

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

POSCO Future M Co., Ltd.,
Petitioner,

v.

TIAX LLC,
Patent Owner.

IPR2024-01229
U.S. Patent No. 9,391,317 B2

**PETITIONER'S REPLY TO
PATENT OWNER'S PRELIMINARY RESPONSE**

Pursuant to the Board's authorization provided November 25, 2024 (Ex_1029), Petitioner submits this Reply to address new evidence in the POPR.

I. INTRODUCTION

Patent Owner asserts that Lampe-Onnerud (Ex_1007) does not necessarily pertain to polycrystalline cathode materials because using single-crystal material purportedly was also well-known in the art. But the Patent Owner's attorney argument finds no support in its new exhibits to the POPR. Exhibits 2001, 2002, and 2004, which all postdate Lampe-Onnerud by 18+ years, disclose that experimenting with using single-crystal material for lithium nickel oxide-based cathodes became a research focus *only recently*, well after Lampe-Onnerud's effective date in 2002. Moreover, these exhibits clearly demonstrate why a POSITA would *not* have considered Lampe-Onnerud's process to be beneficial for synthesizing anything other than polycrystalline material.

II. THE EXHIBITS DISCLOSE THAT IN 2002 CATHODES WERE MADE OF POLYCRYSTALLINE MATERIAL

Citing to Dr. Aurbach's expert declaration, the Petition explains that a POSITA (in the 2002 timeframe) would have understood that Lampe-Onnerud disclosed a process for synthesizing a "plurality of crystallites" with "a grain boundary between adjacent crystallites" having a greater "concentration of cobalt," as recited in the '317 patent claims. Dr. Aurbach, who co-authored more than 800 papers relevant to the field and partnered with major corporations on research and development throughout

the past 25 years, explained that the “base material [in Lampe-Onnerud] is known to be comprised of secondary particles having agglomerated primary particles and grain boundaries” and that “a [POSITA] would have understood that Lampe-Onnerud discloses a polycrystalline particle of cathode active material.” Ex_1002, ¶¶110, 124. Patent Owner’s counsel disagrees and, without relying on any expert opinion, cites to exhibits 2001-2004 as purported support for asserting that single-crystal materials “are commonly used” in cathodes and that this “easily dates back to the time period around Lampe-Onnerud’s 2002 effective filing date.” POPR, 23. But none of these four exhibits shows using single-crystal cathode material in the 2002 timeframe. Indeed, three of them make precisely the opposite point.

Exhibit 2001 (“Lüther”) compared electrochemical conditions for polycrystalline materials used for cathodes – which the authors referred to as still being the “state-of-the-art” when the article was published in 2024 – with “recent approaches” experimenting with using single-crystals instead. Ex_2001, 1-2. Lüther concluded that, since the “[s]tate-of-the-art cathodes in LIBs [(lithium-ion batteries)] ... are based on PC [(polycrystalline)] secondary particles ... [that have] cracks due to mechanical stress alongside grain boundaries, separated SC [(single-crystal)] primary particle NCM appears to be a promising modification.” *Id.*, 8. In other words, twenty-two years after Lampe-Onnerud, Lüther now suggests that manufacturers consider using a single-crystal material for cathodes instead of the well-established polycrystalline material. Notably, none of citations in Lüther that pertains to using

single-crystal material is dated prior to 2015.

Exhibit 2002 (“Zhang”), published in 2020, makes a similar observation about potentially using single-crystal cathode materials. Zhang notes that “[s]ingle-crystal cathode materials for lithium-ion batteries have attracted increasing interest in providing greater capacity retention than their polycrystalline counterparts.” Ex_2002, 1. Zhang refers to using polycrystalline particles as being the “conventional” approach and proposes that “[u]nderstanding the fundamental role of surface chemistry in single-crystal structure stability can offer new insights into Ni-rich, single-crystal, cathode materials.” *Id.*, 2, 8. Zhang also provides no indication that lithium-ion battery cathodes in the 2002 timeframe used a single-crystal material.

Exhibit 2004 (“Li”), published in 2022, also confirms that researchers only recently began to consider replacing polycrystalline cathode materials in favor of single-crystal materials. Li explicitly states that cathode material is “usually in a secondary particle morphology” but that changes in volume during cycling “leads to cracking of the aggregated secondary particles.” Ex_2004, 1. Li explains that, “to deal with the above-mentioned issues, *in recent years*, researchers have developed single crystalline NCM cathode materials,” which “[d]ue to the absence of grain boundaries, single crystalline NCM materials can avoid the intergranular cracking and ... therefore, it has excellent structural stability.” *Id.*, 2 (emphasis added).

Exhibit 2003 (“Song”), published in 2003, is Patent Owner’s only exhibit that is contemporaneous with Lampe-Onnerud. It also does not indicate that single-crystal

cathode materials were commonplace in the early 2000s. Indeed, *Song makes no mention of single-crystal materials*. Patent Owner’s argument hinges on its attorney’s interpretation of Song’s SEM images, which purportedly make “clear” that Song was working from single-crystal materials. POPR, 23. But those images lack the resolution required to differentiate between single-crystal and polycrystalline morphologies. Song also does not include a cross-sectional view of a particle (which, in the ’317 patent, shows that it is a polycrystalline). Ex_1001, FIG. 1.

Single-crystal cathode active materials were considered innovative and unexpected as recently as 2020 as highlighted by Lüther, Zhang, and Li. If Song were directed to single-crystal materials, the reference would mentioned it back in 2003.

III. THE EXHIBITS SHOW WHY LAMPE-ONNERUD SOUGHT TO IMPROVE POLYCRYSTALLINE CATHODE MATERIAL

Lampe-Onnerud endeavored to solve issues that – as the new exhibits describe – were well-known problems for polycrystalline materials, not for single-crystal materials. Lampe-Onnerud explained that the purpose of creating a cobalt gradient is to stabilize and improve the cyclability of lithium nickel oxide-based materials, which “generally are less safe (i.e., exhibit more gassing) than [lithium cobalt oxide-type materials].” Ex_1007, 1:25-27. Lampe-Onnerud defined “[s]afety” to mean “structural stability or structural integrity,” such that a material is unsafe if it “causes gassing at elevated temperatures.” *Id.*, 7:19-22. The purpose of wetting the material with a cobalt solution was to stabilize and improve cyclability with a cobalt

gradient that gradually decreases in concentration relative to the particle's "core."

The exhibits explain that these safety, stability, and cyclability issues were associated with polycrystallines as opposed to single-crystal materials. As Lüther noted, polycrystalline materials deform "along grain boundaries" during charging, and this "'particle breathing' phenomenon" gives rise to "oxygen release" (i.e., gassing) whereas the absence of grain boundaries in single-crystal materials "prohibits the aforementioned failure cascade because the particle integrity is preserved and surface-related degradation reactions are suppressed." Ex. 2001, 1-2. Li also explained that in "the past decades," lithium nickel cobalt manganese oxide has become the most promising cathode material but suffers from "battery gassing issues," where "irreversible phase transition is accompanied by oxygen evolution," resulting in safety issues. Ex. 2004, 1. Zhang noted that conventional polycrystalline material tends to have cycling problems and cracking at grain boundaries "which exacerbated structural collapse and capacity loss," whereas a single-crystal material already has "high levels of structural stability," and without grain boundaries, "single-crystal electrodes were theorized to provide increased oxygen loss resistance ... when compared with polycrystalline materials." Ex. 2002, 2.

Whereas Lüther, Li, and Zhang sought to solve problems by substituting polycrystalline with a single-crystal material, Lampe-Onnerud synthesized polycrystalline to have a cobalt concentration gradient – anticipating the '317 patent.

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Respectfully Submitted,

/s/ Michael A. Oblon

Michael A. Oblon (Reg. No. 42,956)
JONES DAY
51 Louisiana Avenue, NW
Washington, DC 20001
Telephone: (202) 879-3815
moblon@jonesday.com

Matthew Johnson (Reg. No. 59,108)
JONES DAY
500 Grant Street, Suite 4500
Pittsburgh, Pennsylvania 15219-2514
Telephone: (412) 391-3939
mwjohnson@jonesday.com

Haytham Soliman (Reg. No. 80,826)
JONES DAY
717 Texas St # 3300
Houston, Texas 77002
Telephone: (832) 239-3939
hsoliman@jonesday.com

Tova Werblowsky (Reg. No. 82,311)
JONES DAY
250 Vesey Street
New York, New York 10281-1047
Telephone: (212) 326-3939
twerblowsky@jonesday.com

JONES DAY
Counsel for Petitioner

CERTIFICATE OF SERVICE

The undersigned hereby certifies that a copy of the foregoing Reply to the Patent Owner Preliminary Response is being served on December 3, 2024, via email to the following addresses:

weston.gould@dinsmore.com
Oleg.khariton@dinsmore.com

Dated: December 3, 2024

Respectfully Submitted,

/s/ Michael A. Oblon

Michael A. Oblon (Reg. No. 42,956)
Counsel for Petitioner

APPENDIX OF EXHIBITS

EXHIBIT	TITLE
1001	U.S. Patent No. 9,391,317 (“the ’317 Patent”)
1002	Expert Declaration of Doron Aurbach, Ph.D
1003	Declaration of Hyunkuk Noh, Ph.D
1004	Prosecution file history for U.S. Patent No. 9,391,317
1005	Prosecution file history for U.S. Patent No. 9,209,455
1006	U.S. Provisional Application No. 61/575,115
1007	U.S. Pat No. 6,921,609 to Lampe-Onnerud et al., issued July 26, 2005 (“Lampe-Onnerud”).
1008	U.S. Publ. App. No. 2011/0143209 to Park et al., published June 16, 2011, filed December 1, 2010, and issued as U.S. Patent No. 8,586,247, claiming priority to Prov App. No. 61/285,785 filed December 11, 2009 (“Park”)
1009	Japanese Published Application No. JP 2003-059489 to Takagi et al, published February 28, 2003 (“Takagi”)
1010	Certified translation of JP 2003-059489 (“Takagi”)
1011	Zheng et al., Microstructural Changes in $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$ Positive Electrode Material During the First Cycle, Journal of the Electrochemical Society, 158 (4) A357-A362 (2011) (Published February 9, 2011) (“Zheng”)

1012	Fergus, Recent Developments in Cathode Materials For Lithium Ion Batteries, <i>Journal of Power Sources</i> 195 (2010) 939-954 (“Fergus”)
1013	Kalyani et al., Various Aspects of LiNiO ₂ Chemistry: A Review, <i>Science and Technology of Advanced Materials</i> 6 (2005) 689-703 (“Kalyani”)
1014	Volovich et al., Percolation Models of Grain Boundary Wetting in Polycrystalline Materials, <i>Russian Journal of General Chemistry</i> , 2008, Vol. 78, No. 11, pp. 2182-2190 (“Volovich”)
1015	Wittingham, Lithium Batteries and Cathode Materials, <i>American Chemical Society, Chemical Reviews</i> 2004, Vol. 104, No. 10, pp. 4271-4301 (“Wittingham”)
1016	Ghiji et al., A Review of Lithium-Ion Battery Fire Suppression, <i>Energies</i> 2020, 13, 5117 (“Ghiji”) http://doi.org/10.3390/en13195117
1017	Muto et al., Capacity-Fading Mechanisms of LiNiO ₂ -Based Lithium Ion Batteries, <i>Journal of the Electrochemical Society</i> , 156 (5) A371-A377 (2009) (“Muto”)
1018	Liu et al., Comparison of Structure and Electrochemistry of Al- and Fe-doped LiNi _{1/3} Co _{1/3} Mn _{1/3} O ₂ , <i>Electrochimica Acta</i> 51 (2006) 4199-4203 (“Liu”)
1019	Ellis et al., Positive Electrode Materials for Li-Ion and Li-Batteries, <i>Chemistry of Materials Review</i> , 2010, 22, 691-714 (“Ellis”)
1020	Turner et al., Surface Analysis: X-ray Photoelectron Spectroscopy, Auger Electron Spectroscopy, and Secondary Ion Mass Spectrometry, <i>Anal Chem</i> , 1982, Vol. 54, pp. 293-322 (“Turner”)

1021	U.S. Published Patent Appl. No. 2009/0104530 to Shizuka et al
1022	U.S. Publ. App. No. 2011/0033750 to Hosokawa et al., published February 10, 2011, filed March 3, 2010 (“Hosokawa”)
1023	Japanese Published Application No. JP 2010/047466 A (issued as Japanese Patent JP 5451228 B2), Lithium Transition Metal Oxide with Layer Structure, published March 4, 2010 (“Kagei”)
1024	Certified translation of Kagei excerpt
1025	File History for Lampe-Onnerud
1026	US Provisional Application 61/285,785
1027	U.S. Pat No. 7,381,496 to Onnerud et al., issued June 3, 2008 (“Onnerud”).
1028	Declaration of Michael A. Oblon
1029	November 25, 2024 Board authorization to file reply