IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent of:	Ren et al.	
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Title:	SECONDARY LITHIUM ION CELL OR BATTERY, AND PROTECTING CIRCUIT, ELECTRONIC DEVICE, AND CHARGING DEVICE OF THE SAME	

DECLARATION OF MR. LOUIS HRUSKA

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

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

Dated: August 22, 2023 By: Louis Hruska

APPLE-1003

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I. QUALIFICATIONS AND BACKGROUND INFORMATION

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

2. I have over forty years of experience in electrochemical energy storage, including fundamental research, design, testing and manufacturing of battery cells and systems. I hold over twenty patents, issued in a broad range of battery related technologies, including materials, separators, cell design, electrical controls, communication and software. My work experience includes cell design and process development for Lithium cell manufacturing in the time period preceding the claimed priority date of the patent at issue.

3. I have been retained on behalf of Apple Inc. to offer technical opinions relating to U.S. Patent No. 7,749,641 ("the '641 Patent") and prior art references relating to its subject matter. I have reviewed the '641 Patent, and relevant excerpts of the prosecution history of the '641 Patent. I have also reviewed the following references:

- APPLE-1004 Japanese Patent Publication No. 2001-167763 ("Yamaki")
- APPLE-1005 Japanese Patent Publication No. H09-293536 ("Sakamoto")

- APPLE-1006 Japanese Patent Publication No. H07-122298 ("Yasunami")
- APPLE-1007 Japanese Patent Publication No. JP2000260472A ("Koyama")
- APPLE-1008 PCT Publication No. WO 01/29920 ("Richardson")
- APPLE-1009 U.S. Patent No. 5,858,573 ("Abraham")
- APPLE-1012 "High-voltage lithium cathode materials." Hiroo Kawai, Mikito Nagata, Hisashi Tukamoto, Anthony R. West. Journal of Power Sources 81-82 (1999) 67-72 ("Kawai")
- APPLE-1014 "Effect of Li2CO3 additive on gas generation in lithium-ion batteries." Jee-Sun Shin, Chi-Hwan Han, Un-Ho Jung, Shung-Ik Lee, Hyeong-Jin Kim, Keon Kim. Journal of Power Sources 109 (2002) 47-52 ("Shin")
- APPLE-1015 David Linden. Handbook of Batteries. Second ed. McGraw-Hill, Inc., 1995. ("Handbook")
- APPLE-1016 U.S. Patent No. 5,192,629 ("Guyomard")
- APPLE-1017 U.S. Patent No. 6,280,881 ("Wendsjo")
- APPLE-1018 U.S. Patent Pub. No. 2002/0012830 ("Uemura")
- APPLE-1019 U.S. Patent No. 6,506,518 ("Kobayashi")

- APPLE-1020 "Mathematical Modeling of the Lithium Deposition Overcharge Reaction in Lithium-Ion Batteries Using Carbon-Based Negative Electrodes." Pankaj Arora, Marc Doyle, Ralph E. White. Journal of the Electrochemical Society 146 (10) 3543-353 ("Arora")
- APPLE-1021 JPH0562712 ("Satoshi")
- APPLE-1022 JPH10172608 ("Ijiri")
- APPLE-1023 U.S. Patent No. 5,487,960 ("Tanaka")
- APPLE-1024 U.S. Patent No. 6,372,387 ("Kawakami")
- APPLE-1025 U.S. Patent No. 5,587,133 ("Amatucci")
- APPLE-1026 U.S. Patent No. 5,278,000 ("Huang")
- APPLE-1027 European Patent Specification EP 0 746 050 B1 ("Shimizu")
- APPLE-1028 U.S. Patent No. 6,172,482 ("Eguchi")
- APPLE-1029 U.S. Patent No. 6,577,105 ("Iwaizono")
- APPLE-1030 U.S. Patent No. 5,763,119 ("Adachi")

4. I have also reviewed various supporting references and other documentation as further noted in my opinions below.

5. Counsel has informed me that I should consider these materials through the lens of one of ordinary skill in the art related to the '641 Patent at the time of the earliest possible priority date of the '641 Patent, and I have done so

during my review of these materials. The ’641 Patent claims the benefit of priority to a Chinese application filed September 28, 2001 (“the Critical Date”). Counsel has informed me that the Critical Date represents the earliest possible priority date to which the challenged claims of ’641 Patent are entitled, and I have therefore used that Critical Date in my analysis below.

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

7. In writing this declaration, I have considered the following: my own knowledge and experience, including my work experience in the fields of electrochemical engineering, battery design, cell manufacturing, and application engineering, my experience in creating and managing organizations devoted to those subjects, and my experience working with other experts in those fields. In addition, I have reviewed and analyzed various publications and patents, in addition to the materials I cite in this declaration.

8. My opinions, as explained below, are based on my education, experience, and expertise in the fields relating to the ’641 Patent. Unless otherwise stated, my testimony below refers to the knowledge of one of ordinary skill in the art as of the Critical Date, or before. Any figures that appear within this document

have been prepared with the assistance of Counsel and reflect my understanding of the ’641 Patent and the prior art discussed below.

II. OVERVIEW OF CONCLUSIONS FORMED

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

- Claims 5-8 and 11-14 are obvious over Yamaki and Sakamoto;
- Clams 9-10 are obvious over Yamaki, Sakamoto, and Abraham;
- Claims 5-14 are obvious over Yasunami and Koyama;
- Claims 9-10 are obvious over Yasunami, Koyama, and Richardson.

III. LEVEL OF ORDINARY SKILL IN THE ART

10. In my opinion, one of ordinary skill in the art relating to, and at the time of, the invention of the ’641 Patent would have held a Bachelor’s degree in chemistry or chemical engineering, with at least 4 years of experience in cell design, electrochemical techniques, and battery materials development. A Master’s Degree could have substituted for some of the relevant experience. Equivalent training with battery specific experience could accommodate other degreed fields.

11. Based on my experience, I have an excellent understanding of the capabilities of one of ordinary skill relevant to this discussion. I have

hired, trained, and managed many such persons over the course of my career. Based on my knowledge, skill, and experience, I have an understanding of the capabilities of one of ordinary skill. Furthermore, I possess those capabilities myself and did so for some years prior to 2001.

IV. THE '641 PATENT

A. Overview

12. The '641 Patent, relating to secondary lithium ion cells or batteries, states that "[c]ontrary to this [common] opinion, the inventor unexpectedly found that the efficacy of electrode active material is greatly increased by increasing the charge cut-off voltage and property [sic] adjusting the ratio of positive electrode material to negative electrode material of single lithium ion cell." APPLE-1001 (the '641 Patent), 3:3-12. Despite this purported discovery, though, the claims of the '641 Patent merely recite what was already well-known in the art: a battery having a charge cut-off voltage in a certain range, and a ratio of positive to negative electrode material in a certain range. And although the '641 Patent appears to assert that these features lead to desirable, and unexpected, performance characteristics, its experimental results fail to demonstrate that the recited ranges are in any way critical to the performance of lithium secondary batteries.

V. PRIOR ART ANALYSIS

A. [GROUND 1A] – Claims 5-8 and 11-14 are obvious over Yamaki and Sakamoto

1. Overview of Yamaki

13. Yamaki describes a "lithium secondary battery with high energy density and excellent cycle characteristics." APPLE-1004 (Yamaki), Abstract.

14. Yamaki's battery includes "a Co-based oxide to be used as the positive electrode active material and a conductive material to be used in the positive electrode." APPLE-1004 (Yamaki), [0012]. Yamaki explains that by including a positive electrode of this composition in a battery, "the maximum potential of the positive electrode during charging can be from 4.4 V to 4.8 V in terms of metallic Li while also suppressing deterioration in the reversibility of charging and discharging as more charging and discharging cycles are performed." APPLE-1004 (Yamaki), [0014].

15. The positive electrode of Yamaki's battery "contain[s] a Co-based oxide including Li and Co as a positive electrode active material," where "the Co-based oxide contains at least one element selected from Mg, Al, Mn, Ti, and Sr." APPLE-1004 (Yamaki), [0013]. Yamaki explains that the presence of these elements in the Co-based oxide "has the effect of suppressing deterioration in the reversibility of charging and discharging due to a phase transition occurring in the Co-based oxide." *Id.*, [0018]; *see* [0008] ("A factor in cycle deterioration" of

"lithium secondary batteries of the prior art that use LiCoO2 as the positive electrode active material" (*id.*, [0006]) "is that when the potential of the positive electrode exceeds 4.3 V relative to metallic Li, the crystal structure of the LiCoO2 undergoes a rhombohedral-to-monoclinic phase transition, impairing the reversibility of Li ion insertion and ejection."). Yamaki further explains that this suppression of deterioration in the charging behavior of the battery "is because the added element is believed to replace Co or Li or form a solid solution in the crystal structure of the Co-based oxide, slightly distorting the ideal layered crystal structure of the rhombohedral LiCoO2, and acting as points of inhibition in the phase transition." *Id.*, [0019].

16. The positive electrode of Yamaki's battery also includes "a conductive material whose main component is carbon." APPLE-1004 (Yamaki), [0014]. This material "is amorphous carbon or at least a material whose main component is carbon with amorphous carbon in the surface layer." *Id.*, [0024]; *see* Abstract ("the surface layer of the conductive material has an amorphous carbonaceous material"), [0025] ("the conductive material may be formed entirely of amorphous carbon, but an amorphous carbon layer on the surface of a highly crystalline carbon material with high electrical conductivity such as graphite is preferred from the standpoint of conductivity"). The presence of this carbon-containing material "can suppress the oxidative decomposition of the organic

electrolyte even at high charging potential," which also contributes to the ability of the battery to operate at high maximum potential. *Id.*

17. The negative electrode of Yamaki's battery is "a carbon material or a material capable of intercalating Li or forming a compound." APPLE-1004 (Yamaki), [0030]. For instance, the "carbon material" of the negative electrode can be "natural graphite, artificial graphite, or a carbon material obtained by heat-treating heavy oil, coal tar, or pitch-based fibers before being carbonized and pulverized." *Id.*, [0031].

18. Yamaki's battery also includes other components that are generally standard elements of secondary lithium batteries. For instance, the battery includes a separator: "a porous film made of an organic polymer material such as polyethylene or polypropylene, or a gel organic polymer compound is interposed between the positive electrode and the negative electrode to provide insulation." APPLE-1004 (Yamaki), [0032]. It also includes an electrolyte, which is "[a]n organic electrolyte common in the art can be used, for example, such as a lithium salt dissolved in an organic solvent." *Id.*, [0033].

19. The electrode compositions of Yamaki's battery improve "the discharge capacity of the positive electrode active material … by setting the maximum potential of the positive electrode during charging to 4.4 V or more in terms of metallic lithium," *e.g.*, "from 4.4 V to 4.8 V." APPLE-1004 (Yamaki),

[0014]-[0015]. Yamaki explains that as a result of this high maximum potential value, "the amount of positive electrode active material in the battery can be reduced, and the amount of negative electrode active material can be increased," which in turn allows "a lithium secondary battery with higher energy density [to] be obtained in a limited battery volume." *Id.*, [0015]. Importantly, Yamaki notes that these improvements are achieved "while also suppressing deterioration in the reversibility of charging and discharging as more charging and discharging cycles are performed." *Id.*, [0014].

20. Generally, Yamaki explains that lithium secondary batteries of this composition have "an initial discharge capacity for the positive electrode active material of 160 mAh/g or more, and a capacity retention rate of 80% or more when charging and discharging have been repeated 10 times." APPLE-1004 (Yamaki), [0028]. Yamaki's experiments, though, reveal even better performance than what this statement indicates. *See* APPLE-1004 (Yamaki), [0056]-[0057]. For instance, tests demonstrated that lithium secondary batteries with positive electrode active materials composed of Co-based oxides with one or more of Al, Mg, Mn, Sr, or Ti retained between 93-98% capacity after 10 cycles when charged to 4.5 V, as indicated in the highlighted boxes of Table 1, reproduced below. *See* APPLE-1004 (Yamaki), [0057]. By comparison, a "secondary battery produced using a conventional additional element-free Co-based oxide ($LiCoO_2$)," charged to 4.3 V,

retained only 75% capacity after 10 cycles, as shown in the "C. Ex. 1" line of the table below. APPLE-1004 (Yamaki), [0050]; *see* [0058] ("In the configuration of the lithium secondary battery of the prior art shown in Comparative Example 1, setting the charge potential to 4.5 V resulted in low capacity retention and poor cycle characteristics even though the positive electrode active material had a high initial discharge capacity.").

Table 1

	Positive Electrode Active Material	Conductive Material	Charged Positive Electrode Potential (V: Metal Li Reference)	Initial Battery Capacity (mAh)	Initial Discharge Capacity of Positive Electrode Material (mAh/g)	Capacity Retention After 10 Cycles (%)
Ex. 1	$LiCo_{0.97}Al_{0.03}O_2$	(A)	4.5	3.99	182	95
Ex. 2	$LiCo_{0.99}Al_{0.01}O_2$	(A)	4.5	4.12	188	94
Ex. 3	$LiCoMg_{0.03}O_2$	(A)	4.5	4.17	190	98
Ex. 4	$LiCo_{0.97}Mn_{0.03}O_2$	(A)	4.5	4.05	184	97
Ex. 5	$LiCo_{0.99}Sr_{0.01}O_2$	(A)	4.5	4.24	193	93
Ex. 6	$LiCo_{0.97}Al_{0.02}Mn_{0.01}O_2$	(A)	4.5	4.00	182	95
Ex. 7	$LiCo_{0.98}Ti_{0.02}Mg_{0.01}O_2$	(A)	4.5	4.07	184	95
Ex. 8	$LiCo_{0.97}Al_{0.01}Ti_{0.01}Mn_{0.01}O_2$	(A)	4.5	4.00	182	93
Ex. 9	$LiCo_{0.97}Al_{0.03}O_2$	(A)	4.4	3.86	165	97
Ex. 10	$LiCo_{0.97}Al_{0.03}O_2$	(A)	4.6	4.19	211	89
C. Ex. 1	$LiCoO_2$	(B)	4.5	4.15	189	75
C. Ex. 2	$LiCoO_2$	(B)	4.3	3.82	157	95

Yamaki, Table 1 (annotated)

2. Overview of Sakamoto

21. Sakamoto describes "a non-aqueous electrolyte secondary battery" having an electrode composition that results in "a highly reliable positive and negative electrode configuration with excellent charge-discharge cycle performance." APPLE-1005 (Sakamoto), [0001].

22. Sakamoto explains that in existing lithium secondary batteries, if "the reversible capacities on the positive electrode side and the negative electrode side are substantially equal, the charge cycle performance is insufficient due to significant deterioration from charging and discharging" and due to "deposit of lithium … on the negative electrode during overcharge." APPLE-1005 (Sakamoto), [0006]. Sakamoto also notes that "the reversible capacity balance between the negative electrode and the positive electrode has a big impact on these problems." *Id.*

23. Sakamoto aims to address these problems through the design of its battery. Specifically, Sakamoto describes secondary lithium batteries in which "[t]he reversible capacities of the positive electrode and the negative electrode are made to satisfy the range of 1.05 < negative electrode/positive electrode <= 1.30." APPLE-1005 (Sakamoto), [0012]. Sakamoto explains that a battery with a reversible capacity balance in this range does not experience "lithium deposition on the positive electrode … during either overcharge or overdischarge, and a battery with especially low cycle deterioration can be obtained." *Id.*

24. Sakamoto's approach is generally applicable to various "positive electrode active materials," which are "preferably a (noble metal) active material having an electrode potential of 3 V to 4 V or higher." APPLE-1005 (Sakamoto), [0010]; *see* [0009]. For instance, like Yamaki's positive electrode active material,

the positive electrode active material of Sakamoto's secondary battery can be "a transition metal oxide containing lithium such as Li_xCoO_2, Li_xNiO_2, $Li_xMn_2O_4$ or $Li_aM_bL_cO_d$," where "M is one or more transition metals such as Co, Ni, Mn, Fe, Ti" and "L is one or more metals selected from B, Al, In, Si, Ge, Sn, Pb, Mg, Zn, Cu and P." *Id.*, [0012]. The "negative electrode active material" of Sakamoto's battery can be "a silicon oxide or carbonaceous material," such as an "active material having a low electrode potential of 1 V or less with respect to metallic lithium." *Id.*, [0010], [0012]; *see* claim 2.

25. Sakamoto's battery is compatible with conventional electrolytes: "[e]xamples of electrolytes that can be used include organic non-aqueous electrolytes obtained by dissolving a lithium ion dissociative salt such as $LiClO_4$, $LiPF_6$, $LiBF_4$, $LiCF_3SO_3$, $LiC(SO_2CF_3)_3$, $LiN(SO_2CF_3)_2$, etc. serving as a supporting electrolyte in a non-aqueous organic solvent.

26. Sakamoto presents experimental data on the performance of batteries having LiCoO2 as the positive electrode active material and silicon monoxide (SiO) as the negative electrode active material. *See* APPLE-1005 (Sakamoto), [0015], [0016] ("any material can be used for the negative electrode and the positive electrode as long as it is capable of occluding and releasing lithium as described above"). Specifically, Sakamoto prepared several battery samples, each having the same positive and negative electrode compositions and with "a

reversible capacity balance" (negative electrode/positive electrode) ranging from 0.94 to 1.50. While Sakamoto does not describe how these various reversible capacity balances were achieved, a POSITA would have understood that given the consistent composition (and thus consistent capacity per unit mass) across the samples, a target capacity balance is achieved by adjusting the mass of either the positive electrode active material or the negative electrode active material in the sample. This is because the capacity per unit mass of an electrode active material times the mass of that electrode active material in the battery gives the capacity of that electrode active material in the battery: the ratio of the capacities can thus be tailored by adjusting the mass of each electrode active material.

27. Sakamoto's battery samples "were subjected to a charge-discharge cycle with a constant current/constant voltage of 400 mA, a charge upper limit voltage of 4.2 V, a charging time of 2.5 hours, a constant current of 400 mA, and a discharge final voltage of 2.7 V." APPLE-1005 (Sakamoto), [0016]. The "cycle characteristics" were monitored up to 300 charge-discharge cycles to measure capacity retention. *Id.*, [0015]-[0016]. These experiments demonstrated that "a difference in capacity retention was observed depending on the reversible capacity balance of the negative electrode/positive electrode." *Id.*, [0016]. For instance, "Battery A with a reversible capacity balance of 0.94"—that is, below the range of desirable capacity balance values identified by Sakamoto—"fell below 70% after

300 cycles" and was "found to have small deposits of lithium between the separator and the negative electrode plate." *Id.*, [0016]. "Battery D with a reversible capacity balance of 1.18"—that is, inside of the range of desirable capacity balances—"remained above 70%." *Id.*. "Battery F and Battery G, which have a reversible capacity balance greater than 1.3"—that is, above the range of desirable capacity balances—"experience less cycle deterioration, but their capacities are lower after the initial cycle." *Id.* The batteries "with a capacity balance greater than 1.05 were found not to have deposits of lithium." *Id.*

28. I note that Sakamoto does not specify particular limits on the applicability of its teachings regarding the reversible capacity balance of lithium secondary batteries. For instance, while Sakamoto describes batteries with operation at certain charge cut-off voltages (*see id.*, [0016]), it does not preclude the implementation of its reversible capacity balance in batteries operated to different charge cut-off voltages. Moreover, Sakamoto describes implementation of its reversible capacity balance in batteries that are generally consistent with the composition and design objectives of Yamaki's batteries, as I discuss below in Paragraphs [37]-[40] and [49]-[54].

3. The combination of Yamaki and Sakamoto

29. In the Yamaki-Sakamoto combination, a lithium secondary battery is based on the lithium secondary cell described by Yamaki, and includes positive

and negative electrode active materials in amounts that satisfy a reversible capacity balance based on the teachings of Sakamoto.

30. Specifically, the composition of the electrodes and electrolyte of the combination battery is based on the teachings of Yamaki. For instance, the combination battery includes "a positive electrode containing a Co-based oxide including Li and Co as a positive electrode active material," where "the Co-based oxide contains at least one element selected from Mg, Al, Mn, Ti, and Sr," based on the teachings of Yamaki. APPLE-1004 (Yamaki), [0013]. Notably, this positive electrode active material composition is consistent with the composition of Sakamoto's positive electrode active material. *See* APPLE-1005 (Sakamoto), [0012].

31. The positive electrode in the combination battery also includes "a conductive material whose main component is carbon," where "at least the surface layer of the conductive material has an amorphous carbonaceous material," again based on the teachings of Yamaki. APPLE-1004 (Yamaki), [0013]. Again, this is consistent with the composition of Sakamoto's positive electrode, which includes "graphite serving as a conductive agent." APPLE-1005 (Sakamoto), [0015].

32. The negative electrode active material in the combination battery is "a carbon material," as taught by Yamaki and again consistent with the composition

of Sakamoto's battery. APPLE-1004 (Yamaki), [0030]; *see* APPLE-1005 (Sakamoto), [0010].

33. The electrolyte is "an organic electrolyte … such as a lithium salt dissolved in an organic solvent," *e.g.*, as listed by Yamaki. APPLE-1004 (Yamaki), [0033]. Here, too, this electrolyte composition is consistent with the compositions described for Sakamoto's battery. *See* APPLE-1005 (Sakamoto), [0013] .

34. As taught by Yamaki, the combination battery can operate with "a maximum potential of the positive electrode during charging … from 4.4 V to 4.8 V." APPLE-1004 (Yamaki), [0014]. For instance, as taught by Yamaki, the presence of the additional elements in the Co-based oxide of the positive electrode active materail "suppress[es] deterioration in the reversibility of charging and discharging" by inhibiting a phase transition in the positive electrode active material, while the carbon-containing conductive material "suppress[es] the oxidative decomposition of the organic electrolyte." APPLE-1004 (Yamaki), [0018], [0024]. These physical effects of the composition of the positive electrode in the combination battery are a contributor to the ability of the battery to operate at these high maximum potentials. For instance, when a positive electrode is overcharged, the desired full charge material (*e.g.*, CoO2) can be further oxidized.

These higher voltage products do not exhibit the reversible chemistry of the standard charged state, leading to reduced cell capacity and short cycle life.

35. The amounts of the positive electrode active material and negative electrode active material in the combination battery are set based on the teachings of Sakamoto: "[t]he reversible capacities of the positive electrode and the negative electrode are made to satisfy the range of 1.05 < negative electrode/positive electrode <= 1.30." APPLE-1005 (Sakamoto), Abstract. That is, the electrode active materials are provided such that there is more capacity provided by the negative electrode active material than by the positive electrode active material. The successful implementation of this ratio in the combination is enabled, at least in part, by "setting the maximum potential of the positive electrode during charging to 4.4 V or more" (*e.g.*, as taught by Yamaki, and as implemented in the combination battery, which I discussed in the previous paragraph). APPLE-1004 (Yamaki), [0015]. Specifically, the high maximum potential improves "the discharge capacity of the positive electrode active material," which in turn allows "the amount of negative electrode active material [to] be increased," *e.g.*, allows for integration of Sakamoto's teachings into the combination battery. APPLE-1004 (Yamaki), [0015]. By providing the positive and negative electrode active materials in the combination battery in amounts based on the capacity balance described by Sakamoto, the "charge-discharge cycle performance" of the

combination battery is further improved, and safety is enhanced. APPLE-1005 (Sakamoto), Abstract; *see* [0008].

4. Reasons for the combination

36. It is my opinion that a POSITA would have been motivated to provide positive electrode active material and negative electrode active material in the combination battery in amounts sufficient to satisfy the capacity balance described by Sakamoto for at least the following reasons.

37. The first reason why a POSITA would have been motivated to pursue this implementation is to achieve the improved "cycle performance" offered by batteries having the materials ratio described by Sakamoto, particularly because this objective is consistent with the objective of Yamaki—"[t]o provide a lithium secondary battery with … excellent cycle characteristics". APPLE-1005 (Sakamoto), [0017]; APPLE-1004 (Yamaki), Abstract. Sakamoto describes that in a battery having a capacity balance in "the range of 1.05 < negative electrode/positive electrode <= 1.30," "lithium ions can be sufficiently occluded in the negative electrode during charging or overcharging, and deposition of lithium is less likely to occur. As a result, a high-performance battery can be obtained that experiences ***less deterioration after repeated charging and discharging***." APPLE-1005 (Sakamoto), [0007]; *see* [0008] ("By setting the reversible capacity balance of the negative electrode and the positive electrode to the range in the

present invention, lithium is not deposited even when overcharged"), [0012] ("lithium deposition on the positive electrode does not occur either during overcharge or overdischarge, and a battery with especially low cycle deterioration can be obtained"), Abstract ("Lithium ions can be occluded adequately in the negative electrode during charging and overcharging, deposits of lithium are less likely to occur, and the deterioration that occurs during repeated charging and discharging is reduced."), [0006]. Indeed, it was well known that batteries having more negative electrode capacity than positive electrode capacity exhibit good cycle life characteristics. For instance, Arora describes that "most lithium-ion cells are manufactured with excess negative electrode (excess capacity)" so that "at the end of the charge the negative electrode will still be undergoing the normal charging process (lithium intercalation)" rather than "the lithium deposition side reaction on the negative electrode," which can eventually obscure the negative electrode, reducing the cycle life of the battery. APPLE-1020 (Arora), 5.

38. Other examples demonstrating that batteries having more negative electrode capacity than positive electrode capacity exhibit good cycle life characteristics are as follows. For instance, Satoshi describes that in a "non-aqueous electrolyte secondary battery … [having] a theoretical capacity ratio betweeen the positive electrode and the negative electrode [] in the range of 1:1 to 1.3," "the decrease in cycle life due to the decrease in negative electrode capacity

can be suppressed." APPLE-1021 (Satoshi), [0011]. Kobayashi explains that "it is necessary to set the -/+ ratio in the range of at least 1.3 and at most 1.8. According to the present invention, since the deterioration of the positive and negative electrodes can be suppressed, the charging/discharging cycle life and preservation life can be improved." APPLE-1019 (Kobayashi), 3:24-28. Uemura states that "a capacity balance ratio B/A of the total capacity B of the negative electrode material to the total capacity A of the positive electrode material is preferably fixed at a range of 1 to 1.5. If the capacity balance ratio B/A is below 1, lithium ion holding sites on the negative electrode material become insufficient. As the result, branch-shaped or needle-shaped crystal (dendrite crystal) tends to occur during the charge to cause a short circuit phenomenon between the positive electrode and the negative electrode." APPLE-1018 (Uemura), [0033]. The generation of dendrite crystals causes deterioration of the function of the electrode, thus reducing cycle life.

39. Cells with higher negative electrode capacity will continue to store Lithium as intercalated Li atoms with a zero or metallic valence, safely allowing the cathodic current to flow while the positive electrode, having utilized all of the available active material, climbs in voltage. If the negative electrode did not contain this overbalance of Lithium storage capacity, the current would result in

Lithium metal plating, possibly in dendritic form, short circuiting and damaging the cell.

40. In keeping with Yamaki's objective to provide a battery with good cycle characteristics, a POSITA would have been motivated to improve further the cycle characteristics of the battery, and would have adjusted the capacity balance of the positive and negative electrodes to achieve this objective, *e.g.*, based on Sakamoto's teachings. APPLE-1004 (Yamaki), Abstract; *see* [0001], [0012], [0014], [0016], [0018], [0059]-[0060].

41. <u>The second reason</u> why a POSITA would have been motivated to implement the combination battery according to a capacity balance such as that described by Sakamoto is because using such a capacity balance "is safer," *e.g.*, "due to low risk of heat being generated and an explosion occurring." APPLE-1005 (Sakamoto), [0007], [0008]; *see* [0017] ("A higher level of safety because there is no deposit of lithium"). Again, a POSITA would have been further encouraged to pursue this implementation because Yamaki already indicates that such a ratio is appropriate. *See* APPLE-1004 (Yamaki), [0015]. More generally, it was well-known that batteries having more capacity in the negative electrode than in the positive electrode were safer. For instance, Arora describes that "most lithium-ion cells are manufactured with excess negative electrode (excess capacity)" so that "at the end of the charge the negative electrode will still be

undergoing the normal charging process (lithium intercalation)" rather than "the lithium deposition side reaction on the negative electrode," which can eventually obscure the negative electrode, raising the risk of short circuits, which can create safety hazards. APPLE-1020 (Arora), 5. Similarly, Ijiri describes that a battery where "the effective capacity of the negative electrode active material layer is 100-110 with respect to the effective capacity of 100 of the positive electrode active material layer" enables "the object of the present invention" to be achieved, namely, to provide a "safe" battery. APPLE-1022 (Ijiri), [0007], [0017]. As I noted above, Uemura describes the growth of dendrites if there is insufficient negative electrode capacity, which can "cause a short circuit phenomenon." APPLE-1018 (Uemura), [0033].

42. During any battery charge or discharge process, the current is necessarily conducted through both electrodes; for each reaction occurring at the negative electrode, one must occur at the positive. Each electron that leaves the positive electrode during charge is balanced by one entering the negative electrode. For this reason, it is important to maintain a surfeit of capacity at the negative during charge, as the reactions that would occur there if no additional Lithium could be intercalated, are more dangerous than those that would occur at the positive if no additional cobaltite, for example, could be oxidized. Specifically, if no additional Lithium could be stored as a zero valance Li atom within the

carbon matrix, the entering electrons would result in metallic Lithium forming at the surface of the carbon. Metallic Lithium is formed when a lack of space for atoms housed in the carbon matrix causes solid blocks of Lithium atoms to form. This frequently leads to dendritic metal formation – spikes of Lithium that reach toward the opposing electrode, penetrating the separator and causing a short circuit and a possible fire.

43. The third reason why a POSITA would have been motivated to pursue this implementation is to obtain a battery having "[r]esistance to overcharging," which is achievable with batteries satisfying a capacity balance such as that described by Sakamoto—and again, which is consistent with the indications already provided by Yamaki. APPLE-1005 (Sakamoto), [0017]; *see* APPLE-1004 (Yamaki), [0015]).

44. By maintaining an excess capacity, or storage space within the carbon of the negative electrode, the undesirable effects of overcharging can be directed to the positive electrode. The voltage will increase as the scarcity of readily available active material also results in higher IR or resistive losses. This reduces the overall energy efficiency of the cell but does not cause immediate damage. When the potential of the positive electrode does increase, it can result in permanent capacity loss due to over-oxidation of the active material, but that reaction, although

undesirable, is far preferable to an immediate safety issue that could occur if the negative electrode did not possess the excess capacity.

45. The fourth reason why a POSITA would have been motivated to pursue this combination is that a POSITA reviewing Yamaki's disclosure would have understood that an increase in the amount of negative electrode active material is suitable for Yamaki's battery. Yamaki explains that in a battery implementing a "maximum potential" during charging of "from 4.4 V to 4.8 V," Yamaki explains that "the amount of positive electrode active material in the battery can be reduced, and ***the amount of negative electrode active material can be increased***." APPLE-1004 (Yamaki), [0014]-[0015]. Yamaki does not provide any specific disclosure regarding how much of an increase in the amount of negative electrode active material to implement. Thus, a POSITA would have explored batteries having known ratios of positive to negative materials, and specifically would have looked to the teachings of Sakamoto—which also seeks to improve cycle life in lithium secondary batteries—for implementation details. Sakamoto, in the context of a materials system consistent with that of Yamaki's battery, describes a specific range of capacity ratios that satisfy the general guidance already provided by Yamaki. *See* APPLE-1004, [0014]; APPLE-1004 (Yamaki), [0015].

46. The fifth reason why a POSITA would have been prompted to pursue this implementation is because this combination is just the application of known techniques (specifically, a secondary lithium ion battery having more negative electrode capacity than positive electrode capacity) to an already-known system (specifically, Yamaki's secondary lithium ion battery), and it produces predictable results (specifically, a secondary lithium ion battery having negative electrode active material and positive electrode active material in amounts to provide a specific capacity balance). In fact, the '641 Patent confirms that its batteries are just conventional batteries, stating that "the present method … can used to the secondary lithium ion cell or battery prepared according to the method of the prior art, such as the present commercialized secondary lithium ion cell or battery." APPLE-1001 ('641 Patent), 7:51-56; *see* 1:24-28, 8:37-55. And as I have explained here, both batteries with charge cut-off voltages in the claimed range and batteries with ratios of positive to negative electrode materials in the claimed range were encompassed in the realm of such conventional batteries.

47. It is my opinion that a POSITA would have expected to be successful in providing positive electrode active material and negative electrode active material in the combination battery in amounts sufficient to satisfy the capacity balance described by Sakamoto for at least the following reasons.

48. The first reason why a POSITA would have expected success in providing positive and negative active material in the combination battery to satisfy the capacity balance described by Sakamoto is because Yamaki already indicates that increasing the amount of negative electrode active material in its battery is appropriate. See APPLE-1004 (Yamaki), [0015]. Notably, Yamaki suggests this increase in the amount of negative active material specifically in the context of a "maximum potential" (*e.g.*, a charge cut-off voltage; *see* [5a], below) "from 4.4 V to 4.8 V," which indicates that providing more negative than positive electrode capacity in the context of a battery charged to a maximum potential 4.4 V to 4.8 V is a feasible endeavor. APPLE-1004 (Yamaki), [0014].

49. The second reason why a POSITA would have expected success in combining teachings based on both Yamaki and Sakamoto is that the two disclosures share the common objective of improving cycle life of a secondary lithium ion battery. *See* APPLE-1004 (Yamaki), Abstract ("To provide a lithium secondary battery with … excellent cycle characteristics"), [0001], [0012], [0014], [0016], [0018], [0059]-[0060]; APPLE-1005 (Sakamoto), [0017] ("The present invention … has the following effects: A. Excellent cycle performance"), [0006]-[0008], [0012]. Given this shared goal, in the common context of a lithium secondary battery of similar composition, a POSITA would have had a reasonable

expectation that the teachings of Sakamoto would be applicable to a battery such as the one described by Yamaki.

50. The third reason why a POSITA would have expected success in combining teachings based on both Yamaki and Sakamoto is because both describe secondary lithium ion batteries with similar materials systems. This is an indication that the teachings of Sakamoto are compatible with, and applicable to, the battery described by Yamaki. And as I have noted previously, neither Yamaki nor Sakamoto gives any indication that the features of their respective batteries would preclude their being integrated with features of the other. In fact, it is the opposite: Yamaki explicitly encourages a POSITA to look toward a ratio of positive to negative material that is consistent with the teachings of Sakamoto. *See* APPLE-1004 (Yamaki), [0014]; APPLE-1005 (Sakamoto), [0012].

51. For instance, both Yamaki and Sakamoto describe a positive electrode including lithium cobalt oxide as a positive electrode active material and a carbon-based conductive material. *See* APPLE-1004 (Yamaki), Abstract, [0016], [0013], [0018], [0020]-[0023]; APPLE-1005 (Sakamoto), [0012], [0015]. Specifically, Yamaki's "Co-based oxide serving as the positive electrode active material" is "represented by the general formula $Li_xCo_yA_zO_2$ (where A is at least one element selected from Mg, Al, Mn, Ti and Sr." APPLE-1004 (Yamaki), [0020]. Similarly, Sakamoto lists positive electrode active materials including "a transition metal

oxide containing lithium such as Li_xCoO_2, Li_xNiO_2, $Li_xMn_2O_4$ or $Li_aM_bL_cO_d$" where "M is one or more transition metals such as Co, Ni, Mn, Fe, Ti, and L is one or more metals selected from B, Al, In, Si, Ge, Sn, Pb, Mg, Zn, Cu and P." APPLE-1005 (Sakamoto), [0012]. At least some of the lithium cobalt oxides described by both Yamaki and Sakamoto are the same, such as Yamaki's $Li_xCo_yA_zO_2$ where A is Mg or Al, and Sakamoto's $Li_aM_bL_cO_d$ where M is Co and L is Mg or Al.

52. Both Yamaki and Sakamoto also have a carbonaceous material as the negative electrode active material. *See* APPLE-1004 (Yamaki), [0030]-[0031] ("The carbon material" of the negative electrode "can be, for example, natural graphite, artificial graphite, or a carbon material obtained by heat-treating heavy oil, coal tar, or pitch-based fibers before being carbonized and pulverized."); APPLE-1005 (Sakamoto), [0012]. Importantly, Sakamoto indicates that its disclosure is applicable to "any material … for the negative electrode as long as it is capable of occluding and releasing lithium," which is true for Yamaki's negative electrode material. APPLE-1005 (Sakamoto), [0016]; *see* APPLE-1004 (Yamaki), [0030].

53. And both Yamaki and Sakamoto also describe overlapping lists of organic electrolyte with dissolved lithium salt in their secondary lithium ion batteries. *See* APPLE-1004 (Yamaki), [0033]; APPLE-1005 (Sakamoto), [0013].

For instance, Yamaki describes "organic solvents include carbonates such as ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), dimethyl carbonate (DMC), methylethyl carbonate (MEC) and diethyl carbonate (DEC), esters such as γ-butyrolactone (γ-BL) and methyl acetate (EA), and ethers such as 1,3-dioxolane and 1,2-dimethoxyethane" and "Lithium salts dissolved in the organic solvent include $LiClO_4$, $LiCF_3SO_3$, $LiPF_6$, $LiBF_4$, $LiAsF_6$." APPLE-1004 (Yamaki), [0033]-[0035]. Sakamoto describes many of these same compositions, including at least γ-butyrolactone, propylene carbonate, ethylene carbonate (EC), butylene carbonate, dimethyl carbonate, diethyl carbonate for the organic electrolyte and at least $LiClO_4$, $LiPF_6$, $LiBF_4$, $LiCF_3SO_3$ for the dissolved lithium salt. APPLE-1005 (Sakamoto), [0013].

54. Given the overlap in materials between Yamaki's and Sakamoto's secondary lithium ion batteries, a POSITA would have expected the teachings of Sakamoto to be relevant to, and combinable with, the teachings of Yamaki. And as I have noted previously, neither Yamaki nor Sakamoto gives any indication that the features of their respective batteries would preclude their being integrated with features of the other. In fact, it is the opposite: Yamaki explicitly encourages a POSITA to look toward a ratio of positive to negative material that is consistent with the teachings of Sakamoto. *See* APPLE-1004 (Yamaki), [0015].

55. The fourth reason why a POSITA would have expected success is that secondary lithium ion batteries having more negative than positive electrode capacity were well-known in the art. *See* APPLE-1020 (Arora), 5 ("To minimize the lithium deposition reaction most lithium-ion cells are manufactured with excess negative electrode (excess capacity)"); APPLE-1021 (Satoshi), [0011] ("The non-aqueous electrolyte secondary battery of the present invention is designed so that the theoretical capacity ratio between the positive electrode and the negative electrode is in the range of 1:1 to 1.3"); APPLE-1007 (Koyama), [0006] ("a capacity ratio (B/A) between the electrode capacity A of the positive electrode and the electrode capacity B of the negative electrode is 1.3<(B/A)<=2.5"). Indeed, as I discussed previously, such batteries have numerous advantages, *e.g.*, in terms of safety, cycle life, and other performance characteristics. Moreover, it was also well-known in the art to operate secondary lithium ion batteries with a charge cut-off voltage of greater than 4.2 V. *See* APPLE-1023 (Tanaka), 1:25-35 ("Nonaqueous batteries using a lithium-containing transition metal oxide … as a positive electrode active material and a substance capable of intercalating and deintercalating a lithium ion … as a negative electrode active material have a higher cut-off voltage in charging than conventional alkali batteries, usually of from 3.5 to 5 V"); APPLE-1024 (Kawakami), 40:3-14 (describing a "rechargeable lithium battery" in which "the cut-off voltage of the charge was set at 4.5 V");

APPLE-1025 (Amatucci), 2:63-3:3 ("Such a lithium metal cell utilizing a LiCoO2 positive electrode may be reasonably expected to achieve normal operating ranges of about 4.3 to 5.1 V"); APPLE-1012 (Kawai), 1 ("Single cell lithium battery systems can operate over 4.5 V vs. Li/Li^+ on the basis of reversible extraction of lithium from several cathodes based on complex spinel oxides"). Moreover, as I discussed above, Yamaki does not preclude the implementation of its batteries according to Sakamoto's disclosed material ratio, and indeed encourages an implementation consistent with that described by Sakamoto. *See* APPLE-1004 (Yamaki), [0015]. Likewise, Sakamoto does not preclude the use of its material ratio with battery implementations other than that specifically disclosed in Sakamoto; for instance, Sakamoto does not preclude the use of its material ratio in a battery having a charge cut-off voltage in the range described by Yamaki.

5. Analysis

(i) **Claim 5**

[5pre] A secondary lithium ion cell or battery, characterized in that

56. The Yamaki-Sakamoto combination renders obvious a secondary lithium ion cell or battery, for the following reasons.

57. First, the battery based on the Yamaki-Sakamoto combination is a lithium secondary battery, e.g., as described by Yamaki. See APPLE-1004 (Yamaki), Abstract ("lithium secondary battery"), [0001] ("The present invention relates to a lithium secondary battery"), [0013] ("the present invention is a lithium

secondary battery"), [0012]-[0013], [0015], [0017], [0028], [0043], [0045]-[0048], [0060], claim 1.

58. Additionally, the combination battery is a lithium ion battery at least because lithium ions are the charge carriers during charging/discharging cycles of the battery. *See* APPLE-1004 (Yamaki), [0030] ("The negative electrode used in the battery of the present invention is prepared based on a mixture obtained by adding a binder to a carbon material or a material capable of intercalating Li or forming a compound."), [0033] ("An organic electrolyte common in the art can be used, for example, such as a lithium salt dissolved in an organic solvent.").

59. The battery described in Yamaki is described as a Lithium secondary battery. Yamaki identifies the invention as a cell that contains Lithium and can be charged and discharged. There are only two possible constructions that fit this description: Li Ion batteries (cells) and Lithium metal cells. Yamaki clearly describes the negative electrode as being a carbon structure, which can only include Lithium ions as the active material and would not be used in a Lithium metal cell. Additionally, when describing features of its positive electrode, , Yamaki states "the reversibility of inserting and ejecting Li Ions is more fully obtained." APPLE-1004 (Yamaki), [0023]. For both of these reasons, the invention described by Yamaki is a Lithium Ion battery.

[5a] the secondary lithium ion cell or battery has a charge cut-off voltage of greater than 4.2V but less than 5.8 V, and

60. In the combination, Yamaki describes that "the maximum potential of the positive electrode during charging can be from 4.4 V to 4.8 V." APPLE-1004 (Yamaki), [0014]; see [0015]-[0017], [0028], [0052]-[0054], [0057]. This maximum potential is the charge cut-off voltage of the combination battery, as I explain in the next paragraphs.

61. The charge cut-off voltage of a battery is, generally, the maximum voltage to which the battery is configured to be charged. For instance, in "constant current charging," a battery is charged at constant current until reaching its charge cut-off voltage. Once the charge cut-off voltage is reached, charging is stopped, or charging changes to constant voltage charging to complete the charging of the battery. The latter approach is advantageous in that it provides additional capacity to the battery, *e.g.*, as compared to the former approach.

62. Lithium Ion cells are typically charged by a combined constant current/constant voltage technique because this two-step process results in the fastest accumulation of charge that can be accomplished while maintaining a safe voltage. A safe voltage is one that does not damage the cell cycle life or cause overheating or safety concerns. During the initial charge phase, the current is maintained at a constant value (typically limited by the charging equipment or a design value) and the voltage is allowed to "float." The voltage rises as the state of

charge increase, until it reaches a predetermined value. At that point, the voltage is maintained at the predetermined value while the current is allowed to "float" downward, until a time limit or a low current limit is reached, when the charging process is stopped. This two stage process has been found to produce the highest capacity of the cell without exceeding voltages at which damage will occur.

63. In the Yamaki-Sakamoto combination battery, the "maximum potential of the positive electrode" is a charge cut-off voltage at least because this "maximum potential" is the voltage to which the battery is configured to be charged, e.g., the voltage at which the charging changes from constant current charging to constant voltage charging. APPLE-1004 (Yamaki), [0014]. For instance, Yamaki describes that "[t]he maximum potential of the positive electrode during charging corresponds to the potential in constant voltage charging or constant current charging normally required to bring the secondary battery to a fully charged state." Id., [0017]. Given the advantages of the constant current-constant voltage approach to constant current charging, a POSITA would have understood or found obvious that this "maximum potential" of the combination battery (between 4.4 V and 4.8 V), in the context of Yamaki's constant current charging implementation, is the potential at which charging changes from constant current charging to constant voltage charging. Id. That is, the "maximum

potential," as described by Yamaki, is the charge cut-off voltage of the combination battery. *Id.*

64. Indeed, the "maximum potential" of the combination battery is a charge cut-off voltage because it is, more generally, the maximum voltage to which the battery is configured to be charged. APPLE-1004 (Yamaki), [0014]. First, the voltage at which the charging changes from constant current charging to constant voltage charging—the "maximum potential" of the combination battery, as discussed above—is itself the maximum potential to which the battery is configured to be charged, at least because once the battery reaches that voltage in constant current charging mode, the voltage is held constant for further charging. That is, in constant current charging, the battery is not charged to a voltage greater than its "maximum potential." *Id.* Alternatively or additionally, Yamaki explains that "because the voltage of the battery is controlled by the charger during charging, the maximum potential of the positive electrode during charging may be defined in the present invention based on the set charging voltage of the charger." *Id.*, [0017]. To the extent that the voltage set-point of a battery charger can be considered to be a charge cut-off voltage of the battery itself, the "set charging voltage of the charger" is the maximum potential to which the battery is configured to be charged. *Id.* That is, the "maximum potential" as described by Yamaki is the charge cut-off voltage of the combination battery. *Id.*

65. The maximum potential (e.g., the charge cut-off voltage) of the Yamaki-Sakamoto combination battery is "from 4.4 V to 4.8 V," consistent with the teachings of Yamaki. APPLE-1004 (Yamaki), [0014]; see [0015] ("the discharge capacity of the positive electrode active material can be improved by setting the maximum potential of the positive electrode during charging to 4.4 V or more in terms of metallic Li"), [0028] ("A characteristic of the lithium secondary battery in the present invention is … setting the highest potential of the positive electrode during charging to 4.4 V to 4.8 V, preferably 4.4 V to 4.6 V"), [0016] ("The energy density increases as the charge potential of the positive electrode increases, and a potential of up to 4.8 V is theoretically possible. However, from the standpoint of cycle deterioration, the upper limit on the highest potential of the positive electrode in the present invention is preferably 4.6 V or less in terms of metallic Li."), [0052] ("The button-type lithium secondary batteries … were subjected to a charge/discharge test using a repeated cycle in which … the charging positive electrode potential was 4.5 V in terms of metallic Li, the constant voltage was kept at 4.5 V for two hours after reaching 4.5 V"), [0053]-[0054], [0057].

66. Describing the voltage of any electrochemical system requires identifying the reference point from which a voltage is measured. It can be the opposing active electrode or some other known reference. Yamaki describes

voltages "in terms of metallic Li", meaning the stated voltage is the difference between the electrode under investigation (here, the positive electrode) and the standard potential of metallic Lithium. APPLE-1004 (Yamaki), [0015]. In a Li Ion cell, the negative electrode resting potential is very close to a standard metallic Lithium potential, and at low currents, the difference can be negligible. By using this reference, Yamaki has eliminated any doubt or question regarding the system or relative origin of his stated voltages.

67. The charge cut-off voltages that are described by Yamaki (*e.g.*, Yamaki's maximum potential) and incorporated into the combination battery (*e.g.*, 4.4 V to 4.8 V or 4.4 to 4.6 V) fall within the range of charge cut-off voltages recited in claim 5 (greater than 4.2 V but less than 5.8 V). See APPLE-1004 (Yamaki), [0014]-[0016], [0052]-[0054], [0057]. Moreover, Yamaki discloses specific points within the claimed range—e.g., 4.4 V, 4.5 V, and 4.6 V (see APPLE-1004 (Yamaki), [0057]), and 4.4 V, 4.6V, and 4.8 V (the endpoints of the disclosed ranges; see id., [0028]).

68. Thus, based on Yamaki's disclosures of the maximum potential, the Yamaki-Sakamoto combination renders obvious that the combination battery has a charge cut-off voltage of greater than 4.2V but less than 5.8 V. See APPLE-1004 (Yamaki), [0014]-[0016], [0052]-[0054], [0057].

[5b] a ratio of positive electrode material to negative electrode material of the secondary lithium ion cell or battery is from 1:1.0 to 1:2.5, as calculated by a theoretic capacity with a charge cut-off voltage set at 4.2 V.

Positive and negative electrode materials in the combination battery

69. The battery of the Yamaki-Sakamoto combination includes positive electrode material, e.g., based on the positive electrode active material of Yamaki's battery. For instance, in the combination, Yamaki describes "a positive electrode 1 containing a Co-based oxide including Li and Co as a positive electrode active material," where "[t]he Co-based oxide in the present invention contains at least one element selected from Mg, Al, Mn, Ti and Sr." APPLE-1004 (Yamaki), Abstract, [0018]; *see* [0016] ("The positive electrode potential is specified in terms of the physical properties of the Co-based oxide serving as the positive electrode active material"), [0020]-[0023] (The Co-based oxide serving as the positive electrode active material in the present invention can be represented by the general formula $Li_xCo_yA_zO_2$ (where A is at least one element selected from Mg, Al, Mn, Ti and Sr, and x, y, and z satisfy $0.9 \leq x \leq 1.1$, $0.97 \leq y \leq 1.00$, $0.01 \leq z \leq 0.03$)"), [0039] ("An Al-containing Co-based oxide ($LiCo_{0.97}Al_{0.03}O_2$) with an average particle size of 7 μm was used as the positive electrode active material"), [0013], [0015], [0020], [0028]-[0029], [0045]-[0046], [0050], [0056]-[0060], claims 1-2.

70. The battery of the combination also includes negative electrode material, *e.g.*, based on the "negative electrode active material" of Yamaki's

battery. APPLE-1004 (Yamaki), [0015]; *see* [0030] ("The negative electrode used in the battery of the present invention is prepared based on a mixture obtained by adding a binder to a carbon material or a material capable of intercalating Li or forming a compound."), [0031] ("The carbon material can be, for example, natural graphite, artificial graphite, or a carbon material obtained by heat-treating heavy oil, coal tar, or pitch-based fibers before being carbonized and pulverized. Materials capable of intercalating Li or forming compounds include metals such as aluminum, metal oxides including silicon, and composites of these materials including carbon materials."), [0006], [0013], [0016], [0041], Abstract, claim 1.

The ratio of positive electrode material to negative electrode material in the combination battery

71. In the Yamaki-Sakamoto combination battery, "the reversible capacity balance of the positive electrode and the negative electrode" in the combination battery "is set to the range of 1.05 < negative electrode/positive electrode <= 1.30," based on the teachings of Sakamoto. APPLE-1005 (Sakamoto), [0007]. Sakamoto's "reversible capacity balance" is calculated as "the reversible capacity of the negative electrode" divided by "the reversible capacity of the positive electrode." APPLE-1005 (Sakamoto), [0007]; see Abstract ("The reversible capacities of the positive electrode and the negative electrode are made to satisfy the range of 1.05 < negative electrode/positive electrode <= 1.30"), [0008], [0012], [0015]-[0016], claim 1.

72. Sakamoto's experimental examples corroborate that the reversible capacity balance is based on the capacities of the electrodes. For instance, Sakamoto describes fabricating a "prismatic battery" having "a positive electrode capacity of 751.2 mAh, a negative electrode capacity of 821.6 mAh, and a reversible capacity balance … of 1.09." APPLE-1005 (Sakamoto), [0015]. The 1.09 value for the reversible capacity balance is the value of the negative electrode capacity (821.6 mAh) divided by the positive electrode capacity (751.2 mAh).

73. The reciprocal of Sakamoto's reversible capacity balance—*i.e.*, the reversible capacity of the positive electrode divided by the reversible capacity of the negative electrode—is, in the combination battery, the ratio of positive electrode material to negative electrode material, as calculated by a capacity of the electrode active materials, as I explain in the following paragraphs.

74. First, the capacity of an electrode in a lithium secondary battery is provided by the electrode active material. The capacity of an electrode is a term used to describe the number of electrons that can be reversible stored and discharged in that structure, usually expressed in Amp Hours or milliamp hrs. It is a measure of the amount of material that can be utilized during the storage cycle. Electrodes may contain material that is included to enhance the flow of electrons, like conductive carbon, or material that forms pores to allow the egress of other soluble ions, but the active material includes only those compounds that

participate in the electrochemical reactions. Furthermore, only some fraction of the potentially active material participates in the charge cycle, giving rise to an effective or empirical capacity measurement, distinct from the theoretical capacity that could be calculated by including all of potentially active material that is present.

75. This means that the reciprocal of Sakamoto's "reversible capacity balance" (which is the ratio of the capacities of the electrodes) refers to the reversible capacity of the positive electrode active material divided by the reversible capacity of the negative electrode active material in the combination battery. APPLE-1005 (Sakamoto), [0007]. This ratio is expressed in the following equation:

$$Capacity\ balance\ (reciprocal) = \frac{Capacity\ of\ pos.electrode\ active\ material}{Capacity\ of\ neg.electrode\ active\ material}.$$

76. The capacities of a given type of electrode active material in a lithium secondary battery are a function of the capacity per unit mass of the electrode active material (*e.g.*, in units of milliamp hours per gram (mAh/g)) and the amount (*e.g.*, in units of grams) of electrode active material in the battery. This can be understood by straightforward multiplication: the capacity per unit mass times the mass gives a result that is in units of mAh: this is the capacity of the electrode active material.

77. Thus, the reciprocal of Sakamoto's reversible capacity balance (which is the ratio of the capacities of the electrode active materials) is the ratio between (1) the capacity per unit mass of the positive electrode active material (*e.g.*, in mAh/g) times the mass of the positive electrode active material (*e.g.*, in g) in the battery and (2) the capacity per unit mass of the negative electrode active material times the mass of the negative electrode active material in the battery. This is expressed in the following equation:

$$Capacity\ balance\ (reciprocal) = \frac{\left(\frac{mAh}{g} * g\right) pos.electrode\ active\ material}{\left(\frac{mAh}{g} * g\right) neg.electrode\ active\ material}.$$

78. I note that Sakamoto confirms that the reversible capacity balance is based on the amount (*e.g.*, mass) of each electrode active material in the battery. For instance, in discussing prior art batteries, Sakamoto describes a "method of setting the ratio of the amount of the active material in both electrodes so that the reversible capacities on the positive electrode side and the negative electrode side are substantially equal." APPLE-1005 (Sakamoto), [0006]; *see* [0007] (explaining that "the reversible capacity balance of the positive electrode and the negative electrode" is set to address disadvantages of these prior art batteries).

79. Sakamoto's experimental examples corroborate that the reversible capacity balance is based on the amount (*e.g.*, mass) of each electrode active material in the battery. Specifically, Sakamoto describes the fabrication of several

batteries, each having the same composition of their positive and negative electrodes, and thus each having the same capacity per unit mass of their positive and negative electrode active materials. *See* APPLE-1005 (Sakamoto), [0015]. Sakamoto then describes that the various batteries were produced to have reversible capacity balances of 0.94, 1.01, 1.09, 1.18, 1.30, 1.41, and 1.50. *See id.* The way to achieve these different capacity balances using the same materials is to use different amounts (*e.g.*, different masses) of the positive and/or negative electrode active materials in each battery. That is, the amount (mass) of positive and negative electrode active material in the battery, along with the capacity per unit mass of the materials, determines the reversible capacity balance of Sakamoto's batteries.

80. Thus, a POSITA would have understood, or at least found obvious, that Sakamoto's "reversible capacity balance" (reciprocal) in the combination battery is calculated as the product of the capacity of the positive electrode active material per unit mass and the mass of the positive electrode active material in the battery, divided by the product of the capacity of the negative electrode active material per unit mass and the mass of the negative electrode active material in the battery. This ratio, in the combination battery, is the ratio of positive electrode material to negative electrode material as calculated by a capacity of the electrode active materials.

81. Sakamoto describes this reversible capacity balance (*see* APPLE-1005 (Sakamoto), [0015]) for batteries that were tested with "a charge upper limit voltage of 4.2 V," *e.g.*, a charge cut-off voltage of 4.2 V. APPLE-1005 (Sakamoto), [0016]. In this context, a POSITA would have understood, or at least found obvious, that Sakamoto's reversible capacity balances (*e.g.*, as applied to the combination battery) were calculated by a capacity with a charge cut-off voltage set at 4.2 V, *e.g.*, because such a calculation would be directly relevant to the test criteria applied to the batteries. *See* APPLE-1005 (Sakamoto), [0016]. Specifically, the capacity of an electrode at a particular voltage is an indication of the amount of charge that can be stored in and/or extracted from that electrode at that particular voltage; the capacity of an electrode can differ at different voltages. That is, the same electrode material can store a different amount of charge and/or can release a different amount of charge depending on the applied potential. Thus, in designing a battery, the capacities of the electrodes of the battery (and their ratio) are determined based on the expected voltage (e.g., the intended charge cut-off voltage) of the battery, so that the calculation will be relevant to the actual operation of the battery. That is, for instance, a POSITA designing a battery to have a certain ratio of positive to negative electrode material will be interested in that ratio ***at the charge cut-off voltage of the battery***, because that is the voltage that will control charge movement between the electrodes. Moreover, because

many batteries as of the Critical Date were operated with a 4.2 V charge cut-off voltage, it was also conventional for capacity values to be determined and/or provided with respect to a 4.2 V charge cut-off voltage, *e.g.*, because it is at the charge cut-off voltage of the battery that the capacity is relevant.

82. Additionally, a POSITA would have found obvious that Sakamoto's reversible capacity balances (*e.g.*, as applied to the combination battery) are calculated by a theoretic capacity, *e.g.*, at least a theoretic capacity of the positive electrode active material. For instance, it was often conventional in battery design for the capacity of the positive electrode active material (an element of the calculation of Sakamoto's reversible capacity balance) to be provided as a theoretic capacity. Moreover, a POSITA would have recognized advantages in using a theoretic capacity for the calculation (*e.g.*, as opposed to an empirical value determined via experimental testing). For instance, use of a theoretical value rather than an actual value brings efficiencies and predictability to the design process, *e.g.*, in that less experimental characterization need be carried out.

83. Although it is possible to propose a theoretical model for Lithium storage in Carbon structures that results in an atomic ratio of C to Li (typically 6:1 for graphitic carbon), the actual storage capacity of carbon based negative electrodes in Li Ion batteries varies widely. Coke based or non-graphitic carbons do not have the same ordered atomic structure found in graphite and therefore do

not have even a theoretical ratio. In fact, the storage capacity of carbon electrodes is determined empirically and at carefully defined conditions and voltages.

The value of the ratio in the combination

84. In the combination battery, "the reversible capacity balance of the positive electrode and the negative electrode is set to the range of 1.05 < negative electrode/positive electrode <= 1.30." APPLE-1005 (Sakamoto), [0007]; see Abstract ("The reversible capacities of the positive electrode and the negative electrode are made to satisfy the range of 1.05 < negative electrode/positive electrode <= 1.30, and the reversible capacity of the negative electrode is increased"), [0008] ("The present invention increases the negative electrode/positive reversible capacity balance to within the range of 1.05 < negative electrode/positive electrode <= 1.30."), [0008] ("the range is preferably 1.10 or more and 1.20 or less"), [0015] (describing experimental examples: "A prismatic battery C having a positive electrode capacity of 751.2 mAh, a negative electrode capacity of 821.6 mAh, and a reversible capacity balance (negative electrode/positive electrode) of 1.09 was produced using the positive electrode plate and the negative electrode plate described above. … Battery D with a reversible capacity balance of 1.18, Battery E with a reversible capacity balance of 1.30, Battery F with a reversible capacity balance of 1.41, and Prismatic Battery G

with a reversible capacity balance of 1.50 were prepared in the same manner."), [0012], [0016], claim 1.

85. The ratio of positive electrode material to negative electrode material in the combination battery is the reciprocal of Sakamoto's reversible capacity balance, as discussed above. That is, the ratio of positive electrode material to negative electrode material in the combination battery is, e.g., between 1:1.05 and 1:1.30 (Sakamoto's "1.05 < negative electrode/positive electrode <= 1.30") or between 1:1.10 and 1:1.20 (Sakamoto's "1.10 or more and 1.20 or less"). APPLE-1005 (Sakamoto), [0007]-[0008]; *see* [0015] (the reversible capacity balances of Sakamoto's experimental examples, written as the ratio of positive electrode material to negative electrode material in the combination battery, correspond to 1:1.09 (prismatic battery C), 1:1.18 (Battery D), 1:1.30 (Battery E), 1:1.41 (Battery F), and 1:1.50 (Prismatic Battery G)).

86. The ratios described by Sakamoto and incorporated into the combination battery (e.g., 1:1.05 to 1:1.30, or 1:1.10 to 1:1.20) fall within the range of ratios recited in claim 5 (1:1.0 to 1:2.5). See APPLE-1005 (Sakamoto), [0007]-[0008]. Moreover, Sakamoto discloses specific points within the claimed range: e.g., 1:1.09, 1:1.18, and 1:1.30 (see APPLE-1005 (Sakamoto), [0015]), and 1:1.05, 1:1.10, 1:1.20, and 1:1.30 (the endpoints of the disclosed ranges; *see id.*, [0007]-[0008]).

87. Thus, based on the disclosure of Sakamoto, the Yamaki-Sakamoto combination renders obvious a battery having positive electrode material and negative electrode material in a ratio of from 1:1.0 to 1:1.25. *See* APPLE-1005 (Sakamoto), [0014]-[0016], [0052]-[0054], [0057].

(ii) **Claim 6**

[6] A secondary lithium ion cell or battery according to claim 5, characterized in that the secondary lithium ion cell or battery has a charge cut-off voltage within a range from 4.3 V to 5.2 V.

88. My analysis of element [5a] applies also to claim 6. The charge cut-off voltages described by Yamaki and incorporated into the combination battery (*e.g.*, 4.4 V to 4.8 V) fall within the range of charge cut-off voltages recited in claim 6 (4.3 V to 5.2 V). See APPLE-1004 (Yamaki), [0014]-[0016], [0052]-[0054], [0057]. Thus, based on the disclosure of Yamaki, the Yamaki-Sakamoto combination renders obvious that the combination battery has a charge cut-off voltage within a range from 4.3 V to 5.2 V. See APPLE-1004 (Yamaki), [0014]-[0016], [0052]-[0054], [0057].

(iii) **Claim 7**

[7] A secondary lithium ion cell or battery according to claim 5, characterized in that the secondary lithium ion cell or battery has a charge cut-off voltage within a range from 4.3 V to 4.8. V.

89. My analysis of element [5a] applies also to claim 7. The charge cut-off voltages described by Yamaki and incorporated into the combination battery (*e.g.*, 4.4 V to 4.8 V) fall within the range of charge cut-off voltages recited in

claim 7 (4.3 V to 4.8 V). See APPLE-1004 (Yamaki), [0014]-[0016], [0052]-[0054], [0057]. Thus, based on the disclosure of Yamaki, the Yamaki-Sakamoto combination renders obvious that the combination battery has a charge cut-off voltage within a range from 4.3 V to 4.8 V. See APPLE-1004 (Yamaki), [0014]-[0016], [0052]-[0054], [0057].

(iv) **Claim 8**

[8] A secondary lithium ion cell or battery according to claim 5, characterized in that the ratio of positive electrode material to negative electrode material of the secondary lithium ion cell or battery is from 1:1.15 to 1:2.5.

90. My analysis of element [5b] applies also to claim 8. In the combination battery, the ratio of positive electrode material to negative electrode material (*e.g.*, based on the reciprocal of Sakamoto's reversible capacity balance) is, *e.g.*, "1:1.18," "1:1.30," "1:1.41," or "1:1.50." APPLE-1005 (Sakamoto), [0015]. These values fall within the claimed range (1:1.15 to 1:2.5). Thus, based on this disclosure, the Yamaki-Sakamoto combination renders obvious that the combination battery has a ratio of positive electrode material to negative electrode material from 1:1.15 to 1:2.5.

91. I note that the combination battery can also have a range of ratios (1:1.05 to 1.30 or between 1:1.10 and 1:1.20), based on the teachings of Sakamoto, that overlaps with the recited range of ratios (1:1.15 to 1:2.5). I have been informed that a prior art disclosure of range of values that overlaps with a claimed

range of values is considered to render the claimed range obvious, unless there is an indication of criticality or unexpected results associated with the claimed range. Here, as I discuss below, there is no such indication of criticality or unexpected results, and I thus conclude that range of ratios described by Sakamoto and incorporated into the combination battery render obvious the range of ratios recited in claim 8. Thus, based on the disclosure of Sakamoto, the Yamaki-Sakamoto combination renders obvious that the combination battery has a ratio of positive electrode material to negative electrode material from 1:1.15 to 1:2.5.

92. In this case, I have determined that the ’641 Patent does not provide any indication of criticality or unexpected results associated with the specific claimed range of 1:1.15 to 1:2.5. While the ’641 Patent identifies the range of positive electrode material to negative electrode material as “from 1:1.0 to 1:2.5, preferably from 1:1.15 to 1:2.5,” it provides no rationale as to the relevance of the 1:1.15 lower bound of the putative “preferabl[e] range.” APPLE-1001 (the 641 Patent), 3:29-31. In fact, in its experiments, the ’641 Patent tests a collection of batteries having ratios “of 1:1.0, 1:1.05, 1:1.1, 1:1.15, 1:1.2, 1:1.25 and 1:1.3” and provides a collective characterization of their performance: “these cells … maintain 85.72% of capacity after 400 recycles.” *Id.*, 10:29-37 (Example 1); *see also* similar treatment in Examples 2-4. There is no indication, whether by explanation or experiment, of the relevance of the 1:1.15 to 1:2.5 range recited in

claim 8. Indeed, not only does the ’641 Patent not explain any criticality of the 1:1.15 lower limit of this ratio range, it even implicitly suggests, through its experimental treatment, that there is little notable difference between the 1:1.15 ratio and any other of the specifically tested ratios.

(v) Claims 11-14

[11] A secondary lithium ion cell or battery according to claim 5, wherein the secondary lithium ion cell or battery maintains at least 7500[1] of capacity after 400 cycles.

[12] A secondary lithium ion cell or battery according to claim 5, wherein the secondary lithium ion cell or battery maintains at least 75.83% of capacity after 400 cycles.

[13] A secondary lithium ion cell or battery according to claim 5, wherein the secondary lithium ion cell or battery maintains at least 80% of capacity after 400 cycles.

[14] A secondary lithium ion cell or battery according to claim 5, wherein the secondary lithium ion cell or battery maintains at least 80.72% of capacity after 400 cycles.

A POSITA would have found obvious that the Yamaki-Sakamoto battery has the claimed capacity retention

93. It is my opinion that a POSITA would have understood, or at least found obvious, that the Yamaki-Sakamoto battery maintains at least 75% of

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[1] As best understood, I interpret “7500” to be “75%.” This interpretation is based on claims 12-18, which recite similarly structured percentages of capacity maintenance; in particular, I note the similarity between the set of claims 11-14

capacity after 400 cycles. Moreover, it is also my opinion that a POSITA would have understood, or at least found obvious, that the Yamaki-Sakamoto battery maintains any other reasonable but arbitrary percentage of capacity, such as at least 75.83%, 80%, or 80.72% of capacity, after 400 cycles (or after another reasonable but arbitrary number of cycles).

94. The ’641 Patent provides no description of how to achieve the recited capacity retention beyond specifying ranges for the charge cut-off voltage and ratio of electrode materials. *See* APPLE-1001 (the 641 Patent), Examples 1-28. And as I demonstrated above, the combination battery satisfies the underlying compositional (*e.g.*, ratio of positive to negative electrode material) and operational (*e.g.*, charge cut-off voltage) limitations that yield these recited capacity results. Indeed, there is even overlap in the specific materials of the ’641 Patent’s battery (*e.g.*, “lithium cobalt composite oxides” or “lithium manganese cobalt composite oxides” as positive electrode active material, “a carbonaceous substance” as

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and the set of claims 15-18. *See* APPLE-1001 (’641 Patent), claim 15 (“maintains at least 75% of capacity after 400 cycles”). This interpretation is also based on my review of relevant disclosure in the specification of the ’641 Patent, which describes batteries that maintain various percentages of capacity after some number of cycles. *See id.*, Tables 2-3.

negative electrode active material, and "non-aqueous electrolytes … of the type well known in the art") and the materials of the Yamaki-Sakamoto battery. APPLE-1001 (the 641 Patent), 8:10-9:15; *see* my discussion of element [5a] above. Thus, because the disclosures of Yamaki and Sakamoto, incorporated into the combination battery, align with the elements that the '641 Patent indicates as providing the recited capacity retention, I believe that a POSITA would find obvious that the Yamaki-Sakamoto combination battery, on its face, maintains at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles.

95. Alternatively or additionally, I believe that a POSITA would have found it obvious that Yamaki-Sakamoto combination battery maintains at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles, based on the cycle life performance achieved by both Yamaki and Sakamoto and on expectations for battery performance over a larger number of cycles.

96. The Yamaki-Sakamoto combination battery includes a positive electrode composition based on that described by Yamaki, which in itself provides for good cycleability. *See* APPLE-1004 (Yamaki), [0013] ("a lithium secondary battery having a positive electrode containing a Co-based oxide including Li and Co as a positive electrode active material and a conductive material whose main component is carbon, … the lithium secondary battery characterized in that the Co-based oxide contains at least one element selected from Mg, Al, Mn, Ti, and Sr").

Yamaki explains that "such a battery configuration" "suppress[es] deterioration in the reversibility of charging and discharging as more charging and discharging cycles are performed." APPLE-1004 (Yamaki), [0014]. Moreover, the positive and negative electrode active materials in the combination battery are provided in a ratio based on the teachings of Sakamoto, which also provides for good cycleability: "a battery with especially low cycle deterioration can be obtained." APPLE-1005 (Sakamoto), [0012]. As I discuss in the following paragraphs, based on the specific cycle life achievements measured experimentally by both Yamaki and Sakamoto, and also based on well-known cycle life behavior of secondary lithium ion batteries, I believe a POSITA would have found it obvious that the Yamaki-Sakamoto battery would maintain at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles.

97. Yamaki describes that its battery has "a capacity retention rate of 80% or more when charging and discharging have been repeated 10 times." APPLE-1004 (Yamaki), [0028]; see [0059] ("the cycle characteristics were excellent with a capacity retention rate of 80% or higher"). Yamaki also provides experimental examples in which batteries that are charged to a charge cut-off voltage of 4.5 V retain up to 98% capacity after 10 cycles, as shown in the highlighted rows of Table 1. *See* APPLE-1004 (Yamaki), [0057].

Table 1

	Positive Electrode Active Material	Conductive Material	Charged Positive Electrode Potential (V: Metal Li Reference)	Initial Battery Capacity (mAh)	Initial Discharge Capacity of Positive Electrode Material (mAh/g)	Capacity Retention After 10 Cycles (%)
Ex. 1	$LiCo_{0.97}Al_{0.03}O_2$	(A)	4.5	3.99	182	95
Ex. 2	$LiCo_{0.99}Al_{0.01}O_2$	(A)	4.5	4.12	188	94
Ex. 3	$LiCoMg_{0.03}O_2$	(A)	4.5	4.17	190	98
Ex. 4	$LiCo_{0.97}Mn_{0.03}O_2$	(A)	4.5	4.05	184	97
Ex. 5	$LiCo_{0.99}Sr_{0.01}O_2$	(A)	4.5	4.24	193	93
Ex. 6	$LiCo_{0.97}Al_{0.02}Mn_{0.01}O_2$	(A)	4.5	4.00	182	95
Ex. 7	$LiCo_{0.98}Ti_{0.02}Mg_{0.01}O_2$	(A)	4.5	4.07	184	95
Ex. 8	$LiCo_{0.97}Al_{0.01}Ti_{0.01}Mn_{0.01}O_2$	(A)	4.5	4.00	182	93
Ex. 9	$LiCo_{0.97}Al_{0.03}O_2$	(A)	4.4	3.86	165	97
Ex. 10	$LiCo_{0.97}Al_{0.03}O_2$	(A)	4.6	4.19	211	89
C. Ex. 1	$LiCoO_2$	(B)	4.5	4.15	189	75
C. Ex. 2	$LiCoO_2$	(B)	4.3	3.82	157	95

Yamaki, Table 1 (annotated)

98. In the combination, a battery such as that described by Yamaki is implemented to have positive and negative electrode active material in amounts satisfying the "reversible capacity balance" described by Sakamoto. Sakamoto describes that batteries satisfying this "reversible capacity balance" "experience[] less deterioration after repeated charging and discharging." APPLE-1005 (Sakamoto), [0007]; see [0001], [0012], [0016]. That is, Sakamoto indicates that batteries implementing its reversible capacity ratio have improved capacity retention and cycle life. In light of the improved cycle life enabled by Sakamoto's reversible capacity balance, a POSITA would have expected a battery based on the Yamaki-Sakamoto combination to have cycle characteristics at least as good as those of Yamaki's battery, if not even better—that is, a POSITA would have

expected the Yamaki-Sakamoto combination battery to retain at least up to 98% capacity after 10 cycles. *See* APPLE-1004 (Yamaki), [0057].

99. In fact, it was well-known that having more negative electrode capacity than positive electrode capacity was beneficial for cycle life of a secondary lithium battery. I discussed this above in Paragraphs [37]-[39] of this Declaration. Thus, with this understanding in hand, a POSITA would have been confident that incorporating teachings based on Sakamoto into Yamaki's battery—and thus providing more negative electrode capacity than positive electrode capacity—would result in a battery having cycle life at least consistent with (but probably better than) that demonstrated for Yamaki's battery.

100. Yamaki describes a system where the improved positive electrode material ensures an improved cycle life, through the incorporation of additional metal oxides into the Cobaltite matrix. If the cycle life deterioration were to be the result of negative electrode deterioration, or short circuiting caused by metallic Lithium plating, the improvements noted by Yamaki would not be apparent. The combination of Yamaki and Sakamoto however, would describe a system with the improved cycle life of the Yamaki positive electrode along with the negative overbalanced advantage of Sakamoto, ensuring that the negative electrode would not cause reduced cycle life and the Yamaki improvements in the positive would become the controlling factor that would determine a longer cycle life.

101. Unfortunately, Yamaki does not describe results of testing for more than 10 cycles. However, this 10 cycle capacity measurement does not provide a complete picture of the capacity retention capability of the battery. Testing the cycle life of a battery is a time-intensive and expensive process, and often battery designers perform initial testing of only a small number of cycles to gauge battery performance. In the absence of specific testing data regarding the longer term capacity retention of the Yamaki-Sakamoto battery, a POSITA would have found it reasonable to predict its longer life performance—that is, to predict the results of capacity testing after more than 10 cycles—based on the performance of other, similar batteries. This is often what is done at early stages of battery testing.

102. Many secondary lithium batteries that exhibit high capacity retention after a small number of cycles (*e.g.*, consistent with the behavior of the Yamaki-Sakamoto battery) exhibit significant capacity retention even after larger numbers (*e.g.*, hundreds) of cycles. For instance, Guyomard describes $Li_{1+x}Mn_2O_4$ secondary batteries in which "the cell capacities after 500 cycles was only about 10% less than after 5 cycles." APPLE-1016 (Guyomard), 5:32-36. Wendsjo describes that in lithium manganese oxide spinel secondary batteries, "[a]fter 400 cycles, the capacity was 299 mAh, say 84% of the initial capacity." APPLE-1017 (Wendsjo), 5:10-17. Uemura's FIG. 3 is a table that demonstrates that lithium ion secondary batteries last for hundreds of cycles before "the discharge capacity

becomes less than 90% of the initial discharge capacity." APPLE-1018 (Uemura), FIG. 3, [0081]. And the Handbook of Batteries indicates a typical cycle life of 500-1000 cycles for lithium ion batteries. *See* APPLE-1015 (Handbook), 23.10; *see also* FIG. 36.35b, which shows significant capacity retention of lithium ion batteries out to 500 cycles.

103. Given that batteries that behave similarly to the Yamaki-Sakamoto combination battery after a small number of cycles retain a significant percentage of their capacity even after hundreds of cycles, I believe a POSITA would have had good reason to expect the Yamaki-Sakamoto battery to show similar behavior during longer-term cycle life testing. That is, a POSITA would have expected the Yamaki-Sakamoto battery to maintain at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles.

A POSITA would have found it obvious to obtain a Yamaki-Sakamoto combination battery that has the claimed percent capacity retention

104. Separately, I also believe that a POSITA would have found it obvious to obtain a battery consistent with the Yamaki-Sakamoto battery and that maintains a reasonable, but arbitrary percentage of its capacity after some reasonable but also arbitrary number of cycles—for instance, that maintains at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles—based on routine experimentation guided by a goal of improving cycle life.

105. First, as I have previously discussed, both Yamaki and Sakamoto have an objective of improving cycle life. *See* APPLE-1004 (Yamaki), [0014]; APPLE-1005 (Sakamoto), [0012]. More generally, a POSITA would have been well aware of cycle life criteria that would have applied to batteries to enable them to be commercially viable. Namely, a commercially viable battery has to retain a large percentage of its capacity over a large number of cycles—imagine, for instance, a mobile phone whose battery loses half of its capacity after only a handful of cycles! Indeed, the references I discussed previously indicate both that cycle life is a common concern of battery designers, and that significant capacity retention over a long cycle life is common and relevant. *See* APPLE-1016 (Guyomard), 5:32-36; APPLE-1017 (Wendsjo), 5:10-17; APPLE-1018 (Uemura), FIG. 3, [0081]; APPLE-1015 (Handbook), 23.10, FIG. 36.35b. In this context, a POSITA developing the Yamaki-Sakamoto battery would have found it obvious to aim for a battery that is able to retain a large percentage of its capacity over a large number of cycles.

106. With this objective in mind—generating a battery with long cycle life (both as an articulated goal of Yamaki and Sakamoto and as a practical, commercial objective)—a POSITA would have known how to achieve such a goal. Specifically, there are numerous factors that influence cycle life and that a POSITA would have known to test with the aim of obtaining a battery with long

cycle life. With these factors at hand, a POSITA would have just conducted routine experiments, during longer-term cycle life testing, and consistent with the charge cut-off voltage range and ratio of positive to negative electrode material of the combination battery of the Yamaki-Sakamoto combination battery, with the objective of finding the parameter space that allows for a target percent capacity retention over a target number of cycles to be achieved. In fact, I note that none of these factors (other than charge cut-off voltage and ratio of positive to negative electrode material) are specified in the claims. And, the '641 Patent itself indicates that these parameters are merely standard battery parameters: "[e]xcept voltage, all process parameters for charge and discharge are commonly used in the art." APPLE-1001, 9:22-23.

107. In this context, the retention of at least 75%, 75.83%, 80%, or 80.72% of capacity is an arbitrary number, as are the 400 cycles. A POSITA would have known how to achieve a target percent capacity after a certain number of cycles; the factors do not change, nor does the experimental process.

108. For instance, Yamaki demonstrates that charge cut-off voltage is a factor that affects capacity retention and cycle life. *See* APPLE-1004 (Yamaki), [0057] (compare Rows 1 and 9, demonstrating that batteries having the same electrode compositions but charged to different charge cut-off voltages achieve different percent capacity retention). More generally, charge cut-off voltage was

well-known to be a factor that is relevant to cycle life of a battery. For instance, while batteries charged to higher cutoff voltages will have higher discharge capacity, they will generally also show more rapid loss in capacity upon extended cycling. Corroborating that this is the case, Yasunami presents experimental results demonstrating that batteries having the same electrode compositions but that are charged to different charge cut-off voltages retain a certain percent capacity for different numbers of cycles. APPLE-1006 (Yasunami), [0067]-[0071]

109. Capacity retention and cycle life is also affected by the ratio of positive electrode material to negative electrode material. Sakamoto shows this in its experiments. *See* APPLE-1005 (Sakamoto), [0015]-[0016]. More generally, this was also well-known in the art as factor relevant to cycle life of a battery, as I discussed above in Paragraphs [37]-[39] of this Declaration.

110. Driven to improve cycle life (*e.g.*, based on Yamaki and Sakamoto, and/or driven by commercial criteria), and knowledgeable of the impact of charge cut-off voltage and active material ratio on cycle life, a POSITA would have found it obvious to conduct routine experiments adjusting the charge cut-off voltage (while staying within the 4.4 V to 4.8 V range of the Yamaki-Sakamoto battery) and/or electrode material ratio (again, while staying within the 1:1.05 to 1:1.30 range of the combination battery) to optimize the capacity retention of the Yamaki-Sakamoto battery.

111. Other factors, including charge and discharge rate, ambient temperature, electrolyte purity, and the strength (*e.g.*, ionic strength) of the electrolyte also affect capacity retention in battery cycling.

112. Specifically regarding temperature, cells that are cycled at temperatures outside of the typical ambient conditions will exhibit different failure modes, typically leading to shortened lifetime through capacity loss. Extreme low temperatures will reduce the mobility of the ionic species, particularly in the organic electrolyte used in secondary Lithium cells. This reduced mobility leads to a higher degree of concentration polarization and higher IR losses. Since capacity is measured to a predetermined cut-off voltage, these losses will be observed as shorter cycle life. High temperatures increase the rate of side reactions, including electrolyte decomposition that leads to gas formation, venting and loss of electrode activity caused by the presence of non-conductive decomposition products. The effect on cycle life can be immediate or accumulative, but it is always negative. FIG. 36.34 of the Handbook of Batteries provides an example of "Discharges at 0.2C rate at various temperatures," demonstrating the effect of temperature on discharge performance and battery capacity. *See* APPLE-1015 (Handbook), 36.46.

113. The Handbook of Batteries also provides an example of discharges at different rates ranging from 0.2 C to 3 C, demonstrating the effect of discharge rate on battery capacity. APPLE-1015 (Handbook), 36.46, FIG. 36.34(a).

114. Shin explains that "the decomposition and compositional change of electrolytes and electrodes during charge-discharge cycling," which "can result in an irreversible loss of anode capacity, and gradual oxidation and consumption of electrolyte," which "in turn cause[s] capacity decline during cycling." APPLE-1014 (Shin), 1. And Guyomard describes tests in which an electrolyte with "high ionic conductivity" "extend[ed] the cycle life of the batteries." APPLE-1016 (Guyomard), 5:20-35.

115. Still driven to improve cycle life, and now knowledgeable of the impact of these operational factors on cycle life, a POSITA again would have found it obvious to conduct routine experiments adjusting charge rate, discharge rate, temperature, and/or electrolyte characteristics, in an effort to optimize the capacity retention of the Yamaki-Sakamoto battery.

116. Each of these factors—charge cut-off voltage, ratio of electrode materials, charge and discharge rate, temperature, and electrolyte characteristics—was a known contributor to cycle life and capacity retention of lithium secondary batteries. Given that this is a well-mapped space, I believe that a POSITA would have had a reasonable expectation of success in adjusting these factors to find an optimized capacity retention for the Yamaki-Sakamoto battery. A POSITA working to improve the performance of a lithium ion battery and utilizing the concepts contained in Yamaki and Sakamoto would have used a standard

experimental design approach to investigate the effects of cut-off voltage, charge and discharge rate, temperature, and electrolyte characteristics, along with the design features described in the two references. These design optimization programs were standard and well known techniques for maximizing cell performance variables, including cycle life. By using classic design optimization approaches, the POSITA would achieve results, such as a formulation and design that maximized cycle life, with a modest investment in experimentation and time.

117. Interestingly, I note that the ’641 Patent provides no indication of the charge and discharge rate, the ambient temperature, or the electrolyte purity for its capacity retention testing. The Yamaki-Sakamoto battery already has the structure (ratio of positive to negative electrode material) and operation (charge cut-off voltage) of the batteries of the ’641 Patent, and adjusting the charge and discharge rate and ambient temperature would have been a matter of simple experimentation. As to the question of a specific value for percent capacity retention (75%, 75.83%, 80%, or 80.72%) and a specific number of cycles (400)—the ’641 Patent says nothing about why these values are critical or somehow unexpected. Instead, they are just arbitrary numbers. And given the knowledge of a POSITA about the various factors that influence capacity retention, starting from a battery that already meets what is disclosed by the ’641 Patent, it would have been just straightforward experimentation to identify the factors that can achieve this arbitrary standard—

that is, to obtain a battery that maintains at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles.

B. [GROUND 1B] – CLAIMS 9-10 ARE OBVIOUS OVER YAMAKI, SAKAMOTO, AND ABRAHAM

1. Overview of Abraham

118. Abraham describes "the use of redox reagents, dissolved in non-aqueous electrolytes, to provide overcharge protection for cells having lithium metal or lithium-ion negative electrodes." APPLE-1009 (Abraham), Abstract.

119. Storing charge in a secondary battery requires both the formation of oppositely charged species and the separation of those potential reactants. In a LiIon cell, the Lithium Ions are stored in the charged negative electrode as zero valance Lithium that is contained in the carbon structures. If the zero valance lithium was soluble in the electrolyte, it could migrate to the opposing electrode and react with the oxidized lithium cobalt oxide. If the positive electrode material were soluble, it too could migrate to the opposing electrode and react with the Lithium metal atoms contained there. Only by separating the charged materials can the reactants be maintained in that state.

120. When a species is present that is soluble in the electrolyte and can be reduced at the negative electrode or oxidized at the positive, that species can become the key element in a chain of reduction/oxidation that is internal to the cell itself. Moving from one electrode back to the other, this "redox shuttle" species

creates a channel for electrons to short circuit the designed battery process by endlessly accepting and then rejecting electrons by moving between the two electrodes. When this process occurs by the presence of an impurity, it is undesirable. If it is intentionally enabled by the addition of a "shuttle" species, it can be a valuable technique to limit the voltage of the cell during overcharge.

121. Some shuttle species are only effective or active above certain cell voltages, and by choosing the species carefully, the negative consequences of a redox shuttle (energy inefficiency, rapid self-discharge) can be avoided, but the positive consequences of a "safety valve" limiting the cell voltage can be gained.

122. Generally, Abraham's overcharge protection involves a "redox shuttle," which is "a material with an appropriate oxidation potential [] dissolved in the electrolyte." *Id.*, 2:20-23. The "appropriate oxidation potential" of Abraham's redox shuttle is "an oxidation potential slightly higher than the normal charge limit of the cell but lower than the oxidation potential of the electrolyte." *Id.*, 2:36-38. "At a potential slightly above the cell charge limit (upper cutoff voltage), the redox shuttle is activated by its electrochemical conversion," which causes "[t]he cell potential during overcharge [to be] fixed at the oxidation potential of the redox shuttle," thus preventing damage that could otherwise occur in an overcharge situation. *Id.*, 2:23-27. The activation of the redox shuttle is reversible: "the

oxidized products" diffuse "to the anode where they recombine to form the starting material." APPLE-1009 (Abraham), 2:27-29.

123. Abraham identifies "thianthrenes" as example redox shuttles "used for the protection of lithium and lithium-ion secondary cells from the effects of overcharge." APPLE-1009 (Abraham), 3:19-22. The redox shuttle is "added to the electrolyte where it is available for a reversible oxidation-reduction (redox) reaction. *Id.*, 3:33-35. Abraham explains that "[t]he protective redox shuttle reaction is initiated when the cathode reaches the oxidation potential of the redox reagent," and notes that "[j]udicious selection of the particular thianthrene will allow the overcharge protection agent to be tailored to the appropriate cell voltage." APPLE-1009 (Abraham), 3:47-54. For instance, the redox potential of thianthrene itself is between 4.06-4.12 V. *See id.*, 4:27. The redox potential can be shifted "to more positive values" by "modify[ing] the thianthrene structure by replacing hydrogen atoms on the aromatic rings with electron withdrawing groups." *Id.*, 4:7-10. For instance, 2,7-diacetyl thianthrene has redox potentials of "4.3 V for oxidation and 4.2 V for the subsequent reduction." *Id.*, 4:14-17.

124. Abraham also indicates that the usage of thianthrene derivatives is suitable in situations having still higher charge limits (*e.g.*, charge cut-off voltages). Generally, "[p]roper matching of the overcharge protection additive (shuttle) to the electrochemical couple requires sufficient separation between the

completion of lithium extraction from the cathode reaction and the onset of the shuttle activation." APPLE-1009 (Abraham), 5:11-14. In a specific example, when the cathode is "$LiMn_2O_4$," "a shuttle activated at a potential between 4.2 and 4.5V … would ensure complete cathode utilization, yet be well within the voltage stability window of the electrolyte." *Id.*, 5:15-19.

125. I note that Abraham does not specify particular limits on the applicability of its teachings regarding redox shuttles in lithium secondary batteries. For instance, while Abraham describes some specific implementations of redox shuttles, it does not preclude the implementation of its reversible capacity balance in other battery systems, such as batteries having compositions and charge cut-off voltages consistent with those of the Yamaki-Sakamoto combination battery. Indeed, as I discuss below in Paragraph [142]. Abraham even provides guidance for development of redox shuttles for use in batteries having positions and charge cut-off voltages consistent with those of the Yamaki-Sakamoto combination battery.

2. The Yamaki-Sakamoto-Abraham combination

126. In the Yamaki-Sakamoto-Abraham combination battery, a redox shuttle is "added to the electrolyte where it is available for a reversible oxidation-reduction (redox) shuttle reaction," based on the teachings of Abraham. APPLE-1009 (Abraham), 3:33-35. The redox shuttle is a compound, *e.g.*, a thianthrene

derivate, that has been selected "to be tailored to the appropriate cell voltage." *Id.*, 3:47-54. Specifically, the redox shuttle is a compound that has an oxidation potential "slightly higher than" the 4.4 V to 4.8 V charge cut-off voltage of the combination battery. *Id.*, 2:36-38; *see* APPLE-1004 (Yamaki), [0014]. For instance, the redox shuttle can be a "thianthrene structure [modified] by replacing hydrogen atoms on the aromatic rings with electron withdrawing groups" to obtain a compound having an oxidation potential slightly higher than the charge cut-off voltage. APPLE-1009 (Abraham), 4:7-10. I note that Abraham confirms the feasibility of use of redox shuttles in the context of the 4.4-4.8 V charge cut-off voltage range of the combination battery, noting that for a LiMn2O4 cathode, the redox shuttle can be "activated at potentials between 4.2 and 4.5V." *Id.*, 5:15-17.

127. In operation of the combination battery, the battery is charged to its charge cut-off voltage, *e.g.*, a voltage of between 4.4 V and 4.8 V. APPLE-1004 (Yamaki), [0014]. In an overcharge situation, *e.g.*, "[a]t a potential slightly above" the charge cut-off voltage, "the redox shuttle is activated by its electrochemical conversion." APPLE-1009 (Abraham), 2:23-33. This causes "the cathode potential [to be] maintained indefinitely at the oxidation potential of the redox reagent," preventing damage from overcharging. *Id.* When the voltage on the positive electrode decreases, *e.g.*, to a level below the reduction potential of the

redox shuttle, the redox reaction no longer proceeds and normal charging/discharging operation of the battery can resume. *See id.*, 2:27-29.

3. Reasons for the combination

128. It is my opinion that a POSITA would have been motivated to provide overcharge protection additives, *e.g.*, based on Abraham, in the Yamaki-Sakamoto combination battery, for at least the following reasons.

129. The first reason why a POSITA would have been motivated to provide overcharge protection in the combination battery is to avoid degradation of battery performance that can result from an overcharge situation. For instance, a POSITA would have recognized that overcharging can "cause[] irreversible electrolyte side-reactions which deteriorate cell performance," and would have been motivated to avoid such situations to preserve performance of the combination battery. APPLE-1009 (Abraham), 1:54-56; *see* APPLE-1026 (Huang), 1:24-25 ("Cells subjected to overcharge or overdischarge lose capacity irreversibly"); APPLE-1008 (Richardson), 1 ("Overcharging can … reduce rechargeable capacity and cell life … Even moderate overcharging of transition metal oxide cathodes often leads to structural and compositional changes in the oxide matrix which have significant effects on their subsequent cycling behavior"). In particular, a POSITA would have been encouraged to pursue this implementation because Abraham does not specify any particular implementation

limits and thus is relevant to the composition and operation of the Yamaki-Sakamoto battery.

130. Overcharge is a term that inherently implies charging a cell past a design capacity, typically to a voltage that results in cycle life reduction, safety issues, or both. A well designed cell will accommodate some overcharge without immediate failure and even extreme overcharge should not create fire or explosion in a commercial design. If the negative electrode contains excess capacity, overcharge should not create Lithium metal plating or short circuiting. Overcharge will however degrade the cell performance because the positive electrode, when charged past the point where the effective capacity of the electrode has all been reached, will continue to deliver electrons by bringing the Cobalt (for example) to a higher valence, or higher state of oxidation. These higher oxidation states are typically not as conductive or reactive as the desired first stage oxidation, and the highly oxidized Cobalt becomes removed from the charge/discharge cycle and effectively inert. This material, when accumulated to a significant fraction of the positive electrode, will result in lower capacity and shortened cycle life.

131. <u>The second reason</u> why a POSITA would have been motivated to implement overcharge protection in the combination battery is to mitigate the safety risks associate with overcharging. *See* APPLE-1026 (Huang), 1:26-27 ("repeated overcharge and overdischarge of cells results in possible venting or

exploding of cells"); APPLE-1008 (Richardson), 1 ("Overcharging … can create hazardous conditions"), 2 ("Overcharging of batteries can create hazardous conditions, leading to explosion, fire, and/or release of toxic materials"). Again, a POSITA would have been encouraged to pursue this implementation because Abraham does not specify any particular implementation limits and thus is relevant to the composition and operation of the Yamaki-Sakamoto battery.

132. When a cell is overcharged without safety features in the design, or overcharged at a rate or temperature that exceeds safe levels, it can create significant risk of fire or explosion. If the negative electrode has enough excess capacity to prevent Lithium metal plating, there is reduced risk of short circuiting from metallic dendrites. Overcharge can create conditions, typically due to high voltages and temperatures, where electrolyte solvents and salts can become reactants instead of inert media. Electrolyte decomposition will result in gas formation and subsequent pressure increase. Even if vented before bursting, the gasses can be flammable and create fires upon venting.

133. <u>The third reason</u> why a POSITA would have been motivated to pursue a redox shuttle implementation for overcharge protection is because of the advantages of "[c]hemical overcharge protection," *e.g.*, as opposed to "electronic overcharge controllers," again in light of the generic applicability of Abraham's disclosure. APPLE-1009 (Abraham), 1:65-2:1. For instance, electronic

overcharge protection "lower[s] the energy density of the battery and increase[s] battery cost." APPLE-1009 (Abraham), 2:1-2; *see* APPLE-1027 (Shimizu), [0009]. A POSITA would have opted for chemical overcharge protection, *e.g.*, based on Abraham's redox shuttle approach, to avoid reductions in energy density that could come from use of electronic controllers, and to maintain a low cost battery system.

134. The fourth reason why a POSITA would have been motivated to pursue a redox shuttle implementation for overcharge protection—still in view of the general applicability of Abraham's disclosure—is because, in the context of a multi-cell battery, "[c]hemical overcharge protection" advantageously "provide[s] capacity balance among the individual cells and prevent[s] oxidative degradation of the electrolyte." APPLE-1009 (Abraham), 2:3-5. For instance, a POSITA would have pursued this approach to obtain these benefits, and the resulting increase in cycle life for the battery. *See* APPLE-1009 (Abraham), 2:5-19.

135. Multi-cell batteries can be assembled without individual cell voltage monitoring. If the cells include safety improvements that act at an individual cell level, such as pressure relief, thermal disconnect devices, or fused elements to prevent overcurrent, the battery pack may only contain voltage control that acts at the battery or multi-cell level. The safety devices may prevent catastrophic failure, but they will not prevent low levels of overcharge where the voltage of individual

cells exceeds the recommended voltage, but the battery pack voltage remains below a designated cutoff voltage. In this case, repeated excursions above the individual cell maximum will damage the cell, typically resulting in reduced cycle life. If the individual cell contains some type of overcharge protection, like a redox shuttle mechanism that limits the maximum voltage, the cells within the battery pack will harmlessly shunt the overcharge current to the shuttle mechanism, resulting in increased cycle life when compared to a battery pack that does not contain the overcharge protection but is otherwise similar.

136. The fifth reason why a POSITA would have been prompted to pursue this implementation is because this combination is just the application of known techniques (specifically, chemical overcharge protection) to an already-known system (specifically, the Yamaki-Sakamoto combination battery), and it produces predictable results (specifically, a secondary lithium battery having chemical overcharge protection).

137. It is my opinion that a POSITA would have expected to be successful in providing overcharge protection additives, *e.g.*, based on Abraham, in the Yamaki-Sakamoto combination battery, for at least the following reasons.

138. The first reason why a POSITA would have expected success in combining teachings of Abraham with the Yamaki-Sakamoto battery is because both relate to secondary lithium ion batteries with similar materials systems. This

is an indication that the teachings of Sakamoto are compatible with, and applicable to, the battery described by Yamaki.

139. For instance, the electrolytes in which Abraham describes dissolving the thianthrene-based redox shuttles are consistent with the electrolyte composition of the Yamaki-Sakamoto battery. *See* APPLE-1004 (Yamaki), [0033]; APPLE-1009 (Abraham), 3:36-39, 6:4-10. Specifically, the Yamaki-Sakamoto battery has electrolytes such as those described by Yamaki: "organic solvents include carbonates such as ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), dimethyl carbonate (DMC), methylethyl carbonate (MEC) and diethyl carbonate (DEC), esters such as γ-butyrolactone (γ-BL) and methyl acetate (EA), and ethers such as 1,3-dioxolane and 1,2-dimethoxyethane," with lithium salts dissolved in the solvent. APPLE-1004 (Yamaki), [0033]. Abraham identifies some of these same electrolyes: "[t]he electrolyte may be a liquid solution with a single solvent, such as propylene carbonate-1.0M $LiPF_6$, or a mixed-solvent solution such as 50% ethylene carbon ate:50% propylene carbonate-1.0M $LiPF_6$." APPLE-1009 (Abraham), 3:35-39.

140. Both systems also use lithium-containing transition metal oxides for their positive electrode. *See* APPLE-1004 (Yamaki), Abstract, [0018]; APPLE-1009 (Abraham), 5:47-49 ("Thus, although the carbon/spinel $LiMn_2O_4$ couple is mentioned specifically, other cathodes such as $LiCoO_2$ or $LiNiO_2$ might also be

used”). And both systems contain carbonaceous materials for their negative electrode. *See* APPLE-1004 (Yamaki), [0030]-[0031]; APPLE-1009 (Abraham), 5:27-32 (“The anode may be either disordered carbon or graphite”).

141. Given this overlap in materials, a POSITA would have expected the teachings of Abraham to be relevant to, and combinable with, the Yamaki-Sakamoto combination battery. And as I discussed previously, the disclosure of Abraham, as well as those of Yamaki and Sakamoto, are generally applicable, and none provides any indication precluding the combination of their features with features of the other disclosures.

142. <u>The second reason</u> why a POSITA would have expected success in implementing chemical overcharge protection, *e.g.*, as taught by Abraham, into the Yamaki-Sakamoto combination battery—which has a charge cut-off voltage of between 4.4 V and 4.8 V—is because of Abraham’s teachings of the flexibility of its redox shuttle approach. For instance, although Abraham identifies two specific example thianthrene compounds, it notes that “[t]he choice of a particular redox reagent will vary with the cathode material,” and indicates the suitability of its approach for “high voltage cathodes.” APPLE-1009 (Abraham), 5:44-46, 5:63. Moreover, Abraham provides guidance on how to choose a suitable redox reagent: “[p]roper matching of the overcharge protection additive (shuttle) to the electrochemical couple requires sufficient separation between the completion of

lithium extraction from the cathode reaction and the onset of the shuttle activation." APPLE-1009 (Abraham), 5:11-15. And Abraham describes how to make and characterize candidates for such compounds: "In order to shift the redox potential to more positive values, it is necessary to modify the thianthrene structure by replacing hydrogen atoms on the aromatic rings with electron withdrawing groups." APPLE-1009 (Abraham), 4:7-10. The resulting compounds can be "screen[ed] … for use as redox shuttle reagents" using "[c]yclic voltammetry." APPLE-1009 (Abraham), 5:55-57.

143. The third reason why a POSITA would have expected success is because of the widespread use of redox shuttle additives for overcharge protection in secondary lithium ion batteries as of the Critical Date. *See* APPLE-1008 (Richardson), 1-2; APPLE-1030, Abstract.

4. Analysis

(i) **Claim 9**

[9] A secondary lithium ion cell or battery according to claim 5, characterized in that the secondary lithium ion cell or battery has a first overcharging protection voltage of greater than 4.35 V, and an overcharging protection release voltage of greater than 4.15 V.

144. The Yamaki-Sakamoto-Abraham combination battery includes a "redox shuttle … dissolved in the electrolyte" of the combination battery, *e.g.*, based on the teachings of Abraham. APPLE-1009 (Abraham), 2:20-23; *see* Abstract ("This invention features the use of redox reagents, dissolved in non-

aqueous electrolytes, to provide overcharge protection for cells having lithium metal or lithium-ion negative electrodes (anodes)"), 2:48-56 ("The redox reagent is ideally present in an amount sufficient to maintain proper mass transport for the desired steady overcharge current for the cell"), 3:20-35 ("In a preferred embodiment of the invention, thianthrenes (R) are used for the protection of lithium and lithium-ion secondary cells from the effects of overcharge"), 1:13-18, 3:47-54, 5:11-19, claim 1.

145. The overcharging protection voltage of the combination battery corresponds to the "oxidation potential" of the redox shuttle, e.g., because the oxidation potential is the voltage at which "the redox shuttle is activated by its electrochemical conversion," thereby causing "[t]he cell potential during overcharge [to be] fixed at the oxidation potential of the redox shuttle." APPLE-1009 (Abraham), 2:23-27; see 3:47-54 ("The protective redox shuttle reaction is initiated when the cathode reaches the oxidation potential of the redox reagent as in 1). It proceeds with the diffusion of the oxidized species (R') to the anode where it is reformed as in 2). The reaction is sustained"), 5:11-18. The oxidation potential of the redox shuttle in the combination—and thus the overcharging protection voltage of the combination battery—is "slightly higher than the normal charge limit of the cell but lower than the oxidation potential of the electrolyte." APPLE-1009 (Abraham), 2:36-38; *see* 3:47-54 ("The protective redox shuttle reaction is

initiated when the cathode reaches the oxidation potential of the redox reagent"), 5:12-15 ("Proper matching of the overcharge protection additive (shuttle) to the electrochemical couple requires sufficient separation between the completion of lithium extraction from the cathode reaction and the onset of the shuttle activation"). The charge limit of the combination cell, *e.g.*, the charge cut-off voltage, is "from 4.4 V to 4.8 V." APPLE-1004 (Yamaki), [0014]; see [5a], above. Thus, the overcharging protection voltage of the combination battery is "slightly higher than" a voltage that is between 4.4 V and 4.8 V (e.g., slightly higher than a voltage of 4.4 V, 4.5 V, or 4.6 V; see APPLE-1004 (Yamaki), [0057]), i.e., the overcharging protection voltage is greater than 4.35 V. APPLE-1009 (Abraham), 2:36-38; *see* 3:47-54, 5:12-15. Indeed, Abraham describes the use of redox shuttles that are "activated at potentials between 4.2 and 4.5V," and thus that provide an overcharging protection voltage greater than 4.35 V. APPLE-1009 (Abraham), 5:17-18.

146. When the voltage in the battery falls below the oxidation potential of the redox shuttle, the redox shuttle reaction no longer proceeds, and charging/discharging of the battery can resume, *e.g.*, the overcharging protection provided by the redox shuttle is released. *See* APPLE-1009 (Abraham), 2:26-33 ("The cell potential during overcharge is fixed at the oxidation potential of the redox shuttle. This process is supported by diffusion of the oxidized products to

the anode where they recombine to form the starting material. Once the reformed material diffuses back to the cathode, it is oxidized and the cathode potential is maintained indefinitely at the oxidation potential of the redox reagent, until the time that charging is terminated.”). Thus, the overcharging protection release voltage, *e.g.*, the voltage at which overcharging protection is no longer provided, is also the oxidation potential of the redox shuttle. As discussed above, the oxidation potential of the redox shuttle in the combination is greater than 4.35 V, and thus the overcharging protection release voltage of the combination battery is also greater than 4.35 V, *i.e.*, greater than 4.15 V.

147. If a cell contains a redox shuttle whereby the voltage is prevented from exceeding some high value, even greater than 4.35V, the “release” voltage of the designed charging control system could also be higher than standard charge limits. Even if the release voltage was set higher than 4.35V in this example, the cell would be protected from overcharge by the chemical shuttle mechanism that prevents the cell voltage from rising past the point at which the redox reactions of the shuttle chemistry take effect.

148. To the extent that Patent Owner insists on identification of a specific redox shuttle compound, it would have been within the skill of a POSITA to identify a suitable redox shuttle, *e.g.*, one that is “activated at potentials between 4.2 and 4.5V,” for use in the combination battery, based on the guidance provided

by Abraham. APPLE-1009 (Abraham), 5:17-18. Abraham explains that "[t]he choice of a particular redox reagent will vary with the cathode material." APPLE-1009 (Abraham), 5:44-46. Indeed, although Abraham identifies thianthrene and 2,7-diacetyl thianthrene as specific examples, Abraham describes a general procedure to increase the oxidation potential of thianthrene-based compounds by "replacing hydrogen atoms on the aromatic rings with electron withdrawing groups." APPLE-1009 (Abraham), 4:7-10. Moreover, Abraham notes that the resulting compounds can be "screen[ed] for use as redox shuttle reagents" using cyclic voltammetry." APPLE-1009 (Abraham), 5:55-57. Given Abraham's guidance on the selection criteria for the redox shuttle (*e.g.*, its oxidation potential "slightly higher" than the charge cut-off voltage of the battery; APPLE-1009 (Abraham), 2:36-38) and Abraham's instruction on the synthesis and screening of redox shuttle compounds (*see id.*, 4:7-10, 5:55-57), it would have been a matter of straightforward and routine experimentation for a POSITA to identify a specific compound (*e.g.*, a specific thianthrene derivative) for use in the combination battery.

(ii) Claim 10

[10] A secondary lithium ion cell or battery according to claim 9, characterized in that the secondary lithium ion cell or battery has a first overcharging protection voltage of greater than 4.45 V, and an overcharge protection release voltage of greater than 4.25 V.

149. My analysis of claim [9] is applicable to claim [10]. The combination battery has an overcharging protection voltage of the combination battery is "slightly higher than" a voltage that is between 4.4 V and 4.8 V (*e.g.*, slightly higher than a voltage of 4.5 V or 4.6 V; *see* APPLE-1004 (Yamaki), [0057]), *i.e.*, the overcharging protection voltage is greater than 4.45 V. APPLE-1009 (Abraham), 2:36-38. Indeed, Abraham describes the use of redox shuttles that are "activated at potentials between 4.2 and 4.5V," *i.e.*, that have an oxidation potential, and thus provide an overcharging protection voltage, of greater than 4.45 V. APPLE-1009 (Abraham), 5:17-18. Additionally, the overcharging release voltage is greater than 4.25 V, at least because the overcharging release voltage is the oxidation potential of the redox shuttle, which is greater than 4.45 V.

C. [GROUND 2A] – CLAIMS 5-8 ARE OBVIOUS OVER YASUNAMI AND KOYAMA

1. Overview of Yasunami

150. Yasunami describes a "non-aqueous secondary battery having a high battery voltage, high discharge capacity (high energy density), and long charge/discharge cycle life." APPLE-1006 (Yasunami), [0006]; *see* [0001].

151. In Yasunami's battery, "negative electrode active material is a lithium-containing transition metal oxide represented by Li_xMO_j" where M is a transition metal "selected from Ti, V, Mn, Co, Fe, Ni, Nb, and Mo." APPLE-1006 (Yasunami), [0007]. The "positive electrode active material may be a transition metal oxide capable of reversibly occluding and releasing lithium ions," such as "a lithium-containing transition metal oxide represented by Li_yNO_z," where N is a transition metal "selected from Co, Mn, Ni, V, and Fe." *Id.*, [0007], [0021]; *see* [0021] (listing additional metals for the positive electrode active material: "lithium-containing oxides that also contain T, V, Cr, Mn, Fe, Co, Ni, Cu, Mo and/or W"). Yasunami also describes that "[a] conductive agent, a binder, and a filler can be added to the electrode mixture." *Id.*, [0046].

152. The battery also includes "a non-aqueous electrolyte containing a lithium salt." APPLE-1006 (Yasunami), [0009]; *see* [0048] ("The electrolyte generally consists of a solvent and a lithium salt (anion and lithium cation) dissolved in the solvent"). Alternatively, the electrolyte can be a "solid electrolye[]," such as "inorganic solid electrolytes" or "organic solid electrolytes." *Id.*, [0050]; *see* [0051]. Yasunami explains that "[t]here is no particular limitation on the amount of these electrolytes to be added to a battery, and the required amount depends on the amount of positive electrode active material used, the

amount of negative electrode active material used, and the size of the battery." *Id.*, [0049].

153. When the battery is charged and discharged, "the valence of the transition metal [of the positive and negative electrode active materials] is changed by occluding and releasing lithium ions during charging and discharging." APPLE-1006 (Yasunami), [0044]. Yasunami notes that this "is fundamentally different from that of metallic negative electrode active materials, such as lithium metal and lithium alloys, in which lithium is deposited and dissolved during charging and discharging. *Id.*

154. Yasunami's battery can be charged to a "end-of-charge voltage … in the range from 3.5 to 4.7 V," and Yasunami notes that "[i]f it is lower than 3.5 V, the battery voltage will be low, and if it is higher than 4.7 V, the positive electrode active material and the electrolyte may decompose." APPLE-1006 (Yasunami), [0034]. Moreover, Yasunami's battery is "discharged to a battery voltage of 0.5 to 1.5 V during the charge/discharge cycle." *Id.*, [0033].

155. In the context of the capacity of the negative electrode active material, Yasunami places "no particular restrictions on the use ratio for the positive electrode active material and the negative electrode active material," but states that "the ratio is preferably set so that the effective equivalents of each are equal (an effective equivalent is the equivalent that can substantially maintain cycle

performance)." APPLE-1006 (Yasunami), [0019]. Yasunami also indicates that "either the positive electrode active material or the negative electrode active material" can be increased. *Id.*, [0015].

156. Yasunami's discussion of "effective equivalents" refers to the presence of positive and negative electrode active material ***in the battery*** (*e.g.*, as opposed to the precursor materials used during lithium loading; *see* [0018]). This is the case at least because Yasunami refers to these effective equivalents in the context of "maintain[ing] cycle performance." APPLE-1006 (Yasunami), [0019]. The precursor lithium loading processes described in the previous paragraph involve "conducting a discharge in a redox system" or "conducting charging in a redox system." APPLE-1006 (Yasunami), [0018]. The "cycle performance" of the electrode active materials—the context in which the effective equivalents of the materials are discussed—is not a relevant consideration in either of these precursor loading processes: both redox charging and redox discharging are one-directional, non-cyclic processes. By contrast, "maintain[ing] cycle performance" of the electrode active materials is of interest in secondary batteries, where multiple charge/discharge cycles occur; and indeed is of particular interest to Yasunami, whose objective is to provide a battery with "good charge/discharge cycle characteristics." APPLE-1006 (Yasunami),, Abstract, [0019]. In this context, a POSITA would have understood, or at least found obvious, that Yasunami's

disclosure that "the effective equivalents of each [of the positive and negative electrode active materials] are equal" refers to the effective equivalents of the positive and negative electrode active materials in the battery itself. APPLE-1006 (Yasunami), [0019].

157. Moreover, Yasunami's description of the effective equivalents of the positive and negative electrode materials in its battery refers to the capacity of the positive and negative electrode active materials in the battery. Yasunami indicates that the "effective equivalent" of an electrode active material is "the equivalent that can substantially maintain cycle performance." APPLE-1006 (Yasunami), [0019]. The capacity of the electrode active materials in a battery impacts the cycle performance of the battery, *e.g.*, in that an unbalanced capacity between electrodes (*e.g.*, excessive positive electrode capacity) can degrade the ability of the battery to maintain capacity through multiple cycles. Having an "effective equivalent" amount of positive and negative electrode active material thus indicates an equivalent amount of capacity provided by each electrode.

158. As I noted above, in the context of the capacities of the active materials in the battery, Yasunami also indicates that alternative to the "effective equivalent" of the electrode active materials being equal, "either the positive electrode active material or the negative electrode active material" can be increased. APPLE-1006 (Yasunami), [0019]. Moreover, Yasunami describes

generally that "[t]he ratio of the positive electrode active material to the negative electrode active material in a non-aqueous secondary battery of the present invention depends on the types of active materials to be combined." APPLE-1006 (Yasunami), [0032].

159. Yasunami conducted "[c]harging/discharging cycle testing" on batteries having various compositions for their "[l]ithium-containing transition metal oxide negative electrode active materials." APPLE-1006 (Yasunami), [0062], [0066]. The positive electrode active material for each sample was $LiCoO_2$. *See id.*, [0064]. Yasunami's tests were carried out "at a final charge voltage of 3.9 to 4.3 V, a final discharge voltage of 1.8 V, and a current density of 2 mA/cm^2," and "[t]he number of cycles before the discharge capacity reached 60% of the initial value" was measured as a proxy for the "charge/discharge cycle life." *Id.*, [0066]. The results, shown in Tables 1-3, demonstrate that batteries of this composition, and charged to final charge voltages of 3.9 to 4.3 V, reach 60% capacity after approximately 200-300 cycles. *See* [0067], [0069], [0071].

2. Overview of Koyama

160. Koyama describes "a nonaqueous electrolyte secondary battery, such as a lithium secondary battery," and specifically describes a battery "with better charging characteristics than the prior art." APPLE-1007 (Koyama), [0001], [0005]. This improvement is achieved by setting a "capacity ratio (B/A) between

the electrode capacity A of the positive electrode and the electrode capacity B of the negative electrode." *Id.*, Abstract.

161. Like the composition of Yasunami's battery, Koyama's battery includes a "positive electrode active material[]" that is an "inorganic compound[] such as metal oxides of alkali metals and metal chalcogens," such as "Li-containing transition metal oxides such as Li_XMO_Y (where M includes at least one transition metal such as Co, Mn, Ni, V, and Fe)." APPLE-1007 (Koyama), [0008]. The negative electrode active material also can be a "metal oxide[]," such as a lithium-containing transition metal oxide: "Li_XMO_Y (where M includes at least one transition metal such as Ti, V, Mn, Co, and Fe)." *Id.*, [0009]. I note also that Koyama does not specify particular limits on the applicability of its teachings regarding the capacity ratio of lithium secondary batteries. For instance, while Koyama describes batteries with operation at a charge cut-off voltage of 4.2 V (*see id.*, [0021]), it does not preclude the implementation of its reversible capacity balance in batteries operated to different charge cut-off voltages. Moreover, Koyama describes implementation of its capacity ratio in batteries that are generally consistent with the composition of Yasunami's batteries, as I discuss below in Paragraphs [178]-[180].

162. Koyama describes adjusting "the capacity ratio (B/A) between the electrode capacity A of the positive electrode and the electrode capacity B of the

negative electrode" to "1.3<(B/A)≤2.5." APPLE-1007 (Koyama), [0006]. A capacity ratio in this range "s effective in achieving excellent characteristics such as a charge density above 400 (-W/kg)." *Id.*, [0027]. Specifically, "[i]f the capacity ratio (B/A) is less than 1.3, the improvement in charging characteristics is insufficient," while "the capacity ratio (B/A) exceed[ing] 2.5, … result[s] in a decrease in battery capacity and an increase in irreversible capacity." *Id.*, [0007].

3. The Yasunami-Koyama combination

163. In the Yasunami-Koyama combination, a lithium secondary cell is based on the lithium secondary cell described by Yasunami, and includes positive and negative electrode active materials in amounts that satisfy a capacity ratio based on the teachings of Koyama.

164. The composition of the electrodes and electrolyte of the combination battery is based on the teachings of Yasunami. For instance, the combination battery includes a "negative electrode active material" that "is a lithium-containing transition metal oxide represented by Li_xMO_j" where M is a transition metal "selected from Ti, V, Mn, Co, Fe, Ni, Nb, and Mo," based on the teachings of Yasunami. APPLE-1006 (Yasunami), [0007]. Notably, this negative electrode active material composition is consistent with the composition of Koyama's negative electrode active material. *See* APPLE-1007 (Koyama), [0009].

165. Additionally, as taught by Yasunami, the "positive electrode active material may be a transition metal oxide" such as "a lithium-containing transition metal oxide represented by Li_yNO_z," where N is a transition metal "selected from Co, Mn, Ni, V, and Fe." APPLE-1006 (Yasunami), [0007], [0021]. This is consistent with the composition of Koyama's positive electrode. *See* APPLE-1007 (Koyama), [0008].

166. The composition of the electrolyte is also based on the teachings of Yasunami. *See* APPLE-1006 (Yasunami), [0048]-[0051]. Notably, these electrolyte compositions are also consistent with the compositions described for Koyama's battery. *See* APPLE-1007 (Koyama), [0015].

167. The combination battery includes positive electrode active material and negative electrode active material in amounts such that the capacity ratio (B/A) between the capacity of the positive electrode active material and the capacity of the negative electrode active material is in the range of "$1.3<(B/A)\leq 2.5$," based on the teachings of Koyama. APPLE-1007 (Koyama), [0006]. Yasunami indicates its compatibility with such a ratio, noting that "the ratio of the positive electrode active material to the negative electrode active material … depends on the types of active materials to be combined." APPLE-1006 (Yasunami), [0032].

168. In operation, the end-of-charge voltage of the combination battery is between 3.5 V and 4.7 V, as taught by Yasunami. *See* APPLE-1006 (Yasunami),

[0034]. Importantly, the combination battery does not risk decomposition of the positive electrode active material or the electrolyte (*see* APPLE-1006 (Yasunami), [0034]) because the composition of the positive electrode active material and electrolyte are consistent with those described by Yasunami and thus compatible with this range of end-of-charge voltages. Additionally, the combination battery can be "discharged to a battery voltage of 0.5 to 1.5 V during the charge/discharge cycle," consistent with the operation of Yasunami's battery. *Id.*, [0033].

169. Notably, the combination battery maintains the advantages of "high battery voltage, high discharge capacity (high energy density), and long charge/discharge cycle life" that pertain to Yasunami's battery, at least because the combination battery retains the composition of the positive and negative electrode active materials and the end-of-charge voltage that Yasunami identifies as contributing to these advantages. *See* APPLE-1006 (Yasunami), [0006]-[0007].. Moreover, by providing the positive and negative electrode active materials in a ratio based on the teachings of Koyama, the "charging density" of the battery is further enhanced. *See* APPLE-1007 (Koyama), Abstract.

4. Reasons for the combination

170. It is my opinion that a POSITA would have been motivated to provide positive electrode active material and negative electrode active material in the

combination battery, *e.g.*, in amounts based on the teachings of Koyama, for at least the following reasons.

171. The first reason why a POSITA would have been motivated to pursue this implementation is to achieve improvements in charging performance (*e.g.*, charge rate) that are enabled by providing electrode materials that satisfy Koyama's range of capacity ratios, particularly given that Yasunami already indicates that such ratios may be appropriate. *See* APPLE-1007 (Koyama), [0007], [0027]; APPLE-1006 (Yasunami), [0019]. Koyama describes that electrode materials "that keep[] the capacitance ratio (B/A) above 1.3 and below 2.5 is effective in achieving excellent characteristics such as a charge density above 400 (-W/kg)," noting that "[i]f the capacity ratio (B/A) is less than 1.3, the improvement in charging characteristics is insufficient," while "if the capacity ratio (B/A) exceeds 2.5," this "result[s] in a decrease in battery capacity and an increase in irreversible capacity, which in turn reduces the actual capacity that can be drawn upon." APPLE-1007 (Koyama), [0007], [0027]; *see* [0005], [0010], [0027], [0029]. Improved charging performance can enable advantages such as more efficient (*e.g.*, faster) charging of the battery. A POSITA would have been motivated to implement a capacity ratio such as that described by Koyama to obtain a battery with better charging performance, *e.g.*, with the objective of producing a faster charging battery.

172. The description of "charging density" in Koyama uses measurement units of Watts/kg, an unusual but perfectly acceptable way to describe the rate at which energy is applied to the battery to change the state of charge. Traditional descriptions might focus on current (amps), with separate indication of the voltage during the charging process. The combination of amps and volts is in fact the energy or watts measurement that Koyama chooses to provide in the combined form.

173. The second reason why a POSITA would have recognized that the safety advantages of a battery in which there is more capacity in the negative electrode than in the positive—*e.g.*, a battery satisfying the capacity ratio described by Koyama—and would have been motivated to pursue this implementation to enhance the safety of the combination battery. I discussed why having more capacity in the negative electrode than in the positive electrode promotes battery safety in my discussion of the Yamaki-Sakamoto combination; that discussion is applicable here as well. *See* Paragraphs [41]-[42] of this Declaration. I also note that a POSITA would, again, have been encouraged to pursue this implementation based on Yasunami's teaching to pursue such a ratio. *See* APPLE-1006, [0019].

174. The third reason why a POSITA been motivated to provide electrode material based on the teachings of Koyama is because batteries having capacity in the negative electrode than in the positive—*e.g.*, a battery satisfying the capacity

ratio described by Koyama—have good cycle life characteristics. The POSITA would have pursued this combination to obtain a battery with the ability to retain capacity over a long cycle life. I discussed why having more capacity in the negative electrode than in the positive electrode promotes capacity retention in my discussion of the Yamaki-Sakamoto combination; that discussion is applicable here as well. *See* Paragraphs [37]-[39] of this Declaration. For instance, a POSITA implementing Yasunami's battery would have explored batteries having known ratios of positive and negative electrode active materials, and specifically would have looked to the teachings of Koyama for implementation details, because Koyama describes a specific range of ratios satisfying Yasunami's general guidance and in the context of a materials system consistent with that of Yasunami's battery.

175. <u>The fourth reason</u> why a POSITA would have been prompted to pursue this implementation is because this combination is just the application of known techniques (specifically, a secondary lithium ion battery having more negative electrode capacity than positive electrode capacity) to an already-known system (specifically, Yasunami's secondary lithium ion battery), and it produces predictable results (specifically, a secondary lithium ion battery having negative electrode active material and positive electrode active material in amounts to satisfy a specific capacity ratio).

176. It is my opinion that a POSITA would have expected to be successful in providing positive electrode active material and negative electrode active material in the combination battery in amounts based on the teachings of Koyama for at least the following reasons.

177. The first reason why a POSITA would have expected success is because Yasunami makes apparent that selecting a desired amount of positive and negative electrode material, *e.g.*, to achieve performance objectives (*e.g.*, charging performance, safety, and/or cycle life, as discussed above), is consistent with Yasunami's disclosure that "[t]he ratio of the positive electrode active material to the negative electrode active material in a non-aqueous secondary battery of the present invention depends on the types of active materials to be combined." APPLE-1006 (Yasunami), [0032]; *see* [0019] ("There are no particular restrictions on the use ratio for the positive electrode active material and the negative electrode active material … At the time, either the positive electrode active material or the negative electrode active material is preferably increased"). Given Yasunami's flexibility regarding the amounts of the positive and negative electrode active material, a POSITA would have reasonably believed that the specific amounts of materials to satisfy Koyama capacity ratio would be compatible with Yasunami's battery.

178. The second reason why a POSITA would have expected success in combining teachings based on both Yasunami and Koyama is because both describe secondary lithium ion batteries with similar materials systems. This is an indication that the teachings of Koyama are compatible with, and applicable to, the battery described by Yasunami.

179. For instance, both Yasunami and Koyama describe batteries having transition metal oxides for both positive and negative electrode active materials, and with overlapping lists of transition metals as elements in those metal oxides. For instance, Yasunami identifies "a lithium-containing transition metal oxide represented by Li_yNO_z," where N is a transition metal "selected from ***Co, Mn, Ni, V, and Fe***" for its positive electrode active material, while Koyama similarly identifies "Li-containing transition metal oxides such as Li_XMO_Y (where M includes at least one transition metal such as ***Co, Mn, Ni, V, and Fe***)" for its positive electrode active material. APPLE-1006 (Yasunami), [0007]; APPLE-1007 (Koyama), [0008]. Similarly, Yasunami's negative electrode active material is "a lithium-containing transition metal oxide represented by Li_xMO_j" where M is a transition metal "selected from ***Ti, V, Mn, Co, Fe***, Ni, Nb, and Mo," while Koyama's negative electrode active material can be a lithium-containing transition metal oxide "Li_XMO_Y (where M includes at least one transition metal such as ***Ti, V,***

Mn, Co, and Fe)." APPLE-1006 (Yasunami), [0007]; APPLE-1007 (Koyama), [0009].

180. Both Yasunami and Koyama also describe the use of similar organic electrolytes in their batteries. *See* APPLE-1006 (Yasunami), [0048]-[0051]; APPLE-1007 (Koyama), [0015]. For instance, at least ethylene carbonate and diethyl carbonate are described as the organic solvent by both Yasunami and Koyama, and both identify LiPF6 as the lithium salt.

181. Given the overlap in materials between Yasunami's and Koyama's secondary lithium ion batteries, a POSITA would have expected the teachings of Koyama to be relevant to, and combinable with, the teachings of Yasunami. And as I have noted previously, neither Yasunami nor Koyama gives any indication that the features of their respective batteries would preclude their being integrated with features of the other. In fact, it is the opposite: Yasunami explicitly encourages a POSITA to look toward a ratio of positive to negative material that is consistent with the teachings of Koyama. *See* APPLE-1006 (Yasunami), [0019].

182. <u>The third reason</u> why a POSITA would have expected success is that secondary lithium ion batteries having more negative than positive electrode capacity were well-known in the art. *See* APPLE-1020 (Arora), 5; APPLE-1021 (Satoshi), [0011]; APPLE-1005 (Sakamoto), [0007].. Indeed, as I discussed previously, such batteries have numerous advantages, *e.g.*, in terms of safety, cycle

life, and other performance characteristics. Moreover, it was also well-known in the art to operate secondary lithium ion batteries with a charge cut-off voltage of greater than 4.2 V. *See* APPLE-1023 (Tanaka), 1:25-35; APPLE-1024 (Kawakami), 40:3-14; APPLE-1025 (Amatucci), 2:63-3:3; APPLE-1012 (Kawai), 1.

5. Analysis

(i) Claim 5

[5pre] A secondary lithium ion cell or battery, characterized in that

183. The Yasunami-Koyama combination renders obvious a secondary lithium ion cell or battery, for the following reasons.

184. First, the battery based on the Yasunami-Koyama combination is a lithium secondary battery, e.g., based on Yasunami's "a non-aqueous secondary battery having a negative electrode material that is a lithium-containing transition metal oxide represented by Li_xMO_j … and a positive electrode active material that is a lithium-containing transition metal oxide represented by Li_yNO_z"." APPLE-1006 (Yasunami), Abstract; see [0001], [0006]-[0007], [0009], [0032]-[0033], [0073], claims 1-9.

185. Additionally, the combination battery is a lithium ion cell or battery, at least because movement of lithium ions occurs during charging and discharging. For instance, Yasunami describes that in operation of its battery, "the valence of the transition metal" of both the positive electrode active material and the negative

electrode active material "is changed by occluding and releasing lithium ions during charging and discharging." APPLE-1006 (Yasunami), [0044]; *see* [0021] ("The positive electrode active material used in the present invention may be a transition metal oxide capable of reversibly occluding and releasing lithium ions"), [0015] ($LipCoVO_4$ and $LipNiVO_4$ (where p is in the range from 0.3 to 2.2) is an especially preferable negative electrode active material precursor for use in the present invention. The p-value is the value before the start of charging/discharging, and increases or decreases with charging or discharging."), [0026] ("Even more preferred examples of lithium-containing metal oxides in the present invention include $LiyCoO_2$, $LiyNiO_2$, $LiyCoaNi_1$-aO_2, $LiyMn_2O_4$, and LiyCob V_1-bOz," where "the y value is the value before the start of charging and discharging, and increases or decreases due to charging or discharging").

[5a] the secondary lithium ion cell or battery has a charge cut-off voltage of greater than 4.2V but less than 5.8 V, and

The charge cut-off voltage of the combination battery is based on Yasunami's end-of-charge voltage

186. The battery of the combination has "an end-of-charge voltage of 3.5 to 4.7 V," based on the teachings of Yasunami. APPLE-1006 (Yasunami), [0073]; see [0034], [0066], [0068], claim 1 Tables 1-3. This end-of-charge voltage is the charge cut-off voltage of the combination battery, as I discuss in the next paragraphs.

187. A POSITA would have understood, or at least found obvious, that the "end-of-charge voltage," as described by Yasunami and as implemented in the combination battery, is a charge cut-off voltage at least because it is the maximum voltage to which the battery is configured to be charged, e.g., the voltage at which charging of the battery is stopped. APPLE-1006 (Yasunami), [0034]; see [0066]-[0067] ("Charging/discharging cycle testing was performed on the prepared batteries at a final charge voltage of 3.9 to 4.3 V," shown as "End-of-Charge Voltage" in Table 1), [0068]-[0069].

188. In a specific example, a POSITA would have understood, or at least found obvious, that the "end-of-charge voltage," as described by Yasunami and as implemented in the combination battery, is the voltage at which the charging changes from constant current charging to constant voltage charging, as discussed below, and thus that the end-of-charge voltage is the charge cut-off voltage. APPLE-1006 (Yasunami), [0034].

189. Generally, there are two approaches to charging secondary batteries: constant current charging and constant voltage charging, although constant voltage charging alone is typically not used with secondary lithium batteries. Constant voltage charging would require extremely high currents to supply the initial surge in current needed to increase the voltage to a "max voltage" level during the earliest stage of charging. It is more practical to limit the current to a constant,

acceptable value, and monitor the voltage as it increases. In keeping with conventional charging techniques for secondary lithium batteries, a POSITA would have understood, or at least found obvious, that Yasunami uses constant current charging in charging its batteries, and thus that its description of the end-of-charge voltage is in the context of constant current charging.

190. Moreover, as I discussed above for the Yamaki-Sakamoto ground, in constant current charging, once the charge cut-off voltage is reached, either charging is stopped, or charging changes to constant voltage charging for further charging of the battery. The latter approach is advantageous in that it provides additional capacity to the battery, *e.g.*, as compared to the former approach. Consistent with Yasunami's objective of providing a battery with "high discharge capacity," a POSITA would have understood, or otherwise at least found obvious, that Yasunami employs this second charging method, *e.g.*, to obtain these further capacity gains. APPLE-1006 (Yasunami), [0006].

191. Thus, assuming constant current-constant voltage charging for Yasunami's battery, Yasunami's end-of-charge voltage (*e.g.*, the maximum voltage to which the battery is configured to be charged (*see* APPLE-1006 (Yasunami), [0066]-[0067])) is thus the potential at which the charging changes from constant current charging to constant voltage charging. That is, Yasunami's end-of-charge

voltage is, or at least renders obvious, the charge cut-off voltage of the combination battery.

Yasunami's end-of-charge voltage values are a disclosure of, or otherwise render obvious, the claimed range of charge cut-off voltages

192. The end-of-charge voltage (*e.g.*, the charge cut-off voltage) of the combination battery is "3.5 to 4.7 V," consistent with the teachings of Yasunami. APPLE-1006 (Yasunami), [0073]; see [0034] ("The end-of-charge voltage is preferably in the range from 3.5 to 4.7 V, more preferably from 3.7 to 4.5 V, and even more preferably from 3.8 to 4.4 V."), [0066] ("Charging/discharging cycle testing was performed on the prepared batteries at a final charge voltage of 3.9 to 4.3 V."), claim 1 ("A method for charging and discharging a non-aqueous secondary battery" where "an end-of-charge voltage [is] set from 3.5 to 4.7 V"), [0068], Tables 1-3. Table 1 is reproduced below as an example, with charge cut-off voltages within the claimed range highlighted.

[Table 1]

Table 1

No.		Negative Electrode Active Material	End-of-Charge Voltage	End-of-Discharge Voltage Prior to Charging	Energy Density After Discharge to 1.8 V	Cycles (No.)
1	Invention	a	4.3 V	1.0 V	2450 mWH	220
2	"	a	4.2	1.0	2310	255
3	"	a	4.1	0.8	2250	200
5	"	a	4.0	0.6	2120	280
4	"	a	"	1.0	"	290
5	"	a	"	1.3	"	285
6	"	a	"	1.5	"	195
7	"	a	3.9	0.9	1990	305
8	"	b	4.2	1.0	2290	260
9	"	b	4.0	1.0	2030	275
10	"	c	4.2	1.0	2210	190
11	"	c	4.0	1.0	1950	245
A	C. Ex.	a	4.0 V	1.8 V	2120 mWH	120
B	"	b	"	1.8	2030	95
C	"	c	"	1.8	1950	115
D	"	a	"	1.6	2120	140
E	"	a	4.1	0.4	2250	120

193. At least one of the charge cut-off voltages disclosed by Yasunami and incorporated into the combination battery (4.3 V; see APPLE-1006 (Yasunami), [0067] (line 1 of Table 1), [0069] (line 1 of Table 2), [0071] (No. 4 in Table 3)) falls within the range of charge cut-off voltages recited in claim 5 (greater than 4.2 V but less than 5.8 V). Thus, based on the disclosure of Yasunami, the Yasunami-Koyama combination renders obvious a battery having the claimed range of charge cut-off voltages.

194. Alternatively or additionally, the charge cut-off voltage range recited in claim 5 (greater than 4.2 V but less than 5.8 V) overlaps with the range of charge cut-off voltages of described by Yasunami and incorporated into the combination battery (between 3.5 V and 4.7 V). I have been informed that a prior art disclosure of range of values that overlaps with a claimed range of values is

considered to render the claimed range obvious, unless there is an indication of criticality or unexpected results associated with the claimed range. Here, as I discuss below, there is no such indication of criticality or unexpected results, and I thus conclude that range of charge cut-off voltages described by Yasunami and incorporated into the combination battery render obvious the range of charge cut-off voltages recited in claim 5.

195. Specifically, I have determined that the ’641 Patent does not demonstrate that the 4.2 V lower limit of its claimed range of charge cut-off voltages achieves any unexpected results relative to the overlapping charge cut-off voltage range of the combination battery.

196. Notably, the ’641 Patent does not even present results for experiments conducted at any charge cut-off voltage below 4.2 V. Instead, it presents data for lithium ion secondary cells tested with charge cut-off voltages starting at a minimum of 4.2 V and extending up to 5.9 V. *See* APPLE-1001 (the 641 Patent), 10:29-12:55 (Examples 1-13), Table 2 (Examples 14-18), Table 3 (Examples 19-23), Table 4 (Examples 24-28). There are no data demonstrating the behavior of cells charged to a charge cut-off voltage *below* 4.2 V. Without such data, there is no way to conclude whether there is anything critical about this arbitrary 4.2 V threshold.

197. The ’641 Patent does characterize the 4.2 V threshold as being “well accepted as a technical requirement in the industry of manufacture of secondary lithium ion battery,” and states (but does not support this statement with experimental data) that that this industrial “technical requirement” is because, in batteries with a charge cut-off voltage above 4.2 V, “the positive electrode materials and negative electrode materials will undergo structure change, the electrolyte may decompose, and the recycle properties of the cell will be adversely effected.” APPLE-1001 (the 641 Patent), 1:46-52. As I have demonstrated throughout this report, though, this is not true: Yasunami demonstrates that charge cut-off voltages above 4.2 V are feasible. *See* APPLE-1006 (Yasunami), [0034]. And as previously mentioned, there are numerous other examples showing that secondary lithium ion batteries can operate with a charge cut-off voltage of greater than 4.2 V.

[5b] a ratio of positive electrode material to negative electrode material of the secondary lithium ion cell or battery is from 1:1.0 to 1:2.5, as calculated by a theoretic capacity with a charge cut-off voltage set at 4.2 V.

Positive and negative electrode materials in the combination battery

198. The battery of the Yasunami-Koyama combination includes positive electrode material, e.g., based on the positive electrode active material of Yasunami’s battery. For instance, in the combination, Yasunami describes a “positive electrode active material,” which “is a lithium-containing transition metal

oxide represented by Li_yNO_z (where N represents at least one transition metal selected from Co, Mn, Ni, V, and Fe, y is in the range from 0.2 to 1.2, and z is in the range from 1.4 to 3)." APPLE-1006 (Yasunami), [0007]; *see* [0009] ("The non-aqueous secondary battery of the present invention has a basic configuration comprising a positive electrode active material"), [0021] ("The positive electrode active material used in the present invention may be a transition metal oxide capable of reversibly occluding and releasing lithium ions, and is preferably a lithium-containing transition metal oxide."), [0044] ("the positive electrode active materials … are believed to be compounds in which the valence of the transition metal is changed by occluding and releasing lithium ions during charging and discharging"), [0019], [0022]-[0030], [0032], [0035], [0042], [0049], [0058], [0064], [0070]-[0071], [0073], claims 1 and 9.

199. The battery of the combination also includes negative electrode material, *e.g.*, based on Yasunami's "negative electrode active material," which "is a lithium-containing transition metal oxide represented by Li_xMO_j (where M represents at least one transition metal selected from Ti, V, Mn, Co, Fe, Ni, Nb, and Mo, x is in the range from 0.17 to 11.25, and j is in the range from 1.6 to 4.1)." APPLE-1006 (Yasunami), [0007]; *see* [0009] ("The non-aqueous secondary battery of the present invention has a basic configuration comprising … a negative electrode active material"), [0018] ("The negative electrode active material of the

present invention is preferably obtained by inserting lithium ions into a negative electrode active material precursor of a transition metal oxide and/or a lithium-containing transition metal oxide"), [0044] ("the negative electrode active materials … are believed to be compounds in which the valence of the transition metal is changed by occluding and releasing lithium ions during charging and discharging"), [0017], [0019]-[0020], [0028], [0031]-[0032], [0035], [0042], [0045], [0049], [0058], [0062]-[0063], [0073], Tables 1-3, claims 1 and 4-8.

The ratio of positive electrode material to negative electrode material in the combination battery

200. In the Yasunami-Koyama combination battery, positive electrode active material and negative electrode active material are provided in amounts such that the "the capacity ratio (B/A) between the electrode capacity A of the positive electrode and the electrode capacity B of the negative electrode is $1.3<(B/A)\leq 2.5$," based on the teachings of Koyama. APPLE-1007 (Koyama), [0006]. The reciprocal of Koyama's capacity ratio—i.e., the capacity A of the positive electrode over the capacity B of the negative electrode—is, in the combination battery, the ratio of positive electrode material to negative electrode material, as calculated by a capacity of the electrode active materials, as discussed in the following paragraphs.

201. As I discussed above for the Yamaki-Sakamoto combination, the capacity of an electrode in a lithium secondary battery is a function of the capacity

per unit mass of the electrode active material (*e.g.*, in units of mAh/g) and the amount (*e.g.*, in grams) of the electrode active material in the electrode. Thus, the reciprocal of Koyama's capacity ratio is the ratio between (1) the capacity per unit mass of the positive electrode active material (*e.g.*, in mAh/g) times the mass of the positive electrode active material (*e.g.*, in g) in the battery and (2) the capacity per unit mass of the negative electrode active material times the mass of the negative electrode active material. This is expressed in the following equation:

$$Capacity\ ratio\ (reciprocal) = \frac{\left(\frac{mAh}{g} * g\right) pos.electrode\ active\ material}{\left(\frac{mAh}{g} * g\right) neg.electrode\ active\ material}.$$

202. I note that Koyama confirms that its capacity ratio is based on the amount (*e.g.*, mass) of each electrode active material in the battery. For instance, Koyama describes a series of experiments in which the composition of the positive and negative electrodes (and thus the capacity per unit mass of each electrode) remained constant, and "the thickness of [the] negative electrode [] was adjusted" to achieve a target capacity ratio. APPLE-1007 (Koyama), [0017]; *see id.* ("when the thickness of [the] positive electrode [] was 127 μm and the thickness of negative electrode 2 was 100 μm, the capacity ratio (B/A) was 1.40"), [0018] ("the thickness of the negative electrode 2 was changed to 125 μm so that the capacity ratio (B/A) was 1.79"), [0019] ("the thickness of the negative electrode 2 was changed to 161 μm so that the capacity ratio (B/A) was 2.43"), [0020]. The way to

achieve these different capacity ratios using the same materials is to use different amounts (*e.g.*, different masses) of the positive and/or negative electrode active materials in each battery. That is, the amount (mass) of positive and negative electrode active material in the battery, along with the capacity per unit mass of the materials, determines the capacity ratio of Koyama's batteries.

203. That is, a POSITA would have understood, or at least found obvious, that Koyama's "capacity ratio" (reciprocal) in the combination battery is calculated as the product of the capacity of the positive electrode active material per unit mass and the mass of the positive electrode active material in the battery, divided by the product of the capacity of the negative electrode active material per unit mass and the mass of the negative electrode active material in the battery. This ratio, in the combination battery, is the ratio of positive electrode material to negative electrode material, as calculated by a capacity of the electrode active materials.

204. Moreover, in the combination, Koyama describes that the capacity ratio (e.g., the ratio of positive electrode material to negative electrode material) is calculated by a theoretic capacity, and specifically the theoretic capacity of the positive electrode active material. For instance, Koyama states that "the electrode capacity A of positive electrode 1 was derived from the theoretical capacity, while the electrode capacity B of negative electrode 2 was derived from the measured value." APPLE-1007 (Koyama), [0017].

205. Additionally, a POSITA would have understood, or at least found obvious, that Koyama's capacity ratios (*e.g.*, as applied to the combination battery) are calculated by a capacity with a charge cut-off voltage set at 4.2 V. For instance, Koyama notes that, in the context of experiments in which "the samples were charged … to an upper voltage of 4.2 V," "the electrode capacity B of negative electrode 2 was derived from the measured value." APPLE-1007 (Koyama), [0017], [0022]. Because the capacity ratio—which is dependent upon the measured capacity of the negative electrode—is a variable in these tests to 4.2 V, a POSITA would have recognized that the capacity of the negative electrode at 4.2 V is a relevant quantity (e.g., for the reasons I discussed above for Ground 1A, see Paragraph [81] of this document), and thus would have understood or found obvious for the capacity of the negative electrode to be measured at an "upper voltage" (*e.g.*, a charge cut-off voltage) of 4.2 V. APPLE-1007 (Koyama), [0022]. Moreover, a POSITA would have understood this or found it obvious because it was conventional for capacity values to be determined and/or provided with respect to a 4.2 V charge cut-off voltage, as I discussed above for the Yamaki-Sakamoto ground.

The value of the ratio in the combination

206. In the combination battery, positive electrode active material and negative electrode active material are provided in amounts such that the "the

capacity ratio (B/A) between the electrode capacity A of the positive electrode and the electrode capacity B of the negative electrode is 1.3<(B/A)≤2.5," based on the teachings of Koyama. APPLE-1007 (Koyama), [0006]; see [0007] ("a value of 1.4 or higher is preferred"), [0012] ("the capacity ratio (B/A) between the electrode capacity A of the positive electrode 1 and the electrode capacity B of the negative electrode 2 is 1.3<(B/A)≤2.5, specifically, B/A=1.40"), [0017]-[0019], claim 1.

207. The ratio of positive electrode material to negative electrode material in the combination battery is the reciprocal of Koyama's capacity ratio, as discussed above. That is, the ratio of positive electrode material to negative electrode material in the combination battery is, e.g., between 1:1.3 and 1:2.5 (Koyama's "1.3<(B/A)≤2.5") or 1:1.40 (Koyama's "B/A=1.40"). APPLE-1007 (Koyama), [0006], [0012].

208. Koyama's 1:1.3 to 1:2.5 range falls within the range of ratios recited in claim 5 (1:1.0 to 1:2.5). See APPLE-1007 (Koyama), [0006], [0012]. Moreover, Koyama discloses specific points within the claimed range: e.g., 1:1.40 (*see* APPLE-1007 (Koyama), [0012]), and 1:1.10 and 1:2.5 (the endpoints of the disclosed ranges; see id., [0006]).

209. Thus, based on the disclosure of Koyama, the Yasunami-Koyama combination renders obvious a battery having positive electrode material and

negative electrode material in a ratio of from 1:1.0 to 1:1.25. See APPLE-1007 (Koyama), [0006]-[0007], [0012], [0017]-[0019], claim 1.

(ii) **Claim 6**

[6] A secondary lithium ion cell or battery according to claim 5, characterized in that the secondary lithium ion cell or battery has a charge cut-off voltage within a range from 4.3 V to 5.2 V.

210. My analysis of element [5a] applies also to claim 6. Yasunami's 4.3 V charge cutoff voltage falls within the range of charge cut-off voltages recited in claim 6 (4.3 V to 5.2 V). See APPLE-1006 (Yasunami), [0067], [0069], [0071]. Thus, based on the disclosure of Yasunami, the Yasunami-Koyama combination renders obvious that the combination battery has a charge cut-off voltage within a range from 4.3 V to 5.2 V.

211. Alternatively or additionally, Yasunami's range of charge cut-off voltages (e.g., 3.5 to 4.7 V) falls within the range of charge cut-off voltages recited in claim 6 (4.3 V to 5.2 V). See APPLE-1006 (Yasunami), [0034], [0066], [0068], [0073], Tables 1-3, claim 1. Because there is no indication of criticality or unexpected results associated with the claimed range of charge cut-off voltages, as I discuss below, I conclude that the range of charge cut-off voltages described by Yasunami and incorporated into the combination battery render obvious the range of charge cut-off voltages recited in claim 6. Specifically, I note that the '641 Patent measures the percent capacity retention after 400 cycles for various charge

cut-off voltages ranging from 4.2 V to 5.9 V. *See* APPLE-1001 ('641 Patent), 10:29-14:67. But, as demonstrated by example in Table 2 below, the '641 Patent does not measure any difference in behavior, or any onset of surprising or unexpected results, associated with a charge cut-off voltage of 4.3 V. Instead, the capacity retention results are generally consistent: 84.23% in a battery charged to 4.2 V, 85.12% in a battery charged to 4.3 V, and 83.45% in a battery charged to 4.6 V. Similar results are also shown in Tables 3-4, and detailed in the '641 Patent's description of its experiments in 10:29-14:67.

TABLE 2

Example	Charge cut-off voltage (V)	Processing change	Recycle property	Capacity (mAh)
14	4.20	—	Maintaining 84.23% after 400 recycles	375
15	4.30	—	Maintaining 85.12% after 400 recycles	405
16	4.60	The ratio of positive electrode material to negative electrode material is 1:1.6.	Maintaining 83.45% after 400 recycles	575
17	5.80	The ratio of positive electrode material to negative electrode material is 1:2.5, and substance A is added into the electrolyte solution.	Maintaining 82.83% after 400 recycles	567
18	5.90	The ratio of positive electrode material to negative electrode material is 1:2.7, and substance A is added into the electrolyte solution.	Maintaining 54.23% after 30 cycles	312

(iii) **Claim 7**

[7] A secondary lithium ion cell or battery according to claim 5, characterized in that the secondary lithium ion cell or battery has a charge cut-off voltage within a range from 4.3 V to 4.8. V.

212. My analysis of element [5a] applies also to claim 7. Yasunami's 4.3 V charge cutoff voltage falls within the range of charge cut-off voltages recited in claim 6 (4.3 V to 4.8 V). Thus, based on the disclosure of Yasunami, the Yasunami-Koyama combination renders obvious that the combination battery has a charge cut-off voltage within a range from 4.3 V to 4.8 V.

213. Alternatively or additionally, Yasunami's range of charge cut-off voltages (e.g., 3.5 to 4.7 V) falls within the range of charge cut-off voltages recited in claim 6 (4.3 V to 4.8 V). See APPLE-1006 (Yasunami), [0034], [0066], [0068], [0073], Tables 1-3, claim 1. For similar reasons as those I discussed for claim [6], the '641 Patent provides no indication of unexpected results or criticality associated with this range of charge cut-off voltages recited in claim 7, and I thus conclude that the range of charge cut-off voltages described by Yasunami and incorporated into the combination battery render obvious the range of voltages recited in claim 7. Thus, based on the disclosure of Yasunami, the Yasunami-Koyama combination renders obvious that the combination battery has a charge cut-off voltage within a range from 4.3 V to 4.8 V.

(iv) **Claim 8**

[8] A secondary lithium ion cell or battery according to claim 5, characterized in that the ratio of positive electrode material to negative electrode material of the secondary lithium ion cell or battery is from 1:1.15 to 1:2.5.

214. My analysis of element [5b] also applies to claim 8. In the combination battery, the ratio of positive electrode material to negative electrode material (e.g., based on Koyama's "capacity ratio" of "$1.3<(B/A) \leq 2.5$") is from 1:1.3 to 1:2.5, or 1:1.40. APPLE-1007 (Koyama), [0006], [0012]. This range of ratios, as well as the specific disclosed values for the ratio, fall within the range of ratios recited in claim 8 (1:1.15 to 1:2.5). Thus, based on this disclosure, the Yasunami-Koyama combination renders obvious that the combination battery has a ratio of positive electrode material to negative electrode material from 1:1.15 to 1:2.5.

(v) **Claim 9**

[9] A secondary lithium ion cell or battery according to claim 5, characterized in that the secondary lithium ion cell or battery has a first overcharging protection voltage of greater than 4.35V, and an overcharging protection release voltage of greater than 4.15 V.

215. Yasunami describes that the "charger" of its battery "may also be provided with a circuit that incorporates measures against overcharging and overdischarging." APPLE-1006 (Yasunami), [0060]. A charger, and its overcharging protection circuit, such as that described by Yasunami are provided with the Yasunami-Koyama combination battery. A POSITA would have

understood, or at least found to be obvious, that a circuit such as that of Yasunami's charger, and implemented in the combination battery, has an overcharging protection voltage and an overcharging protection release voltage. Indeed, because this is conventional functionality of overcharging protection circuits that enable these circuits to provide overcharging protection. *See* APPLE-1028 (Eguchi), 1:25-30 ("battery packs are usually provided with a battery protection circuit for preventing over-charging or over-discharging of lithium ion secondary batteries"), 4:55-65 ("if the cell voltage reaches an over-charging protection voltage, as a pre-set voltage, the detection control IC 3 outputs … [an] over-charging control signal" to "enable[] prohibition of over-charging to the battery cell"), 5: 50-62. Specifically, the overcharging protection voltage of such a circuit is the voltage at which operation of the circuit is triggered, *e.g.*, to prevent damage to the electrodes that could otherwise result from being in an overcharged state, *e.g.*, as I discussed above for the Yamaki-Sakamoto-Abraham ground. Moreover, I interpret the overcharging protection release voltage is the voltage at which the circuit disengages, *e.g.*, to re-enable charging/discharging operation of the battery.

216. A POSITA would have understood or found obvious that the overcharging protection voltage of the charger is slightly higher than the charge cut-off voltage of the combination battery. This is a conventional configuration for

an overcharging protection circuit, and it enables the battery to reach its full charge cut-off voltage without interruption by the protection circuit—if the overcharging protection voltage were below the charge cut-off voltage of the battery, the overcharging protection would engage before the battery reaches its fully charged voltage. *See* APPLE-1029 (Iwaizono), 10:1-65, 15:35-40 ("if the voltage at which charging has been completed is set on the charger side to 4.20 V, then the charge-prohibiting voltage for the main control means 1 is set to 4.30 V, and the detected set voltage in a prolonged overcharging state for the sub-control means 40 is set to 4.45 V.) As I established above, the charge cut-off voltage of the combination battery is 3.5 to 4.7 V. Thus, when the combination battery is configured to have a charge cut-off voltage within this range, and when the charge cut-off voltage is at least 4.35 V (which falls inside this 3.5-4.7 V range), the overcharging protection voltage of the protection circuit is likewise greater than 4.35 V.

217. A POSITA also would have understood or found obvious that the overcharge protection release voltage of the protection circuit is slightly below the overcharging protection voltage of the circuit. Again, this is a conventional configuration for overcharge protection circuits, and it enables charge/discharge cycling to resume once the positive electrode voltage returns to its normal operational range. *See* APPLE-1029 (Iwaizono), 10:1-65. Thus, when the charge cut-off voltage of the battery is greater than 4.35 V, as I discussed in the preceding

paragraph, the overcharging protection release voltage is at or slightly below 4.35 V, and thus is greater than 4.15 V.

(vi) **Claim 10**

[10] A secondary lithium ion cell or battery according to claim 9, characterized in that the secondary lithium ion cell or battery has a first overcharging protection voltage of greater than 4.45 V, and an overcharge protection release voltage of greater than 4.25 V.

218. My discussion above for claim [9] applies here as well. When the combination battery has a charge cut-off voltage within the disclosed 3.5-4.7 V range and at least 4.45 V, the overcharging protection voltage of the protection circuit is likewise greater than 4.45 V, and the overcharge protection release voltage, which is slightly less than the overcharging protection voltage, is greater than 4.25 V.

(vii) **Claims 11-14**

[11] A secondary lithium ion cell or battery according to claim 5, wherein the secondary lithium ion cell or battery maintains at least 7500 of capacity after 400 cycles.

[12] A secondary lithium ion cell or battery according to claim 5, wherein the secondary lithium ion cell or battery maintains at least 75.83% of capacity after 400 cycles.

[13] A secondary lithium ion cell or battery according to claim 5, wherein the secondary lithium ion cell or battery maintains at least 80% of capacity after 400 cycles.

[14] A secondary lithium ion cell or battery according to claim 5, wherein the secondary lithium ion cell or battery maintains at least 80.72% of capacity after 400 cycles.

A POSITA would have found obvious that the Yasunami-Koyama battery has the claimed capacity retention

219. It is my opinion that a POSITA would have understood, or at least found obvious, that the Yasunami-Koyama battery maintains at least 75% of capacity after 400 cycles. . Moreover, it is also my opinion that a POSITA would have understood, or at least found obvious, that the Yasunami-Koyama battery maintains any other reasonable but arbitrary percentage of capacity, such as at least 75.83%, 80%, or 80.72% of capacity, after 400 cycles (or after another reasonable but arbitrary number of cycles).

220. As I noted above for Ground 1A, the ’641 Patent provides no description of how to achieve the recited capacity retention beyond specifying ranges for the charge cut-off voltage and ratio of electrode materials. *See* APPLE-

1001 (the 641 Patent), Examples 1-28. And as I demonstrated above, the combination battery satisfies the underlying compositional (*e.g.*, ratio of positive to negative electrode material) and operational (*e.g.*, charge cut-off voltage) limitations that yield these recited capacity results. Indeed, there is even overlap in the specific materials of the '641 Patent's battery (*e.g.*, "lithium cobalt composite oxides" or "lithium manganese cobalt composite oxides" as positive electrode active material, "a carbonaceous substance" as negative electrode active material, and "non-aqueous electrolytes … of the type well known in the art") and the materials of the Yasunami-Koyama battery. APPLE-1001 (the 641 Patent), 8:10-9:15; *see* my discussion of element [5a] above. Thus, because the disclosures of Yasunami and Koyama, incorporated into the combination battery, align with the elements that the '641 Patent indicates as providing the recited capacity retention, I believe that a POSITA would find obvious that the Yasunami-Koyama combination battery, on its face, maintains at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles.

221. Alternatively or additionally, I believe that a POSITA would have found it obvious that the Yasunami-Koyama combination battery maintains at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles, based on the cycle life performance achieved by Yasunami's battery, the expected effect that the

combination with Koyaama would have had on cycle life, and on expectations for battery performance over a larger number of cycles.

222. The Yasunami-Koyama combination battery includes lithium-containing transition metal oxides for both the positive electrode and the negative electrode. *See* APPLE-1006 (Yasunami), [0007]. Yasunami explains that these electrode materials are advantageous in achieving "good charge/discharge cycle characteristics." *Id.*, Abstract; *see* [0006] ("It is an object of the present invention to provide a non-aqueous secondary battery having … long charge/discharge cycle life"), [0001]. Yasunami goes on to describe the mechanism by which these compositions improve cycle life. For instance, Yasunami describes that with lithium-containing transition metal oxide as the negative electrode material, "the basic structure" of the material "does not change further due to charging and discharging," and that the lithium-containing transition metal oxide of the positive electrode is "capable of reversibly occluding and releasing lithium ions." *Id.*, [0020]-[0021]. That is, Yasunami's materials can charge and discharge reversibly, without significant structural changes, which allows a battery made of these materials to have a long cycle life with little decay in capacity throughout many cycles. Indeed, Yasunami conducts experiments to demonstrate that this is the case, showing that its batteries, when charged to a charge cut-off voltage of 4.3 V, have a cycle life of 220 cycles (Table 1), 225 cycles (Table 2), or 230 cycles

(Table 2) "before the discharge capacity reached 60% of the initial value." *Id.*, [0066].

223. Moreover, the positive and negative electrode active materials in the combination battery are provided in a ratio based on the teachings of Koyama, such that there is more negative electrode capacity than positive electrode capacity. As I discussed previously in Paragraphs [37]-[39] of this Declaration, it was well-known that having more negative electrode capacity than positive electrode capacity was beneficial for cycle life of a secondary lithium battery. Thus, with this understanding in hand, a POSITA would have been confident that incorporating teachings based on Koyama into Yasunami's battery—and thus providing more negative electrode capacity than positive electrode capacity—would result in a battery having cycle life at least consistent with (but probably better than) that demonstrated for Yasunami's battery. For instance, a POSITA could have expected such a combination battery to maintain at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles.

A POSITA would have found it obvious to obtain a Yasunami-Koyama combination battery that has the claimed percent capacity retention

224. Separately, I also believe that a POSITA would have found it obvious to obtain a battery consistent with the Yasunami-Koyama battery and that maintains a reasonable, but arbitrary percentage of its capacity after some reasonable but also arbitrary number of cycles—for instance, that maintains at least

75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles—based on routine experimentation guided by a goal of improving cycle life.

225. First, as I have previously discussed, Yasunami has an objective of improving cycle life. *See* APPLE-1006 (Yasunami), Abstract ("To provide a non-aqueous secondary battery having … good charge/discharge cycle characteristics"). More generally, as I discussed previously for Ground 1A, a POSITA would have been well aware of cycle life criteria that would have applied to batteries to enable them to be commercially viable; this discussion applies equally to the Yasunami-Koyama combination battery. *See* Paragraphs [37]-[39] of this Declaration; APPLE-1016 (Guyomard), 5:32-36; APPLE-1017 (Wendsjo), 5:10-17; APPLE-1018 (Uemura), FIG. 3, [0081]; APPLE-1015 (Handbook), 23.10, FIG. 36.35b. In this context, a POSITA developing the Yasunami-Koyama battery would have found it obvious to aim for a battery that is able to retain a large percentage of its capacity over a large number of cycles.

226. With this objective in mind—generating a battery with long cycle life (both as an articulated goal of Yasunami and as a practical, commercial objective)—a POSITA would have known how to achieve such a goal. Specifically, there are numerous factors that influence cycle life, including charge cut-off voltage, electrode material ratio, electrolyte characteristics, charge/discharge rate, and ambient temperature, as I discussed above in Paragraphs

[106]-[116] of this Declaration. Again, these factors are applicable to the Yasunami-Koyama battery just as they are to the Yamaki-Sakamoto battery. A POSITA would have known to test with the aim of obtaining a battery with long cycle life. With these factors at hand, a POSITA would have just conducted routine experiments, during longer-term cycle life testing, and consistent with the charge cut-off voltage range and ratio of positive to negative electrode material of the combination battery of the Yasunami-Koyama combination battery, with the objective of finding the parameter space that allows for a target percent capacity retention over a target number of cycles to be achieved.

227. In this context, the retention of at least 75%, 75.83%, 80%, or 80.72% of capacity is an arbitrary number, as are the 400 cycles. A POSITA would have known how to achieve a target percent capacity after a certain number of cycles; the factors do not change, nor does the experimental process.

228. For instance, as I discussed above for Ground 1A, each of these factors—charge cut-off voltage, ratio of electrode materials, charge and discharge rate, temperature, and electrolyte characteristics—was a known contributor to cycle life and capacity retention of lithium secondary batteries. Given that this is a well-mapped space, I believe that a POSITA would have had a reasonable expectation of success in adjusting these factors to find an optimized capacity retention for the Yasunami-Koyama battery. A POSITA working to improve the

performance of a lithium ion battery and utilizing the concepts contained in Yasunami and Koyama would have used a standard experimental design approach to investigate the effects of cut-off voltage, charge and discharge rate, temperature, and electrolyte characteristics along with the design features described in the two references. These design optimization programs were standard and well known techniques for maximizing cell performance variables, including cycle life. By using classic design optimization approaches, the POSITA would achieve results, such as a formulation and design that maximized cycle life, with a modest investment in experimentation and time.

229. Furthermore, as I discussed above, the ’641 Patent provides no indication of the charge and discharge rate, the ambient temperature, or the electrolyte purity for its capacity retention testing. The Yasunami-Koyama battery already has the structure (ratio of positive to negative electrode material) and operation (charge cut-off voltage) of the batteries of the ’641 Patent, and adjusting the charge and discharge rate and ambient temperature would have been a matter of simple experimentation. As to the question of a specific value for percent capacity retention (75%, 75.83%, 80%, or 80.72%) and a specific number of cycles (400)—the ’641 Patent says nothing about why these values are critical or somehow unexpected. Instead, they are just arbitrary numbers. And given the knowledge of a POSITA about the various factors that influence capacity retention,

starting from a battery that already meets what is disclosed by the ’641 Patent, it would have been just straightforward experimentation to identify the factors that can achieve this arbitrary standard—that is, to obtain a battery that maintains at least 75%, 75.83%, 80%, or 80.72% of capacity after 400 cycles.

D. [GROUND 2B] – CLAIMS 9-10 ARE OBVIOUS OVER YASUNAMI, RICHARDSON, AND KOYAMA

1. Overview of Richardson

230. Richardson describes “redox shuttle additives for overcharge protection in rechargeable batteries,” such as “4 V lithium ion batteries. APPLE-1008 (Richardson), 1-2. Richardson explains that “[r]edox shuttle additives provide an internal, automatic overcharge protection mechanism that allows charging current to pass through the cell without overcharging it.” *Id.*, 2.

231. Richardson’s additives are “low molecular weight organic aromatic compounds” “added to the electrolyte of a rechargeable cell” to “function as redox shuttle additives at less severe overcharging conditions.” *Id.*, 2-3. Richardson explains that the redox shuttle additive “is oxidized at the positive electrode to its respective radical cation at certain potentials. The cation diffuses through the electrolyte to the negative electrode, where it is reduced to its original neutral state.” *Id.*, 3. This process “allows the additive(s) to transport an electric charge through the cell without damage to the cell under conditions that would, in the

absence of the additive(s), cause it to lose capacity or fail," thus providing overcharge protection to the cell. *Id.*, 3.

232. These additives have a dual function: "[u]nder more severe overcharging conditions, the cation transport through the electrolyte is insufficient, and the additives are oxidized at the positive electrode to shut off the cell." APPLE-1008 (Richardson), 3. That is, in a severe overcharge situation, the additives act as "shutdown additives," which "render[] the battery harmless on overcharging … even though it ends its useful life." *Id.*, 2. For instance, shutdown additives work by "increase[ing] the resistance of the cell by means of polymerization, precipitation, electrolyte depletion, chemical reaction, or other mechanism," thus "prevent[ing] further charging of the cell." *Id.*, 3.

233. Richardson provides a list of additives that "can act … as redox shuttles under moderate overcharge conditions, and as cell shutdown additives when subjected to more severe overcharging." APPLE-1008 (Richardson), 5. As illustrated in Table II, below, these additives have an "Onset Potential" ranging from 3.72 to 5.05 V. *Id.*, 5-6. The onset potential of an additive is the oxidation potential of the additive, *e.g.*, the potential at which the overcharge protection redox reaction is initiated. *See id.*, 1 ("The redox shuttle approach to overcharge protection employs an electrolyte additive that acts as an internal current shunt when the charging potential exceeds a characteristic onset potential for the

additive”). That is, the “Onset Potential” of these additives is the potential at which the additive begins to act as a redox shuttle. A potential still higher than the onset potential would induce the additive to behave as a shutdown additive. *See id.*, 7 (“The LBNL additives have ***redox shuttle onset potentials*** of about 4.25 V or higher”).

Table II

SD/RS#	Compound Name	CAS No.	Onset Potential	Category
LBNL 1	1,2-difluoro-4,5-dimethoxybenzene	203059-80-7	4.20	A,B
LBNL 2	1,4-difluoro-2,5-dimethoxybenzene	199866-90-5	4.20	A,B
LBNL 3	3,4-dimethoxybenzonitrile	2024-83-1	4.50	B
LBNL 4	piperonylonitrile	4421-09-4	4.52	C
LBNL 5	5-chloro-1,3-benzodioxole	7228-38-8	4.25	C
LBNL 6	5-bromo-1,3-benzodioxole	2635-13-4	4.27	C
LBNL 7	5-nitro-1,3-benzodioxole	2620-44-2	4.57	C
LBNL 8	1,2-dimethoxy-4-nitrobenzene	709-09-1	4.55	B
LBNL 9	3,4,5-trimethoxybenzonitrile	1885-35-4	4.40	B
LBNL 10	octafluoronaphthalene	313-72-4	4.90	A
LBNL 11	2,3-dimethoxybenzonitrile	5653-62-3	4.50	B
LBNL 12	3,5-dimethoxybenzonitrile	19179-31-8	4.45	B
LBNL 13	2,3,4-trimethoxybenzonitrile	43020-38-8	4.50	B
LBNL 14	1,2,3-trimethoxybenzene	634-36-6	4.46	B
LBNL 15	1,2,4-trimethoxybenzene	135-77-3	3.72	B
LBNL 16	3,4-difluoroanisole	115144-40-6	4.57	A,B
LBNL 17	pentafluoroanisole	389-40-2	5.05	A,B
LBNL 18	1,2,4,5-tetrafluorobenzene	327-54-8		A
LBNL 19	1,2,3-trifluorobenzene	1489-53-8		A
LBNL 20	1,3,5-trifluorobenzene	372-38-3		A
LBNL 21	1,2,3,5-tetrafluorobenzene	2367-82-0		A
LBNL 22	2,3,4-trifluorobenzonitrile	143879-80-5		A
LBNL 23	2,3,6-trifluorobenzonitrile	136514-17-5		A
LBNL 24	3,4,5-trifluorobenzonitrile	134227-45-5		A

Richardson, Table 2

234. I note that Richardson does not specify particular limits on the applicability of its teachings regarding redox shuttles in lithium secondary

batteries, and importantly, describes redox shuttles having onset voltages consistent with batteries having the charge cut-off voltages of the Yasunami-Koyama combination battery.

2. The Yasunami-Koyama-Richardson combination

235. In the Yasunami-Koyama-Richardson combination battery, the electrolyte includes one or more redox shuttle additives, *e.g.*, such as those listed in Richardson's Table 2. *See* APPLE-1008 (Richardson), 4 ("electrolyte 28 includes the … shutdown/redox shuttle additives of the invention"), 5-6 ("the following additives in Table II can act [] as redox shuttles under moderate overcharge conditions"). Specifically, a redox shuttle additive is used that has an onset voltage that is higher than the end-of-charge voltage of the combination battery, *e.g.*, higher than a voltage "in the range from 3.5 to 4.7 V." APPLE-1006 (Yasunami), [0034].

236. In operation of the combination battery, the battery is charged to its end-of-charge voltage, *e.g.*, a voltage of between 3.5 and 4.7 V. *See* APPLE-1006 (Yasunami), [0034]. In an overcharge situation, *e.g.*, when the potential of the positive electrode exceeds the end-of-charge voltage and reaches the onset voltage of the redox shuttle additive, the additive undergoes a redox reaction, thus "transport[ing] an electric charge through the cell without damage to the cell," *e.g.*, providing overcharge protection to the battery. APPLE-1008 (Richardson), 3.

When the voltage decreases, *e.g.*, to a level below the onset voltage of the redox shuttle additive, the redox reaction no longer proceeds and normal charging/discharging operation of the battery can resume. *See* APPLE-1008 (Richardson), 2 ("The ability of such additives to extend cell life in lithium and lithium ion cells under conditions of moderate to severe overcharging has been demonstrated"). Or, if the overcharge situation is more severe, the additive acts as a shutdown additive. *See id.*, 2-3.

237. The overcharge protection provided by Richardson's redox shuttle additive can be instead of Yasunami's optional "circuit that incorporates measures against overcharging and overdischarging," or in addition to such a circuit. APPLE-1006 (Yasunami), [0060].

3. Reasons for the combination

238. It is my opinion that a POSITA would have been motivated to provide overcharge protection additives, *e.g.*, as taught by Richardson, in a battery based on the Yasunami-Koyama combination, for at least the following reasons.

239. <u>The first reason</u> why a POSITA would have been motivated to provide overcharge protection in the combination battery is to avoid degradation of battery performance that can result from an overcharge situation. I discussed why providing overcharge protection helps avoid performance degradation in my discussion of the Yamaki-Sakamoto-Abraham combination; that discussion is

applicable here as well. *See* Paragraphs [129]-[130] of this Declaration; APPLE-1009 (Abraham), 1:54-56; APPLE-1026 (Huang), 1:24-25; APPLE-1008 (Richardson), 1. In particular, a POSITA would have been encouraged to pursue this implementation because Richardson does not specify any particular implementation limits and thus is relevant to the composition and operation of the Yasunami-Koyama battery.

240. The second reason why a POSITA would have been motivated to implement overcharge protection in the combination battery is to mitigate the safety risks associate with overcharging. I discussed why providing overcharge protection promotes battery safety in my discussion of the Yamaki-Sakamoto-Abraham combination; that discussion is applicable here as well. *See* Paragraphs [131]-[132] of this Declaration; APPLE-1026 (Huang), 1:26-27; APPLE-1008 (Richardson), 1-2. Again, a POSITA would have been encouraged to pursue this implementation because Richardson does not specify any particular implementation limits and thus is relevant to the composition and operation of the Yasunami-Koyama battery.

241. The third reason why a POSITA would have been motivated to pursue a redox shuttle implementation for overcharge protection is because of the advantages because of the advantages of providing overcharge protection with a redox shuttle, again in light of the generic applicability of Richardson's disclosure.

I discussed the advantages of providing chemical overcharge protection in my discussion of the Yamaki-Sakamoto-Abraham combination; that discussion is applicable here as well. *See* Paragraph [133] of this Declaration; APPLE-1009 (Abraham), 1:65-2:2; APPLE-1027 (Shimizu), [0009].

242. The fourth reason why a POSITA would have been motived to pursue a redox shuttle implementation for overcharge protection—still in view of the general applicability of Richardson's disclosure—is because of the cycle life advantages of this approach in the context of a multi-cell battery. I discussed the role of chemical overcharge protection in multi-cell batteries in my discussion of the Yamaki-Sakamoto-Abraham combination; that discussion is applicable here as well. *See* Paragraphs [134]-[135] of this Declaration; APPLE-1009 (Abraham), 2:3-19.

243. The fifth reason why a POSITA would have been prompted to pursue this implementation is because this combination is just the application of known techniques (specifically, chemical overcharge protection) to an already-known system (specifically, the Yasunami-Koyama combination battery), and it produces predictable results (specifically, a secondary lithium battery having chemical overcharge protection).

244. It is my opinion that a POSITA would have expected to be successful in providing overcharge protection additives, *e.g.*, as taught by Richardson, in a

battery based on the Yasunami-Koyama combination, for at least the following reasons.

245. The first reason why a POSITA would have expected success in combining teachings of Richardson with the Yasunami-Koyama combination battery is because both the Yasunami-Koyama battery and Richardson's battery are secondary lithium ion batteries with similar materials systems. This is an indication that the teachings of Richardson are compatible with, and applicable to, the Yasunami-Koyama battery.

246. For instance, the electrolytes in which Richardson describes dissolving the its redox shuttles are consistent with the non-aqueous electrolyte composition of the Yasunami-Koyama battery. Specifically, both systems are compatible with liquid, non-aqueous electrolytes. *See* APPLE-1006 (Yasunami), [0048]; APPLE-1008 (Richardson), 3 ("These additives could find use in any rechargeable battery utilizing a non-aqueous … electrolyte"), 4 ("In some batteries it may be possible that the electrolyte is liquid"), 6 ("This invention provides new additives for batteries or electrochemical storage cells, particularly for 4 volt cells incorporating liquid or preferably polymer electrolytes"), Abstract.

247. Both systems also use lithium-containing transition metal oxides for their positive electrode. *See* APPLE-1008 (Richardson), 7 ("Lithium manganese oxide cells containing these additives can be charged to their full normal capacity

at 4.2 V"); APPLE-1006 (Yasunami), Abstract (identifying Li_yMnO_z as an example of "a lithium-containing transition metal oxide" for the positive electrode active material).

248. Variations in the specific chemical composition of the positive electrode in different Li Ion cell designs do require different charge voltage limitations, particularly when measure closely. All of them share the same concern however: exceeding the design charge voltage limit will result in damage to the cell, reduced cycle life and possible safety issues. Whatever the specific charge voltage limit, it is possible to identify, either through known reference values or through straightforward experimentation, chemical compounds that can act as redox shuttles at the specific voltage required by that systems unique chemical makeup. There is no relationship between the chemistry of the positive electrode and the chemistry of the redox compound; the only common element is that each can be characterized by a voltage where specific, unrelated reactions will occur.

249. Given this overlap in materials, a POSITA would have expected the teachings of Richardson to be relevant to, and combinable with, those of Yasunami and Koyama. And as I discussed previously, the disclosure of Richardson, as well as those of Yasunami and Koyama, are generally applicable, and none provides any indication precluding the combination of their features with features of the other disclosures.

250. The second reason why a POSITA would have expected success is because Yasunami indicates that the electrolyte of its battery can contain additives: "Other compounds may be added to the electrolyte for the purpose of improving discharge and charge/discharge characteristics." APPLE-1006 (Yasunami), [0053]. Indeed, Yasunami's compounds can be "nitrobenzene derivatives," indicating the compatibility of Yasunami's battery with benzenes, such as 1,2,-dimethoxy-4-nitrobenzene, which are among the redox shuttle additives described by Richardson. APPLE-1006 (Yasunami), [0053]; *see* APPLE-1008 (Richardson), Table II.

251. The third reason why a POSITA would have expected success is because of the widespread use of redox shuttle additives for overcharge protection in secondary lithium ion batteries as of the Critical Date. *See* APPLE-1009 (Abraham), Abstract; APPLE-1030, Abstract.

4. Analysis

(i) **Claim 9**

[9] A secondary lithium ion cell or battery according to claim 5, characterized in that the secondary lithium ion cell or battery has a first overcharging protection voltage of greater than 4.35V, and an overcharging protection release voltage of greater than 4.15 V.

252. The Yasunami-Koyama-Richardson combination battery includes a "redox shuttle additive[]," such as those described by Richardson. APPLE-1008 (Richardson), 3. The overcharging protection voltage of the combination battery is

the “Onset Potential” of the redox shuttle additive, e.g., because when the voltage of the battery reaches the onset potential, the redox shuttle “transport[s] an electrical charge through the cell without damage to the cell” when the onset potential is reached, thereby preventing further voltage increase and providing overcharge protection. APPLE-1008 (Richardson), 3, 5; see 7 (“LBNL additives have redox shuttle onset potentials of about 4.25 V or higher”).

253. A number of the redox shuttle additives described by Richardson have a redox shuttle onset potential (that is, an overcharging protection voltage) of greater than 4.35 V, including 3,4-dimethoxybenzonitrile, piperonylonitrile, 1,2-dimethoxy-4-nitrobenzene, 3,4,5-trimethyoxybenzonitrile, octafluoronaphthalene, 2,3-dimethoxybenzonitrile, 3,5-dimethoxybenzonitrile, 2,3,4-trimethyoxybenzonitrile, 1,2,3-trimethoxybeneze, 3,4-difluoroanisole, and pentafluoroanisole. *See* APPLE-1008 (Richardson), 5-6 (Table II, which I have reproduced below, highlighting each of these additivies). When any of these redox shuttle additives is incorporated into the electrolyte of the combination battery, the overcharging protection voltage of the battery is greater than 4.35 V.

Table II

SD/RS#	Compound Name	CAS No.	Onset Potential	Category
LBNL 1	1,2-difluoro-4,5-dimethoxybenzene	203059-80-7	4.20	A,B
LBNL 2	1,4-difluoro-2,5-dimethoxybenzene	199866-90-5	4.20	A,B
LBNL 3	3,4-dimethoxybenzonitrile	2024-83-1	4.50	B
LBNL 4	piperonylonitrile	4421-09-4	4.52	C
LBNL 5	5-chloro-1,3-benzodioxole	7228-38-8	4.25	C
LBNL 6	5-bromo-1,3-benzodioxole	2635-13-4	4.27	C
LBNL 7	5-nitro-1,3-benzodioxole	2620-44-2	4.57	C
LBNL 8	1,2-dimethoxy-4-nitrobenzene	709-09-1	4.55	B
LBNL 9	3,4,5-trimethoxybenzonitrile	1885-35-4	4.40	B
LBNL 10	octafluoronaphthalene	313-72-4	4.90	A
LBNL 11	2,3-dimethoxybenzonitrile	5653-62-3	4.50	B
LBNL 12	3,5-dimethoxybenzonitrile	19179-31-8	4.45	B
LBNL 13	2,3,4-trimethoxybenzonitrile	43020-38-8	4.50	B
LBNL 14	1,2,3-trimethoxybenzene	634-36-6	4.46	B
LBNL 15	1,2,4-trimethoxybenzene	135-77-3	3.72	B
LBNL 16	3,4-difluoroanisole	115144-40-6	4.57	A,B
LBNL 17	pentafluoroanisole	389-40-2	5.05	A,B
LBNL 18	1,2,4,5-tetrafluorobenzene	327-54-8		A
LBNL 19	1,2,3-trifluorobenzene	1489-53-8		A
LBNL 20	1,3,5-trifluorobenzene	372-38-3		A
LBNL 21	1,2,3,5-tetrafluorobenzene	2367-82-0		A
LBNL 22	2,3,4-trifluorobenzonitrile	143879-80-5		A
LBNL 23	2,3,6-trifluorobenzonitrile	136514-17-5		A
LBNL 24	3,4,5-trifluorobenzonitrile	134227-45-5		A

Table II of Richardson

254. For instance, the redox shuttle additive is typically selected such that it has an onset potential of slightly higher than the charge cut-off voltage of the

combination battery, *e.g.*, so that the redox shuttle reaction does not interrupt normal charging/discharging operation. *See* APPLE-1009 (Abraham), 2:36-38. At least when the charge cut-off voltage of the combination battery is towards the high end of the "3.5 to 4.7 V" range of charge cut-off voltages (for instance, when the charge cut-off voltage is 4.3 V; *see* APPLE-1006 (Yasunami), [0067], [0069]), a POSITA would have recognized the suitability of these redox shuttle additives to provide an overcharging protection voltage greater than the charge cut-off voltage, *e.g.*, greater than 4.35 V. APPLE-1006 (Yasunami), [0034].

255. When the voltage in the battery falls below the oxidation potential of the redox shuttle, the redox shuttle reaction no longer proceeds, and charging/discharging of the battery can resume, *e.g.*, the overcharging protection provided by the redox shuttle is released, as I discussed above with respect to the Yamaki-Sakamoto-Abraham combination. *See* Paragraphs [146]-[147] of this Declaration; APPLE-1009 (Abraham), 2:26-33. Thus, the overcharging protection release voltage, *e.g.*, the voltage at which overcharging protection is no longer provided, is also the oxidation potential of the redox shuttle. As discussed above, the oxidation potential of the redox shuttle in the combination is greater than 4.35 V, and thus the overcharging protection release voltage of the combination battery is also greater than 4.35 V, *i.e.*, greater than 4.15 V.

(ii) Claim 10

[10] A secondary lithium ion cell or battery according to claim 9, characterized in that the secondary lithium ion cell or battery has a first overcharging protection voltage of greater than 4.45 V, and an overcharge protection release voltage of greater than 4.25 V.

256. My analysis of claim [9] is applicable to claim [10]. When the combination includes a redox shuttle additive having an onset potential of greater than 4.45—*e.g.*, 3,4-dimethoxybenzonitrile, piperonylonitrile, 1,2-dimethoxy-4-nitrobenzene, octafluoronaphthalene, 2,3-dimethoxybenzonitrile, 2,3,4-trimethyoxybenzonitrile, 1,2,3-trimethoxybeneze, 3,4-difluoroanisole, or pentafluoroanisole, as described by Richardson, the battery has an overcharging protection voltage of greater than 4.45 V. *See* APPLE-1008 (Richardson), 5-6 (Table II). For instance, at least when the charge cut-off voltage of the combination is towards the high end of the "3.5 to 4.7 V" range of charge cut-off voltages, a POSITA would have recognized the suitability of these redox shuttle additives to provide an overcharging protection voltage of greater than the charge cut-off voltage. APPLE-1006 (Yasunami), [0034]. Additionally, the overcharging release voltage is greater than 4.25 V, at least because the overcharging release voltage is the oxidation potential of the redox shuttle, which is greater than 4.45 V.

VI. LEGAL STANDARDS

A. Terminology

257. I have been informed by Counsel and understand that the best indicator of claim meaning is its usage in the context of the patent specification as understood by one of ordinary skill. I further understand that the words of the claims should be given their plain meaning unless that meaning is inconsistent with the patent specification or the patent's history of examination before the Patent Office. Counsel has also informed me, and I understand that, the words of the claims should be interpreted as they would have been interpreted by one of ordinary skill at the time of the invention was made (not today). Because I do not know at what date the invention as claimed was made, I have used the earliest possible priority date of the '641 Patent as the point in time for claim interpretation purposes (the Critical Date).

B. Legal Standards for Anticipation

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

259. I am informed by Counsel and understand that a challenged claim is unpatentable as "anticipated" under 35 U.S.C. § 102 if it is determined that all the limitations of the claim are described in a single prior art reference. I am informed

by Counsel and understand that, to anticipate a claim, a prior art reference must disclose, either expressly or inherently, each and every limitation of that claim and enable one of ordinary skill in the art to make and use the invention.

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

C. Legal Standards for Obviousness

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

262. I have been informed by Counsel and understand that a claim is unpatentable for obviousness under 35 U.S.C. § 103 (in the pre-AIA form of that statute that applies to the '641 Patent) "if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains." I am

informed by Counsel and understand that obviousness may be based upon a combination of references. I am informed by Counsel and understand that the combination of familiar elements according to known methods is likely to be obvious when it does no more than yield predictable results. However, I am informed by Counsel and understand that a patent claim composed of several elements is not proved obvious merely by demonstrating that each of its elements was, independently, known in the prior art.

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

264. I am informed by Counsel and understand that a patent claim composed of several limitations is not proved obvious merely by demonstrating that each of its limitations was independently known in the prior art. I am informed by counsel for the Patent Owner and understand that identifying a reason those elements would be combined can be important because inventions in many instances rely upon building blocks long since uncovered, and claimed discoveries

almost of necessity will be combinations of what, in some sense, is already known. I am informed by Counsel and understand that it is improper to use hindsight in an obviousness analysis, and that a patent's claims should not be used as a "roadmap."

265. I am informed by Counsel and understand that an obviousness inquiry requires consideration of the following factors: (1) the scope and content of the prior art; (2) the differences between the claims and the prior art; (3) the level of ordinary skill in the pertinent art; and (4) any objective indicia of non-obviousness, such as commercial success, long-felt but unresolved need, failure of others, industry recognition, copying, and unexpected results. I understand that the foregoing factors are sometimes referred to as the "Graham factors."

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

267. I have been informed by Counsel and understand that if a technique has been used to improve one device, and a person of ordinary skill at the time of

invention would have recognized that it would improve similar devices in the same way, using the technique is obvious unless its actual application is beyond his or her skill.

268. I have been informed by Counsel and understand that practical and common sense considerations should guide a proper obviousness analysis, because familiar items may have obvious uses beyond their primary purposes. I have been informed by Counsel and understand that a person of ordinary skill looking to overcome a problem will often be able to fit together the teachings of multiple prior art references. I have been informed by Counsel and understand that obviousness analysis therefore takes into account the inferences and creative steps that a person of ordinary skill would have employed at the time of invention.

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

270. I have been informed by Counsel and understand that a claim can be obvious in light of a single reference, without the need to combine references, if

the elements of the claim that are not found explicitly or inherently in the reference can be supplied by the common sense of one of skill in the art.

271. I have been informed by Counsel and understand that secondary indicia of non-obviousness may include (1) a long felt but unmet need in the prior art that was satisfied by the invention of the patent; (2) commercial success of processes covered by the patent; (3) unexpected results achieved by the invention; (4) praise of the invention by others skilled in the art; (5) taking of licenses under the patent by others; (6) deliberate copying of the invention; (7) failure of others to find a solution to the long felt need; and (8) skepticism by experts. I understand that evidence of secondary indicia of non-obviousness, if available, should be considered as part of the obviousness analysis.

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

273. In sum, my understanding is that prior art teachings are properly combined where one of ordinary skill having the understanding and knowledge reflected in the prior art and motivated by the general problem facing the inventor, would have been led to make the combination of elements recited in the claims. Under this analysis, the prior art references themselves, or any need or problem

known in the field of endeavor at the time of the invention, can provide a reason for combining the elements of multiple prior art references in the claimed manner.

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

VII. CONCLUSION

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

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

APPENDIX A

Louis Hruska

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Tech Executive with comprehensive background in general management, manufacturing, product development, and R&D. CEO/Sr. Executive with demonstrated record of success in business development, P&L management, conceptualizing new products and leading development teams, establishing advanced manufacturing facilities, and applying fundamental new technologies for performance enhancement and cost reduction.

Areas of expertise include:

- Product Development & Commercialization
- Expert Witness Testimony & Deposition
- Technical Sales & Marketing
- Process Development & Manufacturing Engineering
- Global Alliances
- IP Strategy, Licensing, & Defense

PROFESSIONAL EXPERIENCE

Energy Research Group **2008-Present**
Principal
Established consulting practice focusing on strategic planning, acquisition assessment, electronic manufacturing, application engineering, and battery design. Representative engagements have included:

- Led urgent process to find and eliminate failures in high volume GFCI devices at a $1B electronic product manufacturer. Established link between failures and microprocessor, developing data to force $10M settlement from semiconductor provider.

- Assessed technology, staff and market position of a small Lithium Ion battery maker for potential acquisition by a global leader in stationary and automotive battery systems. Served as GM of the acquired organization for 18 months, establishing manufacturing processes, transferring product design skills, and increasing product sales tenfold.

- Provided expert witness reports and testimony in billion-dollar patent infringement cases involving LiIon battery control and communication.

    - Expert for Apple represented by Fish & Richardson vs. Ericsson
    - Expert for Apple represented by Fish & Richardson vs. Qualcomm
    - Expert for LG represented by Fish & Richardson vs. SKI
    - Expert for Apple represented by Erise IP vs. Hitachi Maxell

- Conducted a product and application analysis for a midsized battery company with consumer, military, and automotive businesses. Recommended a focus on automotive/military applications and eliminating consumer products to improve margins and avoid additional investment.

Akermin, Inc. **2007-2008**
President and CEO
Organized, staffed, and developed new business strategy for an innovative technology company with

fuel cell applications. Recruited and trained a senior staff with industry expertise. Established a culture of cooperation and open communication and expanded technical foundation. Attracted new funding and closed on $3M of venture and $1M military funding within 6 months.

Microchip Inc **2002-2006**
PowerSmart Systems Division Director
Responsible for establishing product development, sales, marketing, and manufacturing of a new product line within a growing semiconductor company. Achieved operational breakeven within one year. Participated in corporate technology review and assessment, including smart sensor development, MEMs applications, and semiconductor packaging techniques.

- Identified market opportunity and created business plan to expand Microchip product offering into vehicle and military markets.

- Succeeded with automotive product qualification in a Japanese Tier 1/US OEM environment.

PowerSmart Inc. **1998 – 2002**
Chief Technical Officer and General Manager, PowerSmart Systems

Founding partner and lead technical executive of electronics start-up. Responsible for overall product strategy, manufacturing partnerships, development of engineering organization. Key member of corporate management team, with specific responsibility for building and managing integrated systems business. Played a key role in attracting potential acquirers and completing transaction with Microchip.

- Conceptualized the business based on mutually beneficial spin out of technology from Duracell, played key role in obtaining $12MM initial funding. Built high performing engineering team with mechanical, electrical, and chemical engineering capabilities. Revived key relationships with device OEMs.

- Secured development contracts from major auto companies and the U.S. Army for innovative vehicle battery management systems. Created and led project consortium, which included global leaders in the trucking industry, to fulfill contract deliverables.

- Managed a multi-national alliance for world class mixed signal ASIC design and manufacturing, essential to long term product strategy and business proposition.

- Established product development efforts in automotive sensors, magnetic field measurement, and MEMs sensor integration.

Duracell North American Group, a Division of the Gillette Company 1986 - 1997
Duracell Worldwide Technology Center, Needham MA

Director, Rechargeable Battery Engineering 1995 - 1997

Reported to Chief Technology Officer. Managed staff of 75 in two locations through 5 direct reports. Groups consisted of Product R&D, Mechanical Product Design, Process Engineering, Electronic Product Design and Manufacturing/Engineering.

- Increased the rate of product introduction tenfold, achieving aggressive product cost goals while introducing innovative manufacturing processes and control methods.

- Developed and championed an innovative battery design that provided 50 % more energy at 30% lower cost.

Director, Rechargeable Battery Engineering (Continued)

- Rescued failed product development and supply alliance through immediate identification and management of virtual organization for industrial design.
- Stretched product design team to develop multiple approaches for new battery to satisfy need of major telecom OEM for minimized cost and size, while meeting Duracell's cosmetic and durability requirements for retail.

Manager, Product Development 1992-1995

Reported to VP, Product Development. Led staff of 20 through 4 direct reports for design and manufacturing engineering of product line supporting Duracell's entry into rechargeable batteries.

- Provided technical leadership to develop product and secure adoption of first standard-sized PC battery by a major global PC OEM. Cumulative unit volume approached 500K, with revenues of nearly $25M. Won Division sales recognition award for this achievement.
- Established new technology expertise within Duracell, i.e., chip design and ASIC development, to provide major value-added feature and source of competitive advantage for entire PC battery line. Led to $40 M business within 3 years.
- Led complex technology alliance for electrical interconnect system with global leader in connector technology from concept to fruition, resulting in widely accepted standard offering major advances in thermal management, data communications, and size minimization.

Manager, Manufacturing Engineering 1989 - 1991

Led a diverse engineering project team tasked with dramatically reducing manufacturing cost, improving product quality and increasing production capacity of a purchased facility in Switzerland. Manufacturing technology included high vacuum sputtering, continuous electroplating, high temperature annealing, and polymer processing.

Hired and relocated skilled team of engineering professionals, equipment designers, and quality system experts.

- Supported international marketing and sales efforts with product application testing and concepts.
- Managed plant operations, including quality, production, development, manufacturing engineering.

Project Manager, Advanced Product Development 1988 -1989

Identified the opportunity for a major design improvement in Duracell's core product and proved its manufacturability in the Mexican plant, an effective demonstration site. This innovation was chosen for implementation as a global best design feature for Duracell's total production.

Manager, Process Development 1986 - 1988

Directed a 30-person team to design, specify, and install a $10M manufacturing process for a high-power Lithium battery.

- Introduced new fundamental technologies resulting in major changes in product and process design, enabling proprietary positions in battery design and manufacturing process.
- Met all cost targets, with a payback of less than 10 months despite high risk associated with basic battery chemistry, unproven manufacturing technologies, and magnitude of scale-up.

ELTECH Systems Corporation, Chardon, OH **1981-1985**

R&D Supervisor

Built staff of 10 to engage in ceramic materials and chemical process technology for spin-out.

- Invented proprietary process to improve heat transfer coefficient of titanium seawater heat exchangers, resulting in license income of $1M per year.
- Devised basic process and materials improvements that resulted in $5M annual reduction in power consumption for chemicals manufacturer.
- Developed Industrial Research Institute award-winning zirconia-based asbestos replacement material.

Diamond Shamrock Corporation, Painesville, OH **1976-1981**

Research Engineer

Performed fundamental electro-chemical research directed towards advance battery systems.

EDUCATION

The University of Michigan, Ann Arbor Michigan

- MS, College of Engineering 1976. Chemical and Materials Engineering
- BS, College of Engineering, 1974. Major in Chemical Engineering, Minor in Mechanical Engineering Magna cum Laude; achieved 4.0 GPA in Ch. Engineering coursework.

PATENTS & PUBLICATIONS

- Over 20 patents issued worldwide, covering materials, manufacturing processes, and product designs.
- Contributed to international conferences, authored numerous publications and presentations on product design and market development topics.
- Hruska, L.W., & Savinell, R., “Experimental-Study of an Iron-Redox Battery,” 127 J. Electrochemical Society C356, 1980
- L. W. Hruska and R. F. Savinell, “Investigation of Factors Affecting Performance of the Iron-Redox Battery”, J. Electrochem. Soc. 128, 1981
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PRIOR LITIGATION CONSULTING: RETAINED AND/OR TESTIFIED AT A DEPOSITION OR TRIAL

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- *Maxell Ltd. v. Apple Inc.* (5:19-CV-00036) (EDTX)
- *LG v. SKI* (337-TA-1179) (ITC)
- *Qualcomm Incorporated v. Apple Inc.* (Inv. 337-TA-1065) (ITC)
- *Apple Inc. v. Telefonaktiebolaget LM Ericsson* (IPR2015-01919) (PTAB)
- *Maxim Integrated Products, Inc. v. Silicon Mitus Technology* (5:17-cv-3507) (NDCA)
- *Qualcomm Incorporated v. Apple Incorporated* (3:17-cv-02402) (SDCA)
- *Ericsson v. Apple* (Inv. 337-TA-952) (ITC)
- *Ericsson v. Apple* (Inv. 337-TA-953) (ITC)
- *Ericsson v. Apple* (2:15-cv-00293) (EDTX)
- *Unova Inc. v. Hewlett-Packard* (2:02-cv-03772) (CDCA)
- *Unova Inc. v. Dell* (00-cv-1354) (CDCA)