

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

AMPEREX TECHNOLOGY LTD.
Petitioner,

v.

SEMICONDUCTOR ENERGY LABORATORY CO., LTD.
Patent Owner

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

Issued: August 11, 2020

Filed: June 30, 2017

Inventor: Teruaki Ochiai et al.

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

Case IPR2023-TBD

**PETITION FOR *INTER PARTES* REVIEW
UNDER 35 U.S.C. §312 AND 37 C.F.R. §42.104**

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Unless otherwise noted, with respect to these references, all emphasis is added

LIST OF EXHIBITS

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

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

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

Petitioner Amperex Technology, Ltd. (“Petitioner”) requests *inter partes* review (“IPR”) of claims 1–8 (“Challenged Claims”) of U.S. Patent No. 10,741,828 (“’828 Patent”) (Ex. 1001).

I. INTRODUCTION

The ’828 Patent claims a simple, obvious concept: stabilizing lithium-ion batteries by adding various known metals to the battery’s positive electrode. Ex. 1001, 13:17–38, 13:56–67. Secondary batteries (*i.e.*, rechargeable batteries) comprise positive and negative electrodes separated by a separator. These components are immersed in an electrolyte solution allowing electrons to flow from negative electrode to positive electrode, thereby providing electricity. An external power source recharges the battery by moving electrons from the positive electrode back to the negative electrode. Lithium, the lightest metal in the periodic table, is well-suited for use in such positive electrodes because of its electropositive quality—*i.e.*, ease of losing valence electrons. Many electronic devices (mobile phones, laptop computers, medical equipment) were already powered by lithium-ion batteries at the time of the alleged invention.

It is a well-known problem that lithium-ion positive electrode particles lose capacity (*i.e.*, ability to hold charge) after repeated charging cycles. With each charge and discharge, the crystal structure of the positive electrode may crack or break. Even while not in use, rechargeable batteries experience some loss of capacity

because the surface of the positive electrode active material (“PEAM”) reacts with the electrolyte solution. The ’828 Patent purports to address this known problem. However, prior well-known solutions already existed: adding certain elements to PEAM particles stabilizes them and improves the battery’s long-term capacitance. The ’828 Patent claims just that: using well-known elements to stabilize PEAM particles in a known manner.

As demonstrated herein, the purportedly novel aspects of the ’828 Patent were well-known in the prior art and practiced by persons of skill in the art (“POSITA”) before the claimed invention. References and combinations presented in this Petition showing that the Challenged Claims are unpatentable as obvious were not considered by the Examiner during prosecution. Petitioner respectfully requests that the Challenged Claims be canceled.

II. MANDATORY NOTICES UNDER 37 C.F.R. §42.8(b)

A. Real Party-In-Interest

Amperex Technology Ltd. is the real party-in-interest for Petitioner.

B. Related Matters

Amperex Technology Ltd. has filed an action for declaratory judgment of non-infringement of the ’828 Patent, in the United States District Court for the Eastern District of Virginia (the “Virginia Litigation”). *See Amperex Technology Ltd. v. Semiconductor Energy Laboratory Co., Ltd.*, No. 1:23-cv-272. That matter

may affect, or be affected by, decisions in this proceeding.

C. Counsel

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D. Service Information

Petitioner concurrently submits a Power of Attorney, 37 C.F.R. §42.10(b), and consents to electronic service by email at ATL_IPR@kirkland.com.

III. PAYMENT OF FEES UNDER 37 C.F.R. §42.103

The undersigned authorizes the U.S. Patent and Trademark Office to charge the fee set forth in 37 C.F.R. §42.15(a)(1) for this Petition to Deposit Account No. 506092. Review of all eight claims is requested. The undersigned further authorizes payment for any additional fees that may be due in connection with this Petition, to be charged to the above-referenced deposit account.

IV. CERTIFICATION OF STANDING UNDER 37 C.F.R. §42.104(a)

Petitioner certifies that the '828 Patent is available for IPR. Petitioner is not barred or estopped from requesting IPR on the grounds identified herein. Neither Petitioner, nor any party-in-privity with Petitioner: (1) is the owner of the '828 Patent; (2) has filed a civil action challenging the validity of the '828 Patent; (3) has been served a complaint alleging infringement of the '828 Patent more than one year prior to the present date; or (4) is estopped from challenging the claims on the grounds identified in the petition. The '828 Patent has not been the subject of a prior IPR or a finally-concluded district court litigation.

V. OVERVIEW OF CHALLENGE AND RELIEF REQUESTED

A. Prior Art References

Petitioner's challenge is based on the following prior-art references.

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

2. Hye Jin Lee & Yong Joon Park, *Interface characterization of MgF₂-coated LiCoO₂ thin films*, Solid State Ionics 230, 86–91 (2013) (“**Lee**”) (Ex. 1006) was published in 2013, and is prior art under AIA 35 U.S.C. § 102(a)(1)

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

4. U.S. Patent 9,105,926 (“**Fujiki**”) (Ex. 1008) was filed on July 13, 2010, published on January 27, 2011, and issued on August 11, 2015, and is prior art under AIA 35 U.S.C. § 102(a)(2).

The above prior-art references predate the ’828 Patent—which claims priority to a foreign application filed on July 5, 2016—and were not considered by the Examiner during prosecution. *See generally* Ex. 1002. Even assuming (solely for purposes of this IPR) that the ’828 Patent is entitled to its earliest priority date of July 5, 2016, the above references are prior art. *See also* Ex. 1033 (Babbitt Decl.).

B. Relief Requested

Petitioner requests cancellation of the Challenged Claims as unpatentable under 35 U.S.C. §103. The specific grounds of the challenge are set forth below, supported by the declaration of Dr. Troy Hayes (Ex. 1003).

Ground	Claims	Proposed Statutory Rejection
I	1, 4, 5, 8	Obvious over Shim, either alone or in combination with Lee
II	2, 6	Obvious over Shim (either alone or in combination with Lee) in combination with Yu

III	3, 7	Obvious over Shim in combination with Lee
IV	1, 4, 5, 8	Obvious over Fujiki
V	2, 6	Obvious over Fujiki in combination with Yu
VI	3, 7	Obvious over Fujiki in combination with Lee

VI. DISCRETIONARY DENIAL IS NOT APPROPRIATE HERE

A. The '828 Patent Has Not Been Subject to a Prior Petition

The '828 Patent has not been subject to any prior IPR or PGR petitions. Thus, this is not a “follow-on” petition and there is no basis for the Board to exercise its discretion under 35 U.S.C. §314(a) and 37 C.F.R. §42.108(a). *Gen. Plastic Indus. Co. v. Canon Kubushiki Kaisha*, IPR2016-01357, Paper 19 (P.T.A.B. Sept. 6, 2017).

Petitioner has filed only a single petition challenging the claims of the '828 Patent, avoiding any suggestion that Petitioner has placed a substantial and unnecessary burden on the Board. *See* Trial Practice Guide Update (July 2019).

B. The Presented Grounds and Argument Are Dissimilar to the Art and Arguments Previously Presented to the Office

All factors considered by the Board under 35 U.S.C. §325(d) weigh in favor of institution. *Becton, Dickinson, & Co. v. B. Braun Melsungen AG*, IPR2017-01586, Paper 8 (P.T.A.B. Dec. 15, 2017); *see also Advanced Bionics, LLC v. Med-El Elektromedizinische Geräte GmbH*, IPR2019-01469, Paper 6 at 8 (P.T.A.B. Feb. 13, 2020). The Board has consistently “held that a reference that ‘was neither applied against the claims nor discussed by the Examiner’ does not weigh in favor of

exercising [] discretion under §325(d).” *Fasteners for Retail, Inc. v. RTC Indus., Inc.*, IPR2019-00994, Paper 9 at 7–11 (P.T.A.B. Nov. 5, 2019). The grounds presented herein include obviousness challenges applying Shim and Fujiki as base references. Neither of these references was applied against the Challenged Claims or discussed by the Examiner during prosecution of the ’828 Patent (nor were combinations thereof). Shim was made of record in a post-allowance IDS citing 175 references, but was not applied by the Examiner in any Office Action. The grounds are not cumulative of the references applied during examination because they rely on prior art covering disclosures not found in the references applied during examination. *Bowtech Inc. v. MCP IP, LLC*, IPR2019-00383, Paper 14 at 5 (P.T.A.B. Aug. 6, 2019) (instituting petition where the Examiner did not consider grounds asserted by the petition). To the extent that the Board considers Shim “previously addressed” by the Examiner under § 325(d), it was error for the Examiner to overlook the buried reference. The Examiner did not have the benefit of expert testimony explaining why a POSITA would have understood that the degradation discussed by Shim refers to cracks (*i.e.*, structural defects) within the cathode structure. Ex. 1003, ¶¶ 147–49.

C. The *Fintiv* Factors Favor Institution

The *Fintiv* factors weigh in favor of institution. *See Apple Inc. v. Fintiv, Inc.*, IPR2020-00019, Paper 11 at 6 (PTAB Mar. 20, 2020).

Factor 4 (overlap between issues) weighs in favor of institution. The Virginia Litigation seeks a declaratory of judgment of non-infringement regarding the '828 Patent and does not raise any validity issues. Thus, there is no overlap in issues between this petition and the parallel proceeding.

Factor 1 (existence or likelihood of a stay) is neutral. No evidence suggests a stay will not be granted post-institution. The district court proceedings are in their initial stages with no scheduling order, trial date, or response to the complaint.

Factor 2 (proximity of trial date to IPR statutory deadline) weighs in favor of institution. The district court has not set a trial date and has not set any deadlines in the case. Defendant's responsive pleading (due June 2023) has not yet been entered. The latest Federal Court Management Statistics (from March 2023) indicate the median time from filing to trial in E.D. Va. is 23.2 months. Thus, trial in the Virginia Litigation will likely be later than the projected statutory deadline for a final written decision (December 2024).

Factor 3 (investment in parallel proceeding by court and parties) weighs in favor of institution. As explained above, the parties have invested minimal resources thus far in the parallel district court litigation. Petitioner has diligently filed this petition approximately three months after the Complaint in the Virginia Litigation. Institution of this Petition would reduce the substantial expenses that would result from the related litigation.

Factor 5 (same parties): In view of the other *Fintiv* factors, the parties' identities is of limited weight.

Factor 6 (other circumstances impacting discretion) favors institution, as this Petition presents compelling evidence of unpatentability. Considering the guidance set forth in the *Fintiv* Memorandum at 2–4, this IPR should be instituted.

D. There is Compelling Evidence of Unpatentability

In a June 21, 2022 Memorandum to the Board, the USPTO Director stated that the Board “will not rely on the *Fintiv* factors to discretionarily deny institution . . . where a petition presents compelling evidence of unpatentability,” which “would plainly lead to a conclusion that one or more claims are unpatentable by a preponderance of the evidence.” As set forth below, this Petition meets those criteria.

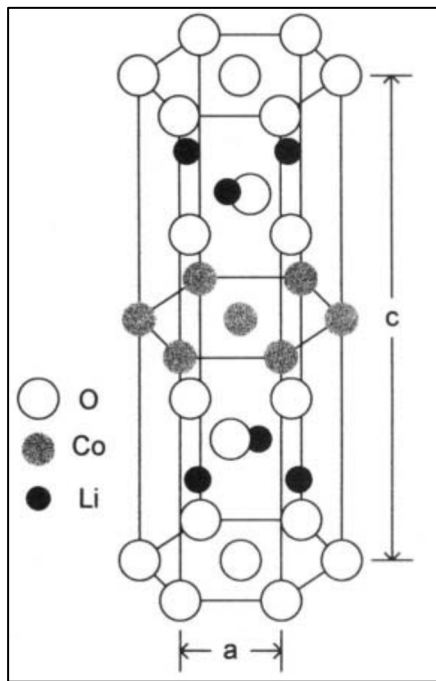
VII. TECHNOLOGY OVERVIEW

A. Structural Defects within Cathodes

A lithium-ion battery has four basic components: a negative electrode (anode), a positive electrode (cathode), a separator between them, and an electrolyte. Lithium-ion batteries typically use microparticles of transition metal oxides, like lithium cobalt oxide (LiCoO₂), as the cathode active material and graphite microparticles as the anode active material. The separator, a thin polymer membrane, allows the migration of lithium ions toward the anode or cathode during battery

operation. Electrolytes are commonly mixtures of solvents and a dissolved lithium salt. Ex. 1003, ¶ 37.

A representation of the atomic structure of LiCoO_2 is shown below:



Ex. 1009, 143. In the cathode, lithium atoms reside within oxygen atoms of the metal oxide. While the battery charges, lithium atoms are removed and proceed into the anode through the electrolyte. During discharge, lithium ions return to the cathode by the reverse electrochemical process. Ex. 1003, ¶ 38.

Additional chemical reactions take place during the manufacturing process. The battery is subjected to “formation cycles,” which are slow and well-controlled charge and discharge cycles where the electrolyte (and additives) form a protective layer on the cathode: the cathode solid electrolyte interphase (“CSEI”). The CSEI

helps protect the electrode active material while allowing lithium-ion transport. Ex. 1003, ¶¶ 40–41.

Cracks in electrode particles are a natural result of the battery’s charging and discharging process. With each such cycle, cathode particles swell and contract due to the insertion and removal of lithium ions. This process causes stress to the crystal structure of the cathode active materials, causing cracks, crevices, fissures, breaks, and/or defects to form in the crystal, CSEI, and other protective films. Ex. 1003, ¶ 44; Ex. 1010 (Wang), 1–8.

B. Addressing Structural Defects in Cathodes

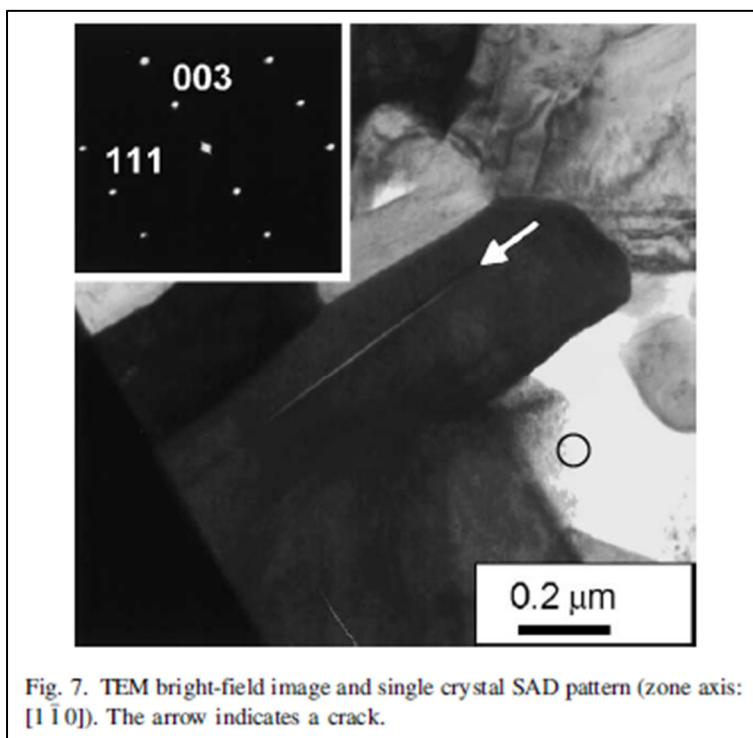
There was (and remains) a continuing desire to make batteries operate at a higher voltage. But charging a battery to a point where more than 50% of lithium is removed from LiCoO_2 decreases the battery’s stability, resulting in increased susceptibility to degradation and failures. But small additions of other elements into the cathode active material stabilize its structure, enabling higher voltage in the battery overall and improving battery safety. Ex. 1003, ¶¶ 46–48.

For example, small additions (“doping”) of magnesium, aluminum, and titanium were known to stabilize the structure of LiCoO_2 . *See* Ex. 1011, 1, 8–9; Ex. 1012, 1–5. Magnesium can be doped into the LiCoO_2 lattice to increase the electronic conductivity of the material. *See* Ex. 1012, 1–5; Ex. 1013. The use of titanium was also well known to improve cathode structural stability by reducing the

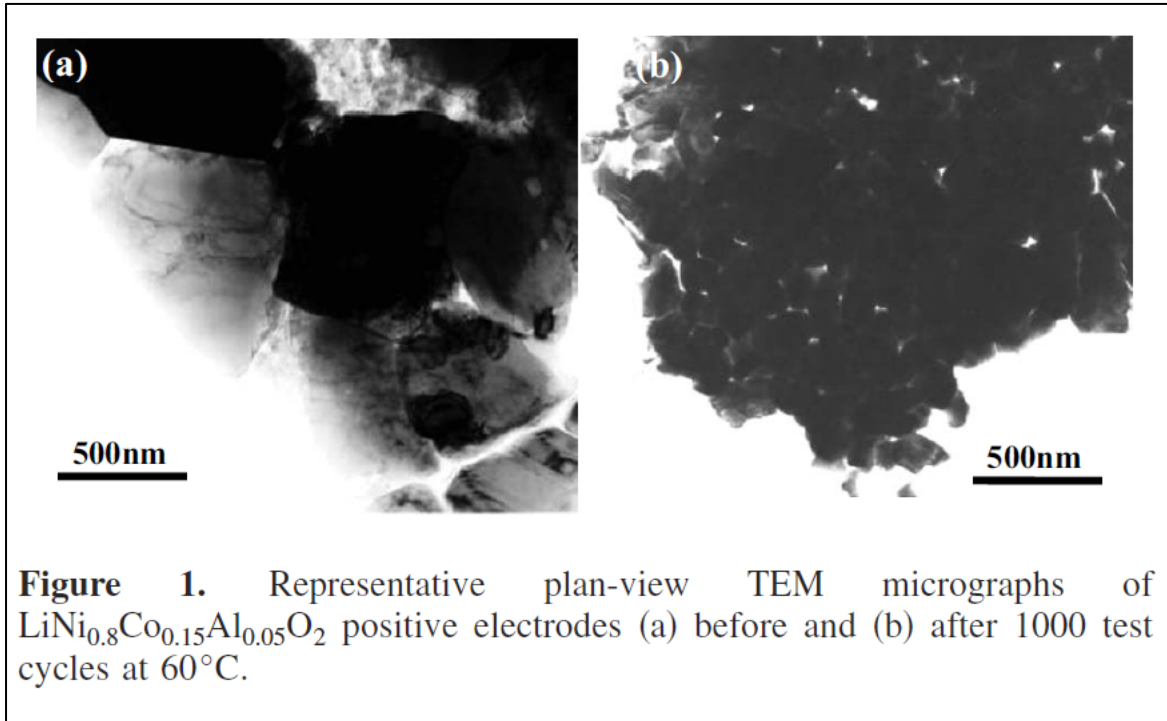
structural distortions that occur during delithiation (*i.e.*, removal of lithium from an electrode in a battery). *See* Ex. 1013, 2.

C. Observing Structural Defects

Cracks and defects in the cathode material structure can be observed via a transmission electron microscopy (“TEM”), a commonplace technology—first demonstrated in the 1930s and made commercially available in the 1940s—in which a beam of electrons is transmitted through a specimen to form an image. Ex. 1010, 3; Ex. 1014, Abstract (“Fracture of grains was observed by TEM in the cathode layer”); Ex. 1015, Fig. 8; Ex. 1026, 2 (TEM image showing “fragment[s]” or “crack[s]” in cycled positive electrode grains); Ex. 1003, ¶¶ 62–63. A TEM image can highlight cracks or defects in the cathode active material particle:

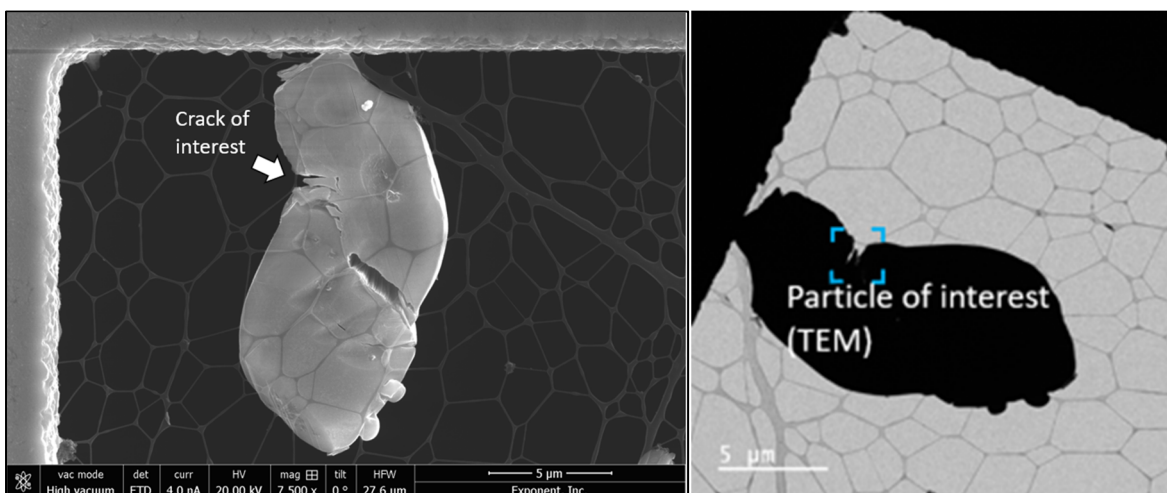


Ex. 1014, 3–4, Fig. 7 (TEM image of a fractured grain with a crack in the crystal pattern).

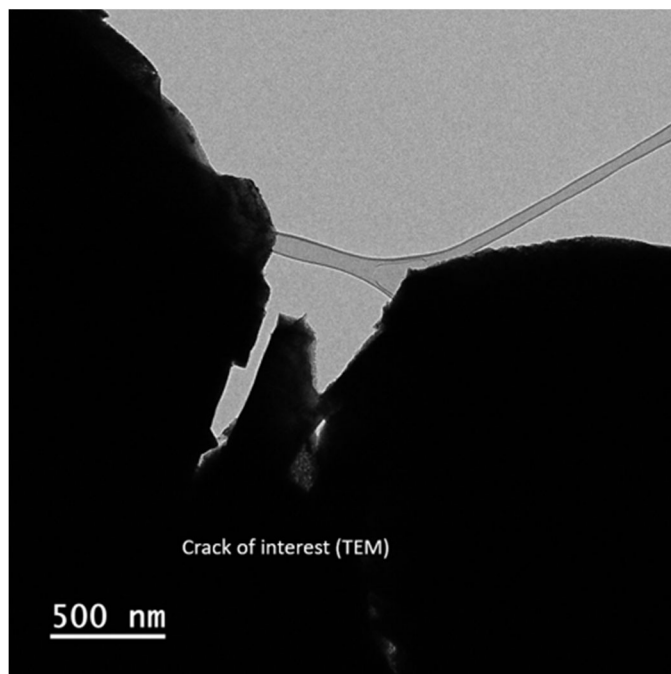


Ex. 1026, 2, Fig. 1 (TEM images showing that “grains were fragmented or cracked”).

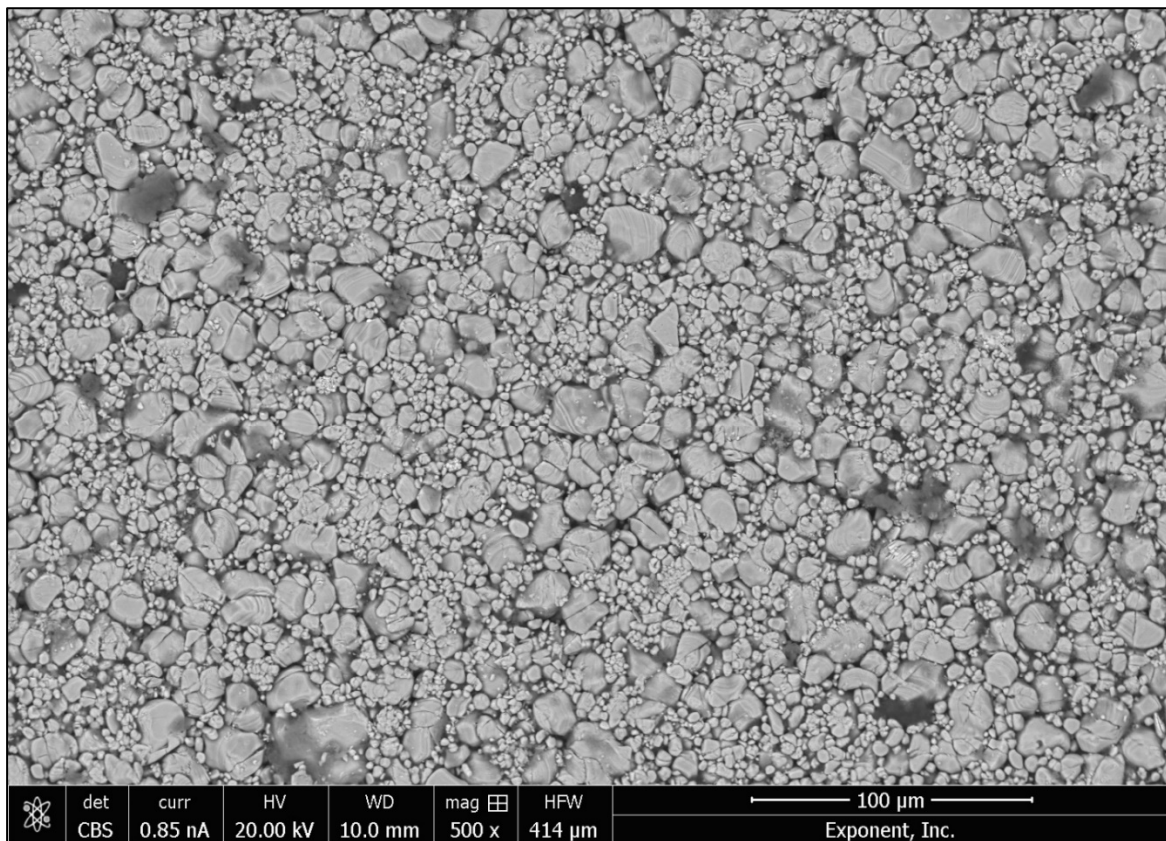
As shown below, Dr. Hayes’s testing confirms that a POSITA would have observed numerous crack portions in PEAM particles using TEM:



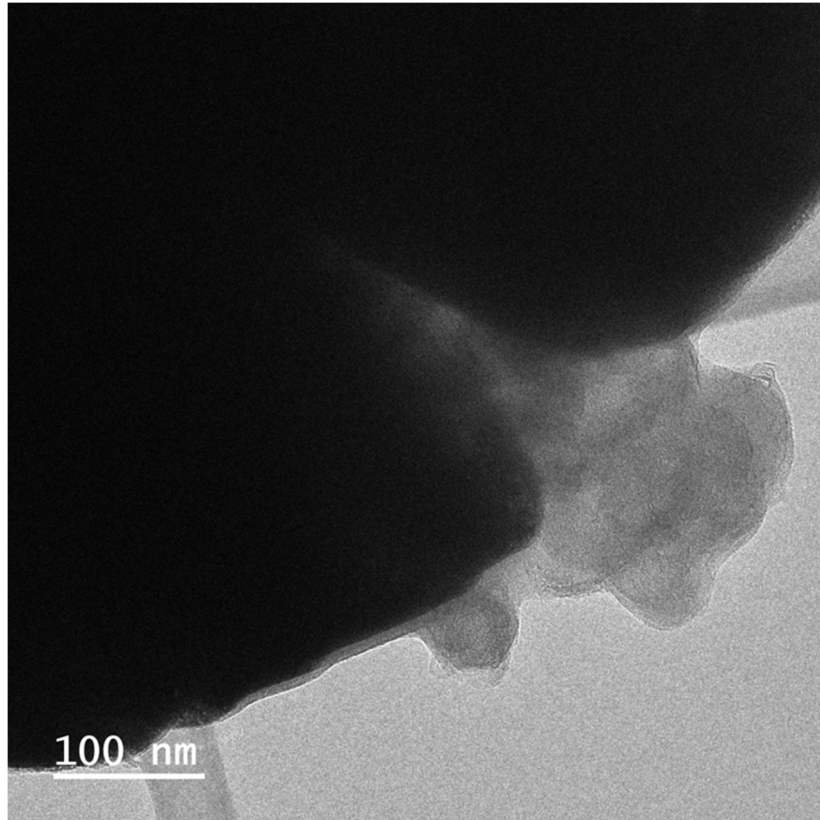
Ex. 1003, ¶¶ 67–68 (SEM (left) and TEM (right) images showing crack in PEAM particle from Samsung Galaxy A8 battery sample dated August 7, 2015).



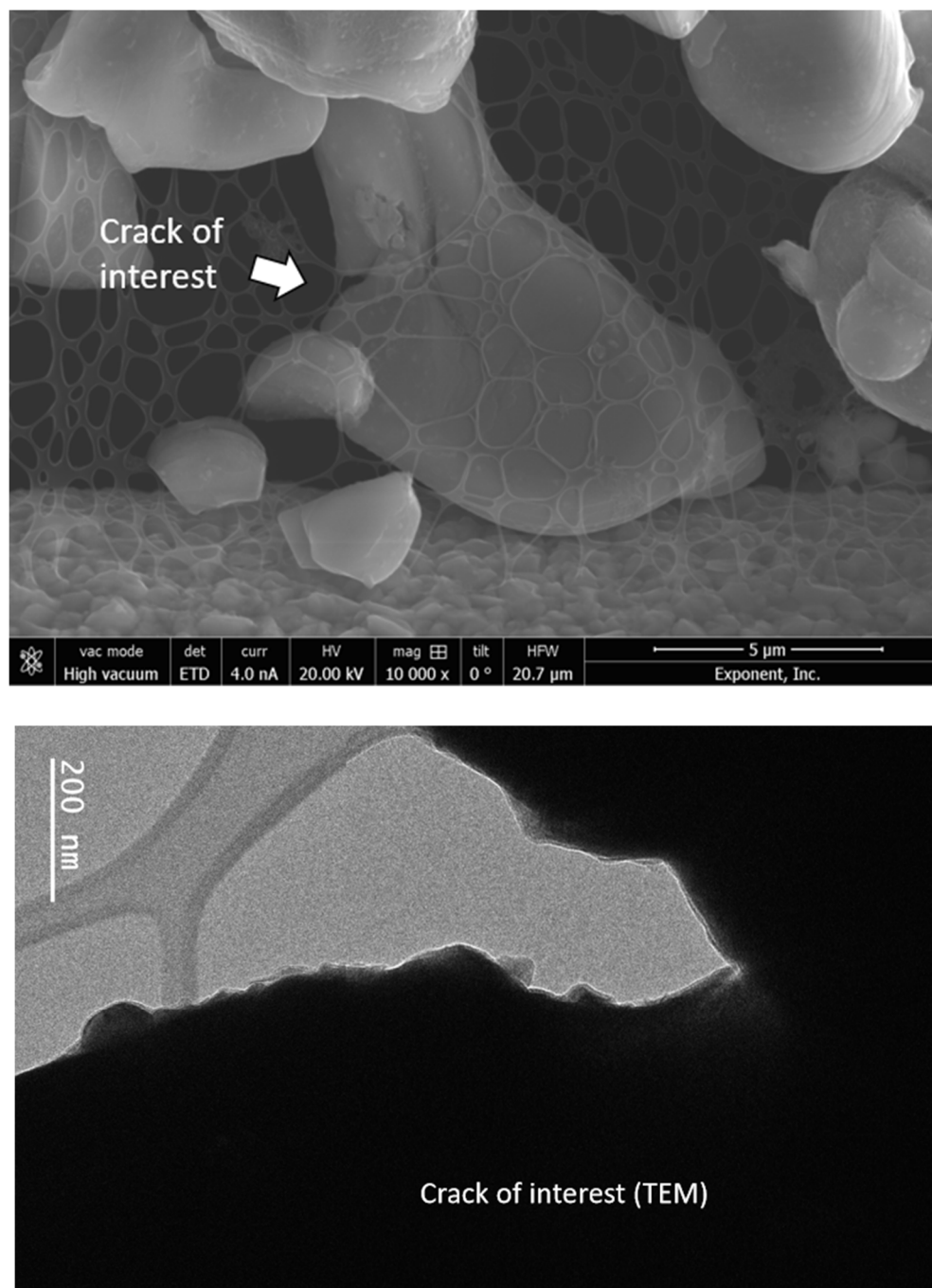
Id., ¶ 68 (TEM image showing crack in PEAM particle from same sample).



Id., ¶ 66 (SEM image of PEAM particles with widespread cracks from same sample).



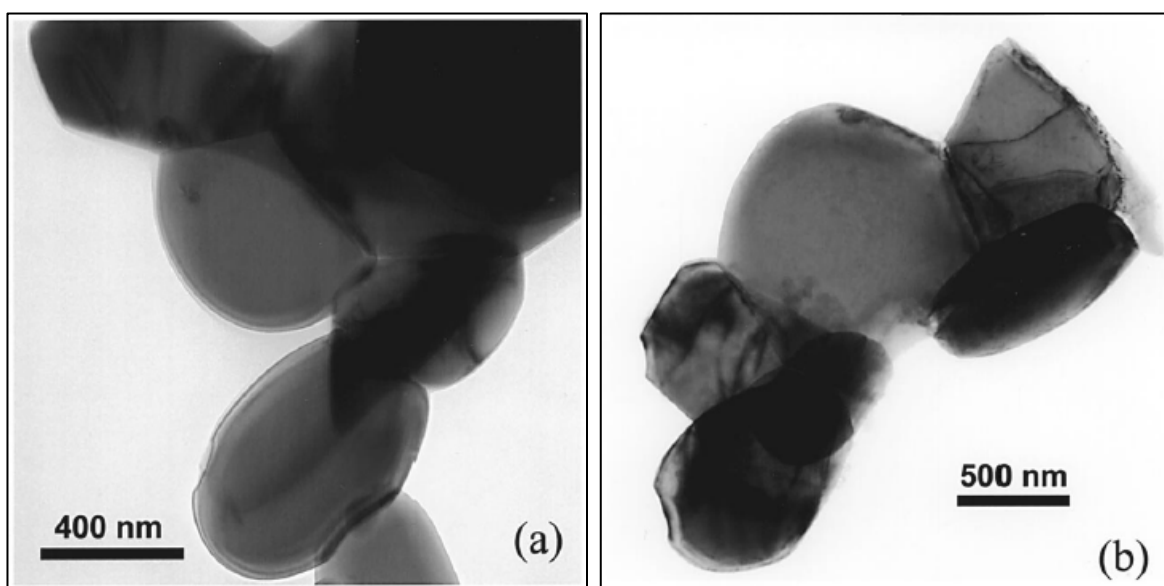
Id., ¶ 75 (TEM image showing crack in PEAM particle from iPhone 5S battery sample dated October 2013).



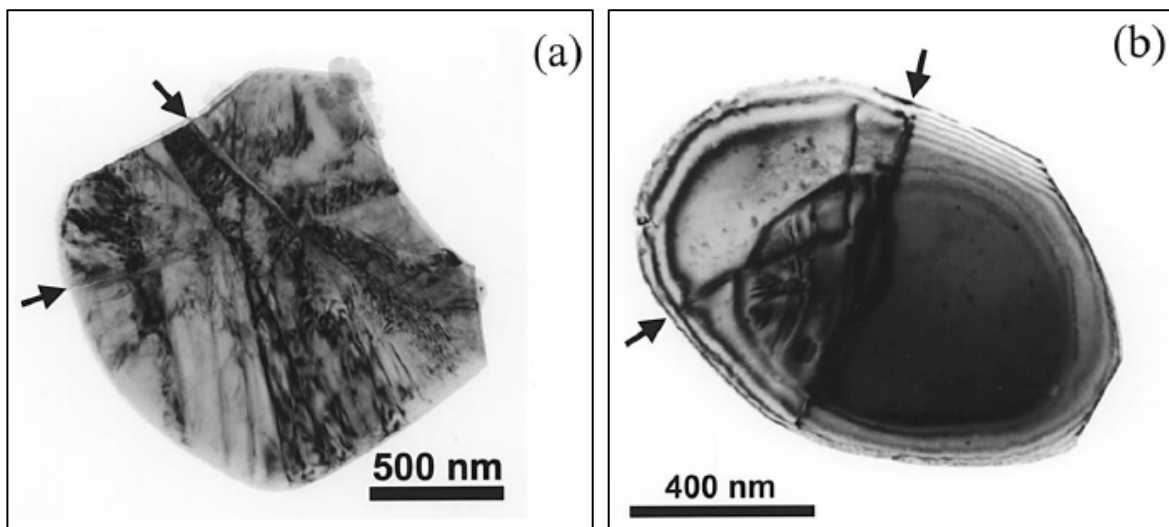
Id., ¶ 74 (SEM (top) and TEM (bottom) images showing crack in PEAM particle from Samsung Galaxy Note 4 battery sample dated April 1, 2016).

Additionally, Wang studied particle-scale damage to LiCoO_2 cathodes caused by electrochemical cycling. Ex. 1010, Abstract. Using TEM imaging, Wang

concluded that LiCoO_2 cathode particles are subject to varying degrees of damage, including fracture. *Id.* Specifically, Wang found that post-cycling, 20–50% of LiCoO_2 particles experienced structural damage, including “microcracks.” *Id.*, 3. Wang used TEM imaging to capture LiCoO_2 particles before electrochemical cycling (below left) and after (below right):



Ex. 1010, 4. As shown below, Wang’s TEM images show LiCoO_2 particles with widespread cracking. *Id.*, 3–4.

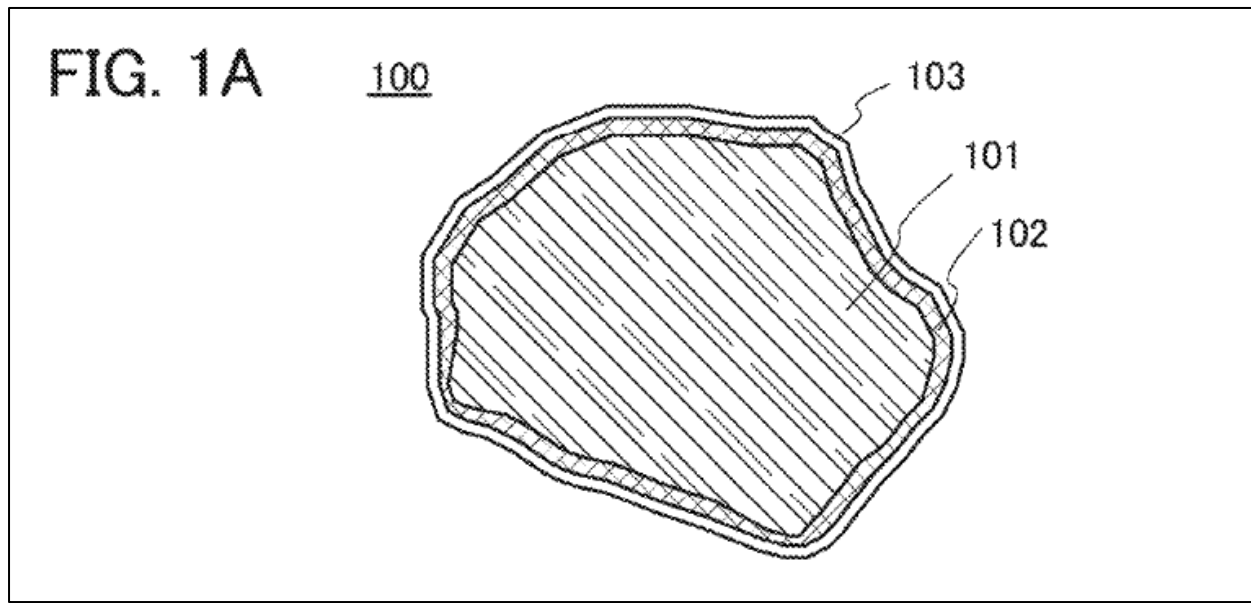


Id., Fig. 5(b) (arrows indicating cracks). Wang’s findings are applicable to LiCoO_2 generally, as “[e]lectrochemical cycling creates a variety of defects in the LiCoO_2 particles in a *typical composite cathode*” and that “[a]ccumulated damage of this kind may therefore be responsible for degradation in LiCoO_2 -based batteries.” *Id.*, 480; Ex. 1003, ¶ 151.

Similarly, Fey studied cracks vis-à-vis metal oxide coatings on LiCoO_2 cathodes. Ex. 1016, 1. Specifically, Fey explains that “[i]t is inevitable that coated surfaces always have defects such as cracks and pinholes in them” and that these defects “may form and close with the application of a load or upon thermal cycling.” *Id.*, 4. Moreover, Fey teaches that “[i]n the case of the coated cathodes . . . the cycling process could have enabled the *particles of the coating material to become ingrained in the crevices and cracks on the cathode surface.*” *Id.*

D. Cathode Particle Dopants and Coating Elements

As shown by the '828 Patent, a PEAM particle (100) can be divided into the “bulk” (101) and one or more coating layers (102, 103):



Ex. 1001, Fig. 1A.

A coating layer mitigates cathode degradation by providing physical protection from the electrolyte solution. Ex. 1003, ¶ 49; Ex. 1005, 1. It was known in the art that elements like magnesium, titanium, and fluorine could be used in coating layers. *See* Ex. 1017, [0011]–[0013] (TiF, MgF₂, TiF₃, and TiF₄ coatings); Ex. 1018, 2:66–3:15 (same); Ex. 1019, 1:24–31 (disclosing one of “Ti, Zr and Hf” used with one of “F or P” on a PEAM surface); Ex. 1020, [0022]–[0023], [0030] (disclosing oxides of “titanium, silicon, magnesium, and zirconium”); Ex. 1008, 4:48–6:1. Further, it was known that multiple coating layers could be used in a

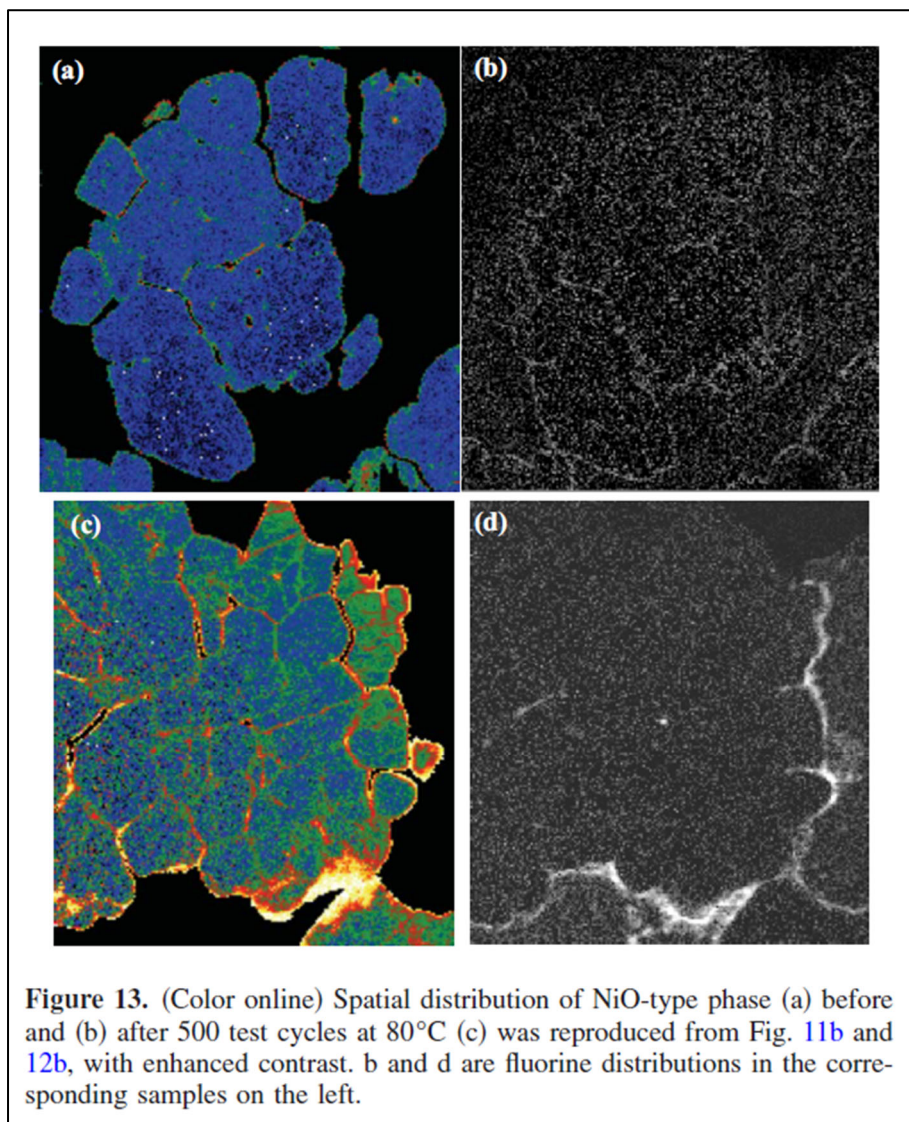
particle. *See* Ex. 1020, [0027]–[0032], [0077] (“[T]he surface layer is arranged on the coating layer”).

Further, it was well known that elements like magnesium and titanium could also be doped (or added) to the bulk of the PEAM to improve cell performance. Ex. 1003, ¶ 48; Ex. 1005, 2; Ex. 1017, [0012] (“M is at least one metal selected from the group consisting of Al, Mg, Fe . . . Ti, and Zr.”); Ex. 1019, 1:24–31.

E. Formation of a Fluorine-Containing Layer

Prior to the ’828 Patent, LiPF_6 was known (and continues) to be a common electrolyte salt for lithium-ion batteries, and it was known that LiPF_6 would lead to the formation of a fluorine-containing layer on the cathode surface. Ex. 1003, ¶ 53.

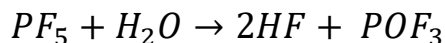
For example, in 2009, Dedryvère’s XPS studies revealed an LiF layer on the cathode surface from the hydrolysis of LiPF_6 . Ex. 1021; *see also* Ex. 1022, 86 (fluorine present “on the surface” of the cathode particle “caused by the formation of LiF derivatives”); Ex. 1024, 1 (“Solid LiF . . . precipitates on the electrode particles[.]”); Ex. 1023, 11. Muto detected fluorine “likely derived from the LiPF_6 electrolyte” “at cracked particle surfaces, which suggests that the electrolyte invaded into the cracked gaps,” as shown below:



Ex. 1026, 6, Fig. 13 (showing fluorine present in crack portions in (b) and (d)).

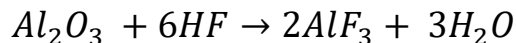
POSITAs were aware that this fluorine-containing layer was beneficial. For example, Carrillo Solano found that such a surface layer “should protect the cathode from reactions with acidic species in the solution.” Ex. 1022, 35; *see also* Ex. 1025, 3 (fluorine-containing solvent “provides an *electrochemically stable surface film* on a LiCoO₂ cathode and *significantly improves the cycle performance* under a high-voltage condition”).

In addition to knowing that electrolytes like LiPF_6 would form a fluorine-containing film on cathodes, POSITAs would have been motivated to fabricate a fluorine-containing coating on the cathodes to protect against hydrofluoric acid (HF) attack. This is because LiPF_6 degrades to form hydrofluoric acid, according to these reactions:



Ex. 1024, 1; *see also* Ex. 1028, 1 (moisture reacts with LiPF_6 to generate HF). It was well understood that hydrofluoric acid detrimentally affects battery performance. *See* Ex. 1003, ¶ 54; Ex. 1028, 1; Ex. 1024, 1–2; Ex. 1005, 2; Ex. 1006, 3–4; Ex. 1023, 11; Ex. 1027, 1.

POSITAs with this concern knew to use metal oxide and metal fluoride coatings. It was understood that metal oxides could scavenge HF to form a metal fluoride layer as indicated below—this was particularly advantageous because such a layer would act as a “permanent physical barrier” to HF:



Ex. 1024, 1–2. Indeed, fluoride-containing coatings “serve[] as a protective layer, preventing direct contact with the reactive electrolyte.” Ex. 1006, 4; *see also id.*, 6; Ex. 1028, 1; Ex. 1029, 2; Ex. 1030, 2; Ex. 1027, 1; Ex. 1003, ¶¶ 55–56.

VIII. THE '828 PATENT

The '828 Patent issued on August 11, 2020, from Application No. 15/638,449 (“’449 Application”), filed on June 30, 2017. It claims priority to several Japanese Applications, the earliest of which was filed on July 5, 2016.

A. Challenged Claims

The '828 Patent has eight claims, all of which are challenged by this Petition.

B. Specification

The '828 Patent admits that lithium-ion batteries had been “actively developed” prior to prosecution. Ex. 1001, 1:32–45. The '828 Patent addresses a well-known mechanism of battery degradation: “[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.*, 4:1–3. The '828 Patent proposes a “coating film” to prevent such deformation, cracking, or breaking. *Id.*, 4:10–12. When the coating film covers a PEAM’s surface, a “decrease in capacity due to charge and discharge cycles can be suppressed.” *Id.*, 3:54–61.

As the '828 Patent admits as background art, such coating films were known. *See id.*, 1:50–53 (“Thus, improvement of a positive electrode active material **has been studied** to increase the cycle performance and the capacity of the lithium ion secondary battery (Patent Document 1 and Patent Document 2).”). In particular, the admitted background art includes JP2012-018914 (“Patent Document 1”), which teaches “[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. 1031, Abstract. Similarly, JP2015-2013432 (“Patent Document 2”) discloses a PEAM with “a ***surface layer*** consisting of a layered structure of lithium cobalt oxide comprising element M (M is Mg and/or Al), and at least a layered structure of lithium cobalt oxide solidified by Ti.” Ex. 1032, Abstract. Further, the ’828 Patent admits that its claimed coating materials were known. Ex. 1001, 22:17–26 (generally known to “synthesiz[e] particles of the composite oxide containing ***lithium, cobalt, magnesium, and fluorine***”).

In view of the robust lithium-battery prior art, the ’828 Patent describes an alleged improvement using different regions inside the PEAM. *Id.*, 2:24–30. For instance, the ’828 describes a PEAM including a first region, preferably an oxide like LiCoO₂; a second region containing a transition metal; and a third region containing a representative element. *Id.*, 2:37–56. These latter two regions may overlap. *Id.*, 2:31–33. If the LiCoO₂ region includes a crystal defect, or crack, the representative element and second transition metal will be present in the crack. *Id.*, 24:24–36.

C. Prosecution History

The application leading to the ’828 Patent was originally filed with 22 claims (none issued). On January 7, 2019, the Examiner issued a Restriction Requirement between claims drawn to a PEAM and claims drawn to a method of forming a

PEAM. Ex. 1002, Vol.4, 785. The Applicant elected claims drawn to a PEAM on March 1, 2019. *Id.*, Vol. 4, 796.

The application received a Non-Final Rejection on March 14, 2019, followed by a Final Rejection on July 12, 2019. Ex. 1002, Vol. 4, 810; Vol. 5, 1122. Despite the Applicant's amendments adding language describing various regions of the PEAM, the Examiner maintained §§ 102 and 103 rejections because certain pending claims were anticipated by Ooyama (Ex. 1020) and obvious over Ooyama in view of Yura (U.S. Pub. No. 2012/0258358), Endo (U.S. Pub. No. 2010/0233542), and/or Takahashi (U.S. Pub. No. 2004/0229123). *Id.*, Vol 4, 810–840; Vol.5, 1122–48. On November 12, 2019, the Applicant filed a Request for Continued Examination, amending certain pending claims, and adding new claims 23–34.

On December 9, 2019, the application received another Non-Final Rejection raising new rejections in view of Yanagihara (WO 2016/017077) and combinations of Yanagihara in view of Kawakami (U.S. Pub. No. 2011/0300441) and/or Endo. Ex. 1002, Vol. 5, 1211. On February 14, 2020, the Applicant cancelled previously pending claims 1–34 and added new claims 35–90, including those that eventually issued in the '828 Patent. Ex. 1002, Vol. 5, 1269–82. In another Final Rejection dated March 2, 2020, the Examiner rejected the majority of the pending claims, but allowed pending claims 56–63 were because the closest prior art, Liu (Ex. 1019), purportedly did not disclose “the distinguishing feature(s)” of “[a] positive electrode

active material layer for a lithium-ion secondary battery, the positive electrode active material layer comprising a positive electrode active material particle comprising a representative element and fluorine present in a portion that includes crystal defects or a crack portion in the positive electrode active material particle.” *Id.*, Vol. 5, 1310. In response, the Applicant canceled the rejected claims and amended the allowed claims to additionally include “observed from a TEM image.” *Id.*, Vol. 5, 1332–36.

D. Person of Ordinary Skill in the Art

A POSITA at the time of the ’828 Patent would have at least a bachelor’s degree in chemistry, chemical engineering, or materials science and at least five years of experience in the field of batteries and battery materials. Individuals with different education and additional experience could still be of ordinary skill in the art if that additional experience compensates for a deficit in their education and experience. Ex. 1003, ¶¶ 83–91.

IX. CLAIM CONSTRUCTION

For the purposes of this proceeding only, terms in the challenged claims should be given their plain and ordinary meaning considering the specification and no explicit construction is needed except for the term addressed below.

A. Crystal Defect (Claim 5)

Petitioner proposes “crystal defect” be construed according to the express and unambiguous definition provided by the ’828 Patent:

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

Ex. 1001, 9:55–58.

Should the Board disagree that the '828 Patent defined “crystal defect,” the ordinary meaning of “crystal defect” as understood by a POSITA at the time of the purported invention should apply. *Phillips*, 415 F.3d at 1303. As Dr. Hayes explains, a POSITA would have understood a crystal defect to be imperfection or interruption of regular arrangement of atoms in a crystalline solid. Ex. 1003, ¶¶ 92–93. The prior art renders the challenged claims obvious under either construction.

X. PRIOR ART

A. Shim

Shim is directed to magnesium-doped PEAMs with lithium titanate (LTO) coatings. Ex. 1005, 1. Shim recognizes several degradation mechanisms of layered cathode materials in lithium-ion batteries, including unwanted chemical reactions between electrolytes and PEAMs. *Id.* Shim teaches an LTO coating to provide physical protection from such degradation. *Id.*, 2.

Shim further teaches doping the active material (LiCoO₂) with magnesium. *Id.* Through doping, magnesium is present throughout the bulk of the active material, and this Mg-doped LiCoO₂ exhibits higher electrical conductivity compared to bare

LiCoO₂. *Id.*, 7. Shim concludes that using magnesium and titanium together contributes to improved electrochemical performance. *Id.*, 8.

B. Lee

Lee is directed to using magnesium fluoride (MgF₂) as a coating for a LiCoO₂ battery cathode. Lee explains “it is well known that acid” (*e.g.*, HF) forms from LiPF₆ degradation in the electrolyte, and that this acid “attacks the surface of the cathode during cycling,” which in turn leads to degradation within the cathode. Ex. 1006, at 3–4. To combat this problem, Lee recognizes that “when the surface of the cathode is covered with a coating layer, the coating material serves as a protective layer, preventing direct contact with the reactive electrolyte.” *Id.*, 4. Lee explains that it was known to use “oxides” or “fluorides” as such coatings, which shield LiCoO₂ from HF. *Id.*, 1, 4–6.

C. Yu

Yu is directed to “partial doping of [LiCoO₂] with metal ions” for “extended cyclability and increase in capacity of lithium-ion cells by enhancing structural stability,” finding that by “introducing a quite small amount of TiO₂ during preparation, the performance of the LiCoO₂ material is improved effectively.” Ex. 1007, 1, 4. Yu characterizes LiCoO₂ materials with varying concentrations of titanium. *Id.*, 1–2. For example, as shown below in Table 3, Yu measures Ti/Co

concentrations of a PEAM at different sputtering times, which correspond to different depths of the PEAM sample:

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

Id., 3, Table 3 (annotated). As shown above, the Ti:Co concentration ratio at 0 seconds of sputtering (*i.e.*, at the surface of the tested sample) is 8.49%. *See id.*

D. Fujiki

Fujiki is directed to improving the capacity retention of lithium-ion batteries with various PEAM compositions and coatings to improve the stability of lithium-ion positive electrodes. Ex. 1008, 4:35–56. These PEAMs contain transition metal A, which can be cobalt (*id.*, 4:48–67); surface element M1, which can be titanium (*id.*, 4:48–56, 5:6–28); and metallic element/compound M2 as a PEAM coating to further stabilize its crystal structure, which can be titanium or a metallic fluoride. *Id.*, 4:48–56, 5:29–42. Fujiki found that using a combination of M1 (on the surface) and M2 (in the coating) resulted in a higher initial capacity and higher capacity retention. *See id.*, 11:46–34:13, 18:61–19:22, 22:28–53, 33:23–27, 33:51–54, 34:8–13, Tables 1–2.

XI. SPECIFIC GROUNDS FOR PETITION UNDER 37 C.F.R. §42.104(B)

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

Shim, either alone or in combination with Lee, renders obvious a lithium-ion secondary battery described by the challenged claims. Ex. 1003, ¶ 121. Lee teaches magnesium fluoride (MgF_2) “as a new coating material to improve the electrochemical properties of the LiCoO_2 film electrode.” Ex. 1006, 1. Lee further teaches that the surface of this particle has a magnesium-cobalt ratio between 0.4 and 1.5. *See id.* As explained below, a POSITA would have been motivated to combine Lee’s teachings with Shim, including by (for example) modifying Shim’s coating to include fluorine or using Lee’s MgF_2 coating with Shim’s particle and lithium titanate coating.

1. Motivation to Combine Shim with Lee

A POSITA would have been motivated to use Lee’s MgF_2 coating with Shim’s particles (for example by modifying Shim’s existing coating or using Lee’s coating with Shim’s particle) for several reasons. **First**, a POSITA would have recognized that Shim and Lee are in the same field of endeavor and directed to the same problem of improving PEAMs for enhanced battery performance. Both references teach the same lithium-ion battery cathode (LiCoO_2) and characterize its properties using similar analytical methods such as galvanostatic cycling and SEM. *See* Ex. 1005, 1, 7; Ex. 1006, 1, 5. Given the references’ complementary teachings,

this combination would have involved routine implementation with no technical risks. Ex. 1003, ¶¶ 122, 128. A POSITA would have had a reasonable expectation of success in combining Lee’s teachings with Shim. It was known that magnesium, titanium, and fluorine could be combined in a coating layer (*e.g.*, titanium fluoride or magnesium fluoride). *See supra*, § VII.D; Ex. 1017, [0011]–[0013] (disclosing TiF, MgF₂, TiF₃, and TiF₄ coatings); Ex. 1018, 2:66–3:15 (similar); Ex. 1019, 1:24–31 (similar); Ex. 1020, [0022]–[0023], [0030]; Ex. 1008, 4:48–6:11. It was also known to use multiple coating layers on the bulk particle, which would have motivated a POSITA to use Lee’s MgF₂ coating with Shim’s lithium titanate-coated particle. *See* Ex. 1020, [0027]–[0032], [0077] (“[T]he surface layer is arranged on the coating layer”); Ex. 1003, ¶ 130.

Second, both references provide express motivations to combine Lee’s teachings with Shim’s particle—including by modifying Shim’s coating to include fluorine or by using Lee’s MgF₂ coating with Shim. *See DyStar Textilfarben GmbH & Co. Deutschland KG v. C.H. Patrick Co.*, 464 F.3d 1356, 1360 (Fed. Cir. 2006) (even an implicit suggestion to combine may satisfy obviousness). Both references discuss using fluoride coatings to address capacity fade due to cathode degradation. *See* Ex. 1005, 2 (“Coating is one of the simplest as well as most effective methods to be considered for minimization of the degradation and consequent capacity fade during cycling.”); Ex. 1006, 1 (“One approach for improving the electrochemical

properties of a cathode is surface modification by coating with stable materials, such as . . . fluorides.”). Further, Shim expressly describes “AlF₃” as a “successful coating material[]” (Ex. 1005, 1), and Lee explains that its MgF₂ coating “was inspired by the fact that other fluorides, such as AlF₃, were very effective in modifying cathode surfaces” and that the “MgF₂ coating layer was very effective in improving the electrochemical properties of the LiCoO₂ film electrode.” Ex. 1006, 1, 6; Ex. 1003, ¶¶ 123–26.

Third, a POSITA would have understood that using a fluorine-containing coating, such as Lee’s MgF₂ layer, in Shim’s particle was a “known technique to improve similar devices [and methods] in the same way” and a “combination of known prior-art elements according to known methods to yield predictable results.” *See KSR Int’l Co. v. Teleflex Inc.*, 550 U.S. 398, 417 (2007). As explained above in Section VII.D, a POSITA would have understood that a fluorine-containing electrolyte (*e.g.*, LiPF₆) degrades to form a fluorine-containing layer on the cathode. *See* Ex. 1024, 1–2. Although this surface film is beneficial (Ex. 1022, 35; Ex. 1025, 3), this degradation can also form detrimental hydrofluoric acid (HF)—as Shim and Lee both recognize. *See* Ex. 1005, 2; Ex. 1006, 3–4; *see also* Ex. 1023, 11; Ex. 1024, 1; Ex. 1027, 1.

A POSITA concerned about HF would have been motivated to use a metal fluoride coating (like Lee’s MgF₂) with Shim’s particle. Specifically, Lee teaches

that “when the surface of the cathode is covered with a coating layer, the coating material serves as a protective layer, preventing direct contact with the reactive electrolyte.” Ex. 1006, 4. Indeed, Lee’s “MgF₂ coating layer seemed to react with the acidic electrolyte and incur damage on cycling instead of the electrode material” and “was effective as a protective layer.” *Id.*, 5–6. A POSITA would have understood it was well-known and desirable to use metal fluorides (such as MgF₂) to protect cathodes from hydrofluoric acid resulting from LiPF₆ degradation. *See* Ex. 1029, 2 (“[F]luorine in the form of metal fluorides such as AlF₃, CaF₂, MgF_[2], LaF₃, CeF₃, FeF₃ is considered to be one of the ***most promising coating materials*** to improve electrochemical performance of cathode, due to . . . preventing the direct contact between lithiated cathode and liquid electrolyte.”); Ex. 1024, 1–2 (fluoride forms a “a permanent physical barrier that restricts penetration of HF”); Ex. 1027, 1; Ex. 1003, ¶¶ 125–27.

2. Independent Claim 1

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

To the extent the preamble is limiting, Shim discloses it. Shim is directed at improving electrochemical performance in lithium-ion batteries and teaches that layered lithium transition metal oxides (like LiCoO₂) are commonly used in PEAMs. Ex. 1005, 1–2. *See* Ex. 1003, ¶¶ 131–32.

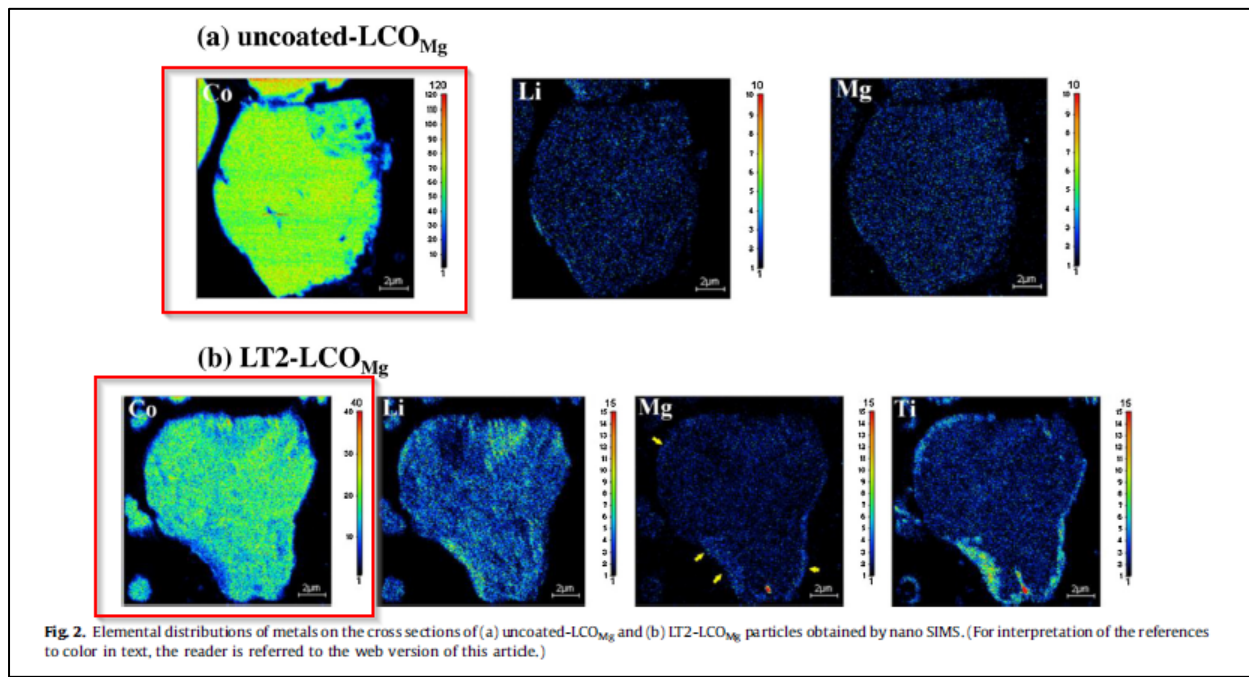
- b. *[Element 1.1]: a positive electrode active material particle comprising*

Shim discloses a PEAM particle—namely a LiCoO_2 particle in a lithium-ion battery. *See* Ex. 1005, 1–2 (specifically disclosing “Mg-doped LiCoO_2 *particles*.”); Ex. 1003, ¶ 133.

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

Shim discloses a PEAM particle comprising cobalt, which the ’828 Patent states is a transition metal. Ex. 1001, 2:53–56. Transition metals are elements found in groups 3–12 of the periodic table, including cobalt. Ex. 1003, ¶ 134.

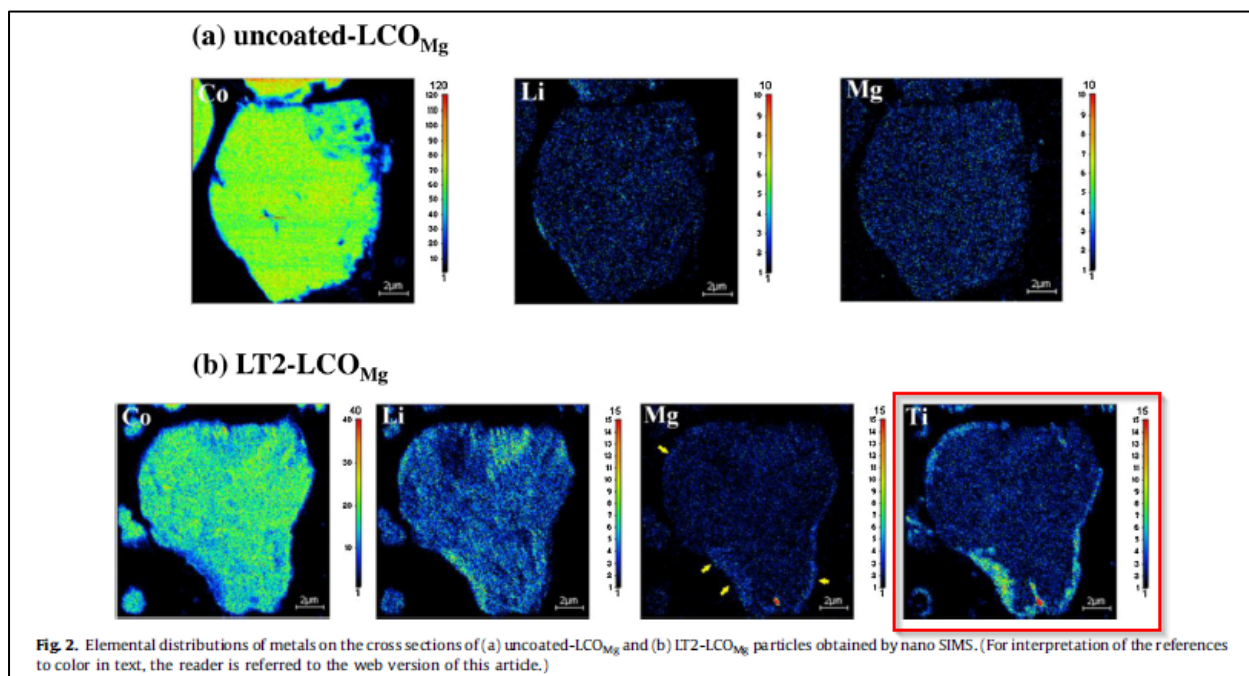
Shim describes PEAMs that comprise “layered lithium *transition metal* oxides” represented as “ LiMO_2 ,” where M consists of “*Co*, Ni, Mn, and so forth.” Ex. 1005, 1. As annotated below, Shim depicts the distribution of cobalt in a PEAM:



Ex. 1005, Fig. 2 (annotated); *see also* Ex. 1003, ¶ 135.

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

Shim discloses that its PEAM particle comprises titanium, which the '828 Patent states is a transition metal. Ex. 1001, 2:53–56. Beyond transitional metal M (described above), Shim further discloses using various forms of lithium titanate ($\text{Li}_x\text{Ti}_y\text{O}_z$) as a coating. *See generally* Ex. 1005. As annotated below, Shim depicts the distribution of titanium in a PEAM:

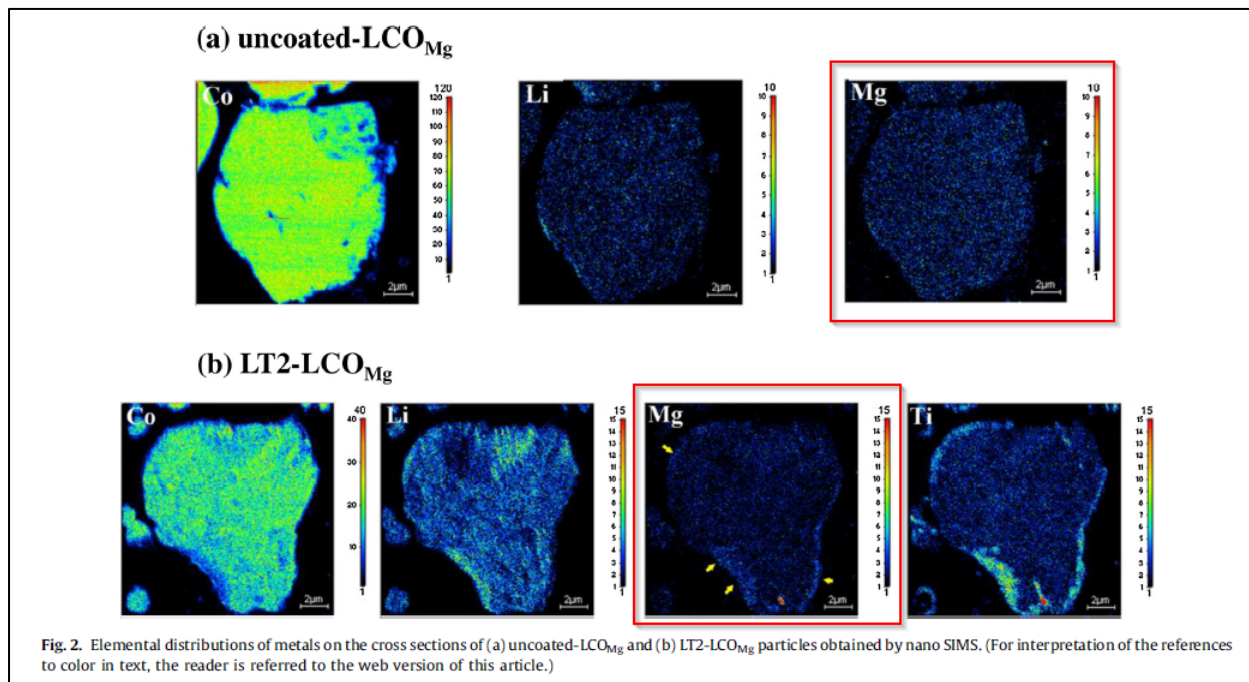


Ex. 1005, Fig. 2 (annotated); *see also* Ex. 1003, ¶¶ 136–38.

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

Shim discloses that its PEAM comprises magnesium, which the '828 Patent expressly states is a representative element. Ex. 1001, 10:29–33, 11:65–12:4, cl. 4. Shim describes PEAM particles doped with magnesium, causing magnesium

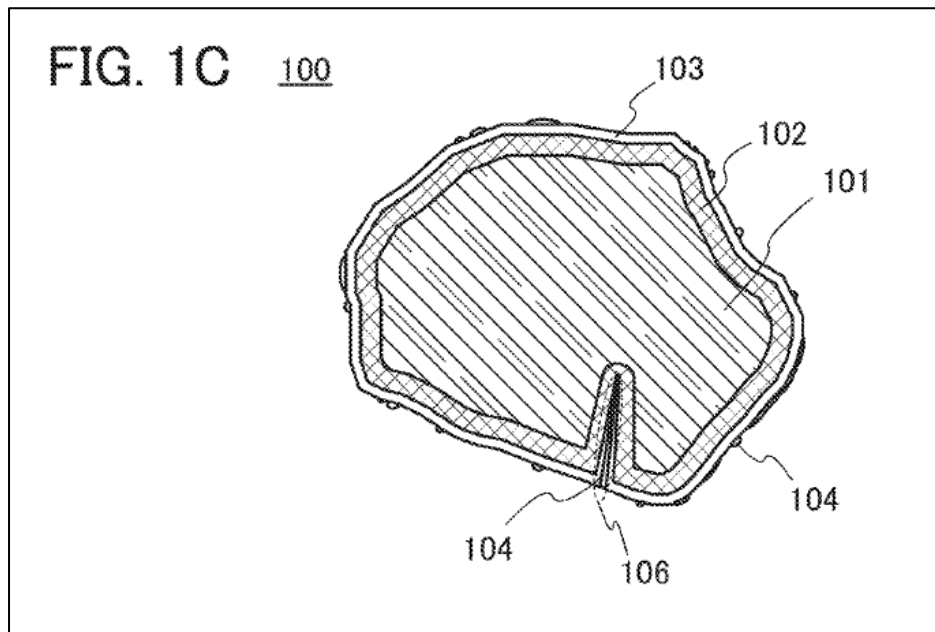
diffusion from the bulk of the particle into the coating layer. Ex. 1005, 2–3. As annotated below, Shim depicts the elemental distribution of magnesium in a PEAM:



Ex. 1005, Fig. 2 (annotated); *see also* Ex. 1003, ¶¶ 139–40.

f. [Element 1.5]: fluorine,

Shim, either alone or in combination with Lee, renders obvious a PEAM particle comprising fluorine. **First**, Shim recognizes that “[c]oating is one of the simplest as well as most effective methods to be considered for minimization of the degradation and consequent capacity fade during cycling,” and identifies aluminum fluoride (AlF₃) as an example of a “successful coating material[.]” Ex. 1005, 1; *see also* Ex. 1008, 6:3–11 (similar use of metallic fluorides). Indeed, a PEAM particle—as used by the ’828 Patent—includes both the bulk and coating:



Ex. 1001, Fig. 1C; *see also id.*, Figs. 1A–B, 9:30–39, 12:45–49 (PEAM 100 includes regions 102 and 103 which are coatings). Thus, Shim renders obvious fluorine used in a PEAM. Ex. 1003, ¶¶ 141–42.

Second, Shim teaches fluorine in the PEAM particle by virtue of its LiPF_6 electrolyte. Ex. 1005, 3. A POSITA would have known that such an electrolyte causes a fluorine-containing film to form on the PEAM’s surface, which is part of the PEAM. Ex. 1003, ¶¶ 143; Ex. 1001, 9:30–30, 12:45–49, Figs. 1A–C.

This film-forming mechanism is well documented in the prior art. Dedryvère found that LiF formed on cathode surfaces from LiPF_6 hydrolysis. Ex. 1021; *see also* Ex. 1023, 12; Ex. 1022, 86; Ex. 1024, 2; Ex. 1026, 6. A POSITA would have further understood that this formation of a fluorine-containing layer from LiPF_6 was beneficial. *See* Ex. 1022, 35 (such surface layers “should protect the cathode from

reactions with acidic species in the solution,” improving the lifespan and capacity of the battery); Ex. 1025, 3 (a fluorine-containing solvents “provide[] an electrochemically stable surface film on a LiCoO_2 cathode and significantly improve[] the cycle performance under a high-voltage condition.”).

Third, to the extent the Board disagrees that Shim alone renders a fluorine-containing coating obvious, a POSITA would have found it obvious to use one—such as Lee’s MgF_2 —with Shim’s particles. As explained above in Section XI.A.1, a POSITA would have been motivated to use MgF_2 to protect Shim’s cathode particles from HF, a degradation byproduct of LiPF_6 . *See* Ex. 1006, 1 (“[T]he MgF_2 coating layer successfully ***protected the surface of the cathode from the reactive electrolyte***, leading to enhanced electrochemical properties for MgF_2 -coated LiCoO_2 films.”). A POSITA would have been especially motivated to combine these references in light of Shim’s express disclosure of LiPF_6 in the electrolyte. *See* Ex. 1005, 3; Ex. 1016, 2; Ex. 1003, ¶¶ 144–45.

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

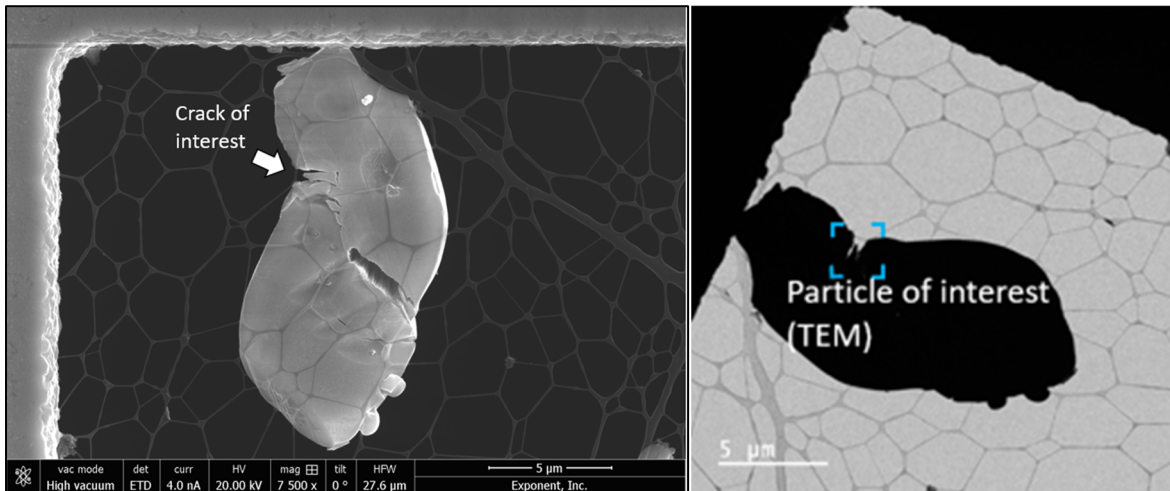
Shim, either alone or in view of Lee, renders this element obvious.

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

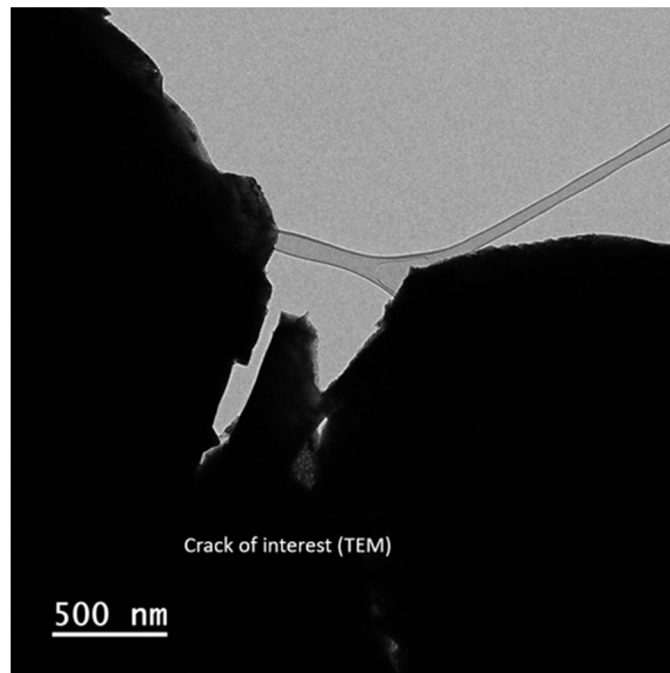
Shim renders obvious a “crack portion” present in its PEAM particles. Shim discloses magnesium-doped PEAM particles with a metal oxide coating with enhanced electrochemical performance with respect to “several different *degradation mechanisms*,” and in particular a “cathode degradation.” Ex. 1005, 1. A POSITA would have understood that the degradation discussed by Shim refers to cracks (*i.e.*, structural defects) within the cathode structure. Ex. 1003, ¶¶ 147–49.

A POSITA would have further understood that such a crack portion within Shim’s PEAM particle would be observable from a TEM image. Shim describes using TEM to examine “structural changes of the materials after charging” (Ex. 1005, 6) and that “TEM images” “demonstrate the irreversible transformation of Li_2TiO_3 and the structure consistency of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ with cycling.” *Id.*, 7, Figs. 5–6. Thus, a POSITA would have understood to use TEM to view structural changes (including cracks) within Shim’s PEAM. Ex. 1003, ¶¶ 150–52.

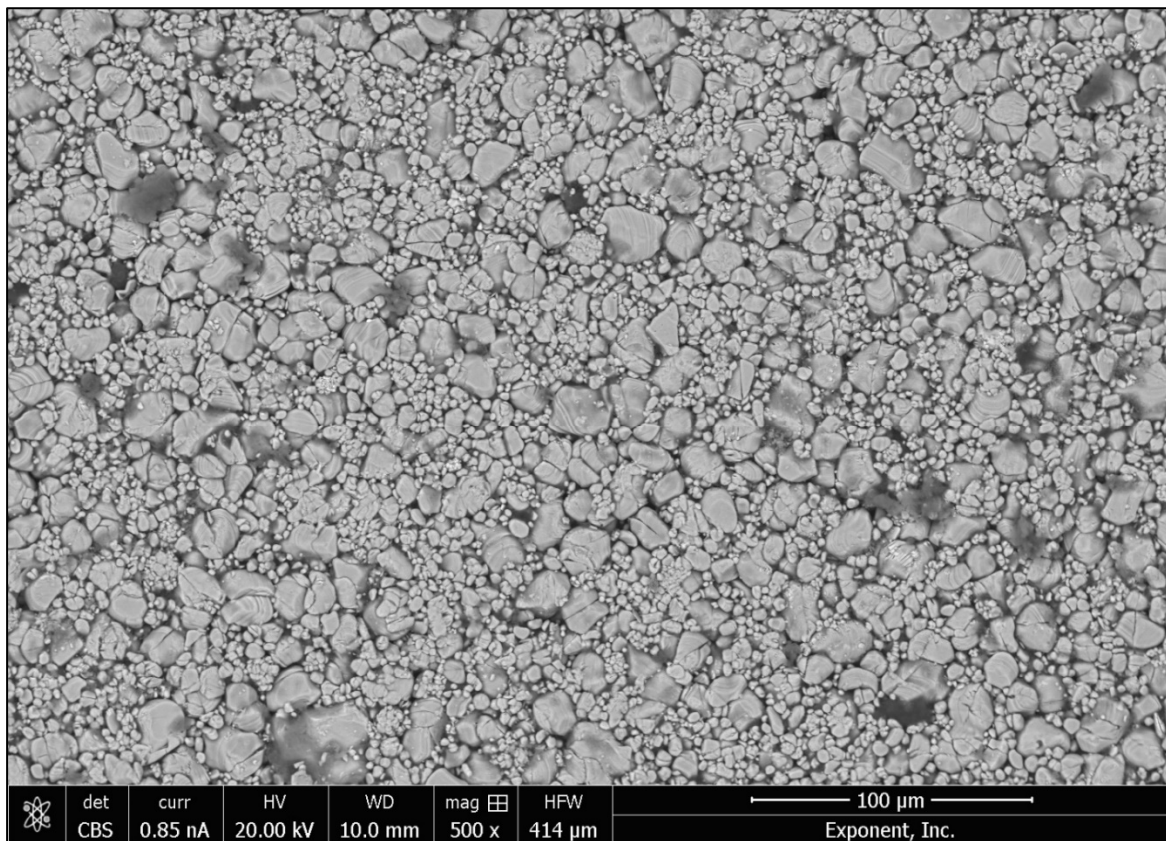
Further, Dr. Hayes tested a variety of commercial mobile phone battery PEAMs using a TEM (all predating the ’828 Patent) to confirm that a POSITA would have observed numerous crack portions in PEAM particles using TEM:



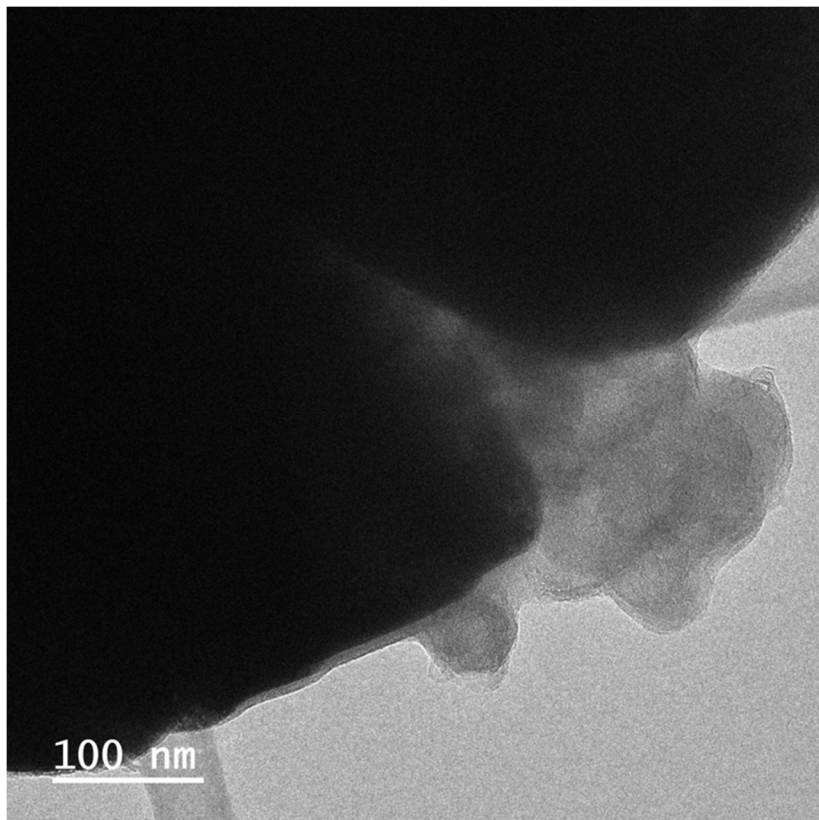
Ex. 1003, ¶¶ 154–55 (SEM (left) and TEM (right) images showing cracks in PEAM particle from Samsung Galaxy A8 battery sample dated August 7, 2015); *see also id.*, ¶ 156.



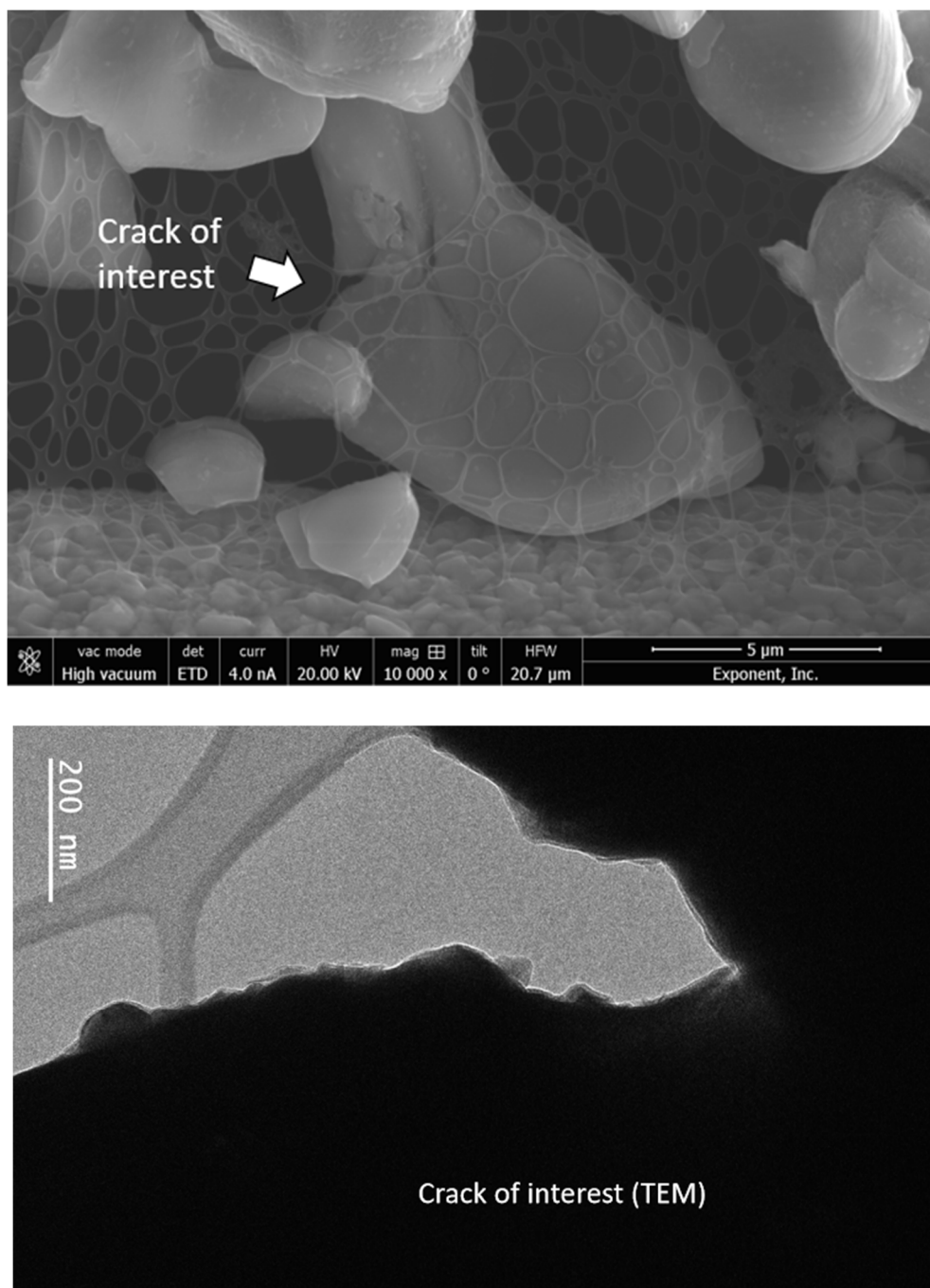
Id., ¶ 155 (TEM image showing crack in PEAM particle from same sample).



Id., ¶ 153 (SEM image of PEAM particles with widespread cracks from same sample).



Id., ¶ 158 (TEM image showing crack in PEAM particle from iPhone 5S battery sample dated October 2013).

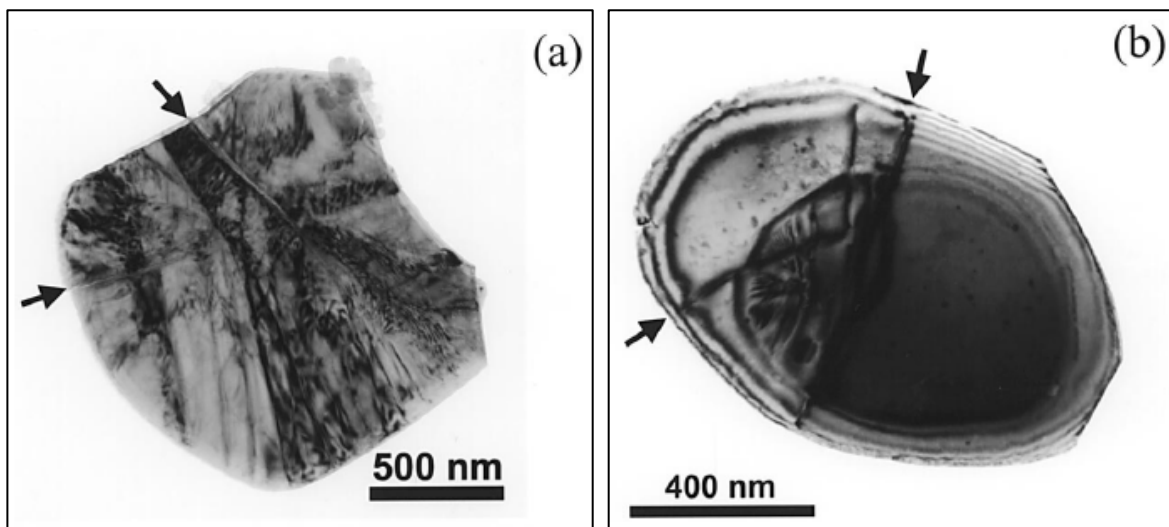
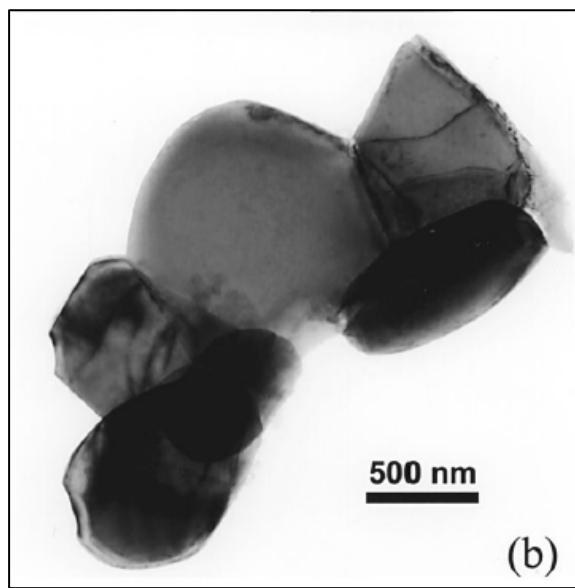


Id., ¶ 157 (SEM (top) and TEM (bottom) images showing crack in PEAM particle from Samsung Galaxy Note 4 battery sample dated April 1, 2016).

To the extent the Board finds Shim does not expressly disclose a crack portion in the PEAM particle observable from a TEM image, a POSITA would have found

it obvious. Specifically, a POSITA would have known that crack portions in PEAMs (like Shim's) were inevitable and could be observed using TEM imaging. For example, Fey (published 10 years before Shim) teaches "[i]t is *inevitable* that coated surfaces always have defects such as *cracks* and pinholes in them" and that such cracks "may form and close with the application of a load or upon thermal *cycling*." Ex. 1016, 4. Similarly, Wang (published 16 years before Shim) teaches "[e]lectrochemical cycling creates a variety of defects in the LiCoO₂ particles in a *typical composite cathode*." Ex. 1010, 8. Thus, a POSITA would have understood that Shim's PEAM degradation caused by cycling (Ex. 1005, 1) refers to cracks. Ex. 1003, ¶ 151.

Moreover, a POSITA would have understood that cracks in Shim's PEAM particles can be observed using TEM imaging. Fey explains that TEM can be used to "examin[e] . . . the microstructure of the coated particles," indicating that the cracks are observable by TEM. Ex. 1016, 1, Fig. 2. As shown below, Wang describes a "crack portion observed from a TEM image in the positive electrode active material particle" (Ex. 1010, 1, 3):

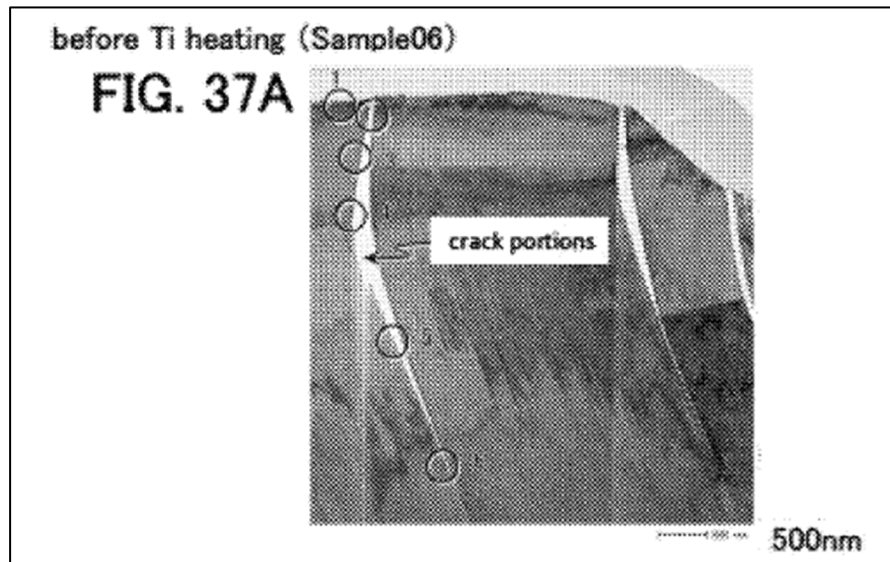


Id., Fig. 3(b), 5(b); *see also* Ex. 1026, 2 (noting that TEM image showed “fragment[s]” or “crack[s]” in cycled positive electrode grains); Ex. 1003, ¶¶ 150, 152.

ii. *Representative element present in a crack portion*

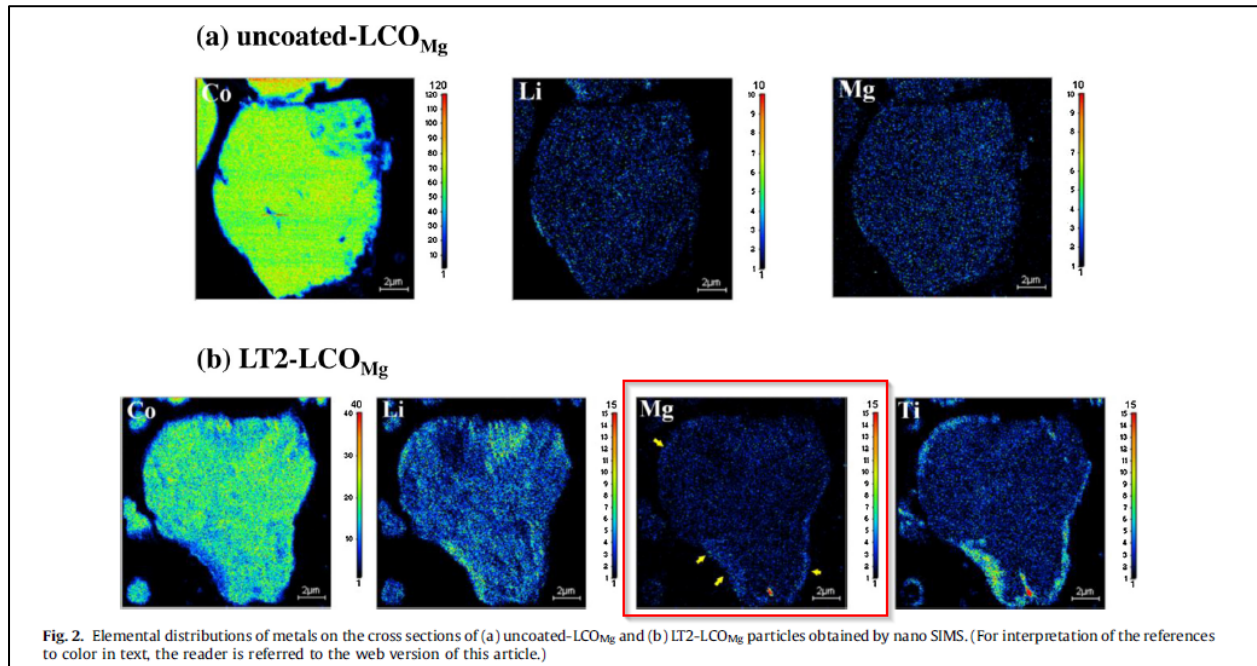
As explained above in Section XI.A.2.e, Shim discloses PEAM particles comprising the representative element magnesium. A POSITA would have further

understood that magnesium is present in a crack portion observed from a TEM image in the PEAM particle for at least two reasons. **First**, Shim discloses doping the bulk of the PEAM particle with magnesium (Ex. 1005, 3), indicating that crack portions of Shim’s PEAM particle (formed in the bulk) necessarily contain magnesium. *See* Ex. 1003, ¶¶ 160–61. As shown below in Figure 37A’s circles, the “crack portions” of Shim’s PEAM particle include both bulk material and voids therein:



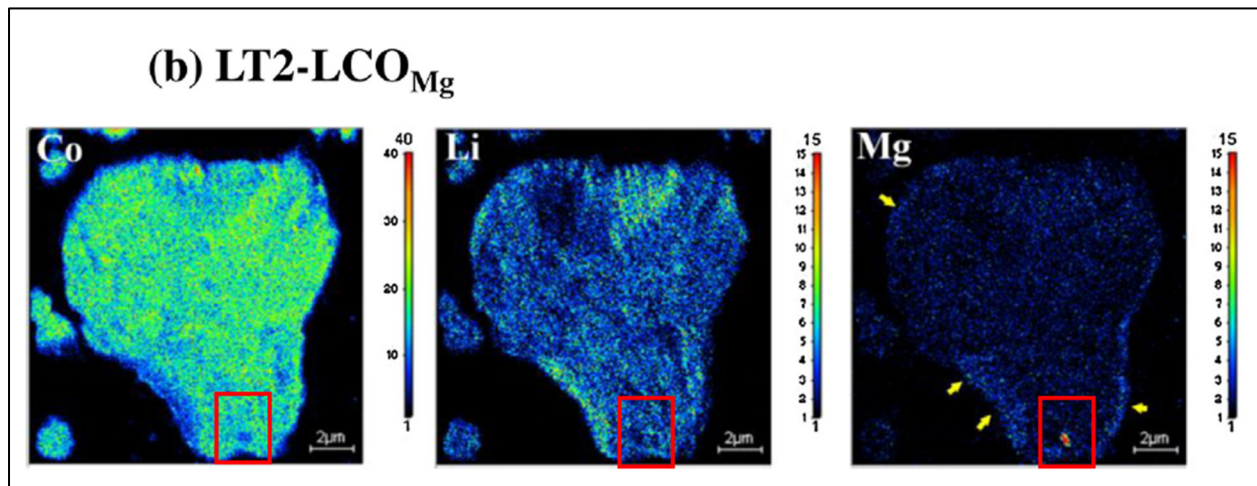
Ex. 1001, Fig. 37A.

Second, a POSITA would have understood: (1) magnesium in the bulk of Shim’s particle diffuses into Shim’s coating; and (2) elements within Shim’s coating will be found in Shim’s crack portions. *See* Ex. 1003, ¶ 162. Specifically, Shim teaches that magnesium’s diffusion into the coating is “clearly evidenced by nano SIM analysis” which shows more magnesium in the coating than the bulk. *See id.*, ¶ 203. Shim annotates this with yellow arrows:



Ex. 1005, Fig. 2 (annotated).

Additionally, a POSITA would have understood that magnesium (after having diffused into the coating) will be become ingrained into the crack portions of Shim's particle. *See* Ex. 1016, 4 (“[P]articles of the coating material [] become ingrained in the crevices and cracks on the cathode surfaces.”). Indeed, as shown below, Shim itself demonstrates this ingrain effect:



Ex. 1005, Fig. 2(b) (annotated). In the crack portion (boxed in red), cobalt and lithium concentrations are low (dark blue patches), while magnesium concentration is high (orange patch). *See id.* Thus, a POSITA would have understood that magnesium in Shim's coating will be found in crack portions. Ex. 1003, ¶ 162. For the same reasons, if Shim's particle is combined with Lee's MgF₂ coating as is outlined in Section XI.A.1, then a POSITA would have understood that the magnesium in this coating will be found in crack portions. *See id.*, ¶ 163.

iii. *Fluorine present in a crack portion*

As described above in Sections XI.A.2.f and XI.A.2.g.ii, Shim (either alone or in combination with Lee) discloses fluorine in Shim's coating, and a POSITA would have understood that such coating material would be present in crack portions. *See* Ex. 1005, Fig. 2(b); Ex. 1016, 4. Thus, because Shim renders obvious a fluorine-containing coating, a POSITA would have understood fluorine would likewise be present in crack portions. Ex. 1003, ¶¶ 164–65. For the same reasons, if Shim's particle is combined with Lee's teaching of a fluorine-containing coating, then a POSITA would have understood that this fluorine would be found in crack portions. *See id.*

3. Dependent Claim 4

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

Shim, either alone or in combination with Lee, renders obvious the lithium-ion secondary battery according to claim 1. *See supra*, § XI.A.1.

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

Shim discloses this element. *See supra*, § XI.A.2.c.

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

Shim discloses this element. *See supra*, § XI.A.2.d.

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

Shim discloses this element. *See supra*, § XI.A.2.e.

4. Independent Claim 5

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

Shim discloses the preamble. *See supra*, § XI.A.2.a.

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

Shim discloses this element. *See supra*, § XI.A.2.b.

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

Shim discloses this element. *See supra*, § XI.A.2.c.

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

Shim discloses this element. *See supra*, § XI.A.2.d.

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

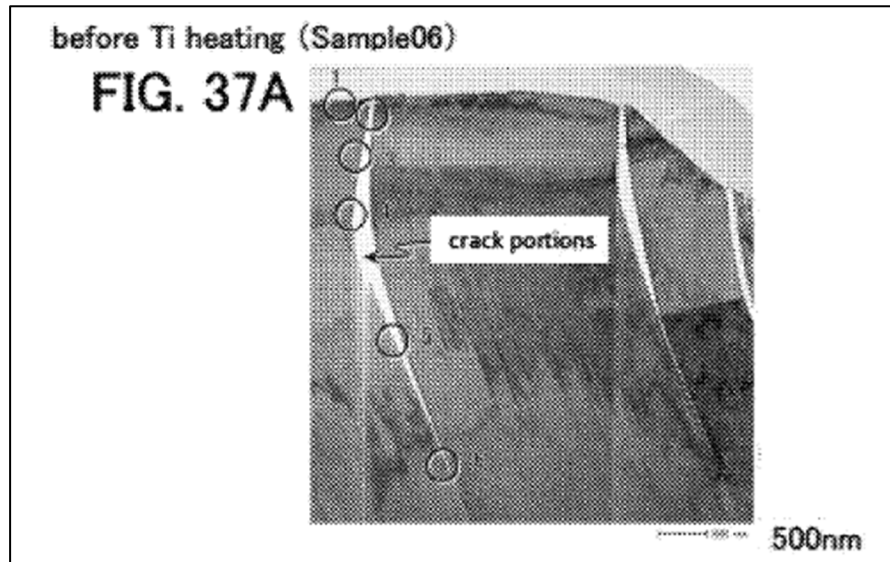
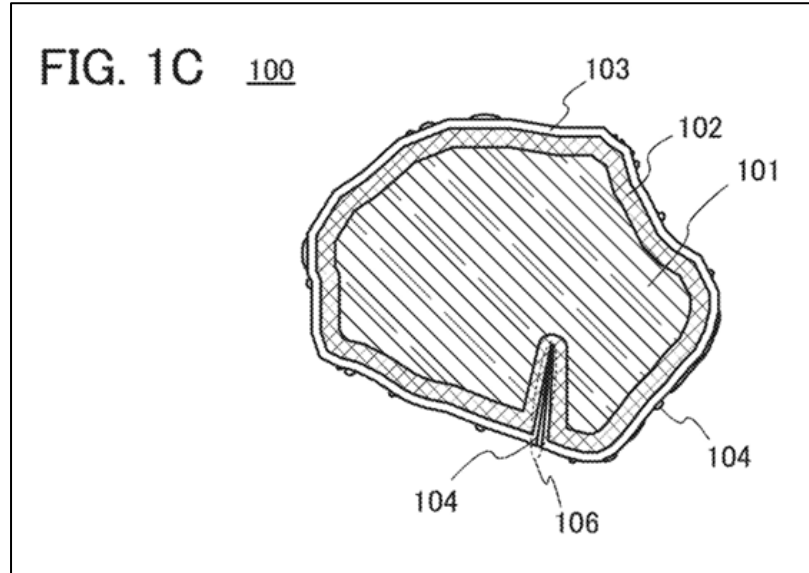
Shim discloses this element. *See supra*, § XI.A.2.e.

f. *[Element 5.5]: fluorine,*

Shim, either alone or in combination with Lee, renders fluorine obvious. *See supra*, § XI.A.2.f.

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

As explained above, the '828 Patent defines a crystal defect as a “defect[] which can be observed from a TEM image,” and specifically includes “a structure in which another element enters crystal, *a cavity*, and the like.” *See supra*, § IX.A; Ex. 1001, 9:55–58. Under this construction, Shim renders this element obvious, because cracks are a type of crystal defect. Independent claim 1 confirm this, as it recites “a crack portion *observed from a TEM image*.” *Id.*, cl. 1. Moreover, the '828 Patent cites a “cavity” as an example of a crystal defect. As shown below, crack portions (including 106) comprise (in part) a cavity within a PEAM particle.



Id., Figs. 1C, 37A; *see also* Ex. 1003, ¶¶ 176–78.

Second, Shim discloses crystal defects under its ordinary meaning (an “imperfection or interruption of regular arrangement of atoms in a crystalline solid”). A POSITA would understand that a “crack portion” necessarily includes crystal defects because the formation of the crack itself interrupts the regularly arrangement of atoms at the edges of the crack. Ex. 1003, ¶¶ 179–80. Thus, under either

construction, because magnesium and fluorine would be present in a “crack portion” observable using TEM, the same would be true for a “crystal defect.” *See, supra* § XI.A.2.g.ii–iii.

Shim also discloses a PEAM particle with a lattice structure that experiences “degradation mechanisms,” including where the “*expanded lattice* suffers from oxygen loss and side reactions with electrolytes.” *See* Ex. 1005, 1. In other words, Shim discloses a degradation mechanism where oxygen is lost from the lattice structure and portions of the lattice react with the electrolyte. These are examples of crystal defects. Ex. 1003, ¶ 181.

5. Dependent Claim 8

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

Shim, either alone or in combination with Lee, renders obvious the lithium-ion secondary battery according to claim 5. *See supra*, § XI.A.4.

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

Shim discloses this element. *See supra*, § XI.A.2.c.

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

Shim discloses this element. *See supra*, § XI.A.2.d.

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

Shim discloses this element. *See supra*, § XI.A.2.e.

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

1. Motivation to Combine Shim and Yu

A POSITA would have been motivated to combine the teachings of Shim (either alone or in combination with Lee) and Yu. Yu teaches a LiCoO_2 PEAM particle doped with titanium (the claimed second transition metal), resulting in a Ti:Co concentration ratio equaling 8.49% at the surface of particle. *See* Ex. 1007, 138 (Table 3). Given Shim and Yu’s complementary teachings, this combination would have involved routine implementation with no technical risks. Ex. 1003, ¶ 186.

First, a POSITA would have recognized that Shim and Yu are directed to improving PEAMs for enhanced battery performance. Both references teach lithium-ion batteries with the same cathode particle (LiCoO_2) treated with the same transition metal, titanium. Ex. 1005, 2; Ex. 1007, 1. Further, both references address capacity fade during cycling due to cathode degradation using doping techniques. *See* Ex. 1005, 2 (“By modifying electronic structure of original active materials, doping is a simple method to efficiently improve cell performances.”); Ex. 1007, 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”). A POSITA would have had a reasonable expectation of success in combining Yu’s teachings with Shim

in this way. It was well-known in the art that magnesium and titanium could be added to the bulk of the PEAM via doping. Ex. 1003, ¶ 187.

Second, both Shim and Yu provide express motivations for using Yu's titanium doping and surface measurement techniques in Shim's PEAM particles. *See DyStar*, 464 F.3d at 1360. Specifically, Shim used magnesium ions as a dopant, recognizing that "because Mg has a valence state of 2^+ rather than Co (3^+) or Li (1^+), the Mg-doped LiCoO_2 exhibits strong electrical conductivity compared to the bare LiCoO_2 and, therefore, it shows excellent cell performances." Ex. 1005, 2. A POSITA would have therefore understood that a dopant with a different valence state than Co (3^+) or Li (1^+) would improve LCO cell performance. Ex. 1003, ¶ 188. Yu teaches just that—using " Ti^{4+} " (with valence state of 4^+) as a dopant. Ex. 1007, 1. Yu further teaches "even a very small amount of Ti . . . can effectively improve the electrochemical performance of the LiCoO_2 material." *Id.*, 4.

Shim provides an express motivation to use Yu's characterization techniques to analyze Shim's particle, as the "[t]hickness of [Shim's] coating layer is too thin (~200 nm) to characterize its detailed structure by XRD." Ex. 1005, 3. Yu teaches characterizing the thin coating layer using XPS, measuring the Ti:Co ratio including at the surface of the particle. Ex. 1007, 3. Thus, a POSITA would have been motivated to use Yu's characterization techniques to better understand the composition of Shim's PEAM particle. Ex. 1003, ¶ 189.

Third, for the above reasons, this combination represents using a “known technique to improve similar devices [and methods] in the same way,” applying “a known technique” (Yu’s titanium doping) to “a known device ready for improvement” (Shim’s PEAM) to yield predictable results, and combining known prior-art elements “according to known methods to yield predictable results.” *KSR*, 550 U.S. at 417. Indeed, Shim acknowledges that “various dopants for LiCoO₂, such as Al, Zr, Sn, B, and so forth, have also been explored” (Ex. 1005, 2), which would motivate a POSITA similarly to use titanium as a dopant. Ex. 1003, ¶ 190.

Fourth, a POSITA would have understood that titanium concentration (and by extension, the ratio of Ti:Co concentration) is a result-effective variable because it was known that titanium “effectively improve[s] the electrochemical performance of the LiCoO₂ material.” Ex. 1007, 4; *see In re Applied Materials*, 692 F.3d 1289, 1295 (Fed. Cir. 2012) (where the prior art discloses overlapping ranges, “it is not inventive to discover the optimum or workable ranges by routine experimentation,” if the parameter is known to be a result-effective variable) (quoting *In re Aller*, 220 F.2d 454, 456 (CCPA 1955)). Thus, the optimum range of Ti:Co concentration ratios could have been discovered by routine experimentation. *Id.* Indeed, a POSITA would have been motivated to use as little titanium as necessary to improve LiCoO₂ performance by enhancing structural stability, understanding that titanium does not participate in the electrochemical reaction and is present at the expense of cobalt,

which is electrochemically active and contributes to the battery's capacity. Ex. 1003, ¶ 190.

2. Dependent Claim 2

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

Shim, either alone or in combination with Lee, renders claim 1 obvious. *See supra*, § XI.A.2.a.

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

Shim (either alone or in combination with Lee) and Yu render this element obvious. As explained above, the first transition metal can be cobalt and the second transition metal can be titanium. *See supra*, §§ XI.A.2.c–XI.A.2.d. Yu measures the relative concentrations of cobalt and titanium using “X-ray photoelectron spectroscopy (XPS)” and “Ar ion etching.” Ex. 1007, 1–2 (“Fig. 2 displays the Ti2p and Co2p [XPS] spectrum of [lithium cobalt titanium oxide]. Ar ion etching was used to examine the concentration depth profiles of the [lithium cobalt titanium

oxide] powder.”).¹ As shown below in Table 3, Yu discloses measured Ti:Co concentration ratios of a PEAM at different sputtering times:

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

Ex. 1007, 3, Table 3 (annotated). Yu makes clear that this data was derived from the XPS spectra of titanium and cobalt, as shown in Figure 2. *Id.*, 2, Fig. 2.

As annotated in red above, at 0 seconds of sputtering time (*i.e.*, at the surface of the PEAM particle) (Ex. 1003, ¶ 194), the ratio of titanium to cobalt is 8.49%, or 0.0849. Ex. 1007 at 3, Table 3. This falls within the claimed range of 0.05 to 0.4, rendering this element obvious. *See Merck & Co. v. Biocraft Lab 'ys.*, 874 F.2d 804, 809 (Fed. Cir. 1989) (claimed ranges do not defeat obviousness where

¹ A POSITA would have understood that Ar ion etching is used with XPS analysis to determine the concentration profile of certain elements through a sample. Ex. 1003, ¶ 193. As argon ions are sputtered (or etched) into the sample, the concentration intensity at various depths of the sample can be analyzed. *Id.* For instance, a longer etching time correlates with a measurement deeper into the sample. *Id.*

“experimentation needed to arrive at the claimed [ranges] was nothing more than routine”).

As explained above in Section XI.B.1, a POSITA would have been motivated to combine the teachings of Yu with Shim. Specifically, a POSITA would have understood that Shim and Yu discuss the same structural degradation problem within the same battery cathode, and that using titanium as a dopant for the lithium cobalt oxide PEAM particle would have increased the battery’s capacity. Ex. 1003, ¶¶ 192–97.

3. Dependent Claim 6

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

Shim, either alone or in combination with Lee, renders claim 5 obvious. *See supra*, § XI.A.4.

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

Shim (either alone or in combination with Lee) and Yu render this element obvious. *See supra*, § XI.B.2.b.

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

1. Dependent Claim 3

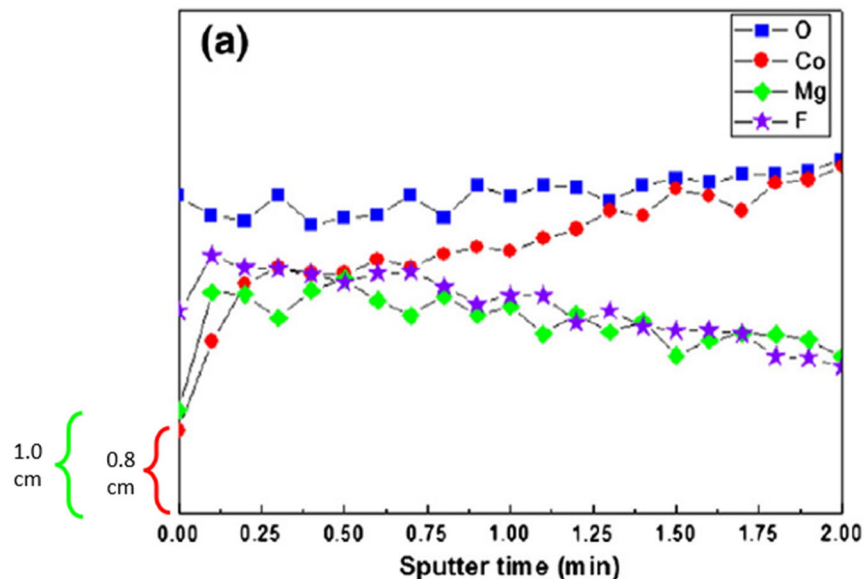
- a. *[Preamble]: The lithium-ion secondary battery according to claim 1,*

Shim, either alone or in combination with Lee, renders claim 1 obvious. *See supra*, § XI.A.2.a.

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

Shim and Lee render this element obvious. As explained above, the first transition metal can be cobalt and the representative element can be magnesium. *See supra*, §§ XI.A.2.c–XI.A.2.d. Lee discloses a LiCoO₂ cathode material coated with MgF₂ and measures the “depth-concentration profiles of the elements (Co, O, Mg, and F)” using “XPS, which is one of the efficient surface analysis tools.” Ex. 1006 at 1–2, 5; Ex. 1003, ¶ 201.

The result of Lee’s XPS analysis of its MgF₂-coated LiCoO₂ (before cycling) is shown below in Figure 5(a):



Ex. 1006, Fig. 5(a) (y-axis annotations added). As shown by the annotated y-axis, at 0 minutes of sputtering (*i.e.*, at the surface of the particle), the ratio of magnesium (green) to cobalt (red) was approximately 1.0 to 0.8, or 1.25. Ex. 1003, ¶¶ 202–04. Moreover, as shown above, within 15 seconds of sputtering (*i.e.*, close to the surface of the particle), the relative concentrations of cobalt and magnesium are approximately equal. Thus, Lee discloses a relative concentration of magnesium to cobalt at the surface of the PEAM particle within the claimed range of 0.4 to 1.5. *Id.*, ¶ 205. *See Merck*, 874 F.2d at 809 (claimed ranges do not defeat obviousness where “experimentation needed to arrive at the claimed [ranges] was nothing more than routine”).

As explained above in Section XI.A.1, a POSITA would have been motivated to combine the teachings of Lee with Shim. Specifically, a POSITA would have

understood that Shim and Lee discuss the same structural degradation problem within the same battery cathode, and that using a metal fluoride coating (such as MgF_2) would have protected the coating from unwanted HF interactions, thereby increasing the battery's capacity. Ex. 1003, ¶ 206. Further, a POSITA would have understood that concentration ratio of Mg:Co is a result-effective variable because increasing magnesium concentration is known to increase the stability of the cathode structure. *See Applied Materials*, 692 F.3d at 1295–96. Thus, the optimum range of Mg:Co concentration could have been discovered by routine experimentation. *Id.*

2. Dependent Claim 7

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

Shim, either alone or in combination with Lee, renders claim 5 obvious. *See supra*, § XI.A.4.

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

Shim and Lee render this element obvious. *See supra*, § XI.B.2.b.

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

1. Independent Claim 1

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

Fujiki discloses the preamble. Fujiki describes a “positive electrode and a nonaqueous electrolyte secondary cell” (*i.e.*, a battery cell), and in particular “a positive electrode active material” “with which a high capacity and excellent charge-discharge cycle characteristics are secured and in which deterioration during use in high-temperature environments is little.” Ex. 1008, 1:14–17, 2:62–67; Ex. 1003, ¶¶ 209–10.

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

Fujiki discloses a PEAM particle. Fujiki is titled “***Positive Electrode Active Material***, Positive Electrode, and Nonaqueous Electrolyte Cell.” *See id.*, Abstract (“Disclosed herein is a positive electrode active material[.]”). Fujiki expressly teaches a positive electrode active material containing “principal transition metal A,” “at least one metallic element M1 different from a principal transition metal element A,” and “a compound of a metallic element M2 different from the metallic element M1.” *Id.*, 3:1–9; *see also id.*, 3:10–30, 3:64–4:47, 10:27–32. Beyond these disclosures, all of Fujiki’s exemplary embodiments of a lithium-ion battery disclose a PEAM particle. *See generally id.*, 11:45–30:17; Table 1, Table 2; Ex. 1003, ¶¶ 211–12.

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

Fujiki's PEAM particle comprises a first transition metal—including at least nickel, cobalt, and/or manganese. Fujiki's PEAM particles comprise “a lithium-containing transition metal oxide containing the principal transition metal element A” (Ex. 1008, Abstract, 3:1–9, 3:10–17, 3:18–30), which comprises “at least one selected from the group consisting of at least Ni, Co and Mn” (*id.*, 4:57–67), which are all transition metals. Ex. 1003, ¶ 213; *see* Ex. 1008, 4:57–67, 11:56–64, 12:29–34, 24:14–20, 29:56–30:17.

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

Fujiki's PEAM particle further comprises a second transition metal—including at least titanium, zirconium, zinc, and/or yttrium. Fujiki teaches a PEAM particle containing “a compound of metallic element M2” (*id.*, Abstract, 3:1–9, 3:10–17, 3:18–30, 4:49–56), which is preferably at least one of “Na, Li, Mg, Zr, Zn, Ba, Bi, Y, Ti, Al, and B” (*id.*, 5:29–32), with zirconium, zinc, yttrium, and titanium being transition metals. Ex. 1003, ¶ 214–16. These M2 transition metals differ from M1 transition metals (namely, cobalt, nickel, and manganese).

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

Fujiki's PEAM particle comprises a representative element—including at least magnesium and/or aluminum. As explained above in Section XI.A.2.e, representative elements are those found in groups 1–2 and 13–17 of the periodic

table. Ex. 1003, ¶ 139. The '828 Patent states that magnesium is a representative element. Ex. 1001, 10:29–33, 11:65–12:4.

Fujiki's PEAM particles contain metallic element M1 which is “preferably at least one element selected from the group consisting of Mn, Fe, Mg, Al, Ni, and Co.” *Id.*, 5:6–19; *see also id.*, 3:1–30, 4:49–56, 5:6–28, 10:26–50, 11:65–12:8. Magnesium and aluminum are representative elements. Ex. 1003, ¶¶ 217–19.

f. *[Element 1.5]: fluorine,*

Fujiki's PEAM particle comprises fluorine, including in Fujiki's disclosures that metallic element M2 can include a metallic fluoride. Fujiki provides several examples of metallic fluorides that can be used in a PEAM particle including aluminum fluoride and yttrium fluoride. *Id.*, 21:36–44, 21:66–22:6, 23:21–29. Fujiki's Example 22 discloses a PEAM comprising a first transition metal cobalt, representative element magnesium, and second transition metal, yttrium fluoride. *Id.*, 22:28–53 (yttrium fluoride “deposited on the surfaces of the particles of the lithium-transition metal compound oxide”).

Fujiki makes clear that “metallic element M2” is preferably not a single element but a compound—namely, “a metallic salt, a metallic oxide or a *metallic halide*.” Ex. 1008, 5:46–49. Fujiki further teaches that M2 “particularly preferabl[y] is a metallic *fluoride*.” *Id.*, 6:3–11. Accordingly, a POSITA would have found a titanium fluoride coating at least obvious, especially because various titanium

fluoride coatings were well known in the art. *See supra*, § VII.D; Ex. 1017, [0011]–[0013] (disclosing TiF, MgF₂, TiF₃, and TiF₄ coatings); Ex. 1018, 2:66–3:15 (same); Ex. 1019, 1:24–31; Ex. 1020, [0022]–[0023], [0030]. Accordingly, Fujiki teaches a PEAM containing fluorine. Ex. 1003, ¶¶ 220–23.

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

Fujiki renders this limitation obvious considering a POSITA's knowledge. *See Koninklijke Philips N.V. v. Google LLC*, 948 F.3d 1330, 1337 (Fed. Cir. 2020) (considering the general knowledge of the POSITA in obviousness analysis, especially in view of expert testimony and corroborating evidence). *See* Ex. 1003, ¶¶ 224–25.

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

Fujiki renders obvious a crack portion observed from a TEM image. Fujiki recognized that “deterioration of a positive electrode active material” presented a problem for lithium-ion batteries. Ex. 1008, 3:34–39, 3:64–4:7 (describing deterioration and instability of the “active material” in LiCoO₂-based batteries), 4:28–34, 5:32–35 (describing ways to “stabiliz[e] the crystal structure of the compound oxide particles [*i.e.*, LiCoO₂] at the time of charging”). A POSITA would

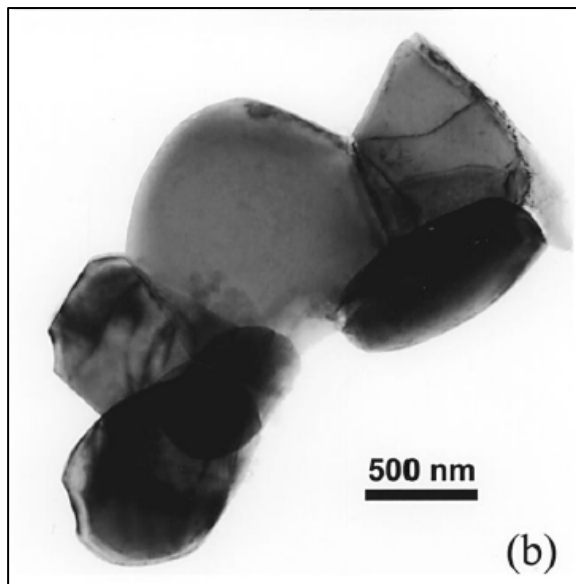
have understood that LiCoO₂ deterioration and the need to “stabiliz[e] the crystal structure” of LiCoO₂ refer to cracking in Fujiki’s PEAM particle. Ex. 1003, ¶ 226.

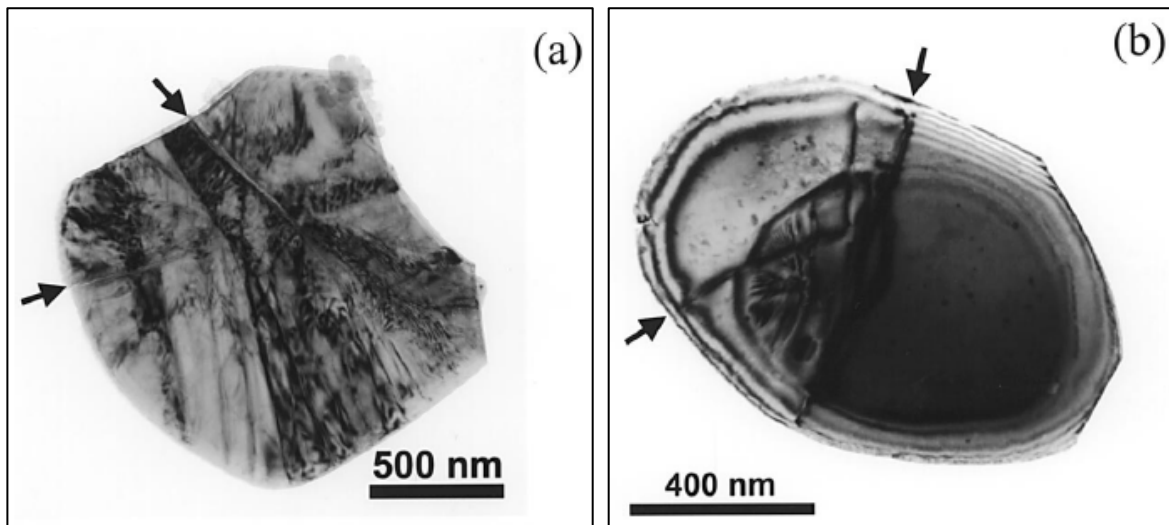
Additionally, Fujiki discloses several methods of observing PEAM particles, including “by a scanning electron microscope (SEM) equipped with an energy dispersive X-ray analyzer (EDX).” Ex. 1008, 12:9–11. A POSITA would have additionally understood that TEM and SEM images alike could be used to observe crack portions in PEAM particles and would have found it obvious to use both techniques to make such observations. Ex. 1003, ¶ 227; Ex. 1010, 3–4; Ex. 1026, 2; *see supra*, § VII.C.

To the extent the Board finds Fujiki does not expressly disclose a crack portion, a POSITA would have found it obvious in light of his/her knowledge. It was well-known that crack portions in PEAMs (like Fujiki’s) were inevitable. For example, at least 10 years before the ’828 Patent, a POSITA would have known that “[i]t is *inevitable* that coated surfaces always have defects such as *cracks* and pinholes in them” and that such cracks “may form and close with the application of a load or upon thermal *cycling*.” Ex. 1016, 4. Similarly, at least 17 years before the ’828 Patent, a POSITA would have known that “[e]lectrochemical cycling creates a variety of defects in the LiCoO₂ particles in a *typical composite cathode*.” Ex. 1010, 8. Consistently with this well-known general concept, Fujiki states that “deterioration of the active material” is a known problem for lithium cobalt oxide

cathodes. Ex. 1008, 3:64–4:7. Thus, a POSITA would have known that Fujiki’s disclosure of deterioration includes inevitable cracks within PEAM particles. Ex. 1003, ¶ 228.

Moreover, a POSITA would have known that the cracks in Fujiki’s PEAM particles may be observed using TEM imaging. For instance, Fey explains that TEM can be used to “examin[e] . . . the microstructure of the coated particles,” indicating that cracks would likewise be observable by TEM. Ex. 1016, 1, Fig. 2. As a further example of a POSITA’s knowledge, Wang describes a “crack portion observed from a TEM image in the positive electrode active material particle” (Ex. 1010, 1, 3):





Id., Fig. 3(b), 5(a)–(b); *see also* Ex. 1026, 2 (TEM image showing “fragment[s]” or “crack[s]” in cycled positive electrode grains); Ex. 1003, ¶¶ 229–36 (Dr. Hayes’s testing confirming the same).

ii. *Representative element present in a crack portion*

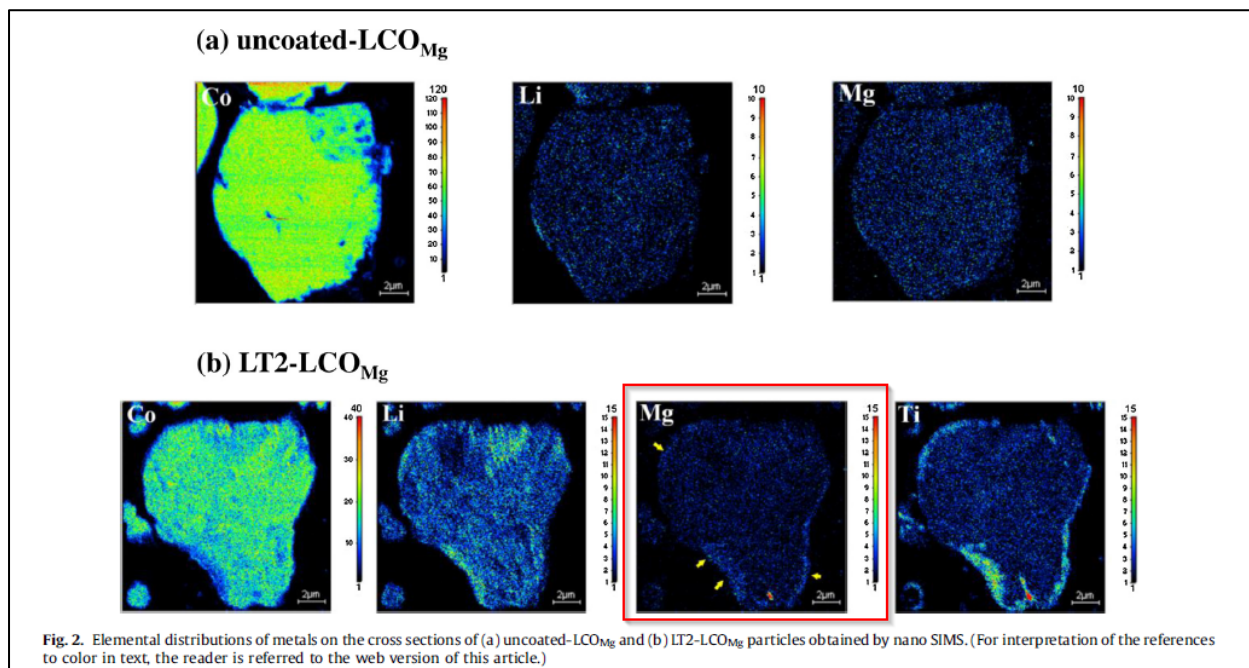
As explained above in Section XI.D.1.e, Fujiki discloses PEAM particles comprising the representative element magnesium. A POSITA would have further understood that magnesium is present in a crack portion observed from a TEM image because Fujiki teaches that magnesium is present throughout the particle—from the surface to the interior. Specifically, Fujiki teaches that “it is effective for the metallic element M1” (*e.g.*, magnesium) to be “in the state of having diffused from the surfaces *into inside portions* near the surfaces of the compound oxide particles so as to show a continuous concentration gradient *toward the centers* of the [PEAM] particles.” Ex. 1008, 5:12–19; 12:19–28 (“[I]t was observed that the magnesium concentration was varying continuously from the surface toward the inside of the

particle.”). In other words, a POSITA would have understood that crack portions of Fujiki’s PEAM particle (*i.e.*, cracks forming in the bulk) contain magnesium because magnesium diffuses into the interior (or bulk) of Fujiki’s particle. *See* Ex. 1003, ¶ 237.

iii. *Fluorine present in a crack portion*

A POSITA would have understood that, due to Fujiki’s coating of PEAM particles with metallic fluorides, fluorine is present in crack portions. Ex. 1003, ¶ 238. Fujiki discloses metallic element M2 as a PEAM coating to stabilize its crystal structure. Ex. 1008, 4:48–56, 5:29–42. As discussed above in Section XI.D.1.e, M2 is preferably a metallic fluoride. *Id.*, 6:3–11, 22:28–53.

A POSITA would have understood that such fluorine in Fujiki’s coating would be present in the crack portions because coating materials were known to become lodged inside such cracks. *See* Ex. 1016, 4 (“[T]he cycling process could have enabled the particles of the coating material to become ingrained in the crevices and cracks on the cathode surface.”). Indeed, this is exemplified with reference to Shim’s Figure 2 below, which shows a high concentration of magnesium and titanium in a crack portion.



Ex. 1005, Fig. 2 (annotated). A POSITA would have understood that magnesium and titanium found in Shim's coating, have become embedded in the cracks of Shim's particle, similar to how fluorine from Fujiki's coating would be present in cracks in Fujiki's particles. *See* Ex. 1003, ¶ 238.

2. Dependent Claim 4

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

Fujiki renders claim 1 obvious. *See supra*, § XI.D.1.

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

Fujiki discloses this element. *See* § XI.D.1.c.

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

Fujiki discloses this element. *See* § XI.D.1.d.

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

Fujiki discloses this element. *See supra*, § XI.D.1.e.

3. Independent Claim 5

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

Fujiki discloses the preamble. *See supra*, § XI.D.1.a.

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

Fujiki discloses this element. *See supra*, § XI.D.1.b.

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

Fujiki discloses this element. *See supra*, § XI.D.1.c.

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

Fujiki discloses this element. *See supra*, § XI.D.1.d.

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

Fujiki discloses this element. *See supra*, § XI.D.1.e.

- f. *[Element 5.5]: fluorine,*

Fujiki discloses this element. *See supra*, § XI.D.1.f.

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

Fujiki renders this limitation obvious. *See supra*, § XI.D.1.g. As explained above in Section XI.A.4.g, at the time of the '828 Patent, a POSITA would have

understood a “crack portion” to be an example of a crystal defect under the ’828 Patent’s express definition. Ex. 1003, ¶ 249. Because Fujiki renders obvious a crack portion observed from a TEM image, it likewise renders obvious a crystal defect observed from a TEM image. Moreover, observing such a crystal defect from a TEM image would have been obvious to try as an identified, predictable solution applying a well-known technique. *See KSR*, 550 U.S. at 417.

Moreover, as Dr. Hayes explains, Fujiki likewise discloses a “crystal defect” under its plain and ordinary meaning, which is an “imperfection or interruption of regular arrangement of atoms in a crystalline solid.” *See supra*, § IX.A; Ex. 1003, ¶¶ 250–51. Fujiki indicates that its LiCoO₂ active material “has a laminar rock salt structure” (Ex. 1008, 4:58–63) which is a lattice structure. Ex. 1003, ¶ 252. Fujiki further teaches that metallic element M2 (which can include titanium and/or a metallic fluoride), has the express “function of *stabilizing the crystal structure* of the compound oxide particles at the time of charging.” Ex. 1008, 5:32–35. Thus, Fujiki recognized that defects occur in the crystal structure of LiCoO₂ and provide mechanisms to mitigate those effects. Ex. 1003, ¶ 252.

As explained above, Fujiki renders obvious that the representative element (magnesium) and the fluorine are present in a portion which includes such crystal defects observed from a TEM image. *See supra*, § XI.D.1.g.

4. Dependent Claim 8

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

Fujiki renders claim 5 obvious. *See supra*, § XI.D.3.

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

Fujiki discloses this element. *See supra*, § XI.D.1.c.

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

Fujiki discloses this element. *See supra*, § XI.D.1.d.

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

Fujiki discloses this element. *See supra*, § XI.D.1.e.

E. Ground 5: Claims 2 and 6 are Rendered Obvious by Fujiki in Combination with Yu

1. Motivation to Combine Fujiki and Yu

A POSITA would have been motivated to combine the teachings of Fujiki and Yu. Ex. 1003, ¶¶ 258–69. Yu discloses doping a LiCoO₂ PEAM particle (such as Fujiki’s) with titanium, which results in a Ti:Co concentration ratio equaling 8.49% (or 0.0849) at the particle’s surface. Ex. 1007, 3, Table 3. Given the references’ complementary teachings, this combination would have involved routine implementation with no technical risks. Ex. 1003, ¶ 258.

First, a POSITA would have recognized that Fujiki and Yu are directed to improving PEAMs for enhanced battery performance. Both references teach lithium-

ion batteries with the same cathode particle—namely, LiCoO_2 —treated with the same transition metal, titanium. Ex. 1008, 5:29–42, 18:63–19:22; Ex. 1007. Further, both references discuss transition metal coating techniques to address the same specific issue—*i.e.*, capacity fade during cycling due to degradation of the cathode structure—with a similar technique: coating the core or bulk PEAM material. *See* Ex. 1008, 34:8–13 (“Thus, where the compound oxide particles coated with the metallic element . . . are used as the positive electrode active material, both a high initial capacity and a high capacity retention could be obtained simultaneously.”). Ex. 1007, 136 (“[P]artial doping of cobalt with metal ions leads to ***extended cyclability and increase in capacity of lithium ion cells*** by enhancing structural stability.”). A POSITA would have had a reasonable expectation of success in combining Yu’s teachings with Fujiki’s—it was well known in the art that titanium could be doped in a PEAM’s bulk. Ex. 1003, ¶¶ 259–64.

Second, Yu provides express motivations to use its titanium doping and surface measurement techniques in Fujiki’s PEAM particles. *See DyStar*, 464 F.3d at 1360. Specifically, Yu explains it was “[a]s well as known” that “partial doping of cobalt with metal ions leads to extended cyclability and increase in capacity of lithium ion cells by enhancing structural stability.” Ex. 1007, 1. Moreover, Yu teaches “that even a very small amount of Ti addition . . . can effectively improve the electrochemical performance of the LiCoO_2 material.” *Id.*, 4.

Third, for the above reasons, this combination represents using a “known technique to improve similar devices [and methods] in the same way”; applying “a known technique” (Yu’s titanium doping) to “a known device ready for improvement” (Fujiki’s PEAM) to yield predictable results; and combining known prior-art elements “according to known methods to yield predictable results.” *KSR*, 550 at 417.

Fourth, a POSITA would have understood that titanium concentration (and the ratio of Ti:Co concentration) is a result-effective variable because it was known that titanium “effectively improve[s] the electrochemical performance of the LiCoO₂ material.” Ex. 1007, 4; *Applied Materials*, 692 F.3d at 1295–96. Thus, the optimum range of Ti:Co concentration ratios could have been discovered by routine experimentation. *Id.* Indeed, a POSITA would have been motivated to use as little titanium as necessary to improve LiCoO₂ performance by enhancing structural stability, understanding that titanium does not participate in the electrochemical reaction and is present at the expense of cobalt, which is electrochemically active and contributes to the battery’s capacity. Ex. 1003, ¶ 264.

2. Dependent Claim 2

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

Fujiki renders claim 1 obvious. *See supra*, § XI.D.1.

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

Fujiki and Yu render this element obvious. As explained above in Section XI.B.2.b (with reference to the combination of Shim and Yu), at the surface of Yu's PEAM particle, the Ti:Co ratio is 0.0849—within the claimed range (0.05–0.4). Ex. 1007, Table 3; Ex. 1003, ¶ 266.

As explained above in Section XI.E.1, a POSITA would have been motivated to combine the teachings of Yu with Fujiki. Specifically, a POSITA would have understood that Fujiki and Yu discuss the same structural degradation problem within the same battery cathode, and that Yu teaches that using titanium as a dopant for the lithium cobalt oxide PEAM particle would have increased the battery's capacity. Ex. 1003, ¶ 267.

3. Dependent Claim 6

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

Fujiki renders claim 5 obvious. *See supra*, § XI.A.4.

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

an XPS analysis and a concentration of the first transition metal is defined as 1.

Fujiki and Yu render this element obvious. *See supra*, § XI.E.2.b.

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

1. Motivation to Combine Fujiki and Lee

A POSITA would have been motivated to combine the teachings of Fujiki and Lee. Ex. 1003, ¶¶ 270–80. Lee teaches using a MgF_2 coating on a LiCoO_2 PEAM particle, resulting in a Mg:Co concentration ratio of approximately 1.0 to 1.25 at the surface of particle. *See* Ex. 1006, Fig. 5(a). As explained below, a POSITA would have had several reasons to combine Lee’s teachings with Fujiki, which would have involved routine implementation with no technical risks. Ex. 1003, ¶ 270.

First, a POSITA would have recognized that Fujiki and Lee are in the same field of endeavor and directed to the same problem of improving PEAM for enhanced battery performances. Both references teach the same lithium-ion battery cathode (LiCoO_2) and characterize this cathode’s properties using similar analytical methods such as SEM/EDS. Ex. 1008, 12:9–18; Ex. 1006, 1, 5. A POSITA would have had a reasonable expectation of success in making this combination. It was well-known that magnesium, titanium, and fluorine could be combined in a coating layer (such as titanium fluoride or magnesium fluoride). *See supra*, § VII.D; Ex. 1017, [0011]–[0013] (disclosing coatings including TiF , MgF_2 , TiF_3 , and TiF_4);

Ex. 1018, 2:66–3:15 (same); Ex. 1019, 1:24–31; Ex. 1020, [0022]–[0023], [0030]; Ex. 1003, ¶ 271.

Second, both references discuss using metallic fluoride coatings to address the same specific issue—improving the structural integrity of the PEAM particles during cycling. *See* Ex. 1008, 5:32–35 (“The metallic element M2 is an element which has the function of stabilizing the crystal structure of the compound oxide particles at the time of charging. It is preferable that the metallic element M2 is in present in the state of coating[.]”); Ex. 1006, 6 (“The MgF_2 coating layer was very effective in improving the electrochemical properties of the LiCoO_2 film electrode.”). Lee’s MgF_2 coating is a “metallic fluoride,” which Fujiki indicates is preferable to stabilize the structure of the PEAM particles. Ex. 1008, 6:3–11; Ex. 1003, ¶ 272.

Third, a POSITA would have understood that using Lee’s MgF_2 layer in Fujiki would have been a known “known technique to improve similar devices [and methods] in the same way” and a “combination of known prior-art elements according to known methods to yield predictable results.” *See KSR*, 550 U.S. at 417. As explained above in Section VII.D and XI.A.1, a POSITA would have understood that a fluorine-containing electrolyte (such as LiPF_6) may degrade to form a fluorine-containing film on the particle and hydrofluoric acid, the latter of which adversely affects the cathode and battery performance. *See* Ex. 1003, ¶ 273.

To this end, a POSITA would have been motivated to use Lee's MgF₂ coating. Specifically, Lee teaches that “when the surface of the cathode is covered with a coating layer, the coating material serves as a protective layer, preventing direct contact with the reactive electrolyte.” Ex. 1006, 4–6 (“[T]he MgF₂ coating layer was effective as a protective layer to prevent the dissolution of the transition metal . . . which may lead to enhanced electrochemical properties.”); *see also* Ex. 1024, 1–2; Ex. 1027, 1; Ex. 1029, 2; Ex. 1003, ¶ 274.

Fourth, a POSITA would have understood that magnesium concentration (and the ratio of Mg:Co concentration) is a result-effective variable because increasing magnesium is known to increase the stability of the cathode structure. Ex. 1006, 6; Ex. 1008, 6:3–1; *Applied Materials*, 692 F.3d at 1295–96. Thus, the optimum range of Mg:Co concentration could have been discovered by routine experimentation. *Id.* Indeed, a POSITA would have been motivated to use as little magnesium as necessary to improve LiCoO₂ performance by enhancing structural stability, understanding that magnesium does not participate in the electrochemical reaction and is present at the expense of cobalt, which is electrochemically active and contributes to the battery's capacity. Ex. 1003, ¶ 275.

2. Dependent Claim 3

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

Fujiki renders claim 1 obvious. *See supra*, § XI.D.1.

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

Fujiki and Lee render this limitation obvious. As explained above in Section XI.C.1.b (with respect to the combination of Shim and Lee), at the surface of Lee's PEAM particle, the Mg:Co ratio is between 1.0 and 1.25—within the claimed range of 0.4–1.5. Ex. 1006, 1–2, 5, Fig. 5(a); Ex. 1003, ¶ 277.

As explained above in Section XI.F.1, a POSITA would have been motivated to combine the teachings of Lee with Fujiki. Specifically, a POSITA would have understood that Fujiki and Lee discuss the same structural degradation problem within the same battery cathode, that Fujiki itself teaches using a “metallic fluoride,” and that Lee teaches using an MgF_2 coating would have increased the battery's capacity, especially considering Fujiki's LiPF_6 electrolyte. Ex. 1003, ¶ 278.

3. Dependent Claim 7

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

Fujiki renders claim 5 obvious. *See supra*, § XI.D.3.

- b. *[Element 7.1]: wherein a relative value of a concentration of the representative element is greater than or equal to 0.4 and less than or equal to 1.5 when a surface of the positive electrode active material particle is subjected to*

an XPS analysis and a concentration of the first transition metal is defined as 1.

Fujiki and Lee render this element obvious. *See supra*, § XI.F.2.a.

XII. SECONDARY CONSIDERATIONS

Petitioner is unaware of any evidence of alleged secondary indicia that would support non-obviousness. To the extent Patent Owner subsequently presents such evidence, Petitioner respectfully requests the right to respond.

XIII. CONCLUSION

Claims 1–8 of the '828 Patent are obvious for the reasons set forth above. Because there is a reasonable likelihood that Petitioner will prevail regarding each of these Challenged Claims, review of claims 1–8 is accordingly requested.

Dated: June 4, 2023

Respectfully submitted,

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CERTIFICATE OF COMPLIANCE

This Petition complies with the type-volume limitations as mandated in 37 C.F.R. § 42.24, totaling 13,928 words. Counsel has relied upon the word count feature provided by Microsoft Word.

/Yimeng Dou/

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CERTIFICATE OF SERVICE

The undersigned hereby certifies that a copy of the foregoing document with accompanying exhibits, as listed in Petitioner's Exhibit List on page 4 of this document, was served via overnight delivery directed to the attorney/agent of record for the patent as identified on USPTO PAIR and associated with USPTO Customer No. 26171 at the following address:

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