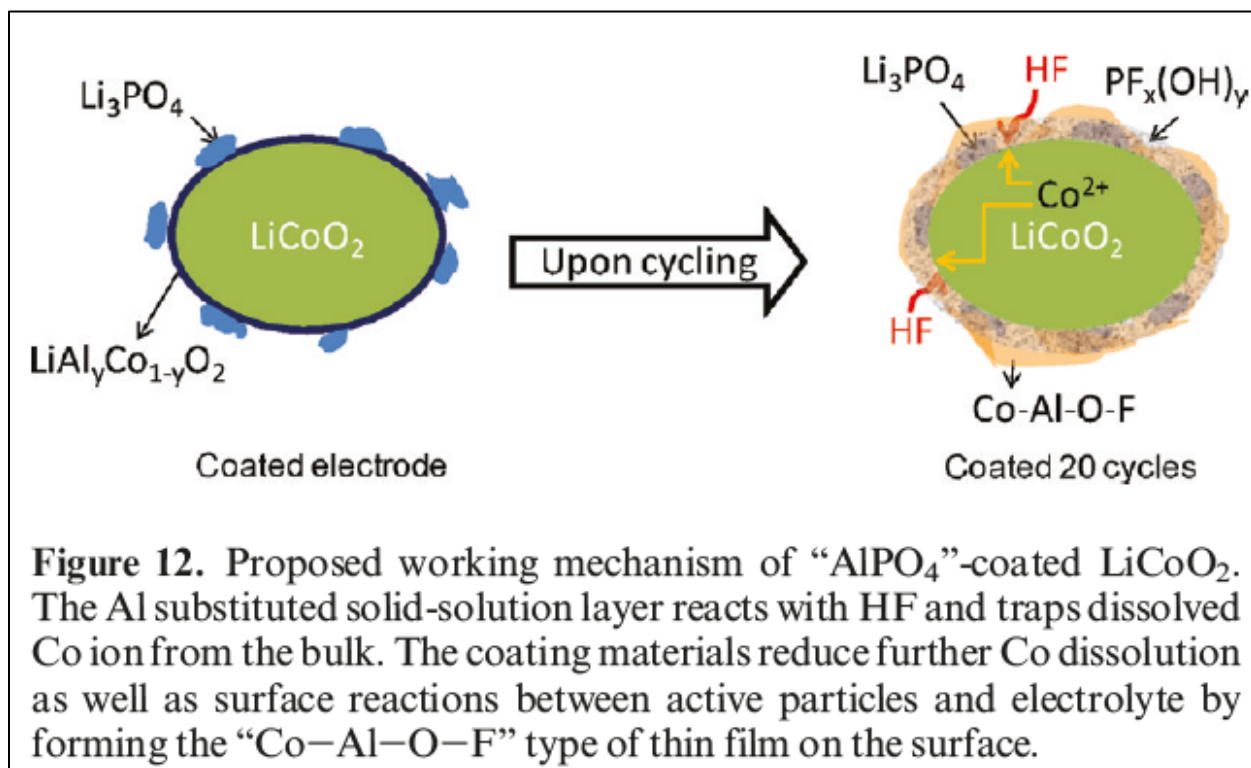
Ex. 1008, Figs. 7A–C (showing an $Fm\bar{3}m$ space group for the coating layer). From these images, and in light of the similar images presented in Kim (see below), a POSITA would have understood that the coating layer is at least 5 nm thick, meaning that the elements of the coating (including titanium, magnesium, and oxygen) can be found in the first 5 nm.

234. Kim includes scale bars on the same and similar images, supporting this point:



Ex. 1009, Figs. 2(d)–(f). As shown above, the coating layer (Fm-3m) is at least 5 nm thick from the surface of the PEAM. Thus, a POSITA would have understood that in Cho’s particle, Mg, O, and Ti may be found in the claimed range.

235. Further, as explained in Section IX.D.2.g, a POSITA would have understood that fluorine is included in Cho’s PEAM, due to reactions with the LiPF_6 electrolyte. For example, the LiPF_6 electrolyte would lead to a “very thin surface layer that predomina[nt]ly consists of LiF and $\text{Li}_x\text{PF}_y\text{O}_z$ ” on the PEAM’s surface. Ex. 1026, 17. Studies, including Lu’s, have shown that this fluorine layer would interact with a particle’s coating layer, as shown below:



Id., Fig. 12 (excerpt). Lu's Figure 12 shows a fluorine-containing electrolyte and an AlPO_4 coating interacting to form a coating with fluorine (from the electrolyte), and O and Al (from the original coating). *Id.* Lu observed that aluminum at the surface of the "coated LiCoO_2 particles promote the formation of Co–Al–O–F species on the particle surfaces." *Id.*, 17. This "observation[] suggest[s] that . . . adding Al_2O_3 in the LiCoO_2 electrode lead to increased surface coverage of Co- and Al-containing fluoride and/or oxyfluoride species during cycling, which can prevent further Co dissolution and side reactions between the electrolyte and the active material and reduce impedance growth during cycling to high voltages." *Id.* In other words, Lu found that fluorine interacted with the elements present in the coating, which was aluminum in Lu's case, and formed a species on the surface of the particle containing aluminum, oxygen, and fluorine. *Id.*

236. Similarly, a POSITA would have understood that Cho's coating (which contains Ti, Mg, and O) would interact with fluorine according to Lu's observations as I explain above to form a species containing titanium, magnesium, oxygen, and fluorine (*i.e.*, metal-containing fluoride and/or oxyfluoride species). Thus, a POSITA would have understood that Ti, Mg, F, and O would all be present at the surface of the PEAM to a depth of 5 nm.

4. Dependent Claim 3

- a. [Element 3.0]: The lithium-ion secondary battery according to claim 1,

237. In my opinion, Cho renders claim 1 obvious, either alone or with Kim.

See supra, § IX.D.2.

- b. [Element 3.1]: wherein the positive electrode further comprises a conductive additive, and

238. In my opinion, Cho discloses this limitation. For example Cho teaches “a cathode active material composition may be prepared by mixing the composite cathode active material, ***a conducting agent***, a binder, and a solvent.” (emphasis added) Ex. 1008, 9:7–9.

- c. [Element 3.2]: wherein the conductive additive comprises carbon fiber, graphene or multilayer graphene.

239. In my opinion, Cho discloses this limitation. Cho states that “[a]ny conducting agent used in the art may be used” (Ex. 1008, 10:13–14), and “[e]xamples of the conducting agent may include carbon black or ***graphite particles*** . . . [or] conductive fibers such as ***carbon fibers*** and metallic fibers.” *Id.*, 10:12–20. A POSITA would have understood that graphite is composed of many layers of graphene. Thus, a conductive additive of at least carbon fiber is disclosed and multilayer graphene would have been obvious to a POSITA in light of the disclosure of graphite.

E. Ground II.B: Claims 4 and 5 Are Rendered Obvious by Cho Alone or in Combination with Kim and/or Blangero

1. Motivation to Combine Cho with Blangero

240. In my opinion, a POSITA would have been motivated to combine Cho's teachings with Blangero's LCO cathode coated with magnesium oxide and titanium oxide, which yields the claimed Ti:Co ratio. Specifically, Blangero and Cho both disclose LCO as a PEAM, and treating the PEAM with the same transition metals, titanium and magnesium, in a surface layer. Ex. 1008, 10:8–9 (“[T]he cathode active material may be . . . LiCoO_2 ”), 1:64–2:11 (the active material may also contain titanium), 5:11–14 (describing “excellent capacity and lifespan characteristics” of Cho's coating), 7:23–31, 9:67–10:2 (“A coating element included in the coating layer may be $\text{Mg}[\cdot]$ ”); Ex. 1005, ¶¶ [0004], [0005], [0094], [0108]–[0110] (describing particles based on LiCoO_2 doped and coated with Mg and Ti).

241. Indeed, both Blangero and Cho describe the use of magnesium and titanium as dopants and coatings for PEAM particles to improve a lithium-ion battery's electrochemical performance. Ex. 1008, 3:63–65 (“[L]ithium transition metal oxide may have excellent capacity characteristics and high energy capacity”); 7:23–40, 9:59–10:7, 19:3–32; Ex. 1005, ¶¶ [0004], [0005]–[0007] (describing coating LiCoO_2 to improve stability while maintaining capacity). Both references teach doping PEAMs with various dopants, including magnesium and titanium. Ex. 1008, 1:64–2:11, 7:23–31; Ex. 1005, ¶¶ [0004], [0087], [0109] (describing “state of the art Mg and Ti doped LiCoO_2 ”).

242. Blangero also teaches that the use of titanium and magnesium dopants results in an “in-situ” coating of a surface layer that “is formed during the second sintering by spontaneous segregation of the dopants from the core.” Ex. 1005, ¶ [0090]. This surface layer includes “enriched in oxidized compounds, like MgO and TiO₂.” Ex. 1005, ¶¶ [0090], [0094]. Blangero teaches that this surface layer is beneficial because the “inert coating layer provides additional safety when the powders are in contact with the electrolyte of the battery.” *Id.*, ¶¶ [0093]–[0094]. This addresses the problem where uncoated cathodes react with the electrolyte which is decomposed and, over time, results in a loss of capacity and a strong increase of resistance in the battery. *Id.* Thus, a POSITA would have been motivated to apply the teachings of Blangero to other LCO particles, including the particles disclosed by Cho.

2. Dependent Claim 4

- a. [Element 4.0]: The lithium-ion secondary battery according to claim 1,

243. In my opinion, Cho renders claim 1 obvious, either alone or with Kim.

See supra, § IX.D.2.

- b. [Element 4.1]: wherein a relative value of a concentration of the titanium is greater than or equal to 0.05 and less than or equal to 0.4 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of cobalt is defined as 1.

244. In my opinion, Cho renders this element obvious in combination with Blangero. A POSITA would have understood that Blangero's particle would have a concentration of Ti:Co in the claimed range. As I explain above in Section IX.B.5, Blangero measures the relative Ti:Co concentration at the particle's surface as 0.0917:

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

Ex. 1005, ¶ [0129], Table 3 (annotations added); *see also id.*, Fig. 4 (showing XPS spectra for Example 1). Table 3 shows that the surface of the particle of examples 1 has a Ti:Co ratio of 1.1/12.0. This equals a ratio of 0.0917 and falls within the claimed range of 0.05 to 0.4, rendering this element obvious. Further, I note that other studies (like Yu) found similar Ti:Co ratios at the surface of LiCoO₂-based particles:

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

Ex. 1011, 3, Table 3 (annotated).

245. As I explain above in Section IX.B.5, a POSITA would have understood that despite enhancing battery stability and performance, titanium is not electrochemically active and therefore a POSITA could have discovered an optimal Ti:Co concentration ratio with routine experimentation. A POSITA would have also known that the Ti:Co concentration is a result-effective variable and that the performance of a battery would be influenced by the titanium-cobalt ratio because titanium is not electrochemically active in the cathode. A POSITA further would have known that the amount of titanium included in a PEAM particle is easily adjusted during cathode synthesis. Accordingly, a POSITA would have understood it may be beneficial to increase the concentration of surface titanium, would have been motivated to experiment with it to improve cathode performance, and could have easily done so.

246. Therefore, in my opinion, Cho and a POSITA's knowledge of Blangero render the claimed range obvious. Alternatively, a POSITA would have been motivated to combine Cho and Blangero for all the reasons explained above in Section IX.E.1.

3. Dependent Claim 5

- a. [Element 5.0]: The lithium-ion secondary battery according to claim 1,

247. In my opinion, Cho renders claim 1 obvious, either alone or with Kim.

See supra, § IX.D.2.

- b. [Element 5.1]: wherein a relative value of a concentration of the magnesium is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of cobalt is defined as 1.

248. In my opinion, Cho renders this element obvious in combination with Blangero. A POSITA would have understood that Blangero renders obvious a particle with the claimed concentration of Mg:Co. As I explain above in Section IX.B.6, Blangero measures the relative Mg:Co concentration at the particle's surface as approximately 0.367:

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

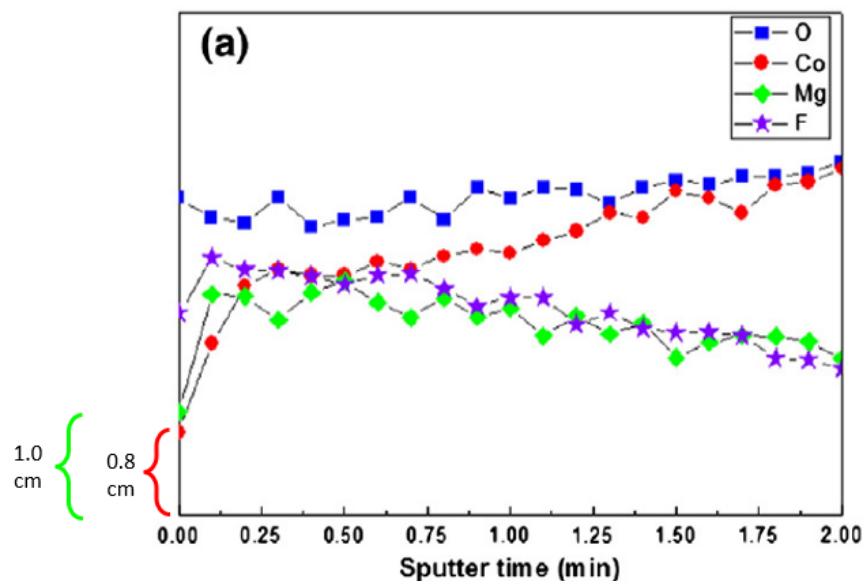
Ex. 1005, ¶ [0108], Table 3; *see also id.*, Fig. 4(b). Table 3 shows that the surface of the particle of examples 1 has a Mg:Co ratio of 4.4/12.0. This equals a ratio of 0.367, which approximates 0.4 in the context of Claim 5 where the range is defined with values expressed as one decimal place. In my opinion, Blangero also discloses a PEAM particle with a higher concentration of magnesium at the surface of the particle than disclosed by Example 1. Example 2 is comprised of “94 mol % of LCO-1 and 6 mol % of 0.25 mol % Ti and 0.5 % mol % Mg doped Co(OH)₂.” *Id.*, ¶ [0111]. Example 1, by comparison, is comprised of “95 mol % of LCO-1 and 5 mol % of 0.25 mol % Ti and 0.5 % mol % Mg doped Co(OH)₂.” *Id.* By increasing from 5 mol % to 6 mol %, the amount of magnesium used in manufacturing Example 2 increases by 20% compared to the amount used in Example 1, while the amount of cobalt decreases by approximately 1% (from 96 mol % to 95 mol %). Accordingly, a POSITA would have understood that the relative concentration of magnesium to cobalt derived from Table 3 is not a floor and instead some of Blangero’s samples would have higher ratios. If, for example, this increase in magnesium were to cause a corresponding 20% increase in surface magnesium and a 1% decrease in surface cobalt, the ratio of Mg:Co would increase from 0.367 to 0.444. In my opinion, this further renders the claimed range obvious.

249. In my opinion, a POSITA would have found the Mg:Co range of 0.4 to 1.5 obvious based on the above ratio taught by Blangero. A POSITA would have

known that magnesium improves performance of LiCoO_2 -based batteries, including Ex. 1008, 5:11–14 (describing “excellent capacity and lifespan characteristics” of Cho’s coating), 7:23–31. 9:67–10:2 (“A coating element included in the coating layer may be Mg[.]”); Ex. 1005, ¶¶ [0093]–[0094] (using Mg or MgO in a coating layer “provides additional safety” to the cathode material from the battery’s electrolyte, preventing “deterioration of the electrochemical performance” and “loss of capacity”).

250. A POSITA would have also known that the Mg:Co concentration is a result-effective variable and that the performance of a battery would be influenced by the magnesium-cobalt ratio because magnesium is not electrochemically active in the cathode. A POSITA further would have known that the amount of magnesium included in a PEAM particle is easily adjusted during cathode synthesis. Moreover, a POSITA would have understood that despite enhancing battery stability and performance, magnesium is not electrochemically active and therefore a POSITA could have discovered an optimal Mg:Co concentration ratio with routine experimentation. Accordingly, a POSITA would have understood it may be beneficial to increase the concentration of surface magnesium, would have been motivated to experiment with it to improve cathode performance, and could have easily done so.

251. I further note that a POSITA would have been aware of other studies (including Lee) showing Mg:Co concentration ratios at the surface of particles within the claimed range, as discussed above in Section IX.B.6 and as shown below:



Ex. 1008, Fig. 5(a) (y-axis annotations added).

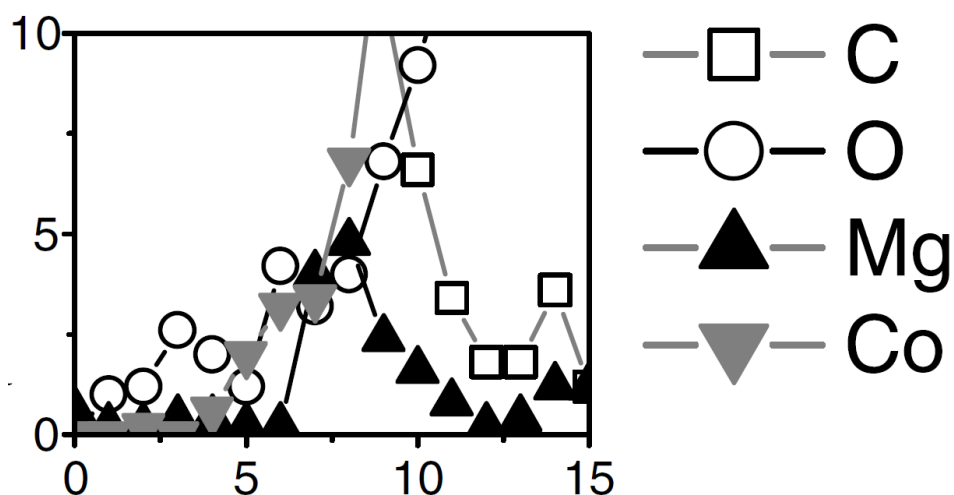
252. Therefore, In my opinion, Cho and a POSITA's knowledge of Blangero render the claimed range obvious. Alternatively, a POSITA would have been motivated to combine Cho and Blangero for all the reasons explained above in Section IX.E.1.

F. Ground II.C: Claims 6–8 Are Rendered Obvious by Cho Alone or in Combination with Kim and/or Yamamoto

1. Motivation to Combine Cho with Yamamoto

253. A POSITA would have been motivated to combine Cho's particle with Yamamoto's MgO coating. In fact, Cho makes clear that Yamamoto's coating (MgO) is compatible with Cho's invention. For instance, Cho teaches that its coating may be "an oxide of a coating element" and that such coating element could be "Mg"—indicating that MgO is a suitable coating material. Ex. 1008, 9:59–10:7; *see also* Ex. 1007, 1 ("The MgO coating improves the durability of the cathode during high-potential cycling."). Additionally, Kim makes clear that the cation-mixing layer (described in both Cho and Kim) is "a pillar layer, effectively improv[ing] the structural stability and electrochemical performance by suppressing the microcracks in the particles and grains during cycling." Ex. 1009, 8. Similarly, Yamamoto describes a coating providing ions serving as "pillars" to stabilize the layered structure of LiCoO_2 . Ex. 1007, 5 ("Mg ions serve as pillars in LiCoO_2 layer[s] to improve the stability at high potential.").

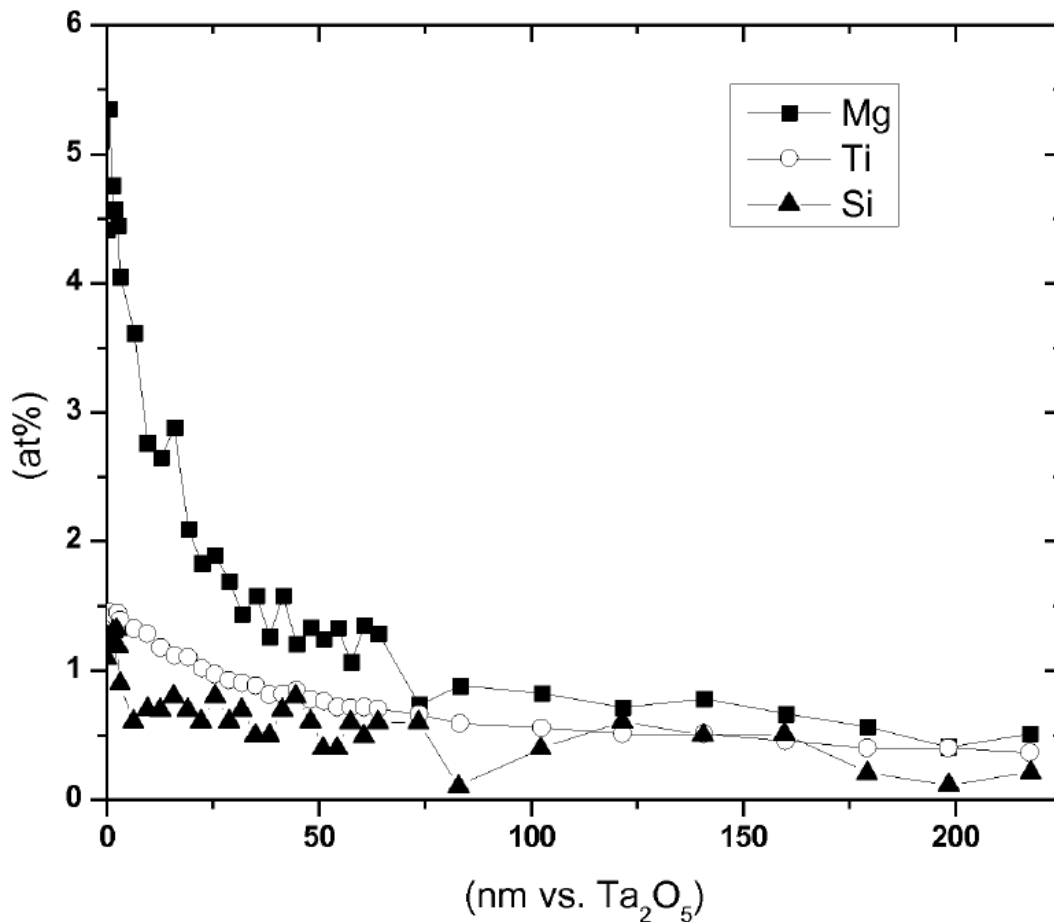
254. As explained in Section IX.C.1, Yamamoto discloses analyzing a PEAM particle using an EDX line scan. Ex. 1007, 2, Fig. 1(b). A POSITA would have understood from Yamamoto that a magnesium coating on the surface of a PEAM will result in a peak Mg concentration near the surface that decreases radially inward toward the center of the PEAM.



Id., Fig. 1 (excerpted).

255. Applying Yamamoto's findings and well-known measurement technique to Cho would have been a routine matter. A POSITA would have further expected to observe that the concentration of magnesium would decrease from the surface to the center of the PEAM as this was a well-documented and observed phenomenon.

256. Other references, including Blangero (Ex. 1005) and Shim 2015 (Ex. 1032) corroborate Yamamoto's findings, as explained in Section IX.C.1 and shown below:



Ex. 1005, Fig. 8, ¶ [0135].

257. Further, both Cho and Yamamoto teach that coating PEAMs with magnesium (as both Yamamoto and Cho describe some form of magnesium oxide) improves the durability and cycling characteristics of the battery. Ex. 1007, 1 (“The MgO coating improves the durability of the cathode during high-potential cycling.”); Ex. 1008, 9:59–10:7 (expressly teaching “an oxide of a coating element,” including “Mg” (*i.e.*, possibly MgO) as a suitable coating layer). As I explain above with respect to Blangero, a POSITA would have been motivated to use enough

magnesium to achieve this improvement, while not using excess magnesium since magnesium is not electrochemically active within a typical operating voltage window of an LiCoO_2 -based cell (in other words, substituting magnesium for cobalt theoretically results in a small loss of mass-specific capacity). With these competing factors in mind, a POSITA would have been motivated to experiment with the concentration of magnesium to determine an optimum concentration profile of magnesium through a PEAM by varying the amount of Mg using routine experimentation.

258. Additionally, a POSITA would have recognized that Cho and Yamamoto are both directed to the same goal of improving performance of PEAMs in lithium-ion batteries. Indeed, both references disclose coatings containing magnesium and oxygen to attain this specific benefit. Ex. 1008, 9:59–10:7 (describing a coating comprising “an oxide of a coating element” such as “Mg”), 5:11–14 (describing “excellent capacity and lifespan characteristics” of Cho’s coating); Ex. 1007, 1 (“The MgO coating improves the durability of the cathode during high-potential cycling. . . . [T]he MgO coating suppresses drastic distortions of local structure at the LiCoO_2 surface.”).

2. Independent Claim 6

a. [Preamble]: A lithium-ion secondary battery comprising

259. To the extent the preamble is limiting, Cho discloses it. *See supra*, § IX.D.2.a.

- b. [Element 6.1]: a positive electrode, the positive electrode comprising a positive electrode active material particle comprising lithium cobaltate,

260. Cho discloses this limitation. *See supra*, § IX.D.2.b.

- c. [Element 6.2]: wherein the positive electrode active material particle comprises a first region and a second region,

261. Cho discloses this limitation. *See supra*, § IX.D.2.c.

- d. [Element 6.3]: wherein the first region comprises a layered rock-salt crystal structure,

262. Cho discloses this limitation. *See supra*, § IX.D.2.d.

- e. [Element 6.4]: wherein the second region comprises a rock-salt crystal structure,

263. Cho, either alone or with Kim, renders this limitation obvious. *See supra*, § IX.D.2.e.

- f. [Element 6.5]: wherein the positive electrode active material particle comprises titanium, magnesium, fluorine, and oxygen in the second region,

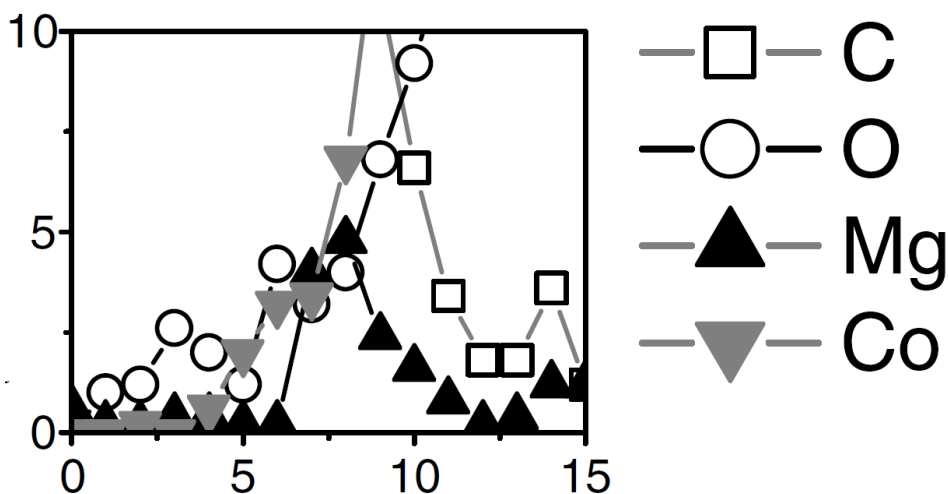
264. Cho, either alone or with Kim, renders this limitation obvious. *See supra*, § IX.D.2.g.

- g. [Element 6.6]: 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, and

265. In my opinion, Cho in combination with a POSITA's knowledge—specifically, knowledge of Yamamoto's EDX line scan studies of MgO coatings on LiCoO_2 —renders this limitation obvious. Alternatively, in my opinion, Cho in combination with Yamamoto renders this limitation obvious.

266. First, Cho renders this limitation obvious in combination with a POSITA's knowledge. The surface of Cho's particle is one end of the second region (*i.e.*, the coating), which Cho discloses may contain magnesium. *See supra* §§ IX.D.3.b; IX.F.2.f. As I explain above in Section IX.C.2.g, Yamamoto further teaches that the second region extends to a depth within the particle where the magnesium concentration (as measured by EDX line analysis) is $\frac{1}{5}$ the maximum peak of Mg in the line analysis, as shown below:



Ex. 1007, Fig. 1 (excerpted). As discussed above in Section IX.C.2.g, Yamamoto indicates that Mg first appears at a depth of 7 nm, reaches a peak at 8 nm (of 5 net counts), and reaches 1/5 of its peak concentration (*i.e.* 1 net count) at a depth of 13 nm. Thus, this teaches the claimed concentration relationship within the second region. *See id.*

267. A POSITA would have found this claimed magnesium concentration relationship obvious in Cho's coating in light of his or her knowledge—specifically, knowledge of Yamamoto's teachings. As shown in the EDX line scan above, a POSITA would have understood a fundamental concept from Yamamoto: that “a Mg containing layer of approximately 5 nm in thickness” (*id.*, 2) (such as MgO) experiences a concentration peak within that 5 nm range, and eventually decreases to 0 farther away from the surface, and into the bulk of the particle. Accordingly, at some given depth, the concentration of magnesium must reach 1/5 of its maximum concentration. Similarly, with respect to Cho's magnesium-containing lithium titanate coating—*e.g.* a coating layer including an oxide of a coating element such as magnesium and titanium “or a mixture thereof” (Ex. 1008, 9:59–6:2)—a POSITA would have understood that magnesium concentration reaches its peak at some given depth, and that at some other given depth, its concentration will reach 1/5 of its peak.

268. Further, it would be obvious to a POSITA that magnesium improves a PEAM's performance. A POSITA would have known that magnesium improves the

electrochemical performance of PEAMs in batteries. *See* Ex. 1008, 5:11–14, 9:67–10:2 (describing “excellent capacity and lifespan characteristics” of Cho’s coating which may include Mg); Ex. 1007, 1. As I explain above with respect to Blangero, a POSITA would have been motivated to use enough magnesium to achieve this improvement, while not over including magnesium since magnesium is inert within a typical operating voltage window of an LiCoO_2 -based cell (in other words, substituting magnesium for cobalt theoretically results in a small loss of capacity). With these competing factors in mind, a POSITA would have been motivated to experiment with the concentration of magnesium to determine an optimum concentration profile of magnesium through a PEAM using routine experimentation.

- h. [Element 6.7]: 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.

269. In my opinion, Cho discloses this limitation. The second region recited by Claim 6 comprises Mg, Ti, F, and O, akin to the second region recited by Claim 2. *See* Ex. 1001, cls. 2, 6; *see also id.* 11:5–12:16 (describing titanium, magnesium, fluorine, and oxygen in a second region over the first region), 12:17–13:57 (describing magnesium and fluorine in a third region over the second region). Thus, for the same reasons as I describe above in Section IX.D.2.h, Cho discloses this limitation.

3. Dependent Claim 7

- a. [Element 7.0]: The lithium-ion secondary battery according to claim 6,

270. In my opinion, Cho renders claim 6 obvious, either alone or with Kim and/or Yamamoto. *See supra*, § IX.F.2.

- b. [Element 7.1]: wherein the titanium, the magnesium, the fluorine, and the oxygen are provided in a range from the surface of the positive electrode active material particle to a depth of 5 nm.

271. In my opinion, Cho discloses this limitation. *See supra*, § IX.D.3.b.

4. Dependent Claim 8

- a. [Element 8.0]: The lithium-ion secondary battery according to claim 6,

272. In my opinion, Cho renders claim 6 obvious, either alone or with Kim and/or Yamamoto. *See supra*, § IX.F.2.

- b. [Element 8.1]: wherein the positive electrode further comprises a conductive additive, and

273. In my opinion, Cho discloses this limitation. *See supra*, § IX.D.4.b.

- c. [Element 8.2]: wherein the conductive additive comprises carbon fiber, graphene or multilayer graphene.

274. In my opinion, Cho discloses this limitation. *See supra*, § IX.D.4.c.

G. Ground II.D: Claims 9 and 10 Are Rendered Obvious by Cho Alone or in Combination with Kim, Yamamoto, and/or Blangero

1. Dependent Claim 9

- c. [Element 9.0]: The lithium-ion secondary battery according to claim 6,

275. In my opinion, Cho renders claim 6 obvious, either alone or with Kim and/or Yamamoto. *See supra*, § IX.F.2.

- d. [Element 9.1]: wherein a relative value of a concentration of the titanium is greater than or equal to 0.05 and less than or equal to 0.4 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of cobalt is defined as 1.

276. In my opinion, this limitation is rendered obvious by Cho either with a POSITA's knowledge or in combination with Blangero. *See supra*, § IX.E.2.b.

2. Dependent Claim 10

- e. [Element 10.0]: The lithium-ion secondary battery according to claim 6,

277. In my opinion, Cho renders claim 6 obvious, either alone or with Kim and/or Yamamoto. *See supra*, § IX.F.2.

- f. [Element 10.1]: wherein a relative value of a concentration of the magnesium is greater than or equal to 0.4 and less than or equal to 1.5 when the surface of the positive electrode active material particle is subjected to an XPS analysis and a concentration of cobalt is defined as 1.

278. In my opinion, this limitation is rendered obvious by Cho either with a POSITA's knowledge or in combination with Blangero. *See supra*, § IX.E.3.b.

X. SECONDARY CONSIDERATIONS

279. I have seen no evidence of, and I am not aware of any, secondary considerations that would tend to show non-obviousness of the Challenged Claims of the '660 Patent, including commercial success, long-felt need, failure of others, skepticism, praise, teaching away, recognition of a problem or copying by competitors. However, should Patent Owner assert any secondary considerations of non-obviousness, I reserve the right to respond.

XI. CONCLUSION

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

Dated: September 1, 2023



Troy A. Hayes, Ph.D., P.E.