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Title:	LITHIUM ION BATTERIES USING DISCRETE CARBON NANOTUBES, METHODS FOR PRODUCTION THEREOF AND PRODUCTS OBTAINED THEREFROM	

DECLARATION OF DR. YU ZHU

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I. QUALIFICATIONS AND BACKGROUND INFORMATION

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

2. I am currently the Associate Director and Professor of the School of Polymer Science and Polymer Engineering at The University of Akron. I am specializing in nanomaterials, polymers, energy storage devices and organic electronics. I received my PhD from the University of Cologne in Germany in 2007, where I worked on polymer synthesis, electrochemistry and organic electronics. From 2008 to 2012, I joined Rice University as a postdoctoral research associate, where my research included synthesis of graphene, carbon nanotube, low-dimensional nanomaterials and their applications in energy storage devices. I became an assistant professor at the University of Akron from August 2012, and was promoted to associate professor in 2018 and full professor in 2021. I have published over 80 peer reviewed articles and invited reviews and am a named inventor on several patent applications. I also serve as the current editor-in-chief of the journal "Colloid and Polymer Science", which was published by Spring-Nature Publisher since 1906.

3. I have a broad range of research interests. One of my key research areas focuses on energy storage, and my laboratory conducts research on lithium

ion batteries, lithium sulfur batteries, solid-state batteries, lithium oxygen batteries, flow batteries and other electrochemical energy storage devices. My lab has intensively worked on the low dimensional materials including graphene, carbon nanotubes, graphene nanoribbons and various functionalized forms of those carbon nanomaterials. In my lab, those carbon nanomaterials were used as various components in energy storage/conversion devices including batteries, supercapacitors and solar cells. Other research projects in my laboratory relate to organic electronics, conducting polymers, solid state electrolytes and polymer composites with low-dimensional filler materials, such as graphene and carbon nanotubes.

4. I have been retained on behalf of LG Energy Solution, Ltd. to offer technical opinions relating to U.S. Patent No. 8,808,909 ("the '909 Patent") and prior art references relating to its subject matter. I have reviewed the '909 Patent, and relevant excerpts of the prosecution history of the '909 Patent. I have also reviewed the following references:

LGES-1001 U.S. Patent No. 8,808,909 ("the '909 Patent")

LGES-1004 U.S. Patent Pub. No. 2004/0160156 ("Ohata")

LGES-1005 "Multi-walled carbon nanotubes functionalized by carboxylic groups: Activation of TiO_2 (anatase) and phosphate olivines ($LiMnPO_4$; $LiFePO_4$) for electrochemical Li-storage." Ladislav

Kavan et al. Journal of Power Sources 195 (2010) 5360-5369 ("Kavan")

LGES-1006 U.S. Patent Pub. No. 2010/0136431 ("Lee")

LGES-1007 U.S. Patent Pub. No. 2010/0124713 ("Farrugia")

LGES-1008 U.S. Patent Pub. No. 2012/0064409 ("Zhamu")

LGES-1009 "Chemical oxidation of multiwalled carbon nanotubes." V. Datsyuk et al. Carbon 46 (2008) 833-840) ("Datsyuk")

LGES-1010 "Surface and structural characterization of multi-walled carbon nanotubes following different oxidative treatments." Kevin A. Wepasnick, et al. Carbon 49 (2011) 24-36 ("Wepasnick")

LGES-1011-1013 Reserved

LGES-1014 "Surface modified multi-walled carbon nanotubes in CNT/epoxy-composites." Florian H. Gojny, Jacek Nastalczyk, Zbigniew Roslaniec, Karl Schulte. Chemical Physics Letters 370 (2003) 820–824 ("Gojny")

LGES-1015 "Sonochemical Oxidation of Multiwalled Carbon Nanotubes." Yangchuan Xing, Liang Li, Charles C. Chusuei, and Robert V. Hull. Langmuir 2005, 21, 4185-4190 ("Xing")

LGES-1025 "The structure and property characteristics of amorphous/nanocrystalline silicon produced by ball milling." T.D. Shen, et al. Journal of Materials Research 10 (1995), 139-149 ("Shen")

LGES-1026 "Crystal-amorphous phase transition induced by ball-milling in

	silicon." E. Gaffet, M. Harmelin. Journal of the Less Common Metals 157 (1990), 201-222 ("Gaffet")
LGES-1027	U.S. Patent No. 9,012,073 ("Kumar")
LGES-1028	U.S. Patent No. 9,190,694 ("Lopez")
LGES-1029	U.S. Patent No. 7,851,085 ("Obravac")
LGES-1030	"Labeling the Defects of Single-Walled Carbon Nanotubes Using Titanium Dioxide Nanoparticles." Xiaohong Li, Jiali Niu, Jin Zhang, Hulin Li, and Zhongfan Liu. The Journal of Physical Chemistry B 107 (2003) 2453-2458 ("X. Li")
LGES-1032	U.S. Patent No. 8,241,794 ("Sheem")
LGES-1033	"TiO2 rutile—An alternative anode material for safe lithium-ion batteries." M. Pfanzelt, P. Kubiak. Journal of Power Sources 106 (2011) 6815-6821 ("Pfanzelt")
LGES-1034	U.S. Patent Pub. No. 2011/0159366A1 ("Nakura")
LGES-1035	U.S. Patent Pub. No. 2009/0117463A1 ("Takezawa")
LGES-1037	"Preparation of Nanocomposites of Metals, Metal Oxides, and Carbon Nanotubes via Self-Assembly." Jing Li, Songbai Tang, Li Lu, and Hua Chun Zeng. Journal of the American Chemical Society 129 (2007) 9401-9409 ("J. Li")
LGES-1038	"Enhanced photocatalytic activity of mesoporous TiO2 aggregates by embedding carbon nanotubes as electron-transfer channel." J. Yu, T. Ma, S. Liu. Phys. Chem. Chem. Phys. 13 (2011) 3491–3501 (Yu)

LGES-1039 "Long-time low-impact ball milling of multi-wall carbon nanotubes." A. Kukovecz, T. Kanyo, Z. Konya, I. Kiricsi. Carbon 43 (2005) 994-1000 ("Kukovecz")

LGES-1040 "Green energy storage materials: Nanostructured TiO_2 and Sn-based anodes for lithium-ion batteries." D. Deng, M. Kim, J. Lee, J. Cho. Energy & Environmental Sciences 2 (2009) 818-837 ("Deng")

LGES-1041 "Hydrothermal synthesis of lithium iron phosphate." J. Chen, M. Whittingham. Electrochemistry Communications 8 (2006) 855-858 ("Chen")

LGES-1042 "The preparation and electrochemical performances of $LiFePO_4$-multiwalled nanotubes composite cathode materials for lithium ion batteries." Y. Feng. Materials Chemistry and Physics 121 (2010) 302-307 ("Feng")

LGES-1043 "Preparation of Single-Walled Carbon Nanotube/Silicon Composites and Their Lithium Storage Properties." J-Y. Eom, H-S. Kwon. Applied Materials & Interfaces 3 (2011) 1015-1021 ("Eom")

LGES-1044 A. Hirsch. "Functionalization of Single-Walled Carbon Nanotubes" *Angew. Chem. Int. Ed.* 2002, 41, No. 11, 1853-1859 ("Hirsch")

5. I have also reviewed various supporting references and other documentation.

6. Counsel has informed me that I should consider these materials through the lens of one of ordinary skill in the art related to the ’909 Patent at the time of the earliest possible priority date of the ’909 Patent, and I have done so during my review of these materials. The ’909 Patent claims the benefit of priority to an application filed June 23, 2011 (“the Critical Date”). Counsel has informed me that the Critical Date represents the earliest possible priority date to which the challenged claims of the ’909 Patent are entitled, and I have therefore used that Critical Date in my analysis below.

7. 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.

8. In writing this declaration, I have considered the following: my own knowledge and experience, including my work experience in the fields of material science, chemical engineering, chemistry, electrochemistry, and polymer science and engineering; my experience in teaching those subjects; and my experience in working with others involved in those fields. In addition, I have analyzed various publications and materials, in addition to other materials I cite in my declaration.

9. My opinions, as explained below, are based on my education, experience, and expertise in the fields relating to the ’909 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 ’909 Patent and the prior art discussed below.

II. OVERVIEW OF CONCLUSIONS FORMED

10. 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 1-2 and 5-8 are obvious over Ohata and Kavan;
- Claims 1-2 and 5-8 are obvious over Lee and Wepasnick; and
- Claims 3-4 are obvious over Lee, Wepasnick, and Zhamu.

III. LEVEL OF ORDINARY SKILL IN THE ART

11. In my opinion, one of ordinary skill in the art relating to, and at the time of, the invention of the ’909 Patent would have had at least a Master’s degree in materials science, chemistry, chemical engineering, a field relevant to polymer science and engineering, or the equivalent, and at least three years of experience in nanomaterials, electrochemistry, batteries, or polymer related activities.

Additional work experience can compensate for less formal education, and vice versa.

12. Based on my experiences, I have a good understanding of the capabilities of one of ordinary skill. Indeed, I have taught, participated in organizations, and worked closely with 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.

IV. SUMMARY OF THE '909 PATENT

13. The '909 Patent relates to "[c]ompositions … useful for lithium ion batteries comprising discrete oxidized carbon nanotubes having attached to their surface lithium ion active materials." LGES-1001, Abstract. However, as I demonstrate here, composites of nanotubes and lithium ion active materials were well-known in the art at the time of filing of the '909 Patent. *See*, *e.g.*, LGES-1004 (Ohata), [0004]; LGES-1005 (Kavan), Abstract; LGES-1006 (Lee), [0040]. And additional features recited in the claims—including the aspect ratio and oxidation levels of the nanotubes—were also conventional characteristics of nanotubes that are insufficient to justify patentability of the claims. *See*, *e.g.*, LGES-1004 (Ohata), [0020]; LGES-1005 (Kavan), 4; LGES-1006 (Lee), [0047]; LGES-1010 (Wepasnick), 6-11.

V. TECHNICAL BACKGROUND

14. Carbon nanotubes are formed of rolled-up graphene sheets. Single-walled carbon nanotubes have a single graphene tube; multi-walled carbon nanotubes are composed of multiple, concentric graphene tubes.

15. Chemical groups can be attached to the sidewalls and/or ends of carbon nanotubes; these carbon nanotubes are then referred to as "functionalized". These chemical groups are often oxygen-containing groups such as carboxyl, hydroxyl, or carbonyl groups. In this case, they are often referred as "oxidized". *See* LGES-1005 (Kavan), Abstract ("Multi-walled carbon nanotubes functionalized by carboxylic groups"); LGES-1010 (Wepasnick), Abstract ("Six commonly used wet chemical oxidants … were evaluated in terms of their effects on the surface chemistry and structure of MWCNTs … X-ray photoelectron spectroscopy (XPS) and energy dispersive spectroscopy (EDX) were used to characterize the extent of surface oxidation, while chemical derivatization techniques used in conjunction with XPS allowed the concentration of carboxyl, carbonyl, and hydroxyl groups at the surface to be quantified"); LGES-1014 (Gojny), 821-822 (describing that oxidation of carbon nanotubes resulted in in "[t]he presence of carboxylic acid groups on the surface"); LGES-1007 (Farrugia), [0028] ("oxidation is carried out by acid-treatment, wherein oxidation chemistry may be used to open the end caps

of both single and multi-walled nanotubes to produce carboxyl, carbonyl and hydroxyl groups at the opened ends and defects on the side walls").

16. A variety of characterization techniques exist for measuring the level of oxidation of carbon nanotubes. One such technique is X-ray photoelectron spectroscopy (XPS), which characterizes the composition of a sample generally near the surface of the sample. *See* LGES-1005 (Kavan), 4 ("The amount of atomic oxygen present on the MWNT-ox surface was also analyzed by X-ray photoelectron spectroscopy"); LGES-1010 (Wepasnick), Abstract ("X-ray photoelectron spectroscopy … were used to characterize the extent of surface oxidation"), 6 ("XPS and EDX were both used to quantify the atomic oxygen concentration following different oxidative treatments"); LGES-1009 (Datsyuk), 2 ("Oxidised [sic] MWCNTs were tested by X-Ray photoelectron spectroscopy in order to quantify and qualify the nature of oxygen present on the MWCNT surface"), 4 ("With reference to functionality grafting, XPS is one of the surface analytical techniques, which can provide useful information on the nature of the functional groups and also on the presence of structural defects on the nanotube surface"). A second surface-sensitive technique is energy-dispersive X-ray spectroscopy (EDX or EDS). *See* LGES-1010 (Wepasnick), Abstract ("energy dispersive spectroscopy (EDX) was used to characterize the extent of surface oxidation"); 6 ("XPS and EDX were both used to quantify the atomic oxygen

concentration following different oxidative treatments"). Thermogravimetric analysis (TGA) is yet another technique that can be used to measure the composition throughout the thickness of a sample. *See* LGES-1009 (Datsyuk), 2 ("Oxidised [sic] MWCNTs were tested by X-Ray photoelectron spectroscopy in order to quantify and qualify the nature of oxygen present on the MWCNT surface. The characterisation [sic] of the oxidised [sic] MWCNTs was completed by thermogravimetric analysis"). Titration techniques can be used to determine select information about nanotube functionalization, such as the presence of certain types of functional groups on the nanotubes. *See* LGES-1005 (Kavan), 2 ("The extent of functionalization was determined by titration … The amount of acid groups in the nanotubes was found to be 2.54×10^{-4} mol g^{-1}"); LGES-1009 (Datsyuk), 6 ("A direct acid-base titration technique using NaOH was performed to determine the concentration of surface groups").

17. Based on characterization results, *e.g.*, obtained by XPS, EDX, and/or TGA analysis, an indication of the oxidation level of a sample of carbon nanotubes can be determined. One way to characterize the oxidation level of nanotubes is simply to determine the amount of oxygen atoms (*e.g.*, the mass of oxygen atoms) in the functional groups that are bound to the nanotubes as compared to the total amount (*e.g.*, mass) of the sample. Another way is to determine the amount of the

functional groups themselves (*e.g.*, the mass of the functional groups) as compared to the total amount (*e.g.*, mass) of the sample.

VI. PRIOR ART ANALYSIS

A. [GROUND 1] – CLAIMS 1-2 AND 5-8 ARE OBVIOUS OVER OHATA-KAVAN

18. It is my opinion that a POSITA would have found claims 1-2 and 5-8 to be obvious over the combination of Ohata and Kavan, as I discuss in the following sections.

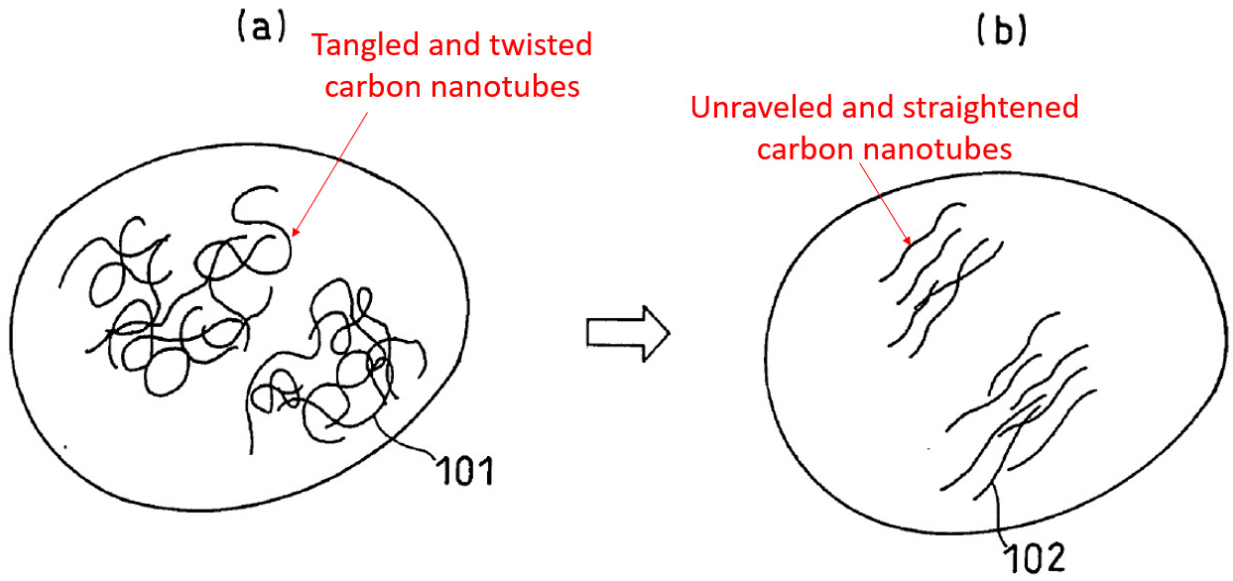
1. Ohata describes a nanotube-containing paste for lithium ion batteries

19. Ohata describes composites of carbon nanotubes and either positive or negative electrode active materials for use as electrodes in lithium ion batteries. These composites form "an electrode superior in conductivity with a small amount of conductive material, yet with high capacity." LGES-1004 (Ohata), [0004].

20. Ohata prepares its composites by first "producing a masterbatch by blending at least carbon nanotubes with a resin," such as a fluorocarbon resin, e.g., PVDF. LGES-1004 (Ohata), [0015], [0022]. In this process, carbon nanotubes that are initially "tangled and twisted with one another" (FIG. 1(a), *infra*) are "unraveled and straightened by blending them with a resin" such that they are "dispersed" in the resin (FIG. 1(b)). *Id.*, [0016]-[0017]. This morphology "form[s]

an excellent conductive path in an electrode" and "serves to increase the mechanical strength of an electrode plate." *Id.*

FIG. 1



Excerpt from Ohata, FIG. 1 (annotated)

21. An "electrode material mixture paste" is then formed by mixing "the masterbatch and an electrode active material with a dispersion medium." LGES-1004 (Ohata), [0023]. The electrode active material can be a positive electrode active material (*e.g.*, lithium metal oxides) or negative electrode active material (*e.g.*, graphite or other carbonaceous material, or an alloy such as a Si—Ni alloy). *Id.*, [0026]-[0027]. The electrode material mixture paste is applied "onto an electrode substrate" to form an "electrode plate" for use in a battery, such as a lithium secondary battery. *Id.*, [0025].

22. Ohata indicates that the size of the nanotubes impacts the conductivity of the electrode, and provides suitable size ranges for the diameter and length of the nanotubes. *See* LGES-1004 (Ohata), [0018]-[0020]. Specifically, Ohata states that "[t]he carbon nanotubes preferably have an aspect ratio … of not less than 100" and "not greater than 1000," explaining that "[w]hen the aspect ratio is too small, excellent conductive paths may not be formed in some electrodes," while "when the aspect ratio is too large, the carbon nanotubes are unlikely to be straightened sufficiently even after a resin is blended thereto." *Id.*, [0020]. The ratio of nanotubes to electrode active material is also relevant for electrode performance: "[t]he amount of the carbon nanotubes contained in the electrode material mixture is, for example, 0.2 to 3 parts by weight per 100 parts by weight of the electrode active material," and "is preferably not greater than 0.8 parts by weight" "[i]n order to obtain a high-capacity electrode." *Id.*, [0024].

2. Kavan describes composite materials containing oxidized nanotubes for use in lithium ion batteries

23. Kavan describes multi-walled carbon nanotubes (MWNTs) functionalized by carboxylic groups, used in composites with either negative electrode active materials or positive electrode active materials, for use as electrodes in lithium ion batteries. *See* LGES-1005 (Kavan), Introduction. Kavan sets out its "central motivation" as a desire to gain an "understanding … of the

MWNT functionalization (by surface oxidation forming carboxyls) for optimum attachment of MWNT towards titania or olivine surfaces." *Id.*, 2.

24. Kavan describes performing "oxidative functionalization" on the MWNTs to produce "MWNT-ox" ("material functionalized by oxidation with HNO_3"). LGES-1005 (Kavan), 2. Titration analysis of the oxidized nanotubes indicates that "[t]he amount of acid groups in the nanotubes was found to be 2.54 x 10^{-4} mol g^{-1} in MWNT-ox, and close to 0 in MWNT-bl."[1] *Id.* Moreover, "[t]he amount of atomic oxygen present on the MWNT-ox surface was also analyzed by X-ray photoelectron spectroscopy" and "gave a value of 7%." *Id.*, 4.

25. Oxidized nanotubes were mixed with "a powder material (TiO_2, $LiMnPO_4$ or $LiFePO_4$)," as were non-functionalized nanotubes as a control, "in a proportion of 1-10wt% of nanotubes in the mixture," and pastes were prepared using the resulting material. LGES-1005 (Kavan), 2. The structure, composition, and electrochemical behavior of the resulting composites were characterized.

26. For the case of TiO_2 as a negative electrode active material, Kavan's results indicate that "the interaction of [oxidized] nanotubes with titania is excellent" and that "[a]ll nanotubes are covered with individual nanocrystals of

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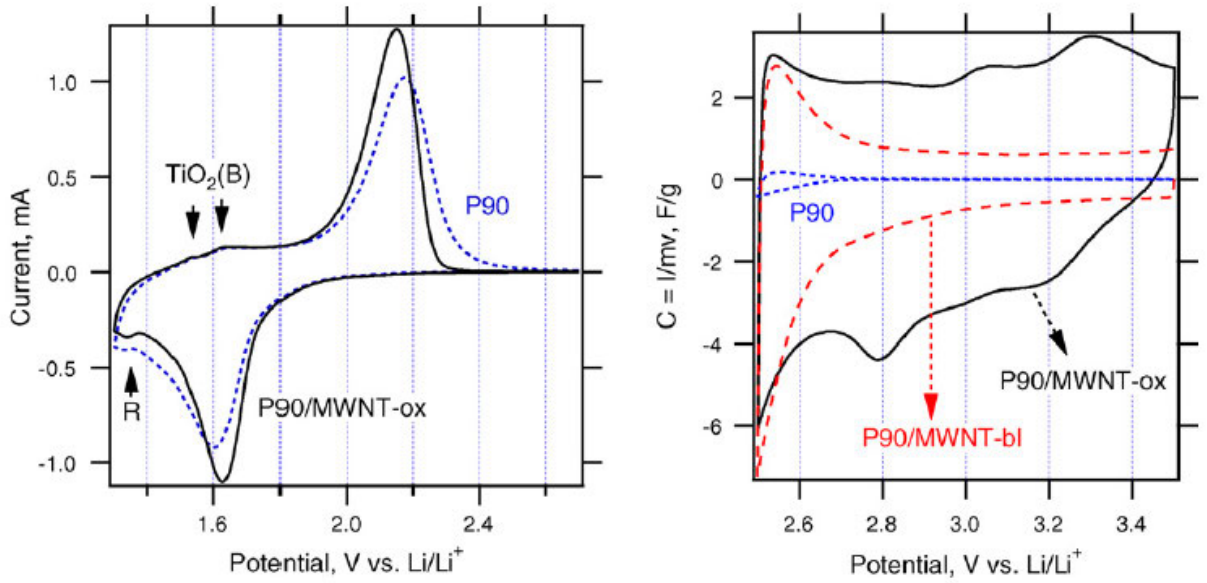
[1] "MWNT-bl" refers to a control sample of "non-functionalized blank material." LGES-1005 (Kavan), 2.

P90 [titania] in a homogeneous nanocomposite." LGES-1005 (Kavan), 3; *see* Fig. 2. These data "support the idea that oxidative functionalization through HNO_3 is crucial for proper anchoring of nanotubes towards TiO_2." *Id.*, 4. Moreover, Kavan states that "[t]here is a clear analogy with the strong interaction of carboxyl-containing molecules … with the TiO_2 surface," which implies that strong binding can be formed between MWNTs' carboxyls and TiO_2. LGES-1005 (Kavan), 2. Such a binding has been reported previously. *See* LGES-1037 (J. Li). In this literature, it reports "carboxylates groups on the CNTs will provide bonding to the TiO_2 … nanoparticles, as revealed by our XPS investigation…" *Id.*, 4.

27. In addition to directly mixing the lithium ion active materials particles with oxidized MWNTs, the '909 Patent also disclosed procedures of synthesized lithium active materials on the surface of oxidized MWNTs using soluble precursors (example 1,2,4,5). However, such procedure has also been reported in the literature. For example, synthesizing TiO_2 on oxidized MWNTs was reported by J. Yu. *See* LGES-1038 (Yu), Abstract, 2. This procedure was generally considered to form chemical bonds between CNT and lithium ion active materials. As stated in J. Yu's report, "Such surface functional groups provide active sites for directed deposition of anatase [TiO_2] nanocrystals around the surface of MWCNTs. For example, the -OH groups on the Ti precursor possibly react with

the -OH and -COOH groups on the MWCNTs surface through esterification to form C-O-Ti or O=C-O-Ti bonds…" *Id.*, 6.

28. The attachment between TiO_2 and nanotubes enhances the electrochemical performance of the combination material. For instance, Kavan provides cyclic voltammetry data in Fig. 5, *infra*, for both TiO_2 electrodes and electrodes composed of a composite of TiO_2 and functionalized nanotubes. These data show that while "the Li-storage capacities of both materials are identical," there are nevertheless electrochemical advantages associated with the TiO_2-nanotube composite. LGES-1005 (Kavan), 5. Specifically, "[t]he beneficial effect of nanotube networking" in the composites of TiO_2 and oxidized nanotubes can be seen in "the shape of cyclic voltammograms of Li-insertion," where for the TiO_2-nanotube composite, "[t]he peak-to-peak separation gets smaller" and "the slopes of insertion onset … and extraction offset … are steeper." *Id.* That is, "the rate of electrochemical charge transfer is higher due to nanotube networking." *Id.*, 9.

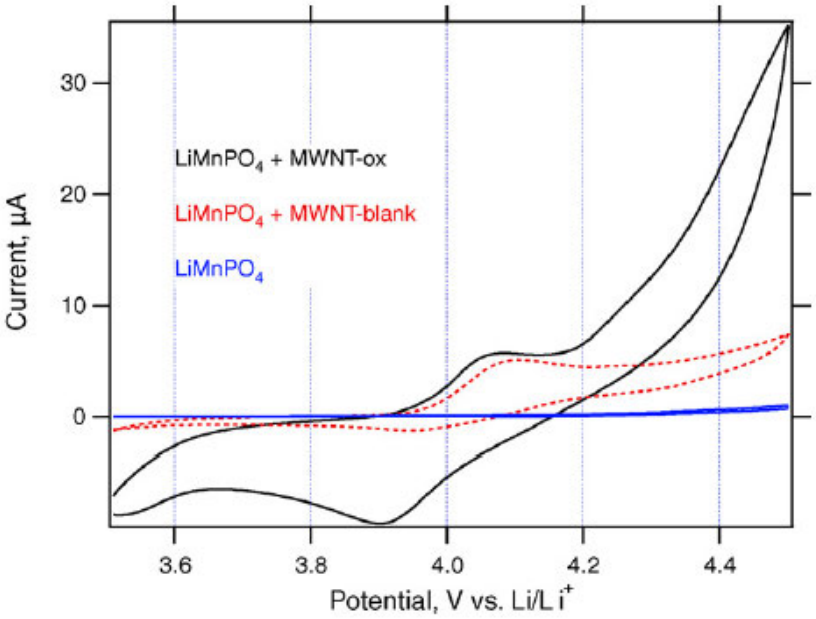


Excerpt from Kavan, Fig. 5

29. The cyclic voltammetry results also reveal new "pseudocapacitive features in the TiO_2/MWNT-ox composite" that are enabled by "the interactions between the oxidized (carboxylated) surface of the carbon nanotubes and TiO_2." LGES-1005 (Kavan), 5. Specifically, Kavan attributes these new features in the cyclic voltammogram of the composite to the creation of "new sites for pseudocapacitive interfacial redox chemistry." *Id.*

30. For the phosphate olivines, Kavan describes that structurally, "naked nanotubes are easily recognized between the individual crystals" of $LiMnPO_4$ or $LiFePO_4$. LGES-1005 (Kavan), 4; *see* Fig. 3. Overall, Kavan concludes that "the functionalization of nanotubes is … crucial for enhancing the electrochemical activity of the composite materials with $LiFePO_4$." *Id.*, 8. For instance, Fig. 7, *infra*, shows cyclic voltammetry results of $LiMnPO_4$ electrodes (blue curve),

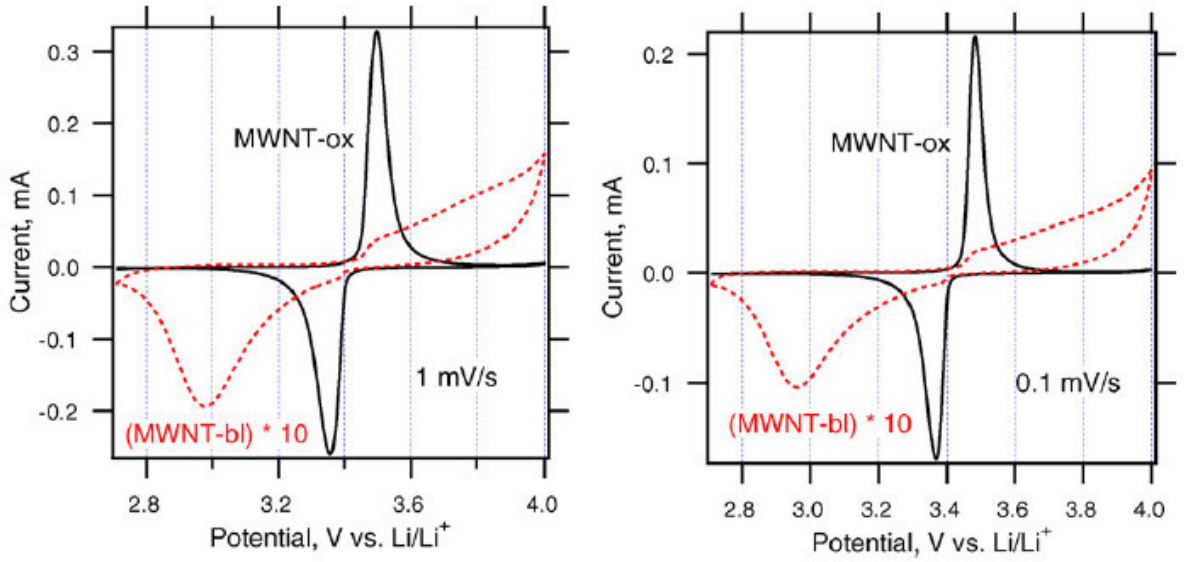
electrodes composed of a composite of $LiMnPO_4$ and non-oxidized nanotubes ("$LiMnPO_4$ + MWNT-blank," red dashed curve) and electrodes composed of a composite of $LiMnPO_4$ and oxidized nanotubes ("$LiMnPO_4$ + MWNT-ox," black curve). These results demonstrate that while "oxidation of $LiMnPO_4$ to MnPO4 is roughly identical for both electrodes" (*i.e.*, both nanotube-containing electrodes), "the oxidative treatment of nanotubes improves the reversibility of $LiMnPO_4$ charge/discharge." *Id.*, 7.



Excerpt from Kavan, Fig. 7

31. Regarding $LiFePO_4$, Kavan states that "the $LiFePO_4$ olivine exhibits excellent performance in a composite with MWNT-ox" and that "the improvement" in electrochemical activation of $LiFePO_4$ "caused by surface oxidation of nanotubes is dramatic." *Id.*, 7. Cyclic voltammograms in Fig. 8,

infra, show results for electrodes composed of a composite of $LiFePO_4$ and non-oxidized nanotubes ("MWNT-blank," red dashed curve) and electrodes composed of a composite of $LiMnPO_4$ and oxidized nanotubes ("MWNT-ox, black solid curves"). *Id.* Kavan characterizes the results for the oxidized nanotube based composite as "impressive" in terms of both "capacity" and "charge/discharge reversibility." *Id.*



Excerpt from Kavan, Fig. 8

32. It is worth noting that the preparation of composites of MWNT-ox and $LiFePO_4$ in Kavan and Example 3 in the '909 Patent are similar. *See* LGES-1005 (Kavan), 2; LGES-1001 ('909 Patent), 5:50-67. In both cases, oxidized CNT and premade $LiFePO_4$ powder mixed in water through sonication. Therefore, the attachment of $LiFePO_4$ nanocrystals on the oxidized CNT is expected to be similar.

3. The Ohata-Kavan combination

33. The Ohata-Kavan combination composition is based initially on the teachings of Ohata. Specifically, "an electrode material mixture containing the masterbatch and an electrode active material" is blended with "a dispersion medium" to form "[a]n electrode material mixture paste." LGES-1004 (Ohata), [0023]. The masterbatch contains carbon nanotubes dispersed in a resin, where the aspect ratio of the nanotubes is between 100 and 1000, as taught by Ohata. *See id.*, [0015], [0020]. The ratio of carbon nanotubes to electrode active material is also consistent with Ohata's teachings, *e.g.*, "[t]he amount of the carbon nanotubes contained in the electrode material mixture is, for example, 0.2 to 3 parts by weight per 100 parts by weight of the electrode active material," and "is preferably not greater than 0.8 parts by weight," which Ohata indicates is sufficient "to obtain a high-capacity electrode." *Id.*, [0024].

34. Based on the teachings of Kavan, the carbon nanotubes used in the masterbatch of the combination are oxidized, as discussed in the next paragraph. *See* LGES-1005 (Kavan), Abstract, 2, 4. Also based on the teachings of Kavan, the electrode material is TiO_2 (for a negative electrode composition) or a phosphate olivine such as $LiMnPO_4$ or $LiFePO_4$ (for a positive electrode composition). *See id.*, Abstract, 2.

35. In the Ohata-Kavan combination, the carbon nanotubes used in preparation of the masterbatch (*see* LGES-1004 (Ohata), [0015]) are functionalized, *e.g.*, "by carboxylic groups," as taught by Kavan. LGES-1005 (Kavan), Abstract. For instance, the nanotubes are exposed to the "oxidative functionalization" process described by Kavan, in which the nanotubes are "functionalized by oxidation with HNO_3" such that "[t]he amount of atomic oxygen present" on the nanotube surface is "7%." *Id.*, 2,4; *see* 2 ("For oxidative functionalization, 1 g of purified nanotubes was mixed with 50 mL of 65% HNO_3 and refluxed for 3 h at 120 ºC under stirring. The nanotubes were filtered on a glass frit, washed with water until the filtrate reached a pH of 6, and dried at 120 ºC for 48 h").

36. These oxidized nanotubes are then "unraveled and straightened by blending them with a resin" to form a masterbatch, consistent with the process described by Ohata. LGES-1004 (Ohata), [0016]. For instance, "the carbon nanotubes contained in the masterbatch" are present in an amount of "5 to 20 parts by weight per 100 parts by weight of the resin." *Id.*, [0021]. In a specific example, based on Ohata, the resin is "a fluorocarbon resin" such as "polyvinylidene fluoride" or "a vinylidene fluoride-hexafluoropropylene copolymer," or a combination of both of them. *Id.*, [0022].

37. As taught by Ohata, "an electrode material mixture containing the masterbatch and an electrode active material" is blended with "a dispersion medium" to form "[a]n electrode material mixture paste." LGES-1004 (Ohata), [0023]. As taught by Ohata, the dispersion medium is "selected according to the resin to be used." *Id.*, [0024]. Ohata indicates that "[t]he positive electrode active material" or "[t]he negative electrode active material to be used" in the electrode material mixture "varies depending on the battery type." LGES-1004 (Ohata), [0026]-[0027]. Consistent with this flexibility, in the Ohata-Kavan combination, a negative electrode material mixture paste is formed using titania (TiO_2), *e.g.*, "[t]itanium dioxide P90 … powder," as the negative electrode active material, as taught by Kavan. LGES-1005 (Kavan), 2. Alternatively, a positive electrode material mixture paste is formed using a phosphate olivine, *e.g.*, $LiFePO_4$ or $LiMnPO_4$, as also taught by Kavan. *Id.*, 2. In this combination electrode material mixture paste, the nanotubes and electrode active material are provided, as taught by Ohata, in amounts such that "[t]he amount of the carbon nanotubes … is, for example, 0.2 to 3 parts by weight per 100 parts by weight of the electrode active material," and "preferably not greater than 0.8 parts by weight." LGES-1004 (Ohata), [0024].

38. The resulting "electrode material mixture paste" can be used to form "[a]n electrode plate," *e.g.*, for a lithium ion battery. LGES-1004 (Ohata), [0025].

For instance, the combination electrode material mixture paste is applied "onto an electrode substrate, which is then dried and rolled" and "cut into a predetermined shape to give an electrode intended." *Id.*, [0025]. Specifically, when TiO_2 is used as the electrode active material, the resulting electrode is a negative electrode for a lithium ion battery. As an alternative, when a phosphate olivine is used as the electrode active material, the resulting electrode is a positive electrode for a lithium ion battery. As I describe in the following sections, either of these implementations renders obvious the claims—that is, either a negative electrode material mixture paste including TiO_2, or a positive electrode material mixture paste including a phosphate olivine.

4. Reasons for the combination

Reasons for incorporation of oxidized nanotubes, and expectation of success

39. It is my opinion that a POSITA would have been motivated to use oxidized nanotubes in the Ohata-Kavan electrode material mixture paste for at least the following reasons.

40. The first reason for using oxidized nanotubes is that, in the specific case when the electrode material mixture paste is a negative electrode material mixture paste having TiO_2 as the active material (*see* LGES-1005 (Kavan), 2), the presence of oxidized nanotubes provides a "beneficial effect of nanotube networking," as illustrated and explained with respect to Kavan's Fig. 5. LGES-

1005 (Kavan), 5. For instance, as I discussed above in my overview of Kavan, "the rate of electrochemical charge transfer is higher" in composites of TiO_2 and oxidized nanotubes as compared to composites with unfunctionalized nanotubes, "due to nanotube networking." *Id.*, 9; *see* Fig. 5. A POSITA would have been motivated to use oxidized nanotubes together with TiO_2 such that the combination electrode material mixture paste would have the ability for fast electrochemical charge transfer.

41. <u>The second reason</u> for using oxidized nanotubes is that, again in the specific case when the electrode material mixture paste is a negative electrode material mixture paste having TiO_2 as the active material (*see* LGES-1005 (Kavan), 2.), oxidized nanotubes "exhibit better affinity towards TiO_2" than do unoxidized nanotubes. LGES-1005 (Kavan), Abstract. This strong interaction between TiO2 and nanotubes is described not only by Kavan (*see* LGES-1005 (Kavan), 4 ("oxidative functionalization through HNO_3 is crucial for proper anchoring of nanotubes towards TiO_2")), but also elsewhere in the literature (*see* LGES-1037 (J. Li), 4 ("carboxylates groups on the CNTs will provide bonding to the TiO_2 … nanoparticles")). A POSITA would have been motivated to use oxidized nanotubes together with TiO_2 to obtain a combination electrode material mixture paste with a robust interaction between the nanotubes and the electrode active material. This strong interaction is beneficial for electrochemical

performance, as Kavan demonstrated via the cyclic voltammetry results shown in Fig. 5. This strong affinity between TiO2 and oxidized nanotubes is also good for providing mechanical stability to the resulting electrode. For instance, an electrode in which the components (*e.g.*, electrode active material and nanotubes) are strongly held together is resistant to deformation and eventual fracture that can occur due to repeated lithium intercalation / de-intercalation, and thus can be expected to have a long cycle life. *See*, *e.g.*, LGES-006 (Lee), [0054] ("the structure of the active material layer 14 can be maintained stably, thereby improving the cycle lifetime characteristics").

42. <u>The third reason</u> for using oxidized nanotubes, now for the specific case when the electrode material mixture paste is instead a positive electrode material mixture paste having a phosphate olivine as the active material (*see* LGES-1005 (Kavan), 2), is that electrochemical performance of an electrode is improved by the presence of oxidized nanotubes. For instance, as I discussed above in my overview of Kavan, oxidized nanotubes in combination specifically with $LiFePO_4$ demonstrate notably good performance: "the $LiFePO_4$ olivine exhibits excellent performance in a composite with MWNT-ox," with "impressive values" for the capacity of the $LiFePO_4$/MWNT-ox composite, as shown at least in Figs. 8 and 10. LGES-1005 (Kavan), 7; *see id.* (describing that in the $LiFePO_4$/MWNT-ox composite "[t]he improvement caused by surface oxidation of

nanotubes is dramatic"), 8 ("It is obvious that the functionalization of nanotubes is, indeed, crucial for enhancing the electrochemical activity of the composite materials with $LiFePO_4$"). Kavan also indicates good electrochemical performance in composites of oxidized nanotubes and $LiMnPO_4$, stating that "the oxidative treatment of nanotubes improves the reversibility of $LiMnPO_4$ charge/discharge." *Id.*, 7. A POSITA would have been motivated to incorporate oxidized nanotubes into the combination positive electrode material mixture paste to achieve these dramatic performance advantages.

43. The fourth reason for using oxidized nanotubes is because it is consistent with, and helpful for achieving, Ohata's goal to have well-dispersed nanotubes in the composite. As I discussed above in my overview of Ohata, Ohata's masterbatch process is designed to generate an electrode material paste with dispersed nanotubes. *See* LGES-1004 (Ohata), [0016] ("The carbon nanotubes are unraveled and straightened by blending them with a resin"). This morphology is desirable because of the electrical and mechanical advantages related to dispersed nanotubes: "The unraveled and straightened carbon nanotubes 102 are expected to form an excellent conductive path in an electrode even if the amount thereof is small. The resin with the unraveled and straightened carbon nanotubes dispersed therein also serves to increase the mechanical strength of an electrode plate." LGES-1004 (Ohata), [0016]-[0017].

44. Oxidized nanotubes are consistent with this objective of having a resin with nanotubes dispersed therein. For instance, oxidized nanotubes are more readily dispersible than unfunctionalized nanotubes, *e.g.*, because the functional groups provide both chemical and physical hindrance against agglomeration. Unfunctionalized nanotubes tend to stick together in clumps due to a phenomenon known as π-π stacking. *See* LGES-1044 (Hirsch), 1854. π-π stacking is a result of interactions between the graphene layers that form the nanotube walls. When nanotubes are functionalized, *e.g.*, with oxygen-containing functional groups, these functional groups disrupt the interactions between nanotubes, thus also disrupting the π-π stacking and preventing the nanotubes from forming clumps. These oxygen-containing groups on the sidewalls of functionalized nanotubes also act as physical barriers that prevent the nanotubes from getting close enough together to be able to form clumps.

45. It was well-known that oxidization of nanotubes generally prevents clumping of the nanotubes. For instance, Gojny explains that "[t]he presence of carboxylic acid groups on the surface" "enables a better dispersion of CNTs," and shows images of "oxidized MWCNTs … [that] are completely dispersed in the [epoxy resin] matrix due to the appearance of carboxylic groups on the surface." LGES-1014 (Gojny), 821-822. Similarly, Xing observed that "treated [oxidized] CNTs had better dispersion" (LGES-1015 (Xing), 4186-4187), and Farrugia

described that functionalization of carbon nanotubes "results in one or more chemical moieties attached to the carbon nanotubes," which "results in better dispersion of the nanotubes throughout the matrix." LGES-1007 (Farrugia), [0027].

46. Because oxidation can facilitate dispersion of nanotubes, which is consistent with one of Ohata's objectives, a POSITA would have been motivated to use oxidized nanotubes in the masterbatch process, *e.g.*, to support and/or enhance, Ohata's goal of a masterbatch with nanotubes dispersed therein.

47. The fifth reason for using oxidized nanotubes in the combination electrode material mixture paste is because oxidative treatments can "remove amorphous carbon and metallic impurities from as-produced CNTs," thus leading to a higher-purity starting material. LGES-1010 (Wepasnick), 2.

48. The sixth reason for using oxidized nanotubes is that doing so is just the application of known techniques (specifically, oxidized nanotubes in an electrode composition) to an already-known structure (Ohata's electrode material paste, which already includes nanotubes), and this combination produces predictable results (specifically, an electrode material paste including oxidized nanotubes).

49. I note that oxidized nanotubes may have a lower conductivity than their unoxidized counterparts. However, use of oxidized nanotubes also brings a

number of performance advantages, such as those I discussed above. I expect that a POSITA would have known how to balance the trade-offs between lower conductivity and the various advantages associated with oxidized nanotubes, and it is my opinion that given the strong advantages that are directly applicable to the context of the Ohata-Kavan combination, a POSITA would have chosen to implement oxidized nanotubes in the combination.

50. It is my opinion that a POSITA would have expected success in incorporating oxidized nanotubes into the Ohata-Kavan electrode material mixture paste for at least the following reasons.

51. First, a POSITA would have expected to be successful in using oxidized nanotubes in combination with Ohata's electrode material mixture paste (which is for a lithium ion battery) because the use of oxidized nanotubes in compositions for lithium ion battery electrodes was conventional. This is demonstrated by Kavan itself. *See* LGES-1005 (Kavan), Abstract, 9. This is also supported by Sheem, which describes a lithium ion battery that "includes an active material and a fiber-shaped or tube-shaped carbon conductive material," which "preferably includes carbon nano fibers or carbon nanotubes," Where "[t]he fiber-shaped or tube-shaped carbon conductive material is oxidized," and "[t]he oxidized fiber-shaped or tube-shaped carbon conductive material includes functional groups selected from the group consisting of carboxyl groups (—COOH), hydroxyl groups

(—OH), carbonyl groups (—COH), and combinations thereof that are attached to the surface thereof"). LGES-1032 (Sheem), 3:28-52.

52. Second, a POSITA would have expected to be successful in using oxidized nanotubes in the context of Ohata's electrode material mixture paste because the composition of Kavan's slurry is generally similar to the composition of Ohata's electrode material paste. For instance, both use a fluorocarbon resin such as polyvinylidene fluoride (PVDF) and a dispersant such as N-methyl-2-pyrrolidone. *See* LGES-1004 (Ohata), [0022] ("As the resin in which the carbon nanotubes are dispersed, a fluorocarbon resin can be used. The fluorocarbon resin is preferably at least one selected from the group consisting of polyvinylidene fluoride and a vinylidene fluoride-hexafluoropropylene copolymer"); LGES-1005 (Kavan), 2 ("The electrodes were prepared as follows: powder precursor material, either pure or carbon-containing … was mixed with 5 wt% of polyvinylidene fluoride (PVDF) dissolved in N-methyl-2-pyrrolidone"). Furthermore, both Ohata and Kavan use a powder of the electrode active material in forming the composite. *See* LGES-1004 (Ohata), [0032] ("A positive electrode material mixture paste was prepared by mixing 3.3 parts by weight of the resultant masterbatch, 100 parts by weight of lithium cobalt oxide ($LiCoO_2$) as an active material and 45 parts by weight of N-methyl-2-pyrrolidone with a mixer"); LGES-1005 (Kavan), 2. These similarities would have provided the POSITA with a reasonable expectation that

oxidized nanotubes, as taught in the context of Kavan's slurry, would be compatible with Ohata's electrode material paste.

Reasons for use of TiO_2 as the electrode active material

53. It is my opinion that a POSITA would have been motivated to use TiO_2 as the negative electrode active material in the Ohata-Kavan electrode material mixture paste for at least the following reasons. In this implementation, the combination electrode material mixture paste can be used to form a negative electrode of a lithium ion battery.

54. The first reason for using TiO_2 as the negative electrode active material is that it is a conventional and well-characterized material for use as a negative electrode active material in lithium ion batteries. For instance, according to Kavan, TiO_2 is among "the top interesting host structures for lithium insertion." LGES-1005 (Kavan), 1. Similarly, Deng states that "TiO_2 is a typical Li-ion intercalation compound." LGES-1040 (Deng), 1.

55. The second reason for using TiO_2 as the negative electrode active material is that the combination of TiO_2 and oxidized nanotubes produces a synergistic effect on electrochemical performance, as demonstrated by Kavan. For instance, Kavan notes that "the electrochemical activity of the composite TiO_2/nanotubes is improved compared to that of the parent pure TiO_2." LGES-1005 (Kavan), 1. More specifically, as I explained above in my overview of

Kavan, Kavan shows that "the electrochemical performance of TiO_2 is improved by nanotube networking," and specifically, that "the composite [of TiO_2] with functionalized nanotubes shows unique faradaic pseudocapacitance." LGES-1005 (Kavan), Abstract; *see* 9 ("The Li-storage capacity of TiO_2 is not enhanced in the composite, but the rate of electrochemical charge transfer is higher due to nanotube networking. … the composite with functionalized nanotubes shows unique faradaic pseudocapacitance, which is specific only for the interaction of functionalized MWNT-ox with the titania surface"). The Ohata-Kavan composition includes nanotubes, and a POSITA would have been motivated to use TiO_2 to obtain enhancements in electrochemical performance that stem from this unique interaction between TiO_2 and the oxidized nanotubes in the Ohata-Kavan electrode material mixture paste.

56. <u>The third reason</u> for using TiO_2 as the negative electrode active material is that TiO_2 has desirable electrochemical performance characteristics, including high energy density, good cycling performance, and rapid charge/discharge velocity. For instance, Pfanzelt notes that nanosized TiO_2 "shows a high capacity, high rate performance and excellent cycling stability," and states that more generally, "TiO_2 materials have become an interesting anode material for lithium-ion batteries with unique properties such as high power densities, cheapness, safety and non-toxicity." LGES-1033 (Pfanzelt), Abstract, 1.

57. The fourth reason for using TiO_2 as the negative electrode active material is that TiO_2 undergoes a relatively small volume change during lithium intercalation and deintercalation, and thus TiO_2 electrodes are structurally durable, *e.g.*, resistant to fracture. For instance, Deng explains that "TiO_2 is a typical Li-ion intercalation compound with a volume change < 4% in the reaction: $TiO_2 + xLi^+ xe \leftrightarrow Li_xTiO_2$ ($0 \leq x \leq 1$)," and further notes that "[t]he negligible lattice changes of TiO_2-based structures upon Li-intercalation and de-intercalation are responsible for the enhanced structural stability and longer cycle life." LGES-1040 (Deng), 1.

58. The fifth reason for using TiO_2 as the negative electrode active material is that TiO_2 is a desirable electrode active material because of its robust safety profile and low toxicity. This is supported by Pfanzelt, which states that nanosized TiO_2 "shows a high capacity, high rate performance and excellent cycling stability. We can clearly prove the safer character of rutile electrodes by thermal analysis." LGES-1033 (Pfanzelt), Abstract; *see* 1 ("TiO2 materials have become an interesting anode material for lithium-ion batteries with unique properties such as high power densities, cheapness, safety and non-toxicity").

59. The sixth reason for using TiO_2 as the negative electrode active material is that TiO_2 is relatively inexpensive, making it an attractive material for use in lithium ion batteries. *See* LGES-1033 (Pfanzelt), 1 ("TiO2 materials have

become an interesting anode material for lithium-ion batteries with unique properties such as high power densities, cheapness, safety and non-toxicity."

60. The seventh reason for using TiO_2 as the negative electrode active material is that doing so is just the application of known techniques (specifically, TiO_2 as an electrode active material in lithium ion battery electrode composition) to an already-known structure (Ohata's negative electrode material paste), and the combination produces predictable results (specifically, a negative electrode material paste including TiO_2 as the electrode active material).

Reasons for use of a phosphate olivine as the electrode active material

61. As an alternative to using TiO_2 as a negative electrode active material, it is separately my opinion that a POSITA would have been motivated to use a phosphate olivine as the positive electrode active material in the Ohata-Kavan electrode material mixture paste for at least the following reasons. In this implementation, the combination electrode material mixture paste can be used to form a positive electrode of a lithium ion battery.

62. The first reason for using phosphate olivines as the positive electrode active material is that phosphate olivines—*e.g.*, specifically $LiFePO_4$—exhibit good electrochemical performance when combined with oxidized nanotubes, as described by Kavan and as I discussed above in my overview of Kavan. Kavan notes that "the $LiFePO_4$ olivine exhibits excellent [electrochemical] performance in

a composite with MWNT-ox," and specifically, that it "exhibits a capacity of 149 $mAhg^{-1}$ … and nearly perfect charge/discharge reversibility of 99-100%." LGES-1005 (Kavan), 7; *see* Abstract ("excellent [electrochemical] performance was found for $LiFePO_4$ (olivine) in composite materials containing only 2wt% of functionalized nanotubes"). This phenomenon of synergy between phosphate olivines (*e.g.*, $LiFePO_4$) and carbon nanotubes also has been observed elsewhere in the literature. For instance, Feng reported that "MWCNTs adding improved the electronic conductivity, the discharge capacity, cycle stability and lithium ion diffusion kinetics of $LiFePO_4$, but MWCNTs adding did not change the orthorhombic olivine-type structure of $LiFePO_4$." LGES-1042 (Feng), Abstract.

63. The second reason for using phosphate olivines as the positive electrode active material is that phosphate olivines have a robust safety profile. Kavan indicates that "phosphate olivines … are promising cathode materials for Li-ion batteries due to high safety." LGES-1005 (Kavan), 1. Nakura provides further details, explaining that "olivine-type lithium phosphates are less likely to promote heat generation inside the battery than lithium cobaltate, since the metals contained therein have a lower reactivity than cobalt. Therefore, non-aqueous electrolyte secondary batteries including olivine-type lithium phosphates are believed to have higher safety than non-aqueous electrolyte secondary batteries including lithium cobaltate." LGES-1034 (Nakura), [0004].

64. The third reason for using phosphate olivines as the positive electrode active material is that phosphate olivines are economically desirable electrode active materials, as indicated by Kavan: "the phosphate olivines … are promising cathode materials for Li-ion batteries due to … low cost." LGES-1005 (Kavan), 1; *see* LGES-1034 (Nakura), [0004] ("Olivine-type lithium phosphates can be stably supplied relatively easily at low costs"). For instance, Chen specifically addresses $LiFePO_4$, stating that "[a] good cathode material has to meet the following criteria: … affordability for consumer electronics …. $LiFePO_4$, with the Olivine structure, … appears to meet many of these requirements." LGES-1041 (Chen), 1. Similarly, Feng states that "[a]mong these cathode materials, olivine structured $LiFePO_4$ have attracted the most interest due to the low cost." LGES-1042 (Feng), 1.

65. The fourth reason for using phosphate olivines as the positive electrode active material is that they are environmentally friendly and relatively non-toxic: "the phosphate olivines … are promising cathode materials for Li-ion batteries due to … environmental tolerability." LGES-1005 (Kavan), 1. For instance, Chen specifically addresses $LiFePO_4$, stating that "[a] good cathode material has to meet the following criteria: … low toxicity. $LiFePO_4$, with the Olivine structure, … appears to meet many of these requirements." LGES-1041 (Chen), 1. Similarly, Feng states that "[a]mong these cathode materials, olivine

structured $LiFePO_4$ have attracted the most interest due to … low toxicity." LGES-1042 (Feng), 1.

66. The fifth reason for using phosphate olivines such as $LiFePO_4$ is that they have good electrochemical performance, such as high capacity, long cycle life, and stability. For instance, Chen specifically addresses $LiFePO_4$, stating that "[a] good cathode material has to meet the following criteria: high capacity that can be retained for up to 1000 cycles; stability that can withstand fast recharge and discharge and other possible extreme conditions …. $LiFePO_4$, with the Olivine structure, … appears to meet many of these requirements." LGES-1041 (Chen), 1; *see id.* ("It [$LiFePO_4$] shows no obvious fading even after several hundred cycles, and shows power capabilities comparable to that of the mixed layered oxides. Its theoretic energy density is 550 W h/kg, higher than obtained in commercial $LiCoO_2$ cells, and moreover it is very stable during discharge/recharge"). Similarly, Feng states that "[a]mong these cathode materials, olivine structured $LiFePO_4$ have attracted the most interest due to … the high approaching to the theoretical, practical capacity of larger than 160 mAh g^{-1}." LGES-1042 (Feng), 1.

67. The sixth reason for using phosphate olivines as the positive electrode active material is that doing so is just the application of known techniques (specifically, a phosphate olivine as an electrode active material in lithium ion battery electrode composition) to an already-known structure (Ohata's positive

electrode material paste), and the combination produces predictable results (specifically, a positive electrode material paste including phosphate olivine as the electrode active material).

Expectation of success in using TiO_2 or phosphate olivines as the electrode active material

68. It is my opinion that a POSITA would have expected success in incorporating TiO_2 as anode or/and a phosphate olivine as an electrode active material for a cathode into the Ohata-Kavan electrode material mixture paste for at least the following reasons.

69. First, a POSITA would have expected to be successful in implementing either TiO_2 (for a negative electrode) or a phosphate olivine (for a positive electrode) in the combination because use of both TiO_2 and phosphate olivines in compositions for lithium ion battery electrodes was conventional, and these materials were well-characterized as electrode active materials. This is indicated by Kavan, as I have discussed above. *See* LGES-1005 (Kavan), 1. The conventional nature of TiO_2 is confirmed by Pfanzelt, which states that "TiO_2 materials have become an interesting anode material for lithium-ion batteries with unique properties such as high power densities, cheapness, safety and non-toxicity." LGES-1033 (Pfanzelt), 1. And the conventional nature of phosphate olivines is confirmed by Nakura ("As an alternative positive electrode active material to lithium cobaltate, lithium phosphates with an olivine structure are

receiving attention." LGES-1034 (Nakura), [0004]) and Chen ("$LiFePO_4$ is a potential cathode candidate for the next generation of secondary lithium batteries." LGES-1041 (Chen), Abstract).

70. Second, a POSITA would have expected to be successful in implementing either TiO_2 or a phosphate olivine in the combination Ohata indicates that various electrode active material compositions can be used with its electrode material mixture paste. For negative electrodes, Ohata states that "[t]he negative electrode active material to be used also varies depending on the battery type." LGES-1004 (Ohata), [0027]. Similarly, for positive electrodes, Ohata again states that "[t]he positive electrode active material to be used varies depending on the battery type." *Id.*, [0026]. Given this flexibility, and the conventional nature of TiO_2 and phosphate olivines as electrode active materials as I indicated above, a POSITA would have expected these materials to be compatible with Ohata's electrode material mixture paste.

71. Third, a POSITA would have expected to be successful in implementing either TiO_2 or a phosphate olivine in the combination because the composition of Kavan's slurry is generally similar to the composition of Ohata's electrode material paste, as I previously discussed when addressing the use of oxidized nanotubes. *See* LGES-1004 (Ohata), [0022], [0032]; LGES-1005 (Kavan), 2. These similarities would have provided the POSITA with a reasonable

expectation that either TiO_2 or phosphate olivines, as taught in the context of Kavan's slurry, could be used with Ohata's electrode material paste.

5. Claim-by-Claim Analysis

(i) Claim 1

[1pre] A composition useful for lithium ion batteries comprising:

72. To the extent the preamble is deemed to be limiting, Ohata-Kavan renders it obvious.

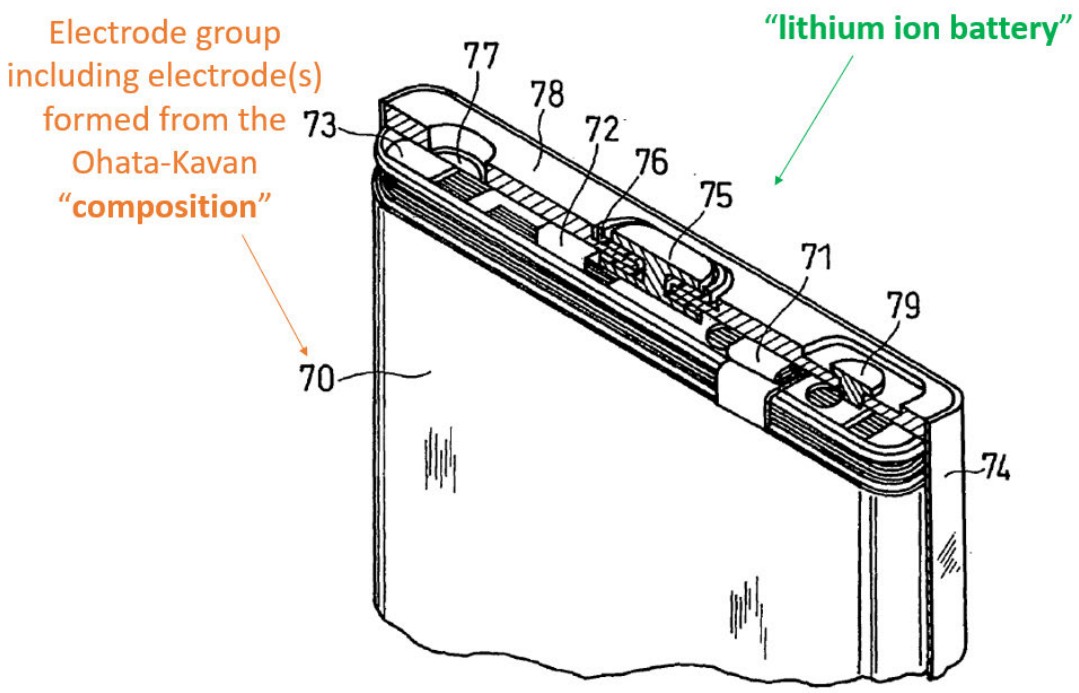
73. First, the Ohata-Kavan combination renders obvious a **composition** based on Ohata's "electrode material mixture paste." LGES-1004 (Ohata), Abstract; *see* [0005]. The combination **composition** includes "a masterbatch comprising at least carbon nanotubes and a resin" and "an electrode active material," as taught by Ohata. *Id.*, [0005]; *see* [0022] ("An electrode material mixture paste can be prepared by blending an electrode material mixture containing the masterbatch and an electrode active material with a dispersion medium"), Abstract, [0016], [0021]-[0022], [0030]-[0032], [0040]-[0042], [0046], claim 1. Consistent with the teachings of Ohata, the electrode active material of the combination composition can be "a negative electrode active material" or "a positive electrode active material." *Id.*, [0007]-[0008]; *see* [0026]-[0027], claims 4-5. Specifically, in the combination **composition**, the negative or positive electrode active material is implemented as TiO_2 or a phosphate olivine, based on

the active material compositions described by Kavan. *See* LGES-1005 (Kavan), Abstract, 2; *see* my discussion of element [1b], *infra*.

74. Moreover, based on Ohata, the combination electrode material mixture paste (a **composition**) is used to produce "[a]n electrode for a battery superior in conductivity," *e.g.*, the composition is **useful for batteries**. LGES-1004 (Ohata), Abstract; *see* [0017] (describing that using Ohata's electrode material mixture, "it is possible to produce a secondary battery excellent in cycle characteristics"), [0005], [0015], claim 1. An example of a **lithium ion battery** is illustrated in Ohata's FIG. 2, *infra*, which shows a "prismatic non-aqueous electrolyte secondary battery" having LGES-1004 (Ohata), [0036]. This secondary battery includes a positive electrode formed from the combination electrode material mixture (*see* LGES-1004 (Ohata), [0037] ("The positive and negative electrodes produced above"), [0032]) and a "non-aqueous electrolyte" including "$LiPF_6$ in a solvent mixture comprising ethylene carbonate (EC) and ethylmethyl carbonate (EMC)." *Id.*, [0039]; *see* [0026] ("The positive electrode active material to be used varies depending on the battery type. As ***the positive electrode active material for a lithium secondary battery***, there can be used, for example, a transition metal oxide"). Alternatively, the battery of FIG. 2 is a **lithium ion battery** including a negative electrode formed from the combination electrode material mixture. *See id.*, [0047] ("With the use of the positive and

negative electrodes thus obtained, a prismatic non-aqueous electrolyte secondary battery analogous to the one in Example 1 was assembled"), [0046], [0027] ("The negative electrode active material to be used also varies depending on the battery type. As ***the negative electrode active material for a lithium secondary battery***, there can be used, for example, a graphite material such as artificial graphite or natural graphite, graphitized mesophase carbon made from coal or petroleum pitch, or other carbonaceous materials such as hard carbon. An alloy material such as an Si—Ni alloy").

FIG. 2



Excerpt from Ohata, FIG. 2 (annotated)

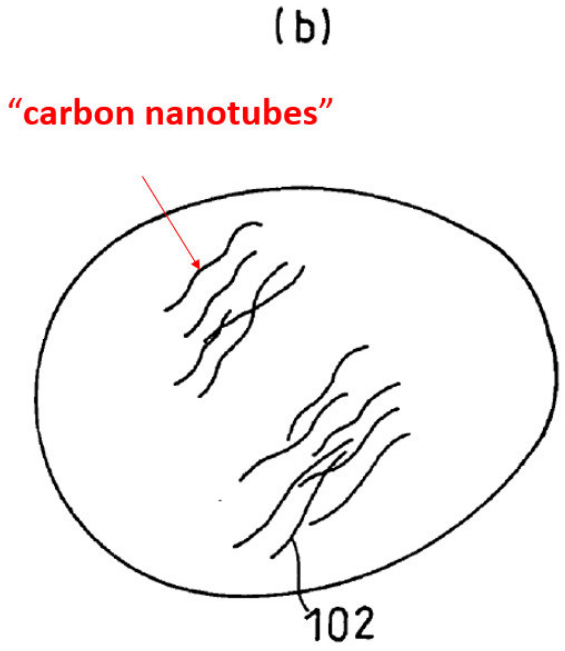
75. Importantly, even with the implementation of TiO_2 or a phosphate olivine as the electrode active material, based on Kavan, the combination composition remains **useful for lithium ion batteries**. TiO_2 (as its derivative form, lithium titanate, most commonly, $Li_4Ti_5O_{12}$, LTO) and phosphate olivines were conventional electrode active materials for lithium ion batteries. *See* LGES-1005 (Kavan), 1 ("The electrochemical lithium storage in carbon nanotube/TiO_2 composites has been studied … reflecting the fact that TiO_2 … belong to the top interesting host structures for lithium insertion"), 2 ("Since TiO_2 is a prospective anode material, and $LiMPO_4$ olivines are promising cathode materials, concerted study of the interaction of MWNT with these three selected materials at defined comparable conditions is relevant for the development of high-power Li-ion batteries"); LGES-1033 (Pfanzelt), 1 ("TiO2 materials have become an interesting anode material for lithium-ion batteries with unique properties such as high power densities, cheapness, safety and non-toxicity"); LGES-1034 (Nakura), [0004] ("As an alternative positive electrode active material to lithium cobaltate, lithium phosphates with an olivine structure are receiving attention")).

[1a] discrete carbon nanotubes

The combination composition includes carbon nanotubes

76. Ohata's "masterbatch" includes "***carbon nanotubes*** and a resin," as shown in Fig. 1(b), *infra*. LGES-1004 (Ohata), Abstract; *see* [0015] ("producing a

masterbatch by blending at least ***carbon nanotubes*** with a resin"), [0016] ("The ***carbon nanotubes*** are unraveled and straightened by blending them with a resin"), [0006] ("The proper amount of the ***carbon nanotubes*** contained in the masterbatch is 5 to 20 parts by weight per 10 parts by weight of the resin"), [0017] ("The resin with the unraveled and straightened ***carbon nanotubes*** dispersed therein"), [0005], [0009]-[0010], [0018]-[0022], [0030], claim 1, claim 3.



Except from Ohata, Fig. 1(b) (annotated)

77. The combination composition (based on Ohata's electrode material mixture paste; *see* [1pre]) is formed by "blending an electrode material mixture containing at least the masterbatch and an electrode active material with a dispersion medium to prepare an electrode material mixture paste." LGES-1004 (Ohata), Abstract; *see* [0005], [0023], [0032], claim 1. As discussed in the

preceding paragraph, the masterbatch includes **carbon nanotubes**. Thus, the **composition** of the Ohata-Kavan combination (based on Ohata's electrode material mixture paste), which includes the masterbatch, thus likewise includes **carbon nanotubes**. *See* LGES-1004 (Ohata), [0024] ("The amount of the carbon nanotubes contained in the electrode material mixture"), [0007]-[0008], [0060]-[0061].

78. Ohata describes an example of producing a negative electrode material mixture paste (**a composition**) from a masterbatch including **carbon nanotubes**. "A negative electrode material mixture paste was produced by mixing 3.3 parts by weight of masterbatch, the same as the one used in the production of the positive electrode in Example 1" (which includes **carbon nanotubes**; *see id.*, [0030]), "100 parts by weight of flake graphite as an active material and 56 parts by weight of N-methyl-2-pyrrolidone as a dispersion medium." *Id.*, [0046].

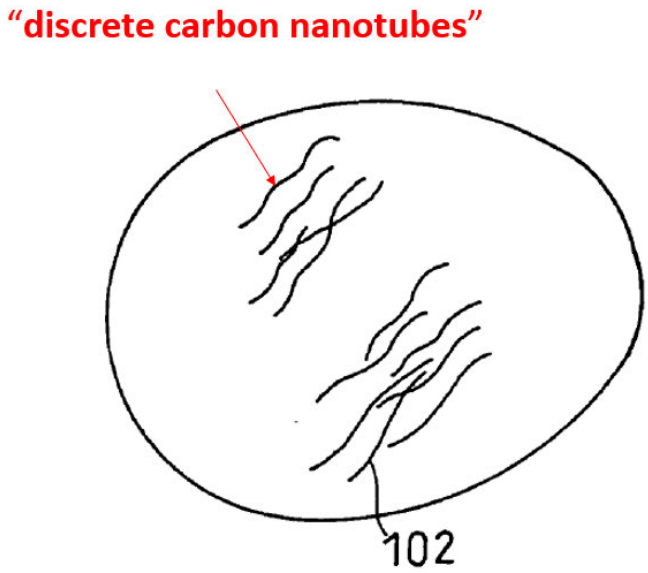
79. Ohata also describes specific examples of producing a positive electrode material mixture paste (**a composition**) from a masterbatch including **carbon nanotubes**: "Fifteen parts by weight of ***carbon nanotubes*** and 85 parts by weight of polyvinylidene fluoride as a resin component were blended in an extruder to produce a masterbatch," and "[a] positive electrode material mixture paste was prepared by mixing 3.3 parts by weight of the resultant masterbatch, 100 parts by weight of lithium cobalt oxide (LiCoO2) as an active material and 45 parts

by weight of N-methyl-2-pyrrolidone." LGES-1004 (Ohata), [0030], [0032]; *see* [0040]-[0042].

The carbon nanotubes in the combination composition are discrete

80. The **composition** of the Ohata-Kavan combination (based on Ohata's electrode material mixture paste) includes a masterbatch, which contains **discrete** nanotubes, as taught by Ohata. Specifically, Ohata describes "producing a masterbatch by blending at least carbon nanotubes with a resin." LGES-1004 (Ohata), [0015]. Prior to this blending, the nanotubes "are tangled and twisted with one another," as illustrated in Ohata's FIG. 1(a). *Id.*, [0016]. In the process of "blending them with a resin" to form the masterbatch, "[t]he carbon nanotubes are ***unraveled and straightened***," as shown in FIG. 1(b), *infra*. *Id.* The result is a "resin with the unraveled and straightened carbon nanotubes ***dispersed therein***," *e.g.*, a masterbatch containing **discrete carbon nanotubes**. *Id.*, [0017]; *see* [0016] ("The unraveled and straightened carbon nanotubes 102 are expected to form an excellent conductive path in an electrode"), [0022] ("As the resin in which ***the carbon nanotubes are dispersed***, a fluorocarbon resin can be used"), [0002] ("in order to reduce the amount of conductive material used, it is proposed to use a masterbatch process, whereby ***a conductive material is uniformly predispersed in a binder***, in the production of a positive electrode"). For instance, a POSITA would have understood or found obvious that these "unraveled and straightened

carbon nanotubes" are **discrete** because they are generally "dispersed" as individual nanotubes in the resin, as described and illustrated by Ohata. LGES-1004 (Ohata), [0017].



Excerpt from Ohata, Fig. 1(b) (annotated)

81. Alternatively/additionally, a POSITA would have understood or found obvious that the nanotubes in the combination composition are **discrete**. As I discuss further for element [1d], *infra*, the nanotubes in the Ohata-Kavan combination are oxidized. At least in some instances, oxidization can help promote non-agglomeration of nanotubes, *e.g.*, oxidized nanotubes can be discrete.

82. The sidewalls and/or ends of oxidized nanotubes are functionalized by species such as carboxyl, carbonyl, or hydroxyl groups that are covalently bound to the nanotubes. *See* LGES-1005 (Kavan), Abstract ("Multi-walled carbon nanotubes functionalized by carboxylic groups"); LGES-1010 (Wepasnick),

Abstract ("Six commonly used wet chemical oxidants … were evaluated in terms of their effects on the surface chemistry and structure of MWCNTs … X-ray photoelectron spectroscopy (XPS) and energy dispersive spectroscopy (EDX) were used to characterize the extent of surface oxidation, while chemical derivatization techniques used in conjunction with XPS allowed the concentration of carboxyl, carbonyl, and hydroxyl groups at the surface to be quantified"); LGES-1014 (Gojny), 821-822 (describing that oxidation of carbon nanotubes resulted in in "[t]he presence of carboxylic acid groups on the surface"); LGES-1007 (Farrugia), [0028] ("oxidation is carried out by acid-treatment, wherein oxidation chemistry may be used to open the end caps of both single and multi-walled nanotubes to produce carboxyl, carbonyl and hydroxyl groups at the opened ends and defects on the side walls"). As I explained above, the presence of these functionalities helps to physically and chemically inhibit the nanotubes from aggregating into clumps, and instead oxidized nanotubes are generally present as individually distinct nanotubes. *See* LGES-1014 (Gojny), 821-822 ("[t]he presence of carboxylic acid groups on the surface" "enables a better dispersion of CNTs" showing images of "oxidised [sic] MWCNTs … [that] are completely dispersed in the [epoxy resin] matrix due to the appearance of carboxylic groups on the surface"); LGES-1015 (Xing), 4186-4187 ("treated [oxidized] CNTs had better dispersion"); LGES-1007 (Farrugia), [0027] (describing that functionalization of carbon nanotubes "results

in one or more chemical moieties attached to the carbon nanotubes," which "results in better dispersion of the nanotubes throughout the matrix").

83. In light of this well-known behavior of oxidized nanotubes, a POSITA would have understood or found obvious that oxidized nanotubes of the combination composition can be present as generally individually distinct nanotubes, *e.g.*, that the nanotubes are **discrete**.

[1b] having crystals or layers of lithium ion active material attached to their surface,

84. The combination composition (based on Ohata's electrode material mixture paste; *see* [1pre]) is formed by "blending an electrode material mixture containing at least the masterbatch and ***an electrode active material*** with a dispersion medium to prepare an electrode material mixture paste." LGES-1004 (Ohata), Abstract; *see* [0005], [0023], [0032], claim 1, claims 8-10. The electrode active material, *e.g.*, either a "negative electrode active material" or a "positive electrode active material," corresponds to **lithium ion active material**. LGES-1004 (Ohata), [0026]-[0027]. The specific composition of this electrode active material as either TiO_2 or a phosphate olivine is based on the teachings of Kavan, as discussed in the following paragraphs.

<u>*Lithium ion active material for anodes*</u>

85. The negative electrode active material (the **lithium ion active material**) in the Ohata-Kavan electrode material mixture paste is titanium dioxide

(TiO_2) based on Kavan's teachings of forming a "paste" including "nanotubes" and "***TiO_2***." LGES-1005 (Kavan), 2; *see* LGES-1004 (Ohata), [0027] ("The negative electrode active material … varies depending on the battery type"); LGES-1005 (Kavan), 2 (describing use of "***Titanium dioxide P90*** … powder" in fabricating samples), 3 ("Fig. 2 shows SEM images of thin film electrodes from … ***TiO_2***/MWNT-ox," in which "[a]ll nanotubes are covered with individual nanocrystals of P90 [***titania***] in a homogeneous nanocomposite"), 9 ("composite materials in which multi-walled carbon nanotubes … were interfaced to … ***TiO_2***"), 5 ("Fig. 5 shows the cyclic voltammogram of … the composites with nanotubes, ***P90***/MWNT-ox"), Abstract ("Multi-walled carbon nanotubes functionalized by carboxylic groups, exhibit better affinity towards ***TiO_2***").

86. TiO_2 is a **lithium ion active material**, as confirmed by Kavan: "***TiO_2*** … belong to the top interesting host structures for ***lithium insertion***." LGES-1005 (Kavan), 1; *see* 2 ("Since ***TiO_2 is a prospective anode material*** …, concerted study of the interaction of MWNT with" TiO_2 "is relevant for the development of high-power Li-ion batteries."). Indeed, that TiO_2 (particularly as the derived form of lithium titanate, $Li_4Ti_5O_{12}$, LTO) is a lithium ion active material was well-known. *See* LGES-1033 (Pfanzelt), 1 ("TiO2 materials have become an interesting anode material for lithium-ion batteries with unique properties such as high power densities, cheapness, safety and non-toxicity"). The '909 Patent identifies

"titanium dioxide" as an example of an "ion active material[]." LGES-1001, 4:30-33.

87. Consistent with the teachings of Kavan, in the combination electrode material paste, "[a]ll nanotubes are covered with individual ***nanocrystals of P90*** in a homogeneous nanocomposite," *e.g.*, the TiO_2 (**lithium ion active material**) is present in the combination composition as **crystals**. LGES-1005 (Kavan), 3; *see* 4 ("industrial powder (P90) is a good source of ***nanocrystals*** which spontaneously self-assemble on the nanotube surface"), *id.* ("the ***P90 crystals*** are ca. 10 nm in size"), 1 ("Carbon nanotubes form conductive composites with inorganic ***nanocrystals***").

88. I note that the process of preparing the combination composition is based on Ohata, which describes "blending an electrode material mixture containing at least the masterbatch and an electrode active material with a dispersion medium." LGES-1004 (Ohata), Abstract. That is, when the electrode active material is TiO_2, the masterbatch (including carbon nanotubes) is blended with the TiO_2 power in a dispersion medium. This blending is performed "with a mixer." *Id.*, [0032]. I do not expect this blending process to have any meaningful impact on the crystallinity of the TiO_2 powder, *i.e.*, the TiO_2 in the combination composition is **crystals**, as I discussed in the preceding paragraph.

89. Moreover, based on Kavan, the nanotubes in the combination electrode material paste have TiO_2 nanocrystals (**crystals of lithium ion active material**) **attached to their surface**. For instance, the TiO_2 nanocrystals are attached to the nanotube surface due at least in part to the surface oxidation of the nanotubes (*see* [1d], *infra*): "oxidative functionalization through HNO_3 is crucial for ***proper anchoring of nanotubes towards TiO_2***"). LGES-1005 (Kavan), 3-4; *see* 2 ("The surface oxidation of carbon nanotubes by HNO_3, H_2SO_4/HNO_3 or H_2O_2, producing among other groups also carboxyls, was sometimes mentioned to improve ***the anchoring of TiO_2***"), *id.* ("detailed understanding is desirable of the MWNT functionalization (by surface oxidation forming carboxyls) for ***optimum attachment of MWNT towards titania … surfaces***"), p.4 ("the interaction of -COOH with phosphate surface might be less efficient, than the ***interaction of carboxyls with titania***"), p.5 ("the ***interactions between the oxidized (carboxylated) surface of carbon nanotubes and TiO_2*** creates new sites for pseudocapacitive interfacial redox chemistry"), p.9 ("***the interaction of functionalized MWNT-ox with the titania surface***"), Abstract ("Multi-walled carbon nanotubes functionalized by carboxylic groups, exhibit ***better affinity towards TiO_2***").

90. Chemical attachment of TiO_2 to oxidized nanotubes has been confirmed elsewhere in the literature. For instance, X. Li demonstrated results

indicating "that TiO_2 nanoparticles were chemically absorbed at SWNT defect sites via an ester-type linkage between carboxylic acid groups at the defect sites and hydroxyl groups at the surface of the TiO_2 nanoparticles." LGES-1030 (X. Li), Abstract. An ester-type linkage is a covalent bond between the carboxyl groups on the surface of the nanotubes and the hydroxyl groups on the surface of the TiO_2 particles. Additionally, J. Li reported XPS data demonstrating that "carboxylates groups on the CNTs will provide bonding to the TiO_2 … nanoparticles." LGES-1037 (J. Li), 4. Here, the XPS data support that there is covalent bonding between the TiO_2 particles and the carboxyl groups on the nanotubes.

91. Based on Kavan's own data and statements regarding the attachment of TiO_2 to the oxidized nanotubes, and supported by literature demonstrating that TiO_2 particles covalently bonded to oxidized nanotubes, I conclude that the TiO_2 particles in Kavan's composition are attached—for instance, via covalent bonding—to the surface of the oxidized nanotubes. Thus, I also conclude that in the Ohata-Kavan composition, which contains oxidized nanotubes (as I discuss for element [1d], *infra*) and TiO_2 particles, the TiO_2 particles are likewise **attached**, *e.g.*, via covalent bonding, to the surface of the oxidized nanotubes.

92. Alternatively/additionally, there are Van der Waals forces between the TiO_2 and the oxygen-containing functionalities on the surface of the nanotubes. To the extent that Van der Waals bonding is considered an attachment, the TiO_2

particles are alternatively/additionally **attached** to the surface of the nanotubes through Van der Waals bonding.

93. Still alternatively/additionally, hydrogen bonds can arise between the TiO_2 particles and hydrogen atoms belonging to the oxygen-containing functionalities on the nanotube surface. *See* LGES-1037 (J. Li), 4 ("hydrogen bonding among the carboxylic, hydroxyl, and/or ether functional groups in both metal oxide and CNTs phases may also be partially responsible for the observed interconnectivity"). To the extent that hydrogen bonding is considered an attachment, the TiO_2 particles are alternatively/additionally **attached** to the surface of the nanotubes through hydrogen bonding.

Lithium ion active material for cathodes

94. Alternatively, the positive electrode active material (the **lithium ion active material**) in the Ohata-Kavan electrode material mixture paste is a phosphate olivine such as $LiMnPO_4$ or $LiFePO_4$ based on Kavan's teachings of forming a "paste" including "nanotubes" and "***$LiMnPO_4$ or $LiFePO_4$***." LGES-1005 (Kavan), 2; *see* LGES-1004 (Ohata), [0026] ("The positive electrode active material … varies depending on the battery type"); LGES-1005 (Kavan), 2 (describing use of "carbon-free olivines, $LiFePO_4$ or $LiMnPO_4$ in fabricating samples), 4 ("Fig. 3 shows the morphological features of our olivine/MWNT-ox composites"), 6 ("Fig. 6 shows a series of cyclic voltammograms of

$LiMnPO_4$/MWNT-ox composite"), 7 ("Fig. 8 shows cyclic voltammograms" for "[t]he composite $LiFePO_4$/MWNT-ox"), 7 ("Fig. 9 shows a series of cyclic voltammograms for a series of composite materials $LiFePO_4$/MWNT-ox"), 8 ("Fig. 11 shows the initial discharge/charge of $LiMnPO_4$/MWNT-ox at varying charge rates from C/10 to C. A similar plot for $LiFePO_4$/MWNT-ox is shown in Fig. 12"), 9 ("composite materials in which multi-walled carbon nanotubes … were interfaced to … $LiMnPO_4$ and $LiFePO_4$").

95. Phosphate olivines such as $LiMnPO_4$ and $LiFePO_4$ are each a **lithium ion active material**, as confirmed by Kavan: 2 ("***$LiMPO_4$ olivines are promising cathode materials***"), 1 ("From the viewpoint of electrochemical lithium storage, the ***phosphate olivines*** … have been extensively studied in the past. They ***are promising cathode materials*** for Li-ion batteries"). Indeed, that these materials are lithium ion active materials was well-known. *See* LGES-1034 (Nakura), [0004] ("As an alternative positive electrode active material to lithium cobaltate, lithium phosphates with an olivine structure are receiving attention"). This is also consistent with the disclosure of the '909 Patent, which identifies a "lithium metal salt with an olivine crystal structure" as an example of an "ion active material[]." LGES-1001, 4:30-33.

96. Consistent with the teachings of Kavan, in the combination electrode material paste, these phosphate olivines (**lithium ion active material**) are present

in the combination composition as **crystals**: "the naked nanotubes are easily recognized between the ***individual crystals of $LiMnPO_4$*** … as well as between ***$LiFePO_4$ crystals***." LGES-1005 (Kavan), p.4; *see id.* ("the ***olivine crystals*** are typically hundreds of nm in size"), p.1 ("Carbon nanotubes form conductive composites with inorganic ***nanocrystals***").

97. As I discussed above when addressing TiO_2, I note that the process of preparing the combination composition is based on Ohata, which describes "blending an electrode material mixture containing at least the masterbatch and an electrode active material with a dispersion medium" "with a mixer." LGES-1004 (Ohata), Abstract, [0032]. Again, I do not expect this blending process to have any meaningful impact on the crystallinity of the phosphate olivine powder, *i.e.*, the phosphate olivine in the combination composition is **crystals**, as I discussed in the preceding paragraph.

98. Moreover, based on Kavan, the nanotubes in the combination electrode material paste have phosphate olivine crystals (**crystals of lithium ion active material**) **attached to their surface**. For instance, the phosphate olivine crystals are attached to the nanotube surface due at least in part to the surface oxidation of the nanotubes (*see* [1d], *infra*): "The surface oxidation of carbon nanotubes … was sometimes mentioned to improve ***the anchoring of … $LiMPO_4$***." LGES-1005 (Kavan), 2; *see id.* ("detailed understanding is desirable of

the MWNT functionalization (by surface oxidation forming carboxyls) for ***optimum attachment of MWNT towards … olivine surfaces***"), 4 ("the interaction of -COOH with phosphate surface"). For instance, the attachment between the phosphate olivine (*e.g.*, $LiFePO_4$ or $LiMnPO_4$) and the surface of the oxidized nanotubes can be a covalent bond, *e.g.*, between the carboxyl groups on the surface of the nanotubes and the transition metals (*e.g.*, Fe or Mn) in the $LiFePO_4$ or $LiMnPO_4$ particles.

[1c] wherein the discrete carbon nanotubes have an aspect ratio of 10 to 500 and

99. In the Ohata-Kavan combination, the nanotubes "have an aspect ratio determined by dividing the average length of the carbon nanotubes by the average diameter of the same of ***not less than 100***" and "preferably ***not greater than 1000***," based on the teachings of Ohata. LGES-1004 (Ohata), [0020]; *see* [0006] ("The carbon nanotubes preferably have … an aspect ratio of not less than 100"), [0009], [0010], claim 2, claims 9-10. For instance, the nanotubes of the combination can have an aspect ratio of ***50*** (LGES-1004 (Ohata), Table 2, *infra*), ***100*** (*id.*, [0030], Table 2), or ***200*** (*id.*, Table 2).

TABLE 2

	Diameter of CNT (μm)	Aspect Ratio
Ex. 10	0.01	1000
Ex. 11	0.05	200

TABLE 2-continued

	Diameter of CNT (μm)	Aspect Ratio
Ex. 12	0.05	50
Ex. 13	0.1	100
Ex. 14	0.1	50

CNT: Carbon nanotube

"aspect ratio of 10 to 500"

Excerpt from Ohata, Table 2

100. In the combination, the specific aspect ratios of ***50***, ***100***, and/or ***200*** (*see* LGES-1004 (Ohata), [0030], Table 2) for nanotubes of the Ohata-Kavan combination fall within the recited range of aspect ratios (**10 to 500**). I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Ohata, I conclude that the Ohata-Kavan combination renders obvious that the nanotubes in the combination composition have **an aspect ratio of 10 to 500**.

101. Alternatively/additionally, the range of aspect ratios ("***not less than 100***" and "***not greater than 1000***") for the nanotubes of the Ohata-Kavan combination overlaps with the recited range of aspect ratios (**10 to 500**). I understand that unless there is evidence of criticality or unexpected results associated with a claimed range, that claimed range is rendered obvious by an

overlapping range in the prior art. I thus conclude that the overlapping range of ratios of the nanotubes of the Ohata-Kavan combination renders obvious the claimed range.

102. I note that the nanotubes in the combination are oxidized, as I discuss in [1d]. Oxidation of nanotubes can sometimes shorten their length. Even to the extent that the nanotubes in the combination are shortened slightly by oxidation, it is reasonable to expect the range of these "shortened" nanotubes still to fall within the claimed range of 10-500.

[1d] oxidation levels from 1% to 15% by weight of the carbon nanotube.

103. The carbon nanotubes in the combination composition are **oxidized** based on the teachings of Kavan. For instance, the nanotubes are "***functionalized by oxidation*** with HNO_3." LGES-1005 (Kavan), 2; *see id.* ("For ***oxidative functionalization***, 1 g of purified nanotubes was mixed with 50 mL of 65% HNO_3 and refluxed for 3 h at 120ºC under stirring"). The result of this oxidation process is "nanotubes functionalized by carboxylic groups." *Id.*, Abstract; *see* 4 ("MWNT-ox exhibits the presence of a new [FTIR] band at 1710cm^{-1} that is attributed to the C=O vibration of the carboxylic groups introduced by oxidative functionalization"), 9 ("multi-walled carbon nanotubes … functionalized by carboxyls"), 2 ("The surface oxidation of carbon nanotubes by HNO_3, H_2SO_4 or H_2O_2, producing among other groups also carboxyls"), 2 ("detailed understanding

of the MWNT functionalization (by surface oxidation forming carboxyls) for optimum attachment of MWNT towards titania or olivine surfaces"), 2 ("simple oxidation of MWNT is an attractive and cheap alternative to our 'nanotube wiring' effect for practical Li-ion batteries"). These oxidized nanotubes are used in preparation of the masterbatch based on Ohata and thus are present in the combination composition.

104. The oxidized nanotubes in the combination composition, functionalized as described by Kavan (*see* LGES-1005 (Kavan), 2), have 7% "atomic oxygen present" on their surface as "analyzed by X-ray photoelectron spectroscopy." *Id.*, 4; *see* 2. From these results, we assume an atomic ratio of carbon to oxygen of 93:7. This assumes that all oxygen detected by XPS represents functional groups covalently attached to the nanotubes, *i.e.*, that the 7 at.% detected by XPS does not include adsorbed H_2O or O_2.

105. As I explained above in my technical overview section, one way that the **oxidation level by weight of the nanotubes** can be determined is by calculating the weight of the oxygen atoms attached to the nanotubes divided by

the total weight of the oxidized nanotubes.[2] In 100 moles atoms of functionalized nanotubes, there are 7 mol (*i.e.*, 7 at.%) oxygen atoms, at 16 g/mol atomic weight; and 93 mol (*i.e.*, 93 at.%) carbon atoms, at 12 g/mol atomic weight. With this method, the oxidation level of Kavan's carbon nanotubes thus can be calculated as follows:

$$\frac{Mass\ of\ oxygen}{Mass\ of\ carbon + mass\ of\ oxygen} * 100$$

$$= \frac{7\ mol\ (16\ g/mol)}{93\ mol\ (12\ g/mol) + 7\ mol\ (16\ g/mol)} * 100$$

= 9.1 wt.% oxygen.

In this regard, Kavan's 7 atomic percent oxygen corresponds to an **oxidation level** of **9.1% by weight of the carbon nanotube**, *e.g.*, 9.1% by weight of the oxidized carbon nanotubes is oxygen.

106. I note that Kavan's titration results indicate that "[t]he amount of acid groups in the nanotubes was found to be 2.54 x 10^{-4} mol g^{-1} in MWNT-ox." LGES-1005 (Kavan), 2. Titration measures the presence of carboxyl groups. As I have discussed above, oxidized nanotubes can include other functional groups

[2] The '909 Patent states that "[t]he term … '% by weight' as used herein is calculated by dividing the weight of a reagent by the total weight of a mixture to which it is added subsequent to the addition of the reagent." LGES-1001, 3:59-62.

beyond carboxyls, *e.g.*, including hydroxyls and carbonyls. Thus, while titration alone can be used as a measure of the concentration of carboxyl groups in a sample of functionalized nanotubes, it is not sufficient to provide a complete characterization of the oxidation level of the nanotubes.

107. This oxidation level value (9.1% by weight) (*see* LGES-1005 (Kavan), 4) for nanotubes of the Ohata-Kavan combination falls within the recited range of oxidation levels (**1% to 15% by weight**). I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Kavan, I conclude that this specific oxidation level disclosed by Kavan, and applied to the nanotubes of the combination, renders obvious a composition in which the nanotubes have **oxidation levels from 1% to 15% by weight of the carbon nanotube**.

(ii) Claim 2

[2] The composition of claim 1, wherein the crystals or layers of lithium ion active material comprise tin, silicon, copper, antimony, aluminum, germanium, titanium, or mixtures thereof.

108. When the composition of the Ohata-Kavan combination is a negative electrode material, the lithium ion active material is $\boldsymbol{TiO_2}$, *i.e.*, the crystals or layers of lithium ion active material comprise **titanium**. *See* my discussion of element [1b], *supra*; *see* LGES-1005 (Kavan), Abstract, 2, 3, 5, 9. I note also that Ohata

indicates that negative electrode active materials can include tin or silicon. *See* LGES-1004 (Ohata), [0027] ("As the negative electrode active material for a lithium secondary battery, … [a]n alloy material such as an Si—Ni alloy or an Sn—Ni alloy can also be used").

(iii) Claim 5

[5] The composition of claim 1, wherein the carbon nanotubes are present in a weight percentage of 0.1% to 10% by weight of the lithium ion active material.[3]

109. In the Ohata-Kavan combination composition, the weight percentage of the carbon nanotubes is specified by Ohata, which teaches that "***[t]he amount of the carbon nanotubes*** contained in the electrode material mixture is, for example, ***0.2 to 3 parts by weight per 100 parts by weight of the electrode active material***. In order to obtain a high-capacity electrode, the amount thereof is preferably ***not greater than 0.8 parts by weight***." LGES-1004 (Ohata), [0024]. Notably, this range of "0.2 to 3 parts by weight" applies to both negative electrode active materials (*see id.*, [0008], [0010], claim 5) and positive electrode active materials (*see id.*, [0007], [0009], claim 4).

110. Ohata's weight percentage range (0.2 to 3 parts by weight carbon nanotubes per 100 parts by weight electrode active material) corresponds to the

[3] I have applied prior art against this limitation as I best understandd it.

carbon nanotubes being present in a weight percentage of 0.2% (0.2 parts nanotubes per 100 parts lithium ion active material) to 3.0% (3:100) by weight of the lithium ion active material. Alternatively/additionally, to the extent the weight percentage is based on the total weight of both nanotubes and lithium ion active material,[4] Ohata's weight percentage range corresponds to the carbon nanotubes being present in a weight percentage of 0.2% (0.2 parts nanotubes per 100.2 parts total) to 2.9% (3:103) by weight.

111. This range of weight percentages disclosed by Ohata (***0.2% to 3.0%***) falls within the claimed **0.1% to 10% by weight** range recited in claim 5. I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Ohata, I conclude that the Ohata-Kavan combination thus renders obvious the claimed range of weight percentages. And even if fewer than all of the Ohata-Kavan nanotubes are discrete nanotubes, a POSITA still would have found it obvious that the weight percentage of discrete nanotubes would be generally

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[4] The '909 Patent states that "[t]he term 'weight percent' or 'percent weight' or '% by weight' as used herein is calculated by dividing the weight of a reagent by the total weight of a mixture to which it is added subsequent to the addition of the reagent." LGES-1001, 3:59-62.

consistent with Ohata's 0.2-3% range, *e.g.*, because given the oxidation level of the nanotubes, a substantial portion of the nanotubes are likely to be discrete.

112. Ohata also provides specific examples of "[p]ositive electrodes" in which "the content of the carbon nanotubes contained in the masterbatch and the amount of the carbon nanotubes or the masterbatch per 100 parts by weight of the positive electrode active material contained in the positive electrode material mixture paste were changed as shown in Table 1." LGES-1004 (Ohata), [0040]. Each of the example weight percentages given in Table 1 (highlighted in purple) falls within the claimed **0.1% to 10% by weight** range recited in claim 5.

TABLE 1

	Amount of CNT Per 100 parts by weight of active material (parts by weight)	Amount of MB per 100 parts by weight of active material (parts by weight)	CNT content in MB (wt %)
Ex. 1	0.5	3.3	15
Ex. 2	0.2	1.3	15
Ex. 3	0.3	2	15
Ex. 4	0.8	5.3	15
Ex. 5	1.0	6.7	15
Ex. 6	1.5	7.5	20
Ex. 7	2.0	10	20
Ex. 8	3.0	15	20

CNT: Carbon nanotube
MB: Masterbatch

113. Specifically, I have mapped the CNT parts by weight for each of Examples 1-8 to a percentage by weight of the lithium ion active material in the

following table. Each of these weight percentage values, for either calculation approach, falls within the range recited in the claim.

	Amount of CNT per 100 parts by weight of active material (from Ohata, Table 1)	Weight of CNTs to weight of lithium ion active material (%)	Weight of CNTs to weight of CNTs and lithium ion active material together (%)
Ex. 1	0.5	0.5% (0.5/100)	0.5% (0.5/100.5)
Ex. 2	0.2	0.2% (0.2/100)	0.2% (0.2/100.2)
Ex. 3	0.3	0.3% (0.3/100)	0.3% (0.3/100.3)
Ex. 4	0.8	0.8% (0.8/100)	0.8% (0.8/100.8)
Ex. 5	1.0	1.0% (1.0/100)	1.0% (1/101)
Ex. 6	1.5	1.5% (1.5/100)	1.5% (1.5/101.5)
Ex. 7	2.0	2.0% (2.0/100)	2.0% (2/102)
Ex. 8	3.0	3.0% (3.0/100)	2.9% (3/103)

114. Furthermore, Ohata provides a specific example in which "[a] negative electrode material mixture paste was produced by mixing 3.3 parts by weight of masterbatch, the same as the one used in the production of the positive electrode in Example 1, 100 parts by weight of flake graphite as an active material

and 56 parts by weight of N-methyl-2-pyrrolidone as a dispersion medium with a mixer." [0046]. The masterbatch has "[f]ifteen parts by weight of carbon nanotubes and 85 parts by weight of polyvinylidene fluoride." *Id.*, [0030]. Thus, this example has a weight percentage of 0.5% weight percentage of carbon nanotubes by weight of the lithium ion active material (3.3 parts by weight masterbatch to 100 parts active material, where 15% by weight of the masterbatch is carbon nanotubes). This value also falls within the claimed **0.1% to 10% by weight** range recited in claim 5.

(iv) Claim 6

[6a] The composition of claim 1, comprising oxidized carbon nanotubes having crystals or layers of lithium ion active materials attached to their surface,

115. The combination composition includes oxidized (as I discussed for element [1d], *supra*) carbon nanotubes (as I discussed for element [1a], *supra*) having crystals or layers of lithium ion active materials attached to their surface (as I discussed for element [1b], *supra*).

[6b] wherein the carbon nanotubes are uniformly dispersed without clustering on a dimension scale less than 1 micrometer.

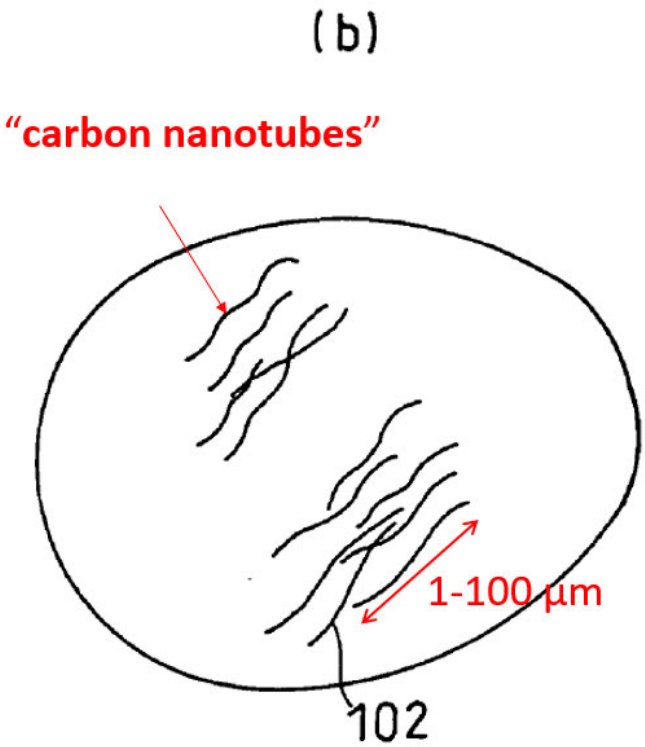
116. Ohata describes that in a masterbatch process, "a conductive material is ***uniformly predispersed*** in a binder." LGES-1004 (Ohata), [0002]. In the combination, this "conductive material" is the oxidized carbon nanotubes, and via

Ohata's masterbatch process, the nanotubes are "***uniformly predispersed***" in the resin. *Id.*, [0002]; *see* [0005] ("producing a masterbatch comprising at least carbon nanotubes and a resin"), [0015] ("producing a masterbatch by blending at least carbon nanotubes with a resin"). This masterbatch, with "uniformly predispersed" nanotubes, is then mixed with an electrode active material and a dispersion medium to form the electrode material mixture paste of the combination. LGES-1004 (Ohata), [0002]; *see* [0023].

117. It is my opinion that a POSITA would have understood, or at least found obvious, that the nanotubes in the Ohata-Kavan combination remain **uniformly dispersed** in the combination paste. For instance, a POSITA would have recognized that the combination manufacturing process based on Ohata, in which the masterbatch is blended with "an electrode active material" and "a dispersion medium" to form an electrode material mixture paste (LGES-1004 (Ohata), [0023]), would allow the nanotubes in the resulting paste to remain **uniformly dispersed**, as they are in the masterbatch.

118. Moreover, a POSITA would have understood, or at least found obvious, that the nanotubes are dispersed **without clustering on a dimension scale less than 1 micrometer**. Ohata's Fig. 1(b), again *infra*, which a POSITA would have understood as representative of the nanotube arrangement in the combination paste, as discussed in the previous paragraph, shows that the

nanotubes are dispersed **without clustering**, *e.g.*, in that the nanotubes are present as generally individual nanotubes. Because there is substantially no clustering, there is also no clustering on a dimension scale of less than 1 micrometer. Moreover, although no scale is provided for the Fig. 1(b) schematic illustration, a POSITA would have been able to estimate the scale based on Ohata's disclosure that the nanotubes "have an average length of 1 to 100 μm." LGES-1004 (Ohata), [0019]. For instance, for nanotubes having a length of 100 μm, the space between the nanotubes is depicted as greater than 1 μm, *e.g.*, the nanotubes are dispersed **without clustering on a dimension scale less than 1 micrometer**. Alternatively/additionally, as confirmed by Ohata's disclosure that the nanotubes are "dispersed," a POSITA further would have understood that the nanotubes in the Ohata-Kavan composition do not form clusters of greater than 1 micrometer in size, *e.g.*, the nanotubes are dispersed **without clustering on a dimension scale less than 1 micrometer**.



Excerpt from Ohata, Fig. 1(b) (annotated)

(v) Claim 7

[7] The composition of claim 1, comprising uniformly dispersible oxidized carbon nanotubes having crystals or layers of lithium ion active materials attached to their surface.

119. The combination composition includes uniformly dispersed, oxidized carbon nanotubes having crystals or layers of lithium ion active materials attached to their surface. *See* claim [6], *supra*, which addresses that the nanotubes are uniformly dispersed oxidized carbon nanotubes having crystals or layers of lithium ion active materials attached to their surface.

120. Moreover, the nanotubes in the Ohata-Kavan combination paste are **dispersible**. The combination composition is a paste (based on Ohata's electrode

material mixture paste) that is "prepared by blending an electrode material mixture containing the masterbatch and an electrode active material with a dispersion medium." LGES-1004 (Ohata), [0023]. That is, the masterbatch (containing the dispersed nanotubes) and the electrode active material are **dispersible** in the dispersion medium at least because the mixture can be blended, *i.e.*, the nanotubes are **dispersible**.

121. I note that some applications of nanotubes rely on vertically aligned arrangements of nanotubes in which nanotubes are grown with an end attached to a substrate. The nanotubes in the Ohata-Kavan combination paste are alternatively/additionally **dispersible** because they are not attached to a substrate in a vertically aligned arrangement.

(vi) Claim 8

[8] The composition of claim 7, wherein the crystals or layers of lithium ion active materials are chemically attached to the surface of the dispersible oxidized carbon nanotubes.

122. In the combination, Kavan describes an "***interaction of carboxyls with titania***." LGES-1005 (Kavan), 4. The carboxyls are among the functional groups on the surface of the **dispersible oxidized carbon nanotubes**, and the titania corresponds to the **crystals of lithium ion active material**. *See* [1b], [1d], *supra*. This "interaction of carboxyls with titania" is an example of a **chemical attachment** of the lithium ion active material to the surface of the nanotubes, *e.g.*,

in that it is a chemical interaction between the carboxyl groups and the titania. *Id.*, 4; *see* Abstract ("Multi-walled carbon nanotubes functionalized by carboxylic groups, exhibit better affinity towards TiO_2 … as compared to that of pristine nanotubes"), p.4 ("oxidative functionalization … is crucial for ***proper anchoring of nanotubes towards TiO_2***"). For instance, the interaction between carboxyl groups and titania can be a covalent bond between the TiO_2 and the oxygen-containing functionalities on the nanotube surface (*e.g.*, the lithium ion active material is **chemically attached** to the nanotubes by way of covalent bonds), as I explained above for element [1b]. *See* LGES-1030 (X. Li), Abstract ("The results indicated that TiO_2 nanoparticles were chemically absorbed at SWNT defect sites via an ester-type linkage between carboxylic acid groups at the defect sites and hydroxyl groups at the surface of the TiO_2 nanoparticles"); LGES-1037 (J.Li), 4 ("carboxylates groups on the CNTs will provide bonding to the TiO_2 … nanoparticles").

123. I also discussed above for element [1b] that the TiO_2 can be attached to the nanotube surface through hydrogen bonding and/or Van der Waals forces. LGES-1037 (J. Li), 4. To the extent either of these mechanisms is considered a chemical attachment, the lithium ion active material is **chemically attached** to the nanotubes by way of hydrogen bonds / Van der Waals forces.

124. Alternatively/additionally, the interaction between carboxyl groups and titania can be a covalent bond between the phosphate olivine and the oxygen-containing functionalities on the nanotube surface (*e.g.*, the lithium ion active material is chemically attached to the nanotubes by way of covalent bonds), as I explained above for element [1b]. *See* LGES-1005 (Kavan), 2, 4.

B. [GROUND 2A] – CLAIMS 1-2 AND 5-8 ARE OBVIOUS OVER LEE-WEPASNICK

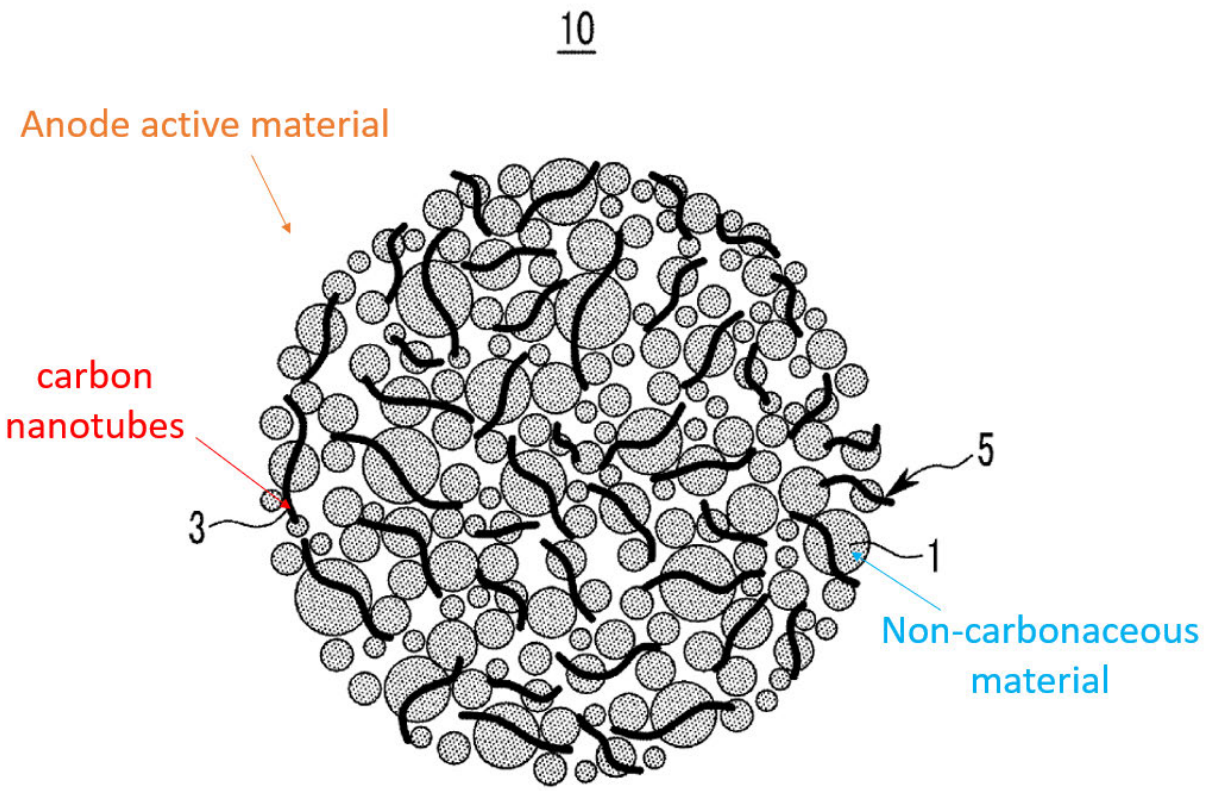
125. It is my opinion that a POSITA would have found claims 1-2 and 5-8 to be obvious over the combination of Lee and Wepasnick, as I discuss in the following sections.

1. Lee describes an anode active material including carbon nanotubes

126. Lee describes "an anode active material" for a lithium ion battery that "exhibit[s] improved cycle lifetime characteristics." LGES-1006 (Lee), [0010].

127. FIG. 1, *infra*, illustrates an example of such an "anode active material," which includes "secondary particles that are formed by the agglomeration of primary nanoparticles 5." LGES-1006 (Lee), [0040]. The primary nanoparticles "include a non-carbonaceous material 1" (blue) "and hollow carbon nanofibers 3" (red) "which are bound to the non-carbonaceous material by Van der Waals forces." *Id.*, [0040]. The non-carbonaceous material is "a silicon-based material" such as silicon or a silicon-metal alloy that acts as lithium ion

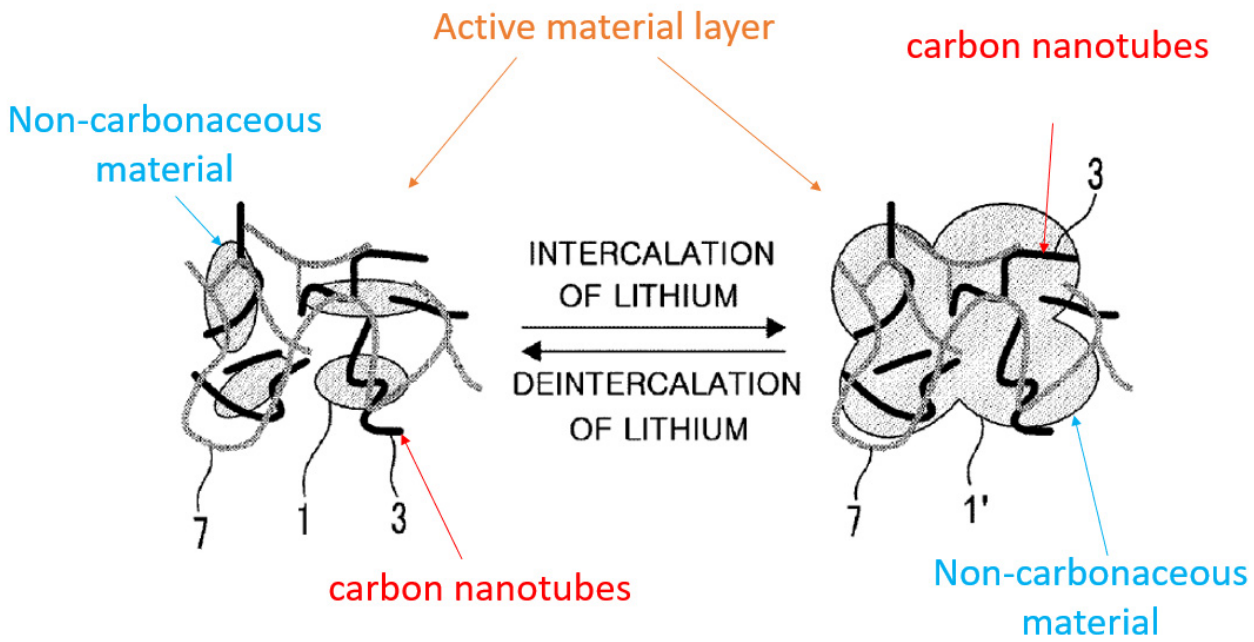
active material. *Id.*, [0043]. The "hollow carbon nanofibers" are "carbon nanotubes." *Id.*, [0046].



Except from Lee, FIG. 1 (annotated)

128. In the anode active material, "the non-carbonaceous material 1 is bound to the hollow carbon nanofibers to form a composite material" that has "a higher electrical conductivity," *e.g.*, to obtain the "higher capacity" of non-carbonaceous lithium ion active materials while compensating for their "lower electrical conductivity" as compared to "conventional carbonaceous anode materials." LGES-1006 (Lee), [0042]-[0043].

129. The anode active material, a polymeric binder, and a solvent are combined "to prepare an anode active material composition" such as a slurry. LGES-1006 (Lee), [0064]; *see* [0085]. This slurry "is coated on a current collector and dried," forming an active material layer of an anode." *Id.*, [0065]; *see* [0049]. The structure of the active material layer of the anode is shown in FIG. 3, *infra*. As illustrated, "a composite of Li and the non-carbonaceous material 1 [] is generated during charging of the anode," denoted by "reference numeral 1′." *Id.*, [0051].



Excerpt from Lee, FIG. 3

130. In Lee's anode active material composition, the polymeric binder has "an electron donor group" that "aids the dispersal of the hollow carbon nanofibers 3" and allows for "the hollow carbon nanofibers 3 [to] be bound with the binder 7,

by charge transfer bonding." *Id.*, [0052]-[0053]. This bonding between nanofibers and binder "enhances interparticle cohesion," such that "even if the volume of the non-carbonaceous material 1 changes, due to the intercalation/de-intercalation of lithium, the structure of the anode active material 10 can be maintained," and thus "the cycle lifetime characteristics may be improved." *Id.*, [0053]; *see* [0054], [0058]. For instance, Lee explains that when "conventional anodes include a non-carbonaceous material, such as Si," are used, intercalation of lithium can "lead[] to abrupt volumetric changes in the crystalline lattice of Si, thereby fracturing the conventional active material," and thus "the discharge capacity markedly decreases." *Id.*, [0059]. In Lee's anode active material layer, by contrast, the binding between the binder and the nanofibers, and the binding between nanoparticles, allows "the structure of the active material layer … [to] be maintained during charging and discharging." LGES-1006 (Lee), [0058].

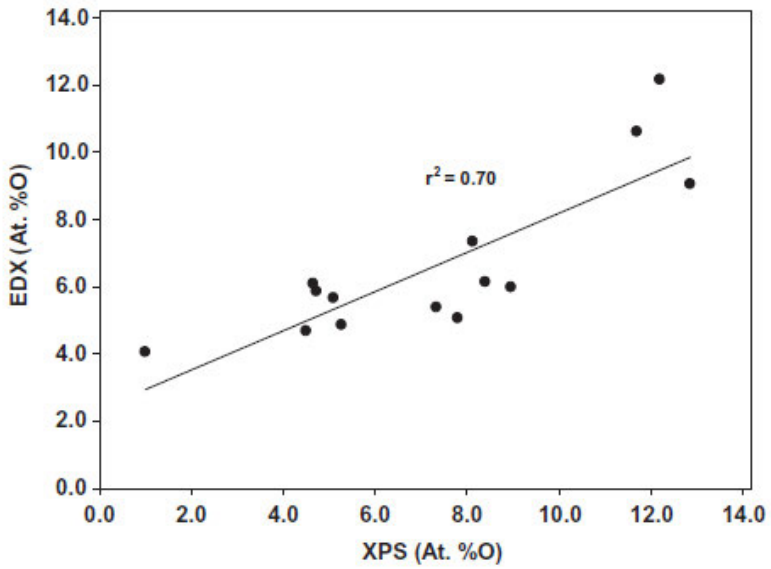
131. Lee's anode active material composition may also contain a conducting agent, which can be "any conducting agent commonly used in lithium batteries," *e.g.*, "carbonaceous materials … metal-based materials … and materials including conductive polymers." *Id.*, [0057]; *see* [0064].

2. Wepasnick characterizes oxidation levels of carbon nanotubes treated by various conventional oxidation treatments

132. Wepasnick is an academic study of the effect of various "commonly used wet chemical oxidants" on "the surface chemistry and structure of MWCNTs." LGES-1010 (Wepasnick), Abstract. Wepasnick explains that the importance of its study is that "knowledge of the concentration and distribution of different oxygen functional groups on the CNT surface following wet chemical oxidative treatments would allow researchers to better control the ultimate structure and properties of the CNT-based materials." *Id.*, 2. For instance, "the surface of CNTs used in reinforced polymer composites are often modified covalently by the addition of polar functional groups, not only to improve dispersion properties but also to enhance chemical interactions with the resin matrix," and this covalent modification is performed by "deliberately grafting oxygen-containing functional groups at the open ends and sidewalls of CNTs." *Id.*; *see* 2 ("In functionalization strategies, the role of oxidation is to create 'foothold' functional groups, such as carboxyl groups, which then serve as attachment points to anchor either larger biomolecular structures or nanoparticles").

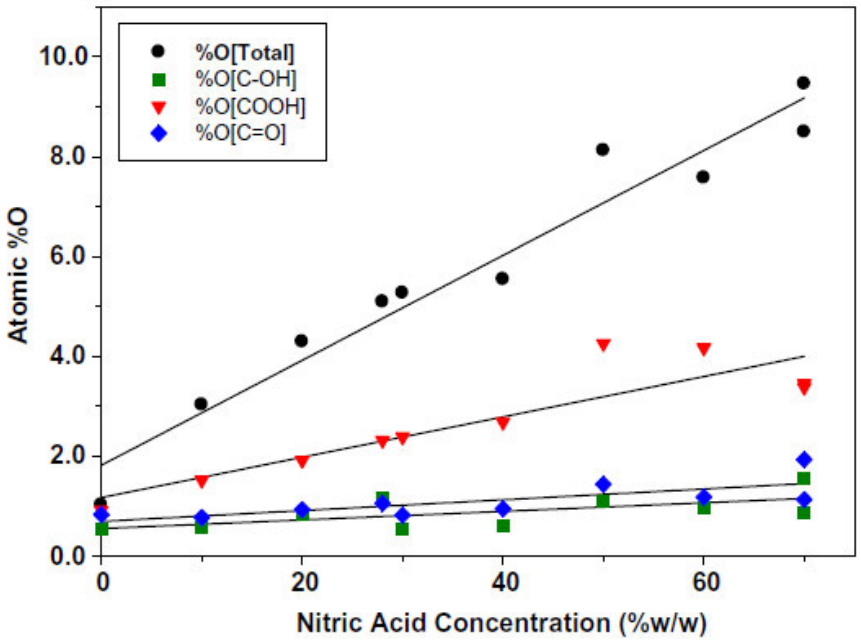
133. Wepasnick's study investigated "the effects of six wet chemical oxidants (HNO_3, H_2SO_4/HNO_3, $KMnO_4$, H_2O_2, O_3, and $(NH_4)_2S_2O_8$) used under

different reaction conditions (e.g., w/w%)." LGES-1010 (Wepasnick), 2-3. The resulting oxidized multiwall nanotubes were characterized by X-ray photoelectron spectroscopy (XPS), energy dispersive X-ray spectroscopy (EDX), and derivatizing reactions to quantify the atomic concentration of oxygen and the concentrations of carboxylic acid, hydroxyl, and carbonyl groups. *Id.*, 3-4. As shown in Wepasnick's Fig. 5, *infra*, "XPS and EDX results are reasonably well correlated … demonstrating that either technique can provide a measure of oxygen incorporation," with both techniques providing results in "close proximity," but EDX identifying a slightly higher atomic percentage of oxygen than XPS for a given sample. *Id.*, 6. The atomic percent oxygen values Wepasnick determines from XPS experiments are determined as the amount of oxygen divided by the amount of carbon and oxygen, *e.g.*, as indicated in Fig. 7. *Id.*, 8.



Excerpt from Wepasnick, Fig. 5

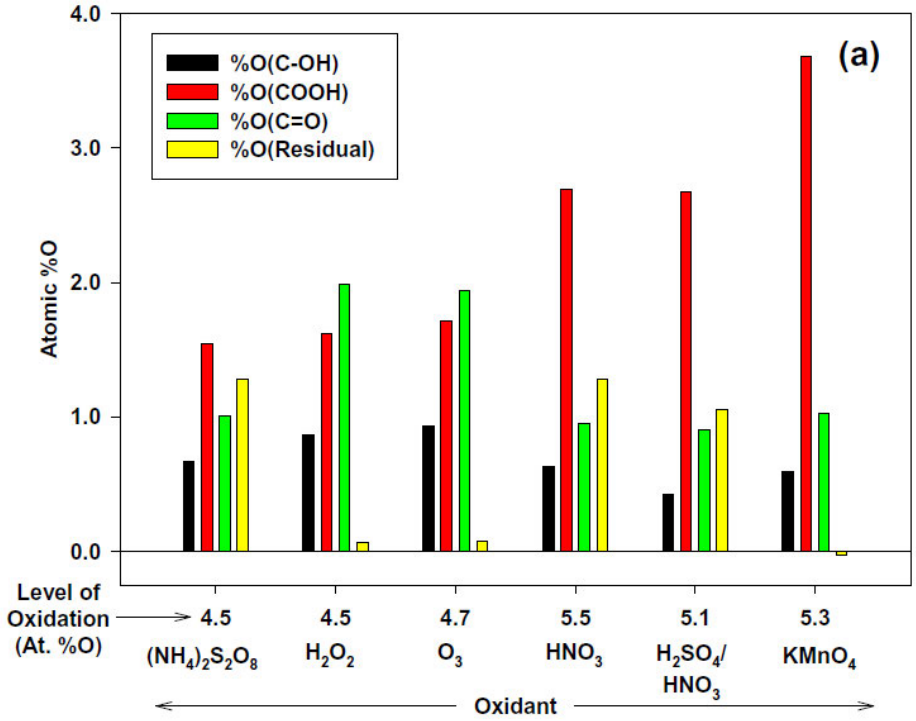
134. Wepasnick's results demonstrate that the identity of the oxidant and the concentration of the oxidant affect both the oxidation levels of the nanotubes and the distribution of the oxygen-containing functional groups on the nanotubes. *See* LGES-1010 (Wepasnick), 8-10. For instance, Wepasnick's study reveals that nanotubes oxidized in different concentrations of nitric acid have oxidation levels ranging from 1.0 atomic % (at.%) to 9.5 at.% (*see* LGES-1010 (Wepasnick), Fig. 6), and different distributions of hydroxyl (C-OH), carboxyl (COOH), and carbonyl (C=O) functional groups (*see id.*, Fig. 7, *infra*). LGES-1010 (Wepasnick), 8.



Excerpt from Wepasnick, Fig. 7

135. Similarly, as illustrated in Wepasnick's Fig. 9(a), also *infra*, oxidation levels ranging from 4.5 at.% to 5.5 at.%, and different distributions of functional

groups, can be obtained depending on the type of oxidant used. LGES-1010 (Wepasnick), 10.



Excerpt from Wepasnick, Fig. 9(a)

136. Structural differences were also observed between nanotubes oxidized with different treatments. *See* LGES-1010 (Wepasnick), 11. For instance, in H_2O_2-treated nanotubes, "the overall level of amorphous carbon was reduced and a few defects were generated in the sidewall," and $KMnO_4$-treated nanotubes had "a large[] fraction of COOH groups" and "a relatively low amount of sidewall damage." LGES-1010 (Wepasnick), 11. Conversely, treatment with H_2SO_4/HNO3 "produced a distortion in the linearity of the MWCNT structure" due to damage extending "into the underlying sidewalls." *Id.*

3. The Lee-Wepasnick combination

137. In the Lee-Wepasnick combination, an anode active material includes carbon nanotubes and a non-carbonaceous material, as described by Lee. The carbon nanotubes are oxidized using one of the "commonly used wet chemical oxidants," as taught by Wepasnick, thereby functionalizing the surface of the nanotubes with "carboxyl, carbonyl, and hydroxyl groups." LGES-1010 (Wepasnick), Abstract. Notably, Wepasnick characterizes the oxidation levels resulting from treatment with each of six different oxidants, under various conditions. *See* LGES-1010 (Wepasnick), Abstract. In the Lee-Wepasnick combination, a POSITA would have chosen an oxidative treatment based on these data to achieve a desired "concentration and distribution of different oxygen functional groups on the CNT surface," thereby allowing the POSITA "to better control the ultimate structure and properties" of the resulting combination anode active material, as discussed *infra*. LGES-1010 (Wepasnick), 2.

138. The oxidized nanotubes are mixed with a non-carbonaceous material "in an organic solvent," and "[t]he resultant is then dried to prepare an anode active material," as taught by Lee. LGES-1006 (Lee), [0060]. In the combination anode active material, the presence of functional groups on the nanotubes facilitates dispersion of the nanotubes and supplements the Van der Waals binding between the non-carbonaceous material and the nanotubes, enhancing the structural stability

and potentially cycle lifetime of the anode active material. *See* LGES-1006 (Lee), [0040] ("The primary nanoparticles 5 include a non-carbonaceous material 1 and hollow carbon nanofibers 3, which are bound to the non-carbonaceous material by Van der Waals forces"), [0044] ("the non-carbonaceous material can bind with the hollow carbon nanofibers 3, which provides excellent cycle lifetime characteristics"); LGES-1010 (Wepasnick), 2 ("In functionalization strategies, the role of oxidation is to create 'foothold' functional groups, such as carboxyl groups, which then serve as attachment points to anchor either larger biomolecular structures or nanoparticles"). For instance, the POSITA would have chosen a particular oxidative treatment based on Wepasnick's characterization data to achieve a "concentration and distribution of different oxygen functional groups on the CNT surface" that allows for a suitable degree of binding between the nanotubes and the non-carbonaceous material. LGES-1010 (Wepasnick), 2.

139. This combination anode active material is mixed with a polymeric binder and, optionally, a conductive agent to form an anode active material composition, which is used to make an anode for a lithium ion battery, consistent with the teachings of Lee. *See* LGES-1006 (Lee), [0064]-[0065], [0085]. Here, too, the presence of oxidized nanotubes is advantageous in providing structural integrity to the anode active material formed from that composition, *e.g.*, via chemical interactions between the nanotubes and the polymeric binder, thus

providing good cycle lifetime characteristics to the composite. *See* LGES-1010 (Wepasnick), 2 ("the surface of CNTs used in reinforced polymer composites are often modified covalently by the addition of polar functional groups, not only to improve dispersion properties but also to enhance chemical interactions with the resin matrix"); LGES-1006 (Lee), [0058] ("The polymeric binder having an electron donor group may enhance the cohesion of anode 20, through the bonding with the hollow carbon nanofibers 3. Thus … the structure of the active material layer 14 can be maintained during charging and discharging").

140. For instance, the POSITA would have chosen a particular oxidative treatment based on Wepasnick's characterization data to achieve a "concentration and distribution of different oxygen functional groups on the CNT surface" that allows for a target objective to be achieved. For instance, oxidation of nanotubes impacts conductivity of the nanotubes, and a POSITA would have chosen an oxidation level that allows the nanotubes to retain a suitable amount of conductivity. Moreover, oxidation of nanotubes enhances dispersibility of the nanotubes, and a POSITA would have considered dispersion in selecting oxidation level. Additionally, a POSITA would have selected the oxidation level such that a suitable degree of binding between the nanotubes and the polymeric binder can be achieved, *e.g.*, to improve the interaction between the nanotubes, the electrode active material, and the binder. LGES-1010 (Wepasnick), 2. In a specific

example, when the polymeric binder of the combination anode active material composition contains "an amino group, an amide group, a thiol group, or a combination thereof," chemical bonding between these groups and the carboxylic acid functionalities on the nanotubes binds the nanotubes to the binder, thereby achieving the structural integrity of the material and achieving Lee's objective of "improved" "cycle lifetime characteristics." LGES-1006 (Lee), [0056], [0053].

4. Reasons for the combination

141. A POSITA would have been motivated to use oxidized nanotubes in the Lee-Wepasnick anode active material for at least the following reasons.

142. The first reason for using oxidized nanotubes is that a POSITA would have recognized the importance of the binding between nanotubes and non-carbonaceous material in achieving "higher electrical conductivity" and "excellent cycle lifetime characteristics" (*see* LGES-1006 (Lee), [0043]-[0044]), and would have been motivated to use oxidized nanotubes because the functional groups on the nanotubes would enhance this bonding. For instance, this is described by Wepasnick, which states that "[i]n functionalization strategies, the role of oxidation is to create 'foothold functional groups, such as carboxyl groups, which then serve as attachment points to anchor … nanoparticles." LGES-1010 (Wepasnick), 2. Similarly, Datsyuk explains that "[r]egulation of the amount of oxygen onto the CNT sidewalls is a prerequisite for building certain functional

nanostructures and devices." LGES-1009 (Datsyuk), 2. Moreover, in the specific context of a composite of oxidized carbon nanotubes and silicon, Eom notes that "[i]t is believed that the surface of the Si particles was oxidized by ball-milling, and the chemical-bonded SiC compound was formed on the interface of the SWCNTs and Si particles," where the nanotubes are oxidized ("effectively purified by an acidic treatment followed by the gas-phase oxidation process." LGES-1043 (Eom), 2-3.

143. The second reason for using oxidized nanotubes is that a POSITA also would have recognized the importance of the binding between nanotubes and the polymeric binder in "improving the cycle lifetime characteristics" of the anode active material composition (*see* LGES-1006 (Lee), [0054]), and similarly would have been motivated to use oxidized nanotubes because the functional groups on the nanotubes would enhance this bonding: "the surface of CNTs used in reinforced polymer composites are often modified covalently by the addition of polar functional groups … to enhance chemical interactions with the resin matrix." LGES-1010 (Wepasnick), 2.

144. The third reason for using oxidized nanotubes is that oxidative treatments can "remove amorphous carbon and metallic impurities from as-produced CNTs," thus leading to a higher-purity starting material. LGES-1010

(Wepasnick), 2; *see* 11 ("the oxidative treatments used in this study remove amorphous carbon").

145. The fourth reason for using oxidized nanotubes is that Wepasnick's characterization study demonstrates that the nature of the oxidant and the treatment conditions affect both the "concentration and distribution of different oxygen functional groups on the CNT surface," and notes these characterization results can allow for "better control [of] the ultimate structure and properties of the CNT-based materials." LGES-1010 (Wepasnick), 2, 11; *see* Table 1, Figs. 6-9. Moreover, the particular oxidative treatment also impacts "the structure of [the] MWCNTs." LGES-1010 (Wepasnick), 11; *see* Fig. 10. A POSITA would have been motivated to select a particular oxidative treatment (*e.g.*, oxidant and treatment conditions) in order to obtain an overall oxidation level and distribution of functional groups that achieves a target performance goal, while also maintaining sufficient structural integrity of the nanotubes. For instance, in the context of the Lee-Wepasnick combination, a POSITA would have been motivated to select an oxidative treatment that produces an oxidation level, and a composition of oxidative species, that facilitates both bonding between the nanotubes and the non-carbonaceous material (*see* LGES-1006 (Lee), [0043]-[0044]) and between the nanotubes and the polymeric binder (*see id.*, [0053]-[0054], [0058]-[0059]).

146. The fifth reason for using oxidized nanotubes is that doing so is just the application of known techniques (specifically, oxidized nanotubes, which are conventionally used in "polymer composites" and as part of "complex nanostructures," LGES-1010 (Wepasnick), 2) to an already-known structure (the nanotubes of Lee's anode active material), and the combination produces predictable results (specifically, an anode active material including oxidized nanotubes).

147. I note that oxidized nanotubes may have a lower conductivity than their unoxidized counterparts. However, nanotubes oxidized at levels such as those described by Wepasnick still retain high conductivity, as evidenced by the strength of C-C bond in the C(1s) XPS spectrum. *See*, *e.g.*, LGES-1010 (Wepasnick), Fig. 6. In addition, the oxidization of MWNTs happened on the surface of the MWNTs, the interior layers of CNTs are still conductive. Moreover, use of oxidized nanotubes also brings a number of performance advantages, such as those I discussed above. I expect that a POSITA would have known how to balance the trade-offs between lower conductivity and the various advantages associated with oxidized nanotubes, and it is my opinion that given the strong advantages that are directly applicable to the context of the Lee-Wepasnick combination, a POSITA would have chosen to implement oxidized nanotubes in the combination.

148. A POSITA would have expected success in using oxidized nanotubes in the Lee-Wepasnick anode active material for at least the following reasons.

149. First, the oxidation treatments studied by Wepasnick, and the oxidation levels that result from these treatments, are commonly used, and indeed Wepasnick acknowledges the conventional nature of these treatments and of their results. *See* LGES-1010 (Wepasnick), Abstract ("Six commonly used wet chemical oxidants … were evaluated"), 7 ("Among common oxidizing methods (defined by the identity of the oxidant and the reaction conditions) used to treat CNTs, we observed marked differences in the extent of MWCNT oxidation"), Table 1 ("Extent of MWCNT oxidation after common oxidative treatments"), 3 ("The experimental details of each wet chemical treatment were based upon established procedures used to either oxidize or purify CNTs"); LGES-1009 (Datsyuk), Abstract, Figs. 4-5.

150. Second, oxidized nanotubes were commonly used in the context of polymer-based composites—of which the Lee-Wepasnick anode active material composition is an example. *See* LGES-1010 (Wepasnick), 2 ("the surface of CNTs used in reinforced polymer composites are often modified covalently by the addition of polar functional groups").

151. Third, oxidized nanotubes are compatible with the composition of the compositions described by Lee. For instance, oxygen functional groups such as

carboxylic acids can bind to silicon, silicon oxide, or silicon-metal alloys, and thus a POSITA would have expected oxidized nanotubes to be capable of bonding to a non-carbonaceous material, as described by Lee. *See* LGES-1006 (Lee), [0043] ("the non-carbonaceous material 1 is bound to the hollow carbon nanofibers"), [0041] ("The non-carbonaceous material may be a silicon-based material, such as silicon (Si), a silicon oxide …, Si—Y, or a mixture thereof"). In fact, Eom demonstrates such bonding between silicon and oxidized carbon nanotubes: "[i]t is believed that the surface of the Si particles was oxidized by ball-milling, and the chemical-bonded SiC compound was formed on the interface of the SWCNTs and Si particles," where the nanotubes are oxidized ("effectively purified by an acidic treatment followed by the gas-phase oxidation process." LGES-1043 (Eom), 2-3. Moreover, oxygen functional groups such as carboxylic acids also can bind to "an amino group, an amide group, [or] a thiol group," and thus a POSITA would have expected oxidized nanotubes to be capable of bonding to the polymeric binder described by Lee, which includes one or more of these species. LGES-1006 (Lee), [0056].

152. Fourth, use of oxidized nanotubes in compositions for lithium ion battery electrodes was conventional. *See* LGES-1005 (Kavan), Abstract, 9; LGES-1032 (Sheem), [0027]-[0031] ("The active material for a rechargeable lithium battery … includes an active material and a fiber-shaped or tube-shaped carbon

conductive material," which "preferably includes carbon nano fibers or carbon nanotubes." "The fiber-shaped or tube-shaped carbon conductive material is oxidized," and "The oxidized fiber-shaped or tube-shaped carbon conductive material includes functional groups selected from the group consisting of carboxyl groups (—COOH), hydroxyl groups (—OH), carbonyl groups (—COH), and combinations thereof that are attached to the surface thereof").

5. Claim-by-Claim Analysis

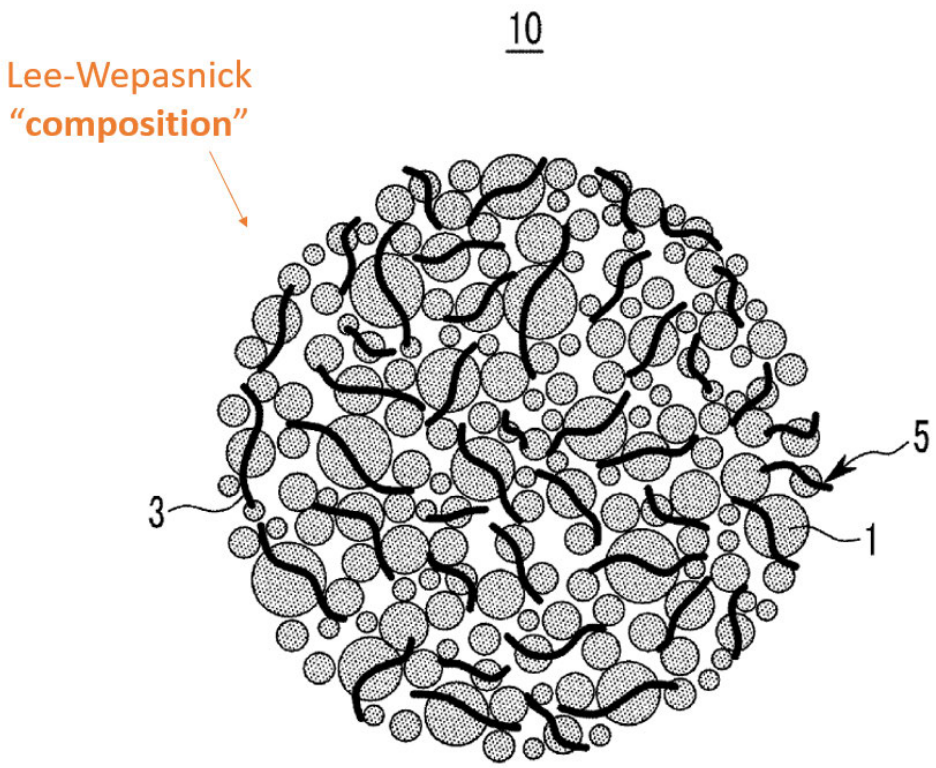
(i) Claim 1

[1pre] A composition useful for lithium ion batteries comprising:

153. To the extent the preamble is deemed to be limiting, Lee-Wepasnick renders it obvious.

In the combination, an anode active material based on Lee is a composition

154. The Lee-Wepasnick combination renders obvious a **composition** based on Lee's "anode active material." LGES-1006 (Lee), Abstract; *see* [0003], [0010]-[0016], FIG. 1 (*infra*).



Except from Lee, FIG. 1 (annotated)

155. The combination **composition** includes "secondary particles that include agglomerated primary nanoparticles, wherein the primary nanoparticles include a non-carbonaceous material that is bound to hollow carbon nanofibers," forming "a composite material." *Id.*, [0014], [0043]; *see* [0060] ("a non-carbonaceous material and hollow carbon nanofibers are mixed in an organic solvent, using a milling process. The resultant is then dried to prepare an anode active material"), FIG. 4 ("mix non-carbonaceous material and hollow carbon fibers in organic solvent using milling to prepare anode active material"), [0084] ("The resulting anode active material included secondary particles that were formed by the agglomeration of primary nanoparticles including Si particles bound

to the carbon nanotubes"), [0015]-[0016], [0039]-[0040], [0063], [0087], claim 1, claim 6, claim 12.

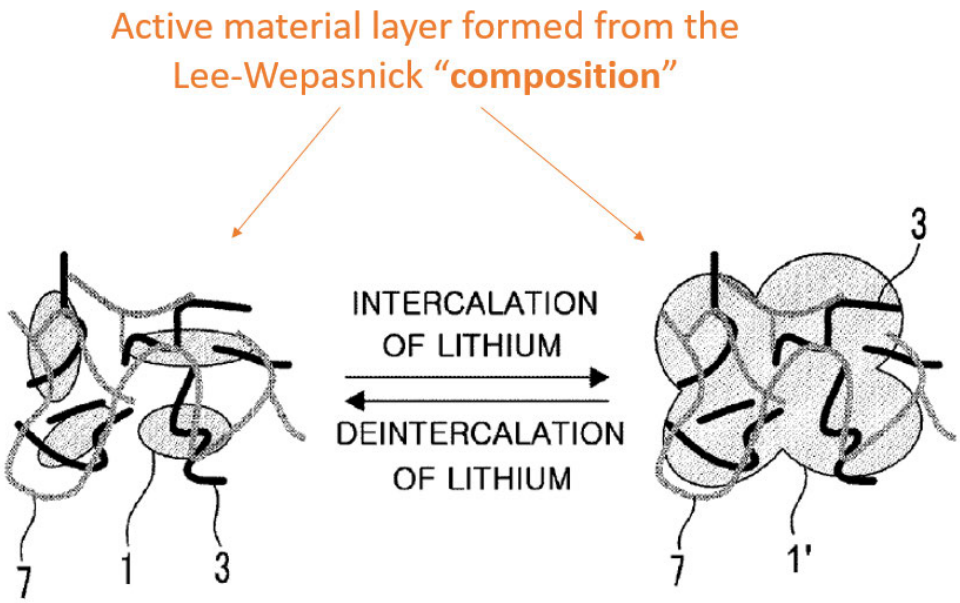
156. The hollow carbon nanofibers (*e.g.*, carbon nanotubes (*see* [1a], *infra*)) in the combination anode active material are oxidized based on the teachings of Wepasnick, as discussed in [1d], *infra*.

In the combination, an anode active material composition based on Lee is a composition

157. Alternatively/additionally, the Lee-Wepasnick combination renders obvious a **composition** based on Lee's "anode active material composition." LGES-1006 (Lee), [0016]. This combination anode active material composition includes "the anode active material" discussed *supra*, "a binder, and a solvent." LGES-1006 (Lee), [0016]; *see* [0064] ("the anode active material and the polymeric binder are mixed in a solvent, to prepare an anode active material composition"), FIG. 4 ("mix anode active material, binder, and conducting agent in solvent to prepare anode active material composition"), [0085] ("Next, the anode active material, artificial graphite as a conducting agent, and polyethyleneimine as a binder were mixed in water, to prepare an anode active material composition in slurry form"), claim 12 ("mixing the anode active material, a binder, and a solvent to prepare an anode active material composition"), claim 18 ("An anode active material composition comprising: secondary particles comprising agglomerated primary nanoparticles, the primary nanoparticles comprising hollow carbon fibers

attached to a silicon-based material; and a polymeric binder having an electron donor group").

158. The anode active material composition of the combination can be coated onto a current collector and dried to form an anode. *See* LGES-1006 (Lee), [0016], [0065] ("The anode active material composition is coated on a current collector and dried, to manufacture an anode"), FIG. 4 ("coat anode active material composition on current collector and dry the composition in vacuum"), [0086] ("The anode active material composition was coated on a copper current collector and dried … to manufacture an anode"), [0003], [0087], claim 12. This process results in "an active material layer … formed on the current collector." *Id.*, [0049], claim 6, FIG. 2. Lee's FIG. 3, *infra*, illustrates "the structure of the active material layer" that is formed from the anode active material composition. *Id.*, [0051]; *see* [0054].



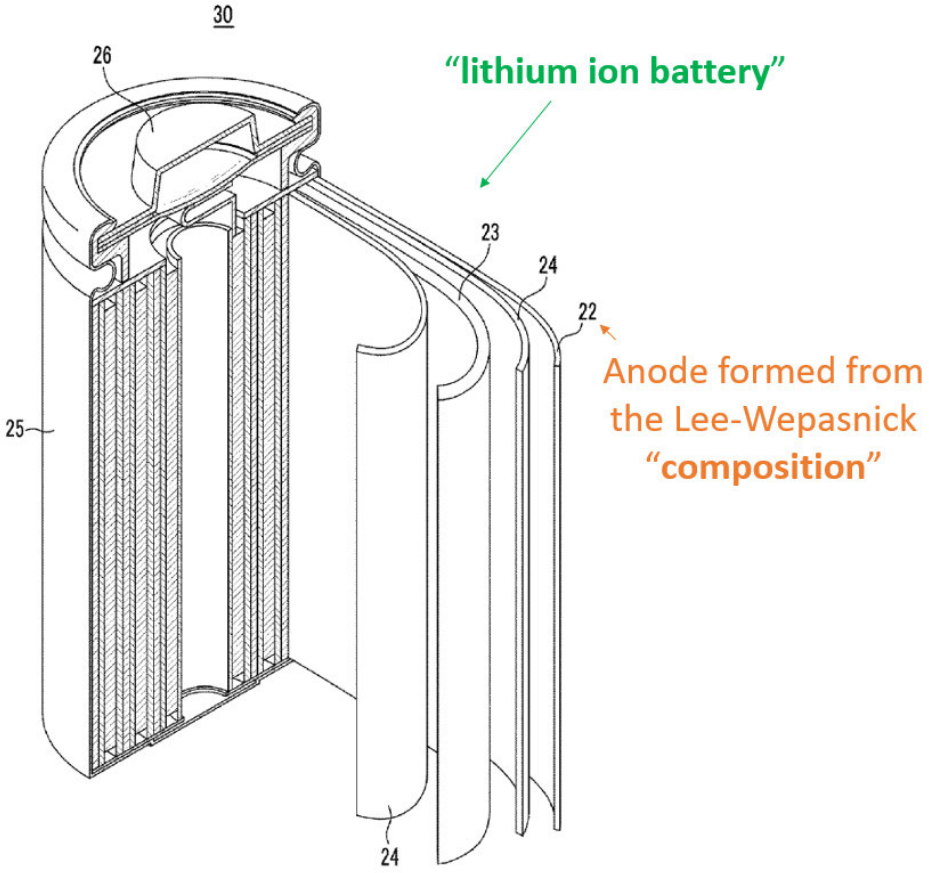
Excerpt from Lee, FIG. 3 (annotated)

The composition of the combination is useful for lithium ion batteries

159. The Lee-Wepasnick combination composition (*e.g.*, either the combination anode active material or the combination anode active material composition) is **useful for lithium ion batteries**, as described by Lee. Specifically, based on Lee, the combination anode active material (**the composition**) is incorporated into an anode active material composition (alternatively/additionally **the composition**), which is "coated onto a current collector … to manufacture an anode." *Id.*, [0065]; *see* [0011] ("***an anode including the anode active material***"), [0058] ("The anode 20 includes the anode active material 20, and thus, exhibits improved electrical conductivity"), [0003], [0016], [0049], [0086], claim 6, claim 12, FIG. 2. The anode (and thus the anode active material and anode active material composition) is an anode of **a lithium**

ion battery. *See* LGES-1006 (Lee), [0013] ("***a lithium battery including the anode***"), [0051] ("FIG. 3 shows the structure of the active material layer" of an anode, in which "a composite of Li and the non-carbonaceous material … is generated during charging of the anode"), [0003], [0017], [0098] ("Coin-type half-cells were manufactured using the anodes manufactured according to Examples 1 and 2 …, a lithium metal as a counter electrode, and an electrolyte. Herein, a solution of 1.3M $LiPF_6$ dissolved in a solvent of ethylene carbonate (EC) and diethylene carbonate (DEC) … was used as the electrolyte"), Abstract, claim 17.

160. FIG. 5, *infra*, shows "a schematic view of a typical structure of ***a lithium battery***" including an anode formed from an anode active material composition (**the composition**) (which in turn is formed from an anode active material, alternatively/additionally **the composition**). LGES-1006 (Lee), [0067]; *see* [0003], [0013], [0017], [0027]. In this **lithium ion battery**, "lithium salt is dissolved in the organic solvent and is a source of ***lithium ions in the battery*** 30, enabling the basic operation of the lithium battery 30. In addition, the lithium salt facilitates the ***migration of lithium ions between the cathode 23 and the anode*** 22." *Id.*, [0079]; *see* [0076] ("The electrolyte includes a nonaqueous organic solvent and a lithium salt. The nonaqueous organic solvent serves as a medium for ***moving ions involved in electrochemical reactions [in] the battery***").



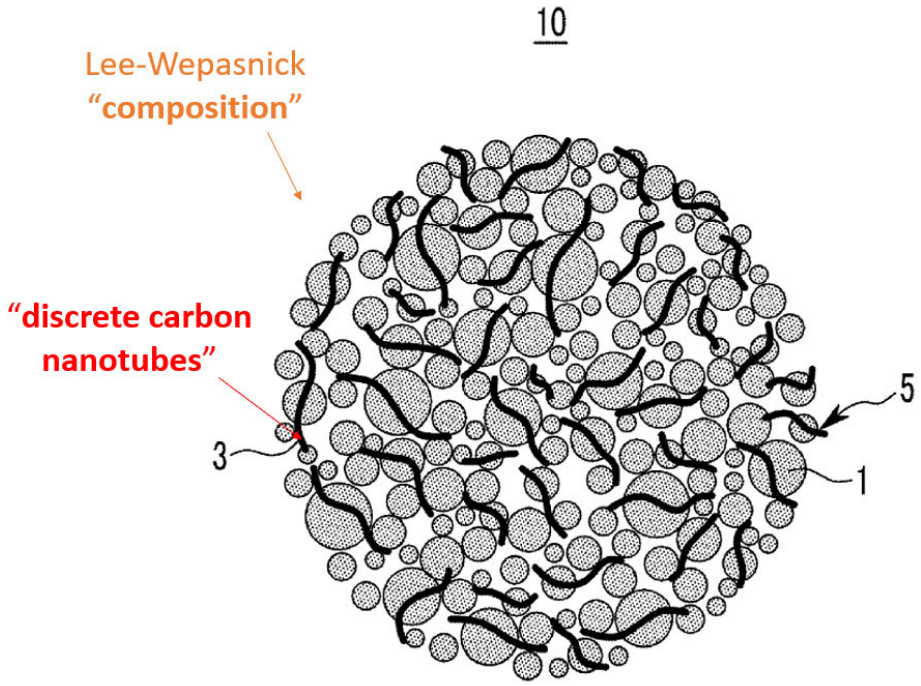
Excerpt from Lee, FIG. 5 (annotated)

[1a] discrete carbon nanotubes

The combination anode active material includes discrete carbon nanotubes

161. The combination **composition** (based on Lee's anode active material; *see* [1pre]) includes "secondary particles that are formed by the agglomeration of primary nanoparticles 5." LGES-1006 (Lee), [0040]. The primary nanoparticles in turn "include a non-carbonaceous material 1 and ***hollow carbon nanofibers*** 3." LGES-1006 (Lee), [0040]. The hollow carbon nanofibers, red in FIG. 1, *infra*, "may be, for example, ***carbon nanotubes***," *i.e.*, the combination anode active material (the **composition**) includes **carbon nanotubes**. *Id.*, [0046]; *see id.*

("Examples of available ***carbon nanotubes*** may be single-wall nanotubes, multi-wall nanotubes, coiled nanotubes, or a combination thereof"), [0020] ("the hollow carbon fibers may be ***carbon nanotubes***"), [0084] ("The resulting anode active material included secondary particles that were formed by the agglomeration of primary nanoparticles including Si particles bound to the ***carbon nanotubes***"), Abstract, [0014], [0087], [0104]-[0105], claims 4 and 8.



Excerpt from Lee, FIG. 1 (annotated)

162. As illustrated in Lee's FIG. 1, the carbon nanotubes in the combination anode active material (the **composition**) are **discrete**, *e.g.*, in that they

are present in the anode active material as generally individually distinct (*e.g.*, non-agglomerated) nanotubes.[5]

163. Alternatively/additionally, a POSITA would have understood or found obvious that the nanotubes in the combination composition are **discrete**. As I discuss further for element [1d], *infra*, the nanotubes in the Lee-Wepasnick combination are oxidized. At least in some instances, oxidization can help promote non-agglomeration of nanotubes, *e.g.*, oxidized nanotubes can be discrete. *See* [1d], *infra*. For instance, as I discussed above for element [1a] of the Ohata-Kavan ground, the sidewalls of oxidized nanotubes are functionalized by species such as carboxylic acid, carbonyl, or hydroxyl groups that are covalently bound to the nanotubes. The presence of these functional groups both physically and chemically prevent the nanotubes from aggregating into clumps and instead allow

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[5] Lee describes fabricating its anode active material by mixing the non-carbonaceous material and the hollow carbon nanofibers "in an organic solvent, using a milling process," such as "bead milling or ball milling." LGES-1006, [0060]. I understand that the accused products are fabricated using a ball milling process. To the extent ball milling and/or bead milling is compatible with discrete carbon nanotubes, it is my opinion that the Lee's process is compatible with discrete nanotubes.

the nanotubes to be generally present as individually distinct nanotubes. *See* LGES-1005 (Kavan), Abstract; LGES-1010 (Wepasnick), Abstract; LGES-1014 (Gojny), 821-822; LGES-1015, 4186-4187; LGES-1007 (Farrugia), [0027]-[0028]. In light of this well-known behavior of oxidized nanotubes, a POSITA would have understood or found obvious that oxidized nanotubes of the combination composition can be present as generally individually distinct nanotubes, *e.g.*, that the nanotubes are **discrete**.

The combination anode active material composition includes discrete carbon nanotubes

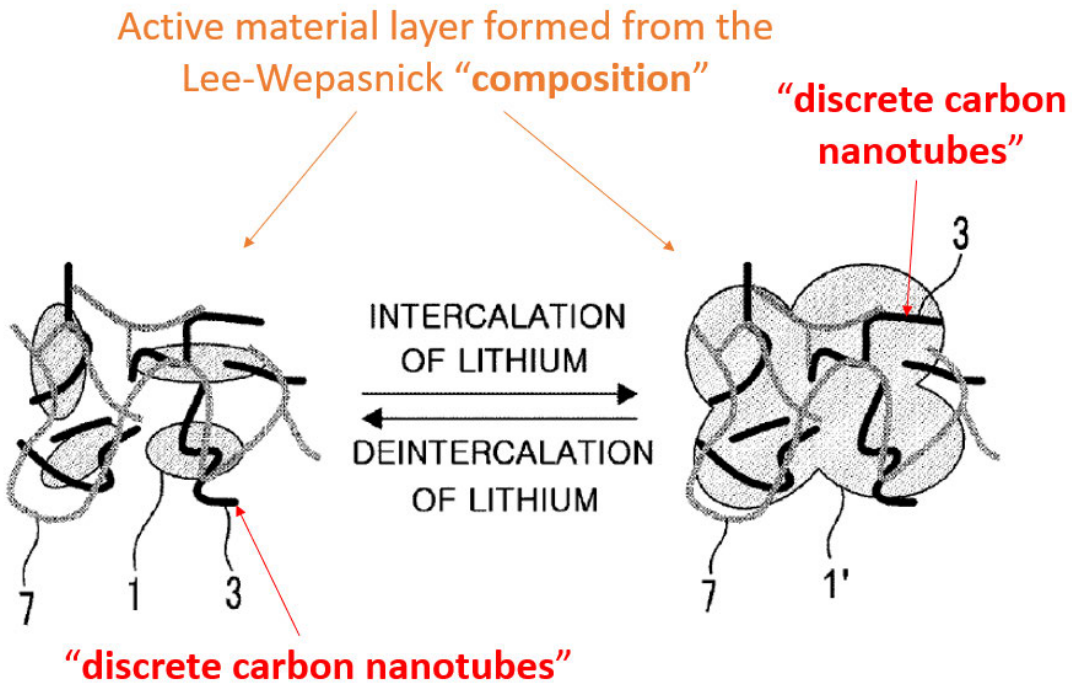
164. Alternatively/additionally, the combination **composition** (based on Lee's anode active material composition; *see* [1pre]) includes **carbon nanotubes**. This is the case at least because the anode active material composition includes the anode active material, which itself includes carbon nanotubes, as discussed *supra*. *See* LGES-1006 (Lee), [0064]-[0065] ("the anode active material and the polymeric binder are mixed in a solvent, to prepare an anode active material composition"), [0016], [0084]-[0085], claim 12.

165. Moreover, Lee provides details confirming the presence of hollow carbon nanofibers (**carbon nanotubes**) in the anode active material composition: "the amount of the ***hollow carbon nanofibers*** may be in a range of about 2 to about 20% … based on the total weight of the anode active material composition." *Id.*, [0064]; *see* [0083]-[0085] ("Silicon … and ***carbon nanotubes*** were mixed …

to prepare an anode active material. … Next, the anode active material, artificial graphite as a conducting agent, and polyethyleneimine as a binder were mixed in water, to prepare an anode active material composition in slurry form"), claim 18 ("An anode electrode material composition comprising: secondary particles comprising agglomerated primary nanoparticles, the primary nanoparticles comprising ***hollow carbon fibers*** attached to a silicon-based material; and a polymeric binder having an electron donor group").

166. The anode active material composition of the combination can be coated onto a current collector and dried to form an anode, resulting in "an active material layer … formed on the current collector." LGES-1006 (Lee), [0049]; *see* [0016], [0057], [0065], [0086]-[0087]. The composition of the active material layer thus is indicative of the composition of the anode active material composition. For instance, Lee's FIG. 3, *infra*, which illustrates "the structure of the active material layer," demonstrates that the anode active material composition includes "hollow carbon nanofibers 3" (red), *e.g.*, **carbon nanotubes**. *Id.*, [0050]; *see* [0015] ("The active material layer comprises a polymeric binder having an electron donor group and an anode active material including secondary particles that include agglomerated primary nanoparticles. The primary nanoparticles include a non-carbonaceous material that is bound to ***hollow carbon nanofibers***"),

[0054] ("In the anode active material layer 14, the binder binds to the ***hollow carbon nanofibers*** 3"), [0053].



Excerpt from Lee, FIG. 3 (annotated)

167. The combination active material layer (the **composition**) includes the anode active material, and thus the nanotubes in the combination active material layer are **discrete** for at least the reasons for which the nanotubes in the anode active material, as discussed *supra*.

168. Alternatively/additionally, the nanotubes in the combination anode active material composition are **discrete** based on Lee's teaching that "[t]he electron donor group" of the polymer binder in the anode active material composition "aids the ***dispersal of the hollow carbon nanofibers***." LGES-1006

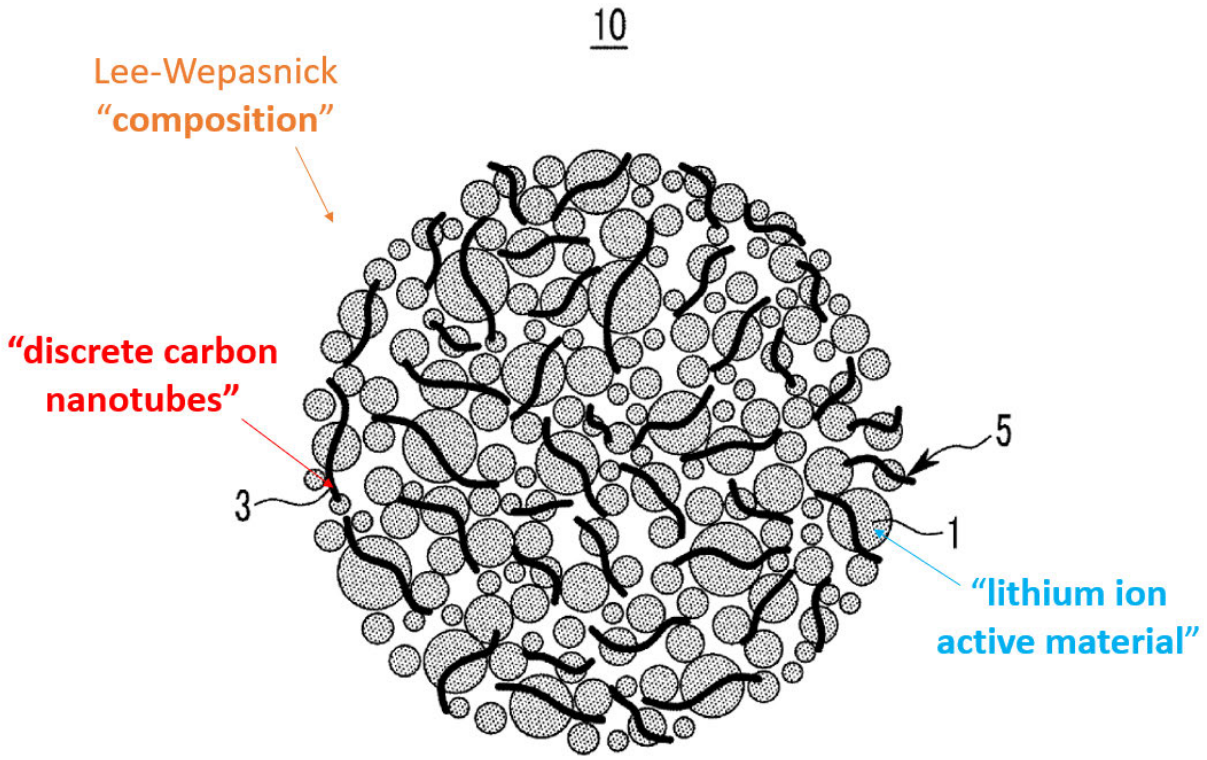
(Lee), [0053]. That is, the nanotubes are generally dispersed as individual nanotubes. *See also* Lee's FIG. 3 schematic illustration of **discrete** nanotubes in the active material layer formed from the anode active material composition.

[1b] having crystals or layers of lithium ion active material attached to their surface,

The combination anode active material includes lithium ion active material attached to the surface of the nanotubes

169. The Lee-Wepasnick combination **composition** (based on Lee's anode active material; *see* [1pre]) includes "secondary particles that are formed by the agglomeration of primary nanoparticles 5." LGES-1006 (Lee), [0040]. The primary nanoparticles in turn "include a ***non-carbonaceous material*** 1 and hollow carbon nanofibers 3." *Id.* The non-carbonaceous material, blue in FIG. 1, *infra*, corresponds to a **lithium ion active material**, as discussed in the next paragraph. *See* LGES-1006 (Lee), Abstract ("The anode active material includes secondary particles formed of agglomerated primary nanoparticles. The primary nanoparticles include a ***non-carbonaceous material*** bound with hollow carbon nanofibers"), [0018] ("***the non-carbonaceous material*** can be selected from the group consisting of silicon (Si), a silicon oxide ((SiOx) where $0 \leq x \leq 2$)), Si—Y, and a mixture thereof. Y may be As, Sb, Bi, Cu, Ni, Mg, In, Zn, Ag, Al, or a combination thereof"), [0039] ("The primary nanoparticles include the ***non-carbonaceous material*** and hollow carbon nanofibers, which are bound to the non-

carbonaceous material by Van der Waals forces"), [0043] ("In the anode active material 10, the ***non-carbonaceous material*** 1 is bound to the hollow carbon nanofibers to form a composite material"), [0044] ("When the average particle size of the non-carbonaceous material 1 is within the above ranges, the ***non-carbonaceous material*** 1 can bind with the hollow carbon nanofibers 3, which provides excellent cycle lifetime characteristics"), [0063] ("as the anode active material is prepared, the ***non-carbonaceous material*** is pulverized into nanoparticles, which bind with the hollow carbon nanofibers, thus forming the primary nanoparticles"), [0084] ("The resulting anode active material included secondary particles that were formed by the agglomeration of primary nanoparticles including ***Si particles*** bound to the carbon nanotubes"), [0014], [0041], claims 1-3, claim 5-6, claims 11-12, claims 18-19.



Excerpt from Lee, FIG. 1 (annotated)

170. In the combination, the non-carbonaceous material is a **lithium ion active material**, *e.g.*, because it is capable of intercalation/deintercalation of lithium ions. For instance, Lee describes that "the volume of the non-carbonaceous material 1 changes, due to the ***intercalation/de-intercalation [of] lithium***." LGES-1006 (Lee), [0053]; *see* [0042]-[0043] (describing that the non-carbonaceous materials in the anode active material "exhibit a higher capacity" and "a higher conductivity" in combination with the carbon nanotubes), [0006]-[0007] ("non-carbonaceous materials, such as silicon (Si), are being studied for use as ***anode active materials***. Such non-carbonaceous materials exhibit a capacity that is at least ten times that of graphite"), [0051] ("***a composite of Li and the non-***

carbonaceous material 1, which is generated during charging of the anode"), FIG. 3. Moreover, the specific non-carbonaceous materials identified by Lee (a material "selected from the group consisting of silicon (Si), a silicon oxide ((SiOx) where 0≤x≤2)), Si—Y, and a mixture thereof. Y may be As, Sb, Bi, Cu, Ni, Mg, In, Zn, Ag, Al, or a combination thereof") are examples of conventional **lithium ion active materials**. LGES-1006 (Lee), [0018]; *see* LGES-1035 (Takezawa), [0004] ("Alloy-based negative electrode active materials that absorb lithium by forming an alloy with lithium are gaining attention as a high capacity negative electrode active material. Known alloy-based negative electrode active materials include, for example, silicon, tin, oxides of these, nitrides of these, and compounds and alloys containing these"). Notably, many of these materials are identified by the '909 Patent as being lithium ion active materials for anodes: "[f]or an anode, the lithium ion active material may comprise carbon, tin, ***silicon***, ***copper***, ***antimony***, ***aluminum***, germanium, titanium or ***mixtures thereof***"). LGES-1001, 2:40-42.

171. In the combination, as described by Lee, the **lithium ion active material** (*e.g.*, based on Lee's non-carbonaceous material) is **attached to the surface of the nanotubes**, as I discuss in the following paragraphs.

172. First, consistent with Lee's disclosure, the lithium ion active material is attached to the surface of the nanotubes by Van der Waals forces: "the hollow

carbon nanofibers … are ***bound*** to the non-carbonaceous material ***by Van der Waals forces***." LGES-1006 (Lee), [0040]; *see* [0043] ("In the anode active material 10, ***the non-carbonaceous material 1 is bound to the hollow carbon nanofibers 3*** to form a composite material. Thus, the anode active material 10 can have a higher electrical conductivity"), [0044] ("When the average particle size of the non-carbonaceous material 1 is within the above ranges, ***the non-carbonaceous material 1 can bind with the hollow carbon nanofibers 3***, which provides excellent cycle lifetime characteristics"), [0063] ("the non-carbonaceous material is pulverized into ***nanoparticles, which bind with the hollow carbon nanofibers***, thus forming the primary nanoparticles"), [0084] ("The resulting anode active material included secondary particles that were formed by the agglomeration of primary nanoparticles including ***Si particles bound to the carbon nanotubes***"), [0014]-[0016], [0039], [0101], Abstract, claim 1, claim 6. To the extent that Van der Waals bonding is considered an attachment, the non-carbonaceous material (*i.e.*, the lithium ion active material) is **attached** to the surface of the nanotubes through Van der Waals bonding.

173. Notably, Van der Waals bonding between the nanotubes and the lithium ion active material occurs even with the presence of oxygen-containing functional groups (the presence of which I explain in element [1d], *infra*). Thus, (again to the extent that Van der Waals forces can be considered an attachment),

the non-carbonaceous material (*i.e.*, the lithium ion active material) is **attached** to the surface of the nanotubes through Van der Waals bonding even when the nanotubes are oxidized, as they are in the Lee-Wepasnick combination.

174. Alternatively/additionally, a POSITA would have recognized that the presence of these oxygen-containing functional groups (again, see my discussion of element [1d]) further facilitates the attachment of the lithium ion active material to the nanotube surface. For instance, Wepasnick generally describes this attachment mechanism, stating that "In functionalization strategies, the role of oxidation is to create 'foothold' functional groups, such as carboxyl groups, which then serve as attachment points to anchor … nanoparticles." LGES-1010 (Wepasnick), 2. More specifically, this **attachment** between the non-carbonaceous material (*i.e.*, the lithium ion active material) and the surface of the oxidized nanotubes can be a covalent bond, as demonstrated by Eom: "It is believed that the surface of the Si particles was oxidized by ball-milling, and the chemical-bonded SiC compound was formed on the interface of the SWCNTs and Si particles," where the nanotubes are oxidized ("effectively purified by an acidic treatment followed by the gas-phase oxidation process." LGES-1043 (Eom), 2-3.

175. Still alternatively/additionally, hydrogen bonds can arise due to force between the non-carbonaceous materials and hydrogen atoms belonging to the oxygen-containing functionalities on the nanotube surface. To the extent that

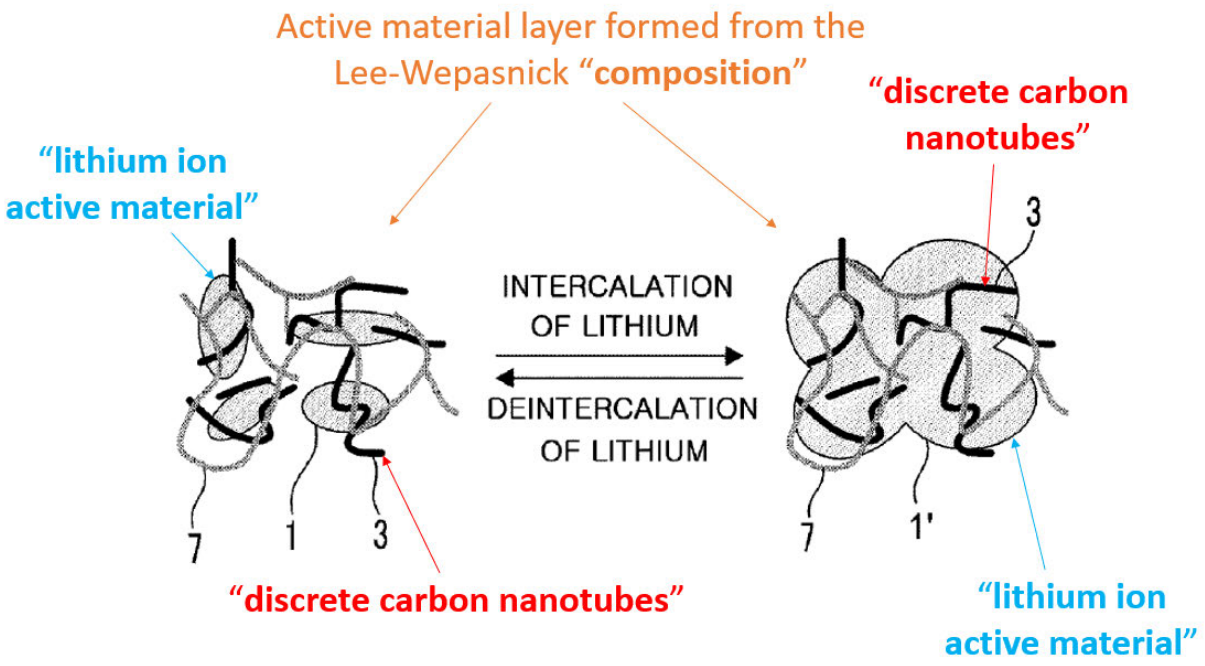
hydrogen bonding is considered an attachment, the non-carbonaceous materials are alternatively/additionally **attached** to the surface of the nanotubes through hydrogen bonding.

The combination anode active material composition includes lithium ion active material attached to the surface of the nanotubes

176. Alternatively/additionally, the Lee-Wepasnick combination composition (based on Lee's anode active material composition; *see* [1pre]) includes **lithium ion active material attached to the surface of the nanotubes**. This is the case at least because the anode active material composition includes the anode active material, which itself includes lithium ion active material attached to the surface of the nanotubes, as discussed *supra*. *See* LGES-1006 (Lee), [0064]-[0065] ("the anode active material and the polymeric binder are mixed in a solvent, to prepare an anode active material composition"), [0016], [0084]-[0085], claim 12. And Lee's discussion of the composition of the anode active material composition confirms the presence of non-carbonaceous material (**lithium ion active material**): *see* [0083]-[0085] ("***Silicon*** … and carbon nanotubes were mixed … to prepare an anode active material. … Next, the anode active material, artificial graphite as a conducting agent, and polyethyleneimine as a binder were mixed in water, to prepare an anode active material composition in slurry form"), claim 18 ("An anode electrode material composition comprising: secondary particles comprising agglomerated primary nanoparticles, the primary

nanoparticles comprising hollow carbon fibers attached to ***a silicon-based material***; and a polymeric binder having an electron donor group").

177. As noted *supra* in [1a], the composition of the active material layer is indicative of the composition of the anode active material composition. *See* LGES-1006 (Lee), [0016]. [0049], [0057], [0065], [0086]-[0087]. Lee's FIG. 3, *infra*, which illustrates "the structure of the active material layer," demonstrates that the anode active material composition includes "non-carbonaceous material 1" (blue), *e.g.*, that the anode active material composition (the **composition** of the combination) includes **lithium ion active material**. *Id.*, [0053]; *see* [0015] ("The active material layer comprises a polymeric binder having an electron donor group and an anode active material including secondary particles that include agglomerated primary nanoparticles. The primary nanoparticles include a ***non-carbonaceous material*** that is bound to hollow carbon nanofibers"), [0051] ("reference number 1′ denotes a composite of Li and the ***non-carbonaceous material 1***, which is generated during charging of the anode 20").



Excerpt from Lee, FIG. 3 (annotated)

The combination composition includes crystals of lithium ion active material

178. In the combination composition (based on either Lee's anode active material or Lee's anode active material composition), a POSITA would have understood, or at least found obvious, that the lithium ion active material (*e.g.*, the non-carbonaceous material) is in the form of **crystals**, as discussed in the following paragraphs.

179. Lee identifies silicon as a specific example of a non-carbonaceous material (*see* Lee, [0041]), and further specifies that silicon as an active material in an anode has a "***crystalline lattice***," *i.e.*, the particles of silicon non-carbonaceous material (**lithium ion active material**) are **crystals**. *Id.*, [0059].

180. Lee acknowledges that in "conventional anodes," crystalline silicon can be problematic, *e.g.*, because "abrupt volumetric changes in the crystalline lattice" can occur, "thereby fracturing the conventional active material." LGES-1006 (Lee), [0059]. However, in Lee's anode active material composition the "bonding" of the binder "with the hollow carbon nanofibers" "enhance[s] the cohesion of [the] anode," thus mitigating these problems with conventional use of crystalline silicon. *Id.*, [0058]; *see* [0059] ("the structure of the active material layer 14 can be maintained during charging and discharging. Thus, when the anode 20 is used, problems arising with conventional anodes including a non-carbonaceous material, such as Si, and/or other problem, can be solved"). Given this context, a POSITA would have understood Lee's disclosure as enabling, and consistent with, the use of **crystals** of lithium ion active material in the combination composition.

181. Alternatively/additionally, to the extent that Lee does not specifically state that the silicon or silicon-metal alloy in the anode active material are **crystals or layers**, a POSITA nevertheless would have found this to be obvious, for at least the following reasons.

182. First, Lee's "method of manufacturing the anode" involves mixing "a non-carbonaceous material" [*i.e.*, the lithium ion active material] "and hollow carbon nanofibers … in an organic solvent, using a milling process" such as "bead

milling or a ball milling." LGES-1006 (Lee), [0060]. A POSITA would have recognized that ball milling produces crystals of material, *e.g.*, polycrystalline or nanocrystalline material.[6] *See* LGES-1025 ("Shen"), Abstract ("The structural transformation of polycrystalline Si induced by high energy ball milling has been studied. … Two phase amorphous and ***nanocrystalline Si has been produced*** by ball milling of polycrystalline elemental Si. … The volume fraction of amorphous Si is about 15% while ***the average size of nanocrystalline grains is about 8 nm***."); LGES-1026 (Gaffet), Abstract ("We present an experimental study of the first reported crystal-amorphous phase transition induced by ball-milling in a pure element: a silicon powder. … The analyses of the X-ray patterns reveal the coexistence of silicon microcrystallites and an amorphous phase"); LGES-1027

[6] The '909 Patent describes a similar manufacturing process: "Lithium iron phosphate … is ground with multiwall carbon nanotubes … in a mortar and pestle." LGES-1001, 5:53-57. To the extent that this disclosure is sufficient to support the existence of **crystals or layers** of lithium iron phosphate attached to the surface of the nanotubes, I believe that a POSITA would have recognized Lee's similar disclosure as also supporting the existence of **crystals or layers** of lithium ion active material.

(Kumar), 20:30-33 ("milling can be desirable from a particle size perspective as well as ***allowing retention of a low level of crystallinity***").

183. Second, use of crystals of lithium ion active materials has performance advantages, *e.g.*, higher electrical conductivity and faster lithiation rate. *See* LGES-1028 (Lopez), 19:24-27 ("Crystalline silicon nanoparticles can be desireable [sic] in some embodiments because of their larger electrical conductivity, relative to amorphous silicon nanoparticles"); LGES-1029 (Obravac), 4:36-39 ("Because the rate of lithiation is generally greater for nanocrystalline material than for amorphous material, it is desirable to include some nanocrystalline material in the alloy composition"), 3:59-4:2.

184. Third, the use of crystals of lithium ion active material in the combination composition was obvious to try. It is one of a finite number of known solutions—namely, either crystals or amorphous particles of lithium ion active material. It was predictable, *e.g.*, in that behavior of silicon and silicon-metal alloy crystals as lithium ion active material was conventional and well-characterized as of the Critical Date. *See* LGES-1043 (Eom), Abstract ("Single-walled carbon nanotube (SWCNT)/silicon composites were produced from the purified SWCNTs and Si powder … and then electrochemically inserted with Li using Li/(SWCNT/Si) cells"). And a POSITA would have had a reasonable expectation of success in using crystals of silicon or silicon-metal alloy.

185. Fourth, a POSITA would have been prompted to pursue this implementation because doing so is just the application of known techniques (specifically, crystals of a lithium ion active material) to an already-known structure (the Lee-Wepasnick anode active material), and the combination produces predictable results (specifically, an anode active material containing crystals of lithium ion active material).

186. A POSITA would have expected success in using crystals of lithium ion active material in the Lee-Wepasnick composition for at least the following reasons.

187. First, Lee indicates that challenges involved in using crystals of lithium ion active material can be overcome through the structure of its anode active material composition. *See* LGES-1006 (Lee), [0058]-[0059].

188. Second, the use of crystals of lithium ion active material was well-characterized, and a POSITA would have known how to structure crystals of lithium ion active material to achieve advantages of the crystallinity.

[1c] wherein the discrete carbon nanotubes have an aspect ratio of 10 to 500 and

189. The carbon nanotubes in the combination composition "have ***an aspect ratio of about 10 to about 10,000***," as taught by Lee. LGES-1006 (Lee), [0047].

190. The claimed range of aspect ratios (**10-500**) overlaps with the range of aspect ratios (***10-10,000***) described by Lee and implemented in the Lee-Wepasnick combination composition. I understand that unless there is evidence of unexpected results or criticality associated with a claimed range, that claimed range is rendered obvious by an overlapping range in the prior art. I thus conclude that the overlapping range of aspect ratios disclosed by Lee and implemented in the Lee-Wepasnick combination renders obvious the claimed range.

191. I note that Lee's fabrication process involves "a milling process" such as "bead milling or ball milling," "at a speed of about 50 to about 60 Hz, for about 1 to about 2 hours." LGES-1006 (Lee), [0060]-[0061]. While some high intensity milling processes can significantly shorten nanotubes, the literature confirms that mild milling, *e.g.*, under parameters such as those described by Lee, will not have a significant impact on nanotube length. For instance, Kukovecz demonstrated that "[t]he length distribution function" of nanotubes exposed to a milling process "changed from the original log-normal to normal after 140 h of milling," and other "morphological descriptors all experienced characteristic changes after ~40 h of milling." LGES-1039 (Kukovecz), Abstract. Because Lee's milling process is significantly shorter—only 1-2 hours—I do not expect the aspect ratio of the nanotubes to change significantly beyond the range disclosed by Lee (10-10,000). However, even to the extent that Lee's nanotubes are shortened slightly due to

milling, it is reasonable to expect the range of Lee's "shortened" nanotubes still to fall within the claimed range of 10-500.

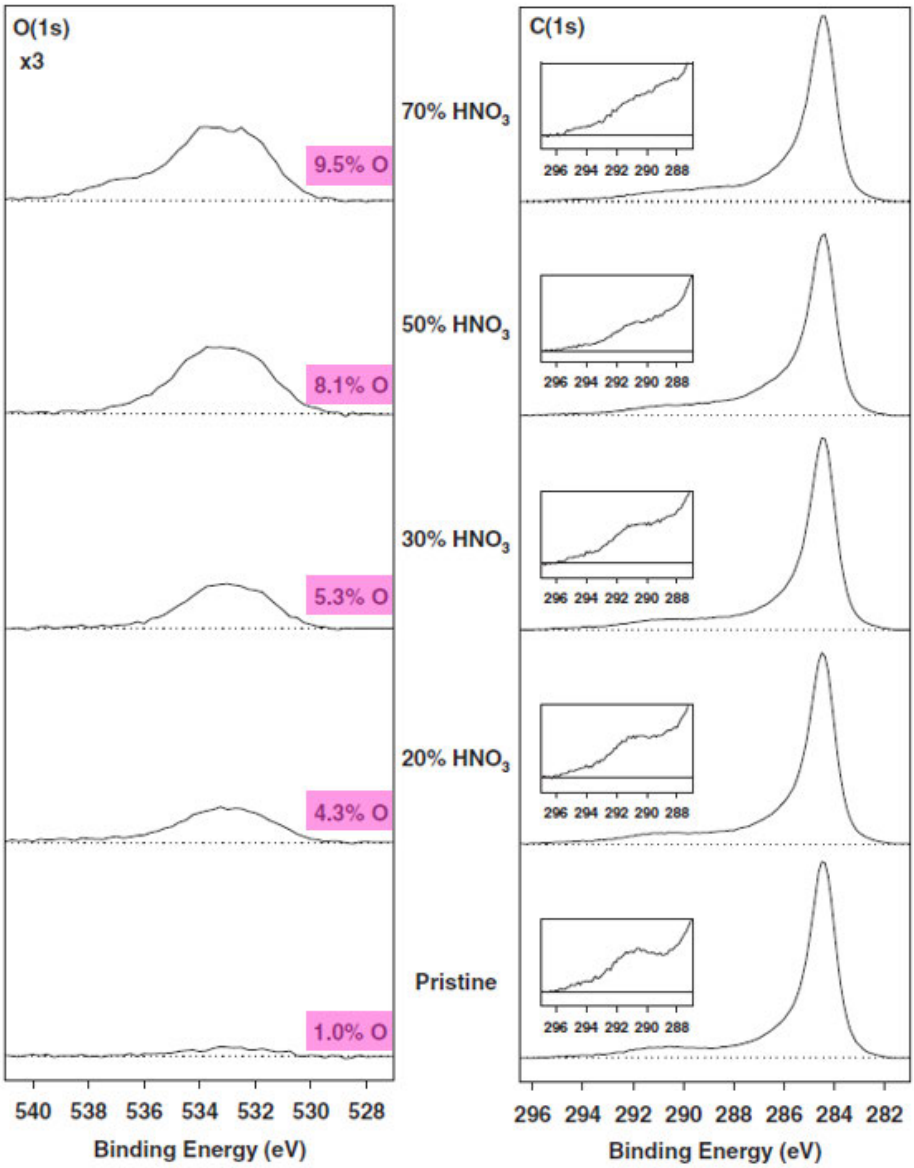
192. I also note that the nanotubes in the combination are oxidized, as I discuss in [1d]. Oxidation of nanotubes can sometimes shorten their length. Again, though, even to the extent that Lee's nanotubes are shortened slightly by oxidation, it is reasonable to expect the range of Lee's "shortened" nanotubes still to fall within the claimed range of 10-500.

[1d] oxidation levels from 1% to 15% by weight of the carbon nanotube.

193. The carbon nanotubes in the combination composition are **oxidized** based on the teachings of Wepasnick. For instance, the nanotubes are oxidized in one of the "commonly used wet chemical oxidants (HNO_3, $KMnO_4$, H_2SO_4/HNO_3, $(NH_4)_2S_2O_8$, H_2O_2, and O_3)," producing "carboxyl, carbonyl, and hydroxyl groups at the surface" of the nanotubes." LGES-1010 (Wepasnick), Abstract; *see id.* ("the distribution of oxygen-containing functional groups was insensitive to the reaction conditions … but was sensitive to the identity of the oxidant"), 2 ("the surface of CNTs … are often modified covalently by the addition of polar functional groups"), 2 ("deliberately grafting oxygen-containing functional groups at the open ends and sidewalls of CNTs is a popular and versatile approach"), 2 ("the role of oxidation is to create 'foothold' functional groups, such as carboxyl groups, which then serve as attachment points").

Nanotubes oxidized using HNO_3 have oxidation levels within the claimed range

194. In one implementation of this oxidative treatment, the nanotubes in the combination composition are oxidized using HNO_3 based on the teachings of Wepasnick. *See* LGES-1010 (Wepasnick), 3. By varying "the w/w% HNO_3 (from 0-70%), "the extent of MWCNT oxidation" (highlighted in Fig. 6, *infra*) can be controlled between 4.3 at.% for nanotubes oxidized in 20% HNO_3 to 9.5 at.% for nanotubes oxidized in 70% HNO_3. *See* LGES-1010 (Wepasnick), 7.



Excerpt from Wepasnick, Fig. 6 (annotated)

195. One way to determine the **oxidation level by weight of the nanotubes** is to calculate the weight of the oxygen atoms attached to the nanotubes divided by the total weight of the oxidized nanotubes (*i.e.*, the weight percent oxygen). In this regard, based on the data in Wepasnick's Fig. 6, the oxidation levels for Wepasnick's HNO_3-oxidized nanotubes range from 5.7% to 12.3% by

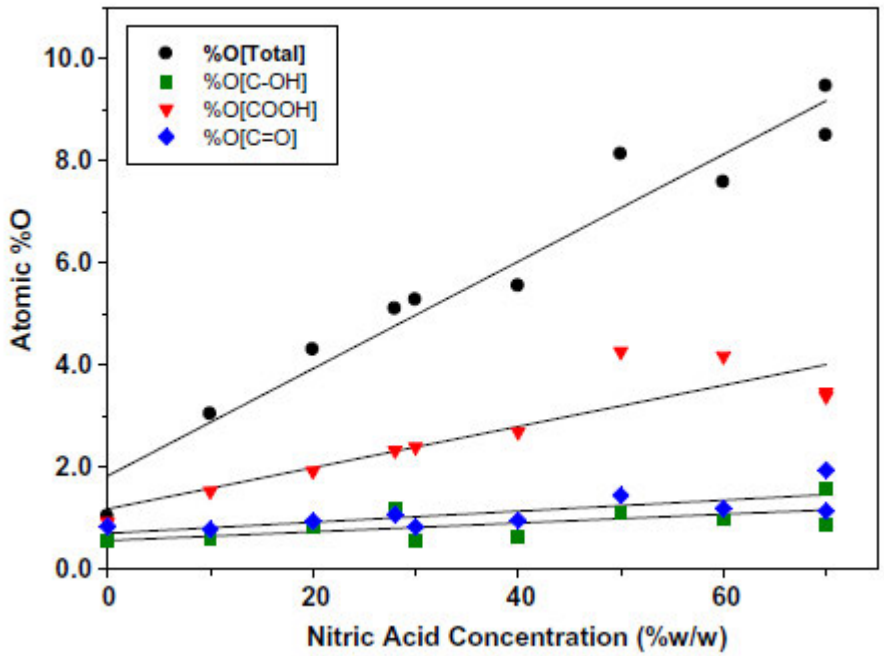
weight of the carbon nanotube (shown in blue and bold in Table 1, *infra*), *e.g.*, 5.7-12.3% by weight of the oxidized carbon nanotubes is oxygen. My calculations are provided as Calculation 1 in Appendix B.

HNO_3	**at.% oxygen**	**Oxidation level (wt.% oxygen)**	**Oxidation level (wt.% functional groups)**
20%	4.3 at.%	**5.7 wt.%**	***7.4 wt.%***
30%	5.3 at.%	**6.9 wt.%**	***7.7 wt.%***
50%	8.1 at.%	**10.5 wt.%**	***13.8 wt.%***
70%	9.5 at.%	**12.3 wt.%**	***14.6 wt.%***

Table 1. Oxidation levels of Wepasnick's HNO_3-oxidized nanotubes. *See* LGES-1010 (Wepasnick), Figs. 6-7. *See* Calculations 1-2 in Appendix B.

196. Each of these oxidation levels by weight for Wepasnick's HNO_3-oxidized nanotubes falls within the recited range of oxidation levels (**1% to 15% by weight**). I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Wepasnick, I conclude that each of these oxidation levels disclosed by Wepasnick, and applied to the nanotubes of the combination, renders obvious a composition in which the nanotubes have **oxidation levels from 1% to 15% by weight of the carbon nanotube**.

197. Wepasnick also demonstrates that "varying the w/w% of HNO_3" affects "the distribution of hydroxyl, carbonyl and carboxylic acid groups." LGES-1010 (Wepasnick), 7; *see* Fig. 7, *infra*.



Excerpt from Wepasnick, Fig. 7

198. Another reasonable way to convert the atomic percent oxygen to an oxidation level by weight is to use this distribution of functional species can be used to calculate the weight percentage of functional groups, *e.g.*, the weight of the -C-OH, -C=O, and -COOH functional groups attached to the nanotubes divided by the total weight of the oxidized nanotubes. This weight percentage of functional groups is alternatively/additionally the **oxidation level by weight of the nanotubes**.

199. In this regard, the oxidation levels for Wepasnick's nanotubes oxidized with 20%, 30%, 50%, and 70% HNO3 are **7.4%, 7.7%, 13.8%, and 14.6%** by weight of the carbon nanotube, respectively (shown in green italics in

Table 1, *supra*), *e.g.*, the -C-OH, -C=O, and -COOH functional groups together constitute 7.4%, 7.7%, 13.8%, or 14.6% by weight of the oxidized carbon nanotubes. My calculations are provided as Calculation 2 in Appendix B.

200. These values (**7.4%, 7.7%, 13.8%, and 14.6%**) fall within the recited range of oxidation levels (**1% to 15% by weight**). I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Wepasnick, I conclude that each of these oxidation levels disclosed by Wepasnick, and applied to the nanotubes of the combination, renders obvious a composition in which the nanotubes have **oxidation levels from 1% to 15% by weight of the carbon nanotube**.

Nanotubes oxidized to 4.5-5.5 atomic percent have oxidation levels within the claimed range

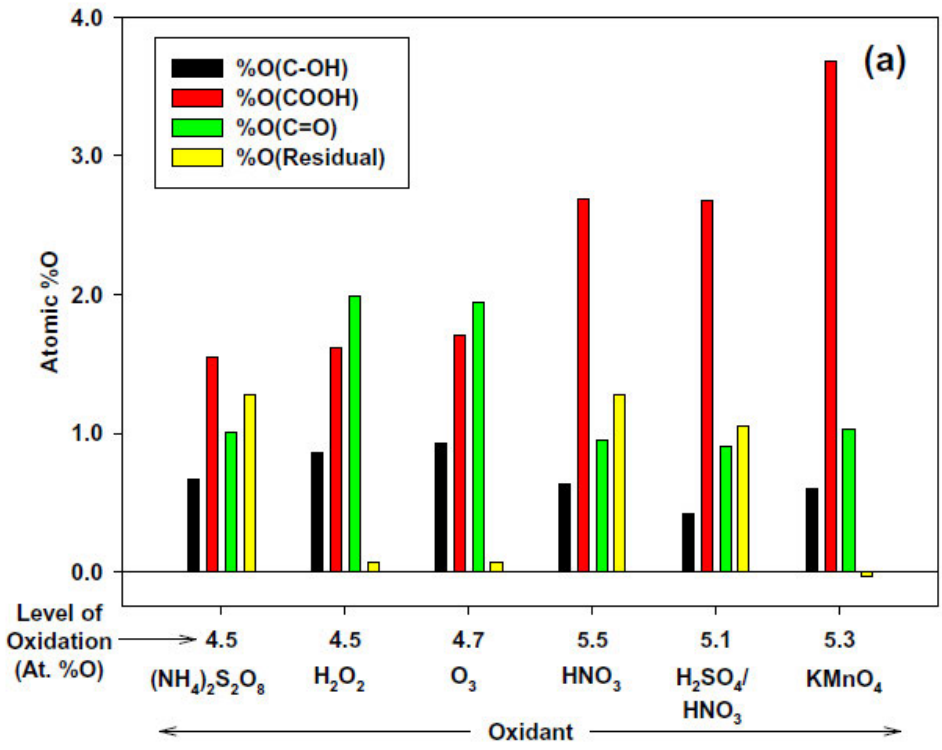
201. In an alternative/additional implementation of this oxidative treatment, the nanotubes in the combination composition are oxidized at an "overall level of oxidation" of "***4.5-5.5 at.% O***" using an oxidant selected from $KMnO_4$, H_2SO_4/HNO_3, HNO_3, O_3, H_2O_2, or $(NH_4)_2S_2O_8$. LGES-1010 (Wepasnick), 8, Fig. 9.

202. When the **oxidation level by weight of the nanotubes** is determined by calculating the weight of the oxygen atoms attached to the nanotubes divided by the total weight of the oxidized nanotubes (*i.e.*, the weight percent oxygen),

Wepasnick's nanotubes oxidized to 4.5-5.5 at.% oxygen correspond to an **oxidation level by weight** of **5.9-7.2%** by weight of the carbon nanotube, *e.g.*, 5.9-7.2% by weight of the oxidized carbon nanotubes is oxygen. My calculations are provided as Calculation 3 in Appendix B.

203. This range of oxidation levels (5.9-7.2% by weight) falls within the recited range of oxidation levels (**1% to 15% by weight**). I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Wepasnick, I conclude that each of these oxidation levels disclosed by Wepasnick, and applied to the nanotubes of the combination, renders obvious a composition in which the nanotubes have **oxidation levels from 1% to 15% by weight of the carbon nanotube**.

204. Furthermore, Wepasnick demonstrates that "distribution of oxygen-containing functional groups is sensitive to the oxidant." Wepasnick, 8; *see* Fig. 9(a), *infra*.



Excerpt from Wepasnick, Fig. 9(a)

205. This distribution of functional species can be used to calculate the weight percentage of functional groups, which is alternatively/additionally the **oxidation level by weight of the nanotubes**. In this regard, the oxidation levels of the nanotubes represented in Wepasnick's Fig. 9(a) range from **6.5-10.1%** by weight of the carbon nanotube (shown in red and bold in Table 2, *infra*), *e.g.*, the C-OH, -C=O, and -COOH functional groups together constitute 6.5-10.1% by weight of the oxidized carbon nanotubes. Alternatively/additionally, when residual oxygen is also counted in calculation of the oxidation levels of the nanotubes, the oxidation levels of the nanotubes represented in Wepasnick's Fig. 9(a) range from **7.9-10.1%** by weight of the carbon nanotube (shown in purple italics in Table 2,

infra), *e.g.*, the C-OH, -C=O, and -COOH functional groups and residual oxygen together constitute 7.9-10.1% by weight of the oxidized carbon nanotubes. My calculations are provided as Calculations 4 and 5 in Appendix B.

Oxidant	**at.% oxygen**	**Oxidation level** (without residual oxygen)	**Oxidation level** (with residual oxygen)
$(NH_4)_2S_2O_8$	4.5 at.%	**6.5 wt.%**	***7.9 wt.%***
H_2O_2	4.5 at.%	**9.7 wt.%**	***9.8 wt.%***
O_3	4.7 at.%	**9.9 wt.%**	***10.0 wt.%***
HNO_3	5.5 at.%	**8.0 wt.%**	***9.6 wt.%***
H_2SO_4/HNO_3	5.1 at.%	**7.6 wt.%**	***8.9 wt.%***
$KMnO_4$	5.3 at.%	**10.1 wt.%**	***10.1 wt.%***

Table 2. Oxidation levels of Wepasnick's oxidized nanotubes. *See* LGES-1010 (Wepasnick), Fig. 9. *See* Calculations 4-5 in Appendix B.

206. Each of the oxidation levels calculated based on Wepasnick's Fig. 9 and tabulated in Table 2, *supra*, falls within the recited range of oxidation levels (1% to 15% by weight). I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Wepasnick, I conclude that each of these oxidation levels disclosed by Wepasnick, and applied to the nanotubes of the combination, renders obvious a composition in which the nanotubes have **oxidation levels from 1% to 15% by weight of the carbon nanotube**.

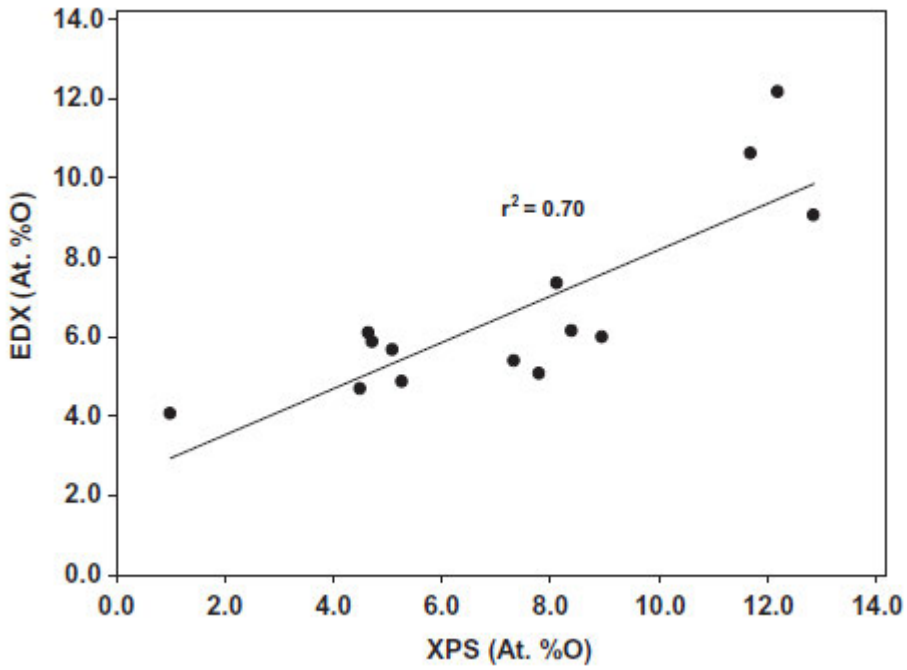
Wepasnick's EDX results corroborate the XPS results used to determine oxidation levels

207. The oxidation levels discussed *supra* are based on Wepasnick's measurements of atomic oxygen concentration using XPS. *See* LGES-1010 (Wepasnick), Figs. 5-9. As I discussed *supra* for Ground 1, while the '909 Patent does not identify a specific characterization technique to be used for determining oxidation levels, a POSITA would have recognized or otherwise found obvious that XPS characterization of oxidation levels is a reasonable and well-known way to determine oxidation levels for nanotubes such as those disclosed by Wepasnick. *See* LGES-1005 (Kavan), 4 ("The amount of atomic oxygen present on the MWNT-ox surface was also analyzed by X-ray photoelectron spectroscopy"); LGES-1009 (Datsyuk), 4 ("With reference to functionality grafting, XPS is one of the surface analytical techniques, which can provide useful information on the nature of the functional groups and also on the presence of structural defects on the nanotube surface").

208. Nevertheless, as I explained above in my technical overview section, other experimental techniques are available for measuring oxidation levels of carbon nanotubes. For instance, Wepasnick used both XPS and EDX (energy dispersive X-ray spectroscopy) "to quantify the atomic oxygen concentration" on the nanotubes, and demonstrated that "XPS and EDX results are reasonably well correlated" and in "close proximity." LGES-1010 (Wepasnick), 6. While still

other characterization methods exist, a POSITA would have understood that, in the context of Wepasnick's nanotubes, both XPS and EDX data provide reliable results for determining oxidation levels.

209. Indeed, the data in Wepasnick's Fig. 5, *infra*, confirm that the nanotubes I discussed *supra* (which have oxidation levels falling within the claimed range as determined by XPS) generally also have oxidation levels falling within the claimed range as determined by EDX. Specifically, the oxidation levels by weight, based on the atomic percent oxygen ranging from 4 at.% to 12 at.% as determined by EDX from Fig. 5 below, range from 5.3% to 15.4%, when calculated as the weight percent of oxygen on the nanotubes. My calculations are provided as Calculation 6 in Appendix B.



Excerpt from Wepasnick, Fig. 5

210. Some of these values fall within the recited range of oxidation levels (**1% to 15% by weight**). I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Wepasnick, I conclude that each of these oxidation levels disclosed by Wepasnick, and applied to the nanotubes of the combination, renders obvious a composition in which the nanotubes have **oxidation levels from 1% to 15% by weight of the carbon nanotube**.

(ii) Claim 2

[2] The composition of claim 1, wherein the crystals or layers of lithium ion active material comprise tin, silicon, copper, antimony, aluminum, germanium, titanium, or mixtures thereof.

211. In the Lee-Wepasnick combination, the anode active material and/or anode active material composition (the **composition**; *see* [1pre]) includes "non-carbonaceous material" (**lithium ion active material**) which "can be selected from the group consisting of ***silicon (Si), a silicon oxide ((SiOx) where 0≤x≤2)), Si—Y***, and a mixture thereof. Y may be As, ***Sb***, Bi, ***Cu***, Ni, Mg, In, Zn, Ag, ***Al***, or a combination thereof.," *i.e.*, the **lithium ion active material comprises silicon** (Si), **copper** (Cu), **antimony** (Sb), and/or **aluminum** (Al). LGES-1006 (Lee), [0018]; *see* [0084] ("The resulting anode active material included secondary particles that were formed by the agglomeration of primary nanoparticles including ***Si particles*** bound to the carbon nanotubes"), claims 5, 11, 19.

(iii) Claim 5

[5] The composition of claim 1, wherein the carbon nanotubes are present in a weight percentage of 0.1% to 10% by weight of the lithium ion active material.

212. The ratio of carbon nanotubes to lithium ion active material in the combination is based on the teachings of Lee.

213. In Example 1, Lee describes that "the proportions of the anode active material, the conducting agent and the binder were 55% (***50% of Si and 5% of***

carbon nanotubes, by weight), 30%, and 15%, respectively, by weight." LGES-1006 (Lee), [0085]. The composition of this example (5% by weight carbon nanotubes, 50% by weight Si) corresponds to the carbon nanotubes being present in a weight percentage of 10% by weight of the lithium ion active material. Alternatively/additionally, to the extent the weight percentage is based on the total weight of both nanotubes and lithium ion active material (*see* [5] of Ground 1), the Example 1 composition corresponds to the carbon nanotubes being present in a weight percentage of **9.1% by weight** (5 parts nanotubes per 55 parts total).

214. In Example 2, Lee describes that "no conducting agent was used, and the proportions of the anode active material and the binder were 85% by weight (***76.5% by weight of Si and 8.5% by weight of carbon nanotubes***) and 15% by weight, respectively." *Id.*, [0087]. In this example, to the extent the weight percentage is based on the total weight of both nanotubes and lithium ion active material, the carbon nanotubes are present in a weight percentage of **10% by weight** (8.5 parts nanotubes per 85 parts total).

215. These specific examples of weight percentages (***9.1% and 10%***) fall within the claimed **0.1% to 10%** range of weight percentages. I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Lee, I conclude that these specific weight percentages disclosed by Lee, and

applied to the combination, render obvious the claimed range of weight percentages.

216. Alternatively/additionally, in the combination, Lee describes that "[i]n the anode active material 10, a mixing ratio of the non-carbonaceous material 1 to the hollow carbon nanofibers 3 may be in ***a range of about 2:1 to about 50:1, by weight***, and particularly, about 5:1 to about 10:1, by weight. If the proportion of the hollow carbon nanofibers 3 is too high, irreversible reactions may occur during charging and discharging. On the other hand, if the proportion of the hollow carbon nanofibers 3 is too low, satisfactory effects may not be attained." LGES-1006 (Lee), [0048]. The 2:1-50:1 range of ratios corresponds to a range of weight percentages of 2% to 50% (or 2% to 33%, to the extent the weight percentage is based on the total weight of both nanotubes and lithium ion active material). The 5:1 to 10:1 range of ratios corresponds to a range of weight percentages of 10% to 20% (or 9% to 17%, to the extent the weight percentage is based on the total weight of both nanotubes and lithium ion active material). *See also id.*, claim 7 ("wherein an amount of the hollow carbon nanofibers is in a ***range of from about 2% to about 20%, by weight***, based on the total weight of the active material layer"), claims 3 and 16.

217. The claimed range of weight percentages (**0.1% to 10%**) overlaps with the range of weight percentages described by Lee (***2-50%*** or ***2-33%*** or ***9-17%***)

and implemented in the Lee-Wepasnick combination composition. I understand that unless there is evidence of unexpected results or criticality associated with a claimed range, that claimed range is rendered obvious by an overlapping range in the prior art. I thus conclude that the overlapping range of weight percentages disclosed by Lee and implemented in the Lee-Wepasnick combination renders obvious the recited range of weight percentages.

(iv) Claim 6

[6a] The composition of claim 1, comprising oxidized carbon nanotubes having crystals or layers of lithium ion active materials attached to their surface,

218. *See* [1b], [1d].

[6b] wherein the carbon nanotubes are uniformly dispersed without clustering on a dimension scale less than 1 micrometer.

219. As discussed in [1a], the nanotubes in the combination anode active material (the **composition**) are present as generally individually distinct nanotubes, *e.g.*, they are **dispersed** in the composition. *See* [1a]. Based on Lee's FIG. 1, *infra*, which illustrates "the structure of an anode active material," and also based on the nature of the nanotubes in the combination as dispersed, a POSITA would have understood, or at least found obvious, that the nanotubes are **uniformly dispersed without clustering** in the combination anode active material. Because

there is substantially no clustering in the composition, a POSITA would have understood that there is also no clustering on a scale of less than 1 micrometer.

(v) Claim 7

[7] The composition of claim 1, comprising uniformly dispersible oxidized carbon nanotubes having crystals or layers of lithium ion active materials attached to their surface.

220. The combination composition includes uniformly dispersed, oxidized carbon nanotubes having crystals or layers of lithium ion active materials attached to their surface. *See* claim [6], *supra*, which addresses that the nanotubes are uniformly dispersed oxidized carbon nanotubes having crystals or layers of lithium ion active materials attached to their surface.

221. Moreover, the nanotubes in the combination composition are **dispersible** because the combination composition is capable of being mixed. For instance, to form the anode active material (the **composition**; *see* [1pre]), "a non-carbonaceous material and hollow carbon nanofibers are ***mixed*** in an organic solvent." LGES-1006 (Lee), [0060]. To form the anode active material composition (alternatively/additionally the **composition**; *see* [1pre]), "the anode active material and the polymeric binder are ***mixed*** in a solvent." *Id.*, [0064].

222. I note that some applications of nanotubes rely on vertically aligned arrangements of nanotubes in which nanotubes are grown with an end attached to a substrate. The nanotubes in the Lee-Wepasnick are alternatively/additionally

dispersible because they are not attached to a substrate in a vertically aligned arrangement.

(vi) Claim 8

[8] The composition of claim 7, wherein the crystals or layers of lithium ion active materials are chemically attached to the surface of the dispersible oxidized carbon nanotubes.

223. I discussed above for element [1d] that that the presence of oxygen-containing functional groups facilitates the attachment of the lithium ion active material to the nanotube surface, for instance, enabling the formation of covalent bonds. *See* LGES-1010 (Wepasnick), 2 ("In functionalization strategies, the role of oxidation is to create 'foothold' functional groups, such as carboxyl groups, which then serve as attachment points to anchor … nanoparticles"). In the specific context of silicon and oxidized carbon nanotubes, Eom notes that "[i]t is believed that the surface of the Si particles was oxidized by ball-milling, and the chemical-bonded SiC compound was formed on the interface of the SWCNTs and Si particles," where the nanotubes are oxidized ("effectively purified by an acidic treatment followed by the gas-phase oxidation process." LGES-1043 (Eom), 2-3. Through these covalent bonds, the lithium ion active material is **chemically attached** to the surface of the nanotubes.

224. Alternatively/additionally, as I also discussed above for element [1d], Lee describes that "the primary nanoparticles include the non-carbonaceous

material and hollow carbon nanofibers, which are ***bound to the non-carbonaceous material by Van der Waals forces***," *i.e.*, the lithium ion active material and the carbon nanotubes are bound by Van der Waals forces. LGES-1006 (Lee), [0039]; *see* [0040] ("The primary nanoparticles 5 include a non-carbonaceous material 1 and hollow carbon nanofibers 3, which are bound to the non-carbonaceous material by ***Van der Waals forces***"), [0084] ("The resulting anode active material included secondary particles that were formed by the agglomeration of primary nanoparticles including ***Si particles bound to the carbon nanotubes***"), [0014]-[0016], [0043]-[0044], [0063], [0101], Abstract, claim 1, claim 6. To the extent that Van der Waals forces can be considered a chemical attachment (*e.g.*, in that they are attractive chemical interactions between entities), the lithium ion material is **chemically attached** to the nanotubes via Van der Waals forces.

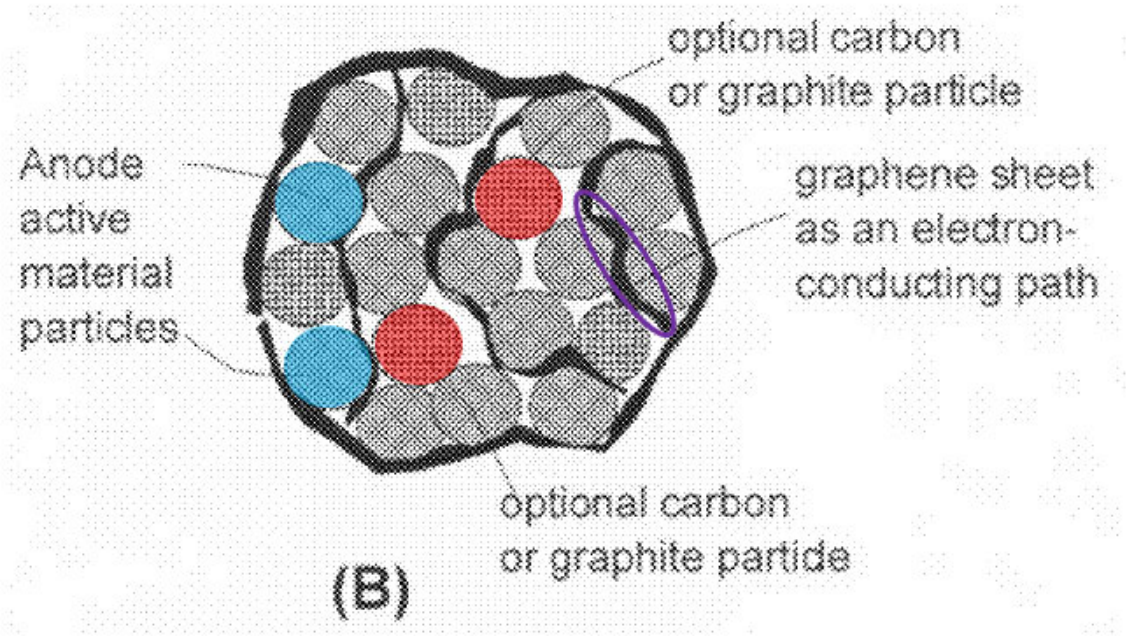
225. Alternatively/additionally, I also discussed above for element [1b] that the lithium ion active material can be attached to the nanotube surface through hydrogen bonding. To the extent hydrogen bonding is considered a chemical attachment, the lithium ion active material is **chemically attached** to the nanotubes by way of hydrogen bonds.

C. [GROUND 2B] – CLAIMS 3 AND 4 ARE OBVIOUS OVER LEE-WEPASNICK-ZHAMU

226. It is my opinion that a POSITA would have found claims 3 and 4 to be obvious over the combination of Lee, Wepasnick, and Zhamu, as I discuss in the following sections.

1. Zhamu describes anode active materials containing graphene

227. Zhamu describes a "nano graphene-enhanced particulate for use as a lithium-ion battery anode active material." LGES-1008 (Zhamu), Abstract. Zhamu's particulates, illustrated in FIG. 3(B), *infra*, includes "a single or a plurality of graphene sheets" (purple) "and a plurality of fine anode active material particles" (blue). *Id.*, [0071]. The particulates also include "a carbon or graphite material" in "the interior of the particulate," in "a fine particle or thin coating form." *Id.*, [0071]. Zhamu's anode active materials allow for "a reversible specific capacity of greater than 1000 mAh/g for longer than 500 cycles" to be achieved, and they "exhibit a high tap density." *Id.*, [0059]. Moreover, "[t]he embracing graphene sheets also appear to be capable of preventing the electrolyte from detrimentally reacting with the anode active material." *Id.*



Excerpt from Zhamu, FIG. 3(B) (annotated)

2. The Lee-Wepasnick-Zhamu combination

228. Lee's anode active material composition includes "[a] conducting agent," and notes that the conducting agent can be "any conducting agent commonly used in lithium batteries," *e.g.*, "***carbonaceous materials, such as natural graphite, artificial graphite***, carbon black, acetylene black, ketchen black, carbon fibers, and the like." LGES-1006 (Lee), [0057]. In the Lee-Wepasnick-Zhamu combination composition, this carbonaceous conducting agent is implemented as graphene based on the teachings of Zhamu. Specifically, the anode active material composition of the combination includes the anode active material of the Lee-Wepasnick combination (*see* [1pre]-[1d], *supra*), which includes a non-carbonaceous material and carbon nanotubes, and "further

include[s] a conducting agent" (LGES-1006 (Lee), [0057]), where the conducting agent is "a single or a plurality of graphene sheets," based on Zhamu. LGES-1008 (Zhamu), [0071]. For instance, the graphene sheets can be "graphene sheets containing no or low oxygen content" or "GO [graphite oxide] of various oxygen contents." LGES-1008 (Zhamu), [0073].

3. Reasons for the combination

229. It is my opinion that a POSITA would have been motivated to implement the conducting agent in the anode active material composition of the Lee-Wepasnick-Zhamu combination as graphene for at least the following reasons.

230. The first reason for using graphene as the conducting agent in the combination is that graphene as a conductive additive offers advantageous thermal management properties, as described by Zhamu. For instance, Zhamu explains that "[g]raphene was recently discovered to exhibit the highest thermal conductivity of all existing materials. In addition to the electrical conductivity, this high thermal conductivity is clearly an advantageous property that could not be achieved by other types of conductive additives." LGES-1008 (Zhamu), [0072]. This high thermal conductivity has implications for battery performance: "A high thermal conductivity implies a high heat dissipation rate. This is an important feature since the charge and discharge operations of a battery produce a great

amount of heat. Without a fast heat dissipation rate, the battery cannot be charged or discharged at a high rate." *Id.*, [0146].

231. The second reason for using graphene as the conducting agent in the combination is that graphene provides structural integrity to the composition: "Graphene sheets are of high strength, high electrical conductivity, and high thermal conductivity. Single layer graphene was recently found to exhibit the highest intrinsic strength and highest intrinsic thermal conductivity among all existing materials. High strength imparts good structural integrity to the resulting electrode." LGES-1008 (Zhamu), [0145]. Particularly in the context of the Lee-Wepasnick composition, in which "cohesion" of the composition is a focus, a POSITA would have been motivated to obtain these further mechanical benefits stemming from the incorporation of graphene. LGES-1006 (Lee), [0053]. Indeed, a POSITA would have recognized the direct applicability of graphene to particle cohesion in that, as described by Zhamu, graphene sheets are "capable of effectively cushioning the large volume changes of electro-active materials such as Si." LGES-1008 (Zhamu), [0151].

232. The third reason for using graphene as the conducting agent in the combination is that graphene has desirable electrical conductivity properties, consistent with Lee's desire to incorporate an electrically conductive additive as the conducting agent. *See* LGES-1006 (Lee), [0057]. For instance, Zhamu states

that "[g]raphene sheets have an electrical conductivity (up to 20,000 S/cm) that is several orders of magnitude higher than that of CB [carbon black] or AB [acetylene black] (typically 0.01-10 S/cm)," and moreover describes that graphene sheets provide "a robust 3-D network of electron-conducting paths and high conductivity." LGES-1008 (Zhamu), [0147]-[0148].

233. The fourth reason for using graphene as the conducting agent in the combination is that the presence of graphene facilitates fabrication of "electrodes with a high tap density and long-term cycling stability." LGES-1008 (Zhamu), [0148].

234. The fifth reason for using graphene as the conducting agent in the combination is that a POSITA would have been prompted to pursue this implementation because doing so is just the application of known techniques (specifically, graphene in an anode active material composition) to an already-known structure (the Lee-Wepasnick anode active material composition), and the combination produces predictable results (specifically, an anode active material composition containing graphene).

235. It is my opinion that a POSITA would have expected success in using graphene as the conducting agent in the Lee-Wepasnick-Zhamu anode active material composition for at least the following reasons.

236. First, a POSITA would have expected success in incorporating graphene into the combination because Lee confirms that "[t]he conducting agent may be any conducting agent commonly used in lithium batteries," and graphene is one such conducting agent. LGES-1006 (Lee), [0057].

237. Second, a POSITA would have expected success in incorporating graphene into the combination because the composition of the particulates of Zhamu's anode active material is generally similar to the composition of the Lee-Wepasnick anode active material composition. For instance, silicon, silicon oxide, or a silicon-metal alloy are identified as suitable active materials for both. *See* LGES-1006 (Lee), [0043]; LGES-1008 (Zhamu), [0041]-[0044]. Moreover, both include further carbon-based additives: the Lee-Wepasnick composition includes carbon nanotubes, while Zhamu's particulates include a further "carbon or graphite material." LGES-1006 (Lee), [0040]; LGES-1008 (Zhamu), [0071].

4. Claim-by-Claim Analysis

(i) Claim 3

[3] The composition of claim 1, further comprising graphene or oxygenated graphene.

238. In the Lee-Wepasnick-Zhamu combination anode active material composition, the "conducting agent" is implemented as "a single or a plurality of graphene sheets," *e.g.*, the combination composition includes **graphene**. LGES-1006 (Lee), [0057]; LGES-1008 (Zhamu), [0039]; *see* Abstract, [0002], [0040],

[0050]. Zhamu also describes use of "graphene oxide," *e.g.*, **oxygenated graphene**. LGES-1008 (Zhamu), [0073] ("Graphene sheets may be oxidized to various extents during their preparation, resulting in graphite oxide (GO) or graphene oxide").

(ii) Claim 4

[4] The composition of claim 3, wherein the weight ratio of graphene to carbon nanotube is in the range of 0.1:99.9 to 99.9:0.1.

239. In the Lee-Wepasnick-Zhamu combination anode active material composition, Lee indicates that "[t]he amount of the conducting agent may be appropriately adjusted, according to an intended application." LGES-1006 (Lee), [0057]; *see* [0064] ("The amounts of the binder and the conducting agent may be appropriately adjusted and are not particularly limited"), [0085], [0091]. In a specific example, Lee describes in Example 1 that the proportions of the anode active material, the conducting agent and the binder were 55% (50% of Si and 5% of carbon nanotubes, by weight), 30%, and 15%, respectively, by weight, excluding the solvent," *e.g.*, the weight ratio of conducting agent (implemented as graphene based on Zhamu) to carbon nanotube is 30:5, or **6:1**. LGES-1006 (Lee), [0085]. This weight ratio (**6:1**) falls within the recited range of weight ratios (0.1:99.9 to 99.9:0.1). I understand that when the prior art discloses a value that falls within a range recited in a claim, that disclosure is relevant to the patentability of the claim, and given this disclosure of Lee, I conclude that this range disclosed

by Lee, and applied to the combination composition, renders obvious a composition having the recited range of weight ratios.

240. I note that in implementing graphene rather than graphite, a POSITA may make slight adjustments to the weight percentage. However, given the broad range of weight ratio values recited in the claim, I do not expect any such adjustments to be significant enough to push the weight ratio of the combination outside of the claimed range.

VII. LEGAL PRINCIPLES

A. Anticipation

241. I have been informed that a patent claim is invalid as anticipated under 35 U.S.C. § 102 if each and every element of a claim, as properly construed, is found either explicitly or inherently in a single prior art reference. Under the principles of inherency, if the prior art necessarily functions in accordance with, or includes the claimed limitations, it anticipates.

242. I have been informed that a claim is invalid under 35 U.S.C. § 102(a) if the claimed invention was known or used by others in the U.S., or was patented or published anywhere, before the applicant's invention. I further have been informed that a claim is invalid under 35 U.S.C. § 102(b) if the invention was patented or published anywhere, or was in public use, on sale, or offered for sale in this country, more than one year prior to the filing date of the patent application

(critical date). And a claim is invalid, as I have been informed, under 35 U.S.C. § 102(e), if an invention described by that claim was described in a U.S. patent granted on an application for a patent by another that was filed in the U.S. before the date of invention for such a claim.

B. Obviousness

243. I have been informed that a patent claim is invalid as "obvious" under 35 U.S.C. § 103 in light of one or more prior art references if it would have been obvious to a POSITA, taking into account (1) the scope and content of the prior art, (2) the differences between the prior art and the claims, (3) the level of ordinary skill in the art, and (4) any so called "secondary considerations" of non-obviousness, which include: (i) "long felt need" for the claimed invention, (ii) commercial success attributable to the claimed invention, (iii) unexpected results of the claimed invention, and (iv) "copying" of the claimed invention by others.

244. I have been informed that a claim can be obvious in light of a single prior art reference or multiple prior art references. To be obvious in light of a single prior art reference or multiple prior art references, there must be a reason to modify the single prior art reference, or combine two or more references, in order to achieve the claimed invention. This reason may come from a teaching, suggestion, or motivation to combine, or may come from the reference or references themselves, the knowledge or "common sense" of one skilled in the art,

or from the nature of the problem to be solved, and may be explicit or implicit from the prior art as a whole. I have been informed that the combination of familiar elements according to known methods is likely to be obvious when it does no more than yield predictable results. I also understand it is improper to rely on hindsight in making the obviousness determination.

VIII. CONCLUSION

245. 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.

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

Dated: 6/7/2024 By: Gu Zhu

APPENDIX A

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PROFESSIONAL HISTORY

The University of Akron – School of Polymer Science and Polymer Engineering

2021/6 – present	Professor
2018/6 – 2021/6	Associate Professor
2012/8 – 2018/6	Assistant Professor
2016/5 - present	Deputy Director, Akron Functional Materials Center
2023/7 -present	Associate Director of School of Polymer Science and Polymer Engineering

Rice University - Department of Chemistry

2008-2012	Postdoctoral Research Associate Advisor: James M. Tour

EDUCATION

Ph.D. in Chemistry, University of Cologne, Germany, 2007
Advisor: Bernd Tieke
"New Diketopyrrolopyrrole(DPP)-Based Conjugated Polymers Prepared upon Palladium Catalyzed Polymerization and Electropolymerization Reactions"
B.S. in Chemical Engineering, Nanjing University of Technology, China, 1997

AWARDS AND HONORS

Outstanding Research Award, University of Akron (2022)
Air-Force Summer Faculty Fellow (2020-2023)
Emerging Young Investigator (Journal of Material Chemistry A) 2017
National Science Foundation (NSF) CAREER award 2015
Beyond Lithium Ion VIII Conference, best poster, 2015
ACS PRF New Doctoral Investigator (DNI) award 2014
Outstanding Undergraduate Student, Nanjing University of Technology, 1997

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PROFESIONAL SOCIETIES

American Chemical Society (ACS): Divisions of Energy and Fuel, Polymer Materials Science & Engineering, Polymer Chemistry,
The Electrochemical Society (ECS)
Materials Research Society (MRS)
American Physical Society (APS)

REFEREED PUBLICATIONS
(> 10500 citations, H-Index 46, *corresponding author)
Google Scholar link:
https://scholar.google.com/citations?hl=en&user=sScmG5wAAAAJ&view_op=list_works&sortby=pubdate

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[70] Kewei Liu, Yanfeng Xia, Chung-fu Cheng, Xuhui Xia, Feng Zou, Bryan D. Vogt* and Yu Zhu* "Microwave Processed, Onion-like Carbon and Fluoropolymer Passivated Lithium Metal Electrode for Enhanced Li Stripping/Plating Performance" *ACS Applied Energy Materials,* 2019, 2, 11, 7933-7941. (impact factor: 6.024, citations=1)

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[38] Sarang M. Bhaway, Yu-Ming Chen, Yuanhao Guo, Pattarasai Tangvijitsakul, Mark D. Soucek, Miko Cakmak, Yu Zhu, Bryan D. Vogt* "Hierarchical Electrospun and Cooperatively Assembled Nanoporous Ni/NiO/MnOx/Carbon Nanofiber Composites for Lithium Ion Battery Anodes" *ACS Applied Materials & Interfaces* 2016, 8 (30), 19484-19493 (impact factor: 9.229, citations=28)

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[22] Yu Zhu, James M Tour* "Organic Light-Emitting Diodes Non-oxide Boost" *Nature Photonics* 2012, 6, 72-73 (impact factor: 38.771, citations=11)

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[20] Bostjan Genorio, Wei Lu, Ayrat M Dimiev, Yu Zhu, Abdul-Rahman O Raji, Barbara Novosel, Lawrence B Alemany, James M Tour* "In Situ Intercalation Replacement and Selective Functionalization of Graphene Nanoribbon Stacks" *ACS Nano* 2012, 6, 4231-4240 (impact factor: 15.881, citations=127)

[19] Zhengzong Sun, Abdul-Rahman O Raji, Yu Zhu, Changsheng Xiang, Zheng Yan, Carter Kittrell, ELG Samuel, James M Tour* "Large-Area Bernal-Stacked Bi-, Tri-, and Tetralayer Graphene" *ACS Nano* 2012, 6, 9790-9796 (impact factor: 15.881, citations=169)

[18] Changsheng Xiang, Wei Lu, Yu Zhu, Zhengzong Sun, Zheng Yan, Chi-Chau Hwang, and James M. Tour* "Carbon Nanotube and Graphene Nanoribbon-Coated Conductive Kevlar Fibers" *ACS Applied Materials & Interfaces* 2012, 4, 131-136. (impact factor: 9.229, citations=85)

[17] Zheng Yan, Jian Lin, Zhiwei Peng, Zhengzong Sun, Yu Zhu, Lei Li, Changsheng Xiang, E Loïc Samuel, Carter Kittrell, James M Tour* "Toward the Synthesis of Wafer-Scale Single-Crystal Graphene on Copper Foils" *ACS Nano* 2012, 6, 9110-9117 (impact factor: 15.881, citations=629)

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[16] Zheng Yan, Jun Yao, Zhengzong Sun, Yu Zhu, James M Tour* "Controlled Ambipolar-to-Unipolar Conversion in Graphene Field-Effect Transistors Through Surface Coating with Poly(ethylene imine)/Poly(ethylene glycol) Films" *Small* 2012, 8, 59-62 (impact factor: 13.281, citations=36)

[15] Yu Zhu, Zhengzong Sun, Zheng Yan, Zhong Jin, James M Tour* "Rational Design of Hybrid Graphene Films for High-Performance Transparent Electrodes" *ACS Nano* 2011, 5, 6472-6479 (impact factor: 15.881, citations=314)

[14] Yu Zhu, Wei Lu, Zhengzong Sun, Dmitry V Kosynkin, Jun Yao, James M Tour "High Throughput Preparation of Large Area Transparent Electrodes Using Non-Functionalized Graphene Nanoribbons" *Chemistry of Materials* 2011, 23, 935-939 (impact factor: 9.811, citations=29)

[13] Zhengzong Sun, Cary L Pint, Daniela C Marcano, Chenguang Zhang, Jun Yao, Gedeng Ruan, Zheng Yan, Yu Zhu, Robert H Hauge, James M Tour* "Towards Hybrid Superlattices in Graphene" *Nature Communications* 2011, 2,559 (impact factor: 14.919, citations=146)

[12] Zheng Yan, Zhiwei Peng, Zhengzong Sun, Jun Yao, Yu Zhu, Zheng Liu, Pulickel M Ajayan, James M Tour* "Growth of Bilayer Graphene on Insulating Substrates" *ACS Nano* 2011, 5, 8187-8192 (impact factor: 15.881, citations=304)

[11] Zheng Yan, Zhengzong Sun, Wei Lu, Jun Yao, Yu Zhu, James M Tour* "Controlled Modulation of Electronic Properties of Graphene by Self-Assembled Monolayers on SiO2 Substrates" *ACS Nano* 2011, 5, 1535-1540 (impact factor: 15.881, citations=112)

[10] Bernd Tieke*, A Raman Rabindranath, Kai Zhang, Yu Zhu "Conjugated Polymers Containing Diketopyrrolopyrrole Units in The Main Chain" *Beilstein Journal of Organic Chemistry* 2010, 6, 830-845 (impact factor: 2.883, citations=158)

[9] Yu Zhu, James M Tour* "Graphene Nanoribbon Thin Films Using Layer-by-Layer Assembly" *Nano Letters* 2010, 10, 4356-4362. (impact factor: 11.189, citations=77)

[8] Zhengzong Sun, Zheng Yan, Jun Yao, Elvira Beitler, Yu Zhu, James M Tour* "Growth of Graphene from Solid Carbon Sources" *Nature* 2010, 468, 549-552 (impact factor: 49.962, citations=1404)

[7] Yu Zhu, Amanda L Higginbotham, James M Tour* "Covalent Functionalization of Surfactant-Wrapped Graphene Nanoribbons" *Chemistry of Materials* 2009, 21, 5284-5291 (impact factor: 9.811, citations=150)

[6] Zhong Jin, Jay R Lomeda, B Katherine Price, Wei Lu, Yu Zhu, James M Tour* "Mechanically Assisted Exfoliation and Functionalization of Thermally Converted Graphene Sheets" *Chemistry of Materials 2009*, 21, 3045-3047 (impact factor: 9.811, citations=104)

[5] Yu Zhu, Kai Zhang, Bernd Tieke* "Electrochemical Polymerization of Bis(3,4-ethylenedioxythiophene)-Substituted 1,4-Diketo-3,6-diphenyl-pyrrolo 3,4-c pyrrole (DPP) Derivative" *Macromolecular Chemistry and Physics* 2009, 210, 431-439 (impact factor: 2.527, citations=37)

[4] A Raman Rabindranath, Yu Zhu, Kai Zhang, Bernd Tieke* "Purple Red and Luminescent Polyiminoarylenes Containing The 1, 4-Diketo-3,6-diphenylpyrrolo[3,4-c]pyrrole (DPP) Chromophore" *Polymer* 2009, 50, 1637-1644 (impact factor: 4.430, citations=32)

[3] Yu Zhu, A Raman Rabindranath, Thomas Beyerlein, Bernd Tieke* "Highly Luminescent 1, 4-Diketo-3,6-diphenylpyrrolo[3,4-c]pyrrole-(DPP-) Based Conjugated Polymers Prepared upon Suzuki Coupling" *Macromolecules* 2007, 40, 6981-6989 (impact factor: 5.985, citations=167)

[2] Yu Zhu, Ingo Heim, Bernd Tieke* "Red Emitting Diphenylpyrrolopyrrole (DPP)-based Polymers Prepared by Stille and Heck Coupling" *Macromolecular Chemistry and Physics* 2006, 207, 2206-2214 (impact factor: 2.527, citations=63)

[1] A Raman Rabindranath, Yu Zhu, Ingo Heim, Bernd Tieke*, "Red Emitting N-Functionalized Poly(1,4-diketo-3,6-diphenylpyrrolo[3,4-c]pyrrole)(Poly-DPP): A Deeply Colored Polymer with Unusually Large Stokes Shift" *Macromolecules* 2006, 39, 8250-8256 (impact factor: 5.985, citations=110)

PRESENTATIONS

Invited Seminars and invited presentation in conferences

1) "Novel Metal Complex Catholytes for Aqueous Redox Flow Batteries" MRS Meeting Spring, April 24, 2024, Seattle.
2) "Novel Metal Complex Catholytes for Aqueous Redox Flow Batteries" International Battery Seminar & Exhibit, 2023, March 20-23, 2023, Orlando.
3) "Toward Superionic Conductive Polymer Electrolytes for Lithium Ion Batteries" Oak Ridge National Laboratory Soft Matter Symposium 2022, October 27, 2022
4) "Molecular Packing of π-Conjugated Molecules through Fused Hydrogen Bond-mediated Self-assembly", Polymer Physics & Polymer Spectroscopy Webinar, University of Halle, Germany, June, 25, 2020

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5) "Functional Polymers for Energy and Electronics Applications" Michigan State University, East Lansing, MI, January 9, 2020
6) "Highly-conductive, Low-cost Polymer Adhesive Composites" 2019 DOE DuraMAT Workshop, Denver, August 27, 2019
7) "Impact-resistant electrolyte with nanofillers for safe Li-ion batteries"2019 Fall ACS National Meeting, San Diego, 2D Materials & Beyond: Innovative Materials, Assemblies & Devices for Energy & Fuel, August, 26, 2019
8) "Polymer-templated, Porous Composites for Energy Storage" 2019 Fall ACS National Meeting, San Diego, ENFL: Atomic-level Understanding & Design of Materials & Processes for Energy Applications, August 25, 2019
9) "Porous Composites for Electrochemical Energy Storage"63 ANNUALMAY CONFERENCE Microscopy Society of NE Ohio, May 22, 2019
10) "High-performance, Transition-Metal Phosphide Catalysts for Oxygen-evolution Reaction" 2019 Spring ACS National Meeting, Orlando, ENFL: Applied Electrocatalysis for Renewable Energy & Synthesis, April 1, 2019
11) "Novel Organic Dye based Redox Molecules for Aqueous Organic Redox Flow Batteries" 2019 Annual Meeting-International Coalition for Energy Storage and Innovation (IECSI-PPSS), January 6, 2019
12) "Novel Electrically Conductive Adhesive Materials with Multiple Fillers" 2018 DOE DuraMAT Workshop, SLAC, August 29, 2019
13) "Metal Organic Frameworks (MOFs) Derived Nanostructures for Electrochemical Energy Storage" 2018 Fall ACS National Meeting, Boston, 2D Materials: Innovative Materials and Devices for Energy and Fuel, August 20, 2018
14) University of Akron, Polymers Appreciation Days and the College of Polymer Science and Polymer Engineering's 30th Anniversary Celebration, "Conjugated Polymers with Fused Hydrogen Bond", May 10, 2018
15) "Phase Diagram Guided Rational Design of Superionic Conductive, Electrochemically Stable Dual-salt Polymer Electrolyte ", 2018 Annual Meeting-International Coalition for Energy Storage and Innovation (ICESI), January 17, 2018
16) Nanjing University of Technology, Department of Chemical Engineering "Design and Fabrication of Well-organized Nanomaterials for Energy Applications", January 14, 2018
17) "Design and Fabrication of Well-organized Carbon Nanomaterials for Energy Applications"American Carbon Society Graphene Symposium/Workshop November 15, 2017
18) "Highly-conductive, Low-cost Polymer Adhesive Composites with Complex Dimensional Fillers" 2017 DOE DuraMAT, BAPVC BAPVC Workshop, November 13, 2017

19) "High Ionic Conductivity Solid Polymer Electrolyte (SPE) for All-Solid-State Li-Ion Battery" MRS 2017 Fall Meeting, November 28, 2017,
20) "Advanced Silicon Composite Anodes for Lithium Ion Battery", International Workshop on Thin-Films for Electronics, Electro-Optics, Energy & Sensors: TFE3S 2017, Dayton, OH, June 26, 2017
21) "2D Conductive Metal Organic Framework as Sulfur Host for Li-S Batteries", 254th ACS National Meeting, Washington DC, August 22, 2017
22) "A Dual-salt Superionic Conductive Polymer Electrolyte for Solid State Lithium Ion Battery"Beyond Lithium Ion Symposium-10 (BLI-X), June 27, 2017
23) Case Western Reserve University, Department of Macromolecular Science and Engineering " Design and Fabrication of Materials with Organized Structure for Organic Electronics and Energy Applications", April 21, 2017
24) Northeastern University, Department of Mechanical & Industrial Engineering, " Design and Fabrication of Well-organized Nanomaterials for Energy Applications" April 12, 2017
25) Tufts University, Department of Chemistry, "Molecular Packing of π-Conjugated Small Molecules and Polymers through Fused Hydrogen Bond-mediated Self-assembly", April 11, 2017
26) National Tsing Hua University, Taiwan, Department of Chemical Engineering, "Design and Fabrication of Well-organized Nanomaterials for Energy Applications", Mar. 29, 2017
27) National Chiao Tung University, Taiwan, Department of Applied Chemistry, " Design and Fabrication of Materials with Organized Structure for Organic Electronics and Energy Storage Applications", Mar. 28, 2017
28) Taxes A&M University, College Station, Department of Chemistry, "Design and Fabrication of Well-organized Nanomaterials for Energy Applications", Mar. 24, 2017
29) Case Western Reserve University, Department of Chemistry "Design and Fabrication of Well-organized Nanomaterials for Energy Applications", Mar. 9, 2017
30) NIST, Material Measurement Laboratory, "Molecular Packing of π-Conjugated Small Molecules and Polymers through Fused Hydrogen Bond-mediated Self-assembly", Feb. 17, 2017
31) University of Houston, Department of Electrical and Computer Engineering, "Design and Fabrication of Well-organized Nanomaterials for Energy Applications", Jan. 27, 2017
32) Rice University, Department of Chemistry, "Design and Fabrication of Well-organized Nanomaterials for Energy Applications", Jan. 25, 2017
33) Hydrogen Bonding Assembled Organic 2D Semiconductors", 252nd ACS National Meeting, Philadelphia PA, Aug. 25, 2016

34) "Patternable conjugated polymers with latent hydrogen-bonding on the main chain", 47th ACS Central Regional Meeting, May 19, 2016
35) "Controlled Synthesis of Nanomaterials for Novel Applications" ACS Akron Section Annual Meeting, May 11, 2016
36) "Controlled Synthesis of Nanomaterials for Novel Applications", APS Akron Physics Club Annual Meeting, April 25, 2016
37) Binghamton University, Department of Physics, "Design and Fabrication of Materials with Organized Structure for Organic Electronics and Energy Storage Applications" Nov. 16, 2016
38) Kenyon College, Department of Chemistry, "Controlled Synthesis of Nanomaterials and Their Application in Energy Storage" Nov. 10, 2016
39) Kent State University, Department of Chemistry, "Controlled Synthesis of Nanomaterials and Their Application in Energy Storage", Nov. 4, 2016
40) Sandia National Lab, Advanced Materials Laboratory, "Synthesis and fabrication of functional (nano)materials from applications in energy", June 16, 2016
41) Pacific Northwest National Lab, Electrochemical Materials & Systems Group "Advanced Polymers for Electrochemical Energy Storage", May 27, 2016
42) "Hybrid Nanomaterials for Supercapacitor Application" 228th ECS National Meeting, San Diego CA, Oct. 12, 2015
43) "Hybrid 2D Materials for Electrochemical Energy Storage Applications", 249th ACS National Meeting, Denver CO, Mar. 24, 2015
44) Central Michigan University, Department of Physics "Controlled Synthesis of Nanomaterials and Their Application in Energy Storage", Sept. 15, 2015
45) PolyOne Corporation "Controlled synthesis and Assembly of Carbon Based Materials for New Applications", Aug. 16, 2013
46) John Carroll University, Department of Chemistry, "Controlled Synthesis and Assembly of Carbon Nanomaterials for New Applications", Feb. 18, 2015
47) GrafTech International Inc, "Nanocarbon Materials for Energy Storage", April 14, 2015
48) "Macro Porous Graphene from Hollow Ni Templates via Polymer Templates with Bi-continuous Structure", 248th ACS National Meeting, San Francisco CA, Aug. 14, 2014
49) "Hybrid 2D Materials for Energy Conversion Device", 247th ACS National Meeting Spring, Dallas TX, Mar. 19, 2014
50) NASA Glenn Research Center, "Controlled Synthesis and Assembly of Carbon Based Nanomaterials for New Applications", July 8, 2014
51) University of Cologne, Department of Physical Chemistry "Controlled Synthesis and Assembly of Carbon Nanomaterials for New Applications", Jan. 24, 2014

Contributed presentation (oral presentation or poster, presenter's name is underlined).

1) Clinton Taubert, Kun Chen, Bryan Vogt, Hongyou Fan, Yu Zhu "Highly-Conductive, Low-Cost Polymer Adhesive Composites with Complex Dimensional Fillers", PV Reliability Workshop, Denver, February 27, 2019, poster (Best Poster Award)
2) Feng Zou, Yu Zhu, "Metal Organic Frameworks Derived Hierarchical Hollow NiO/Ni/Graphene Composites for Lithium and Sodium Storage" 18th International Meeting on Lithium Batteries, June 19-24, 2016, poster.
3) Kewei Liu, Yu Zhu, "A Surface Enhanced Raman Spectroscopy (SERS)-Active Cathode for Lithium-Oxygen Battery Discharge Product Investigation" 18th International Meeting on Lithium Batteries, June 19-24, 2016, poster
4) Yu Zhu, "Advanced Polymer Binder for Silicon Anodes Based High Energy Density Lithium Ion Battery" 18th International Meeting on Lithium Batteries June 19-24, 2016, poster (Best Poster Award)
5) Yu-Ming Chen, Yu Zhu, "Utilization of Nitrogen Doped Carbonized Metal Organic Framework for High Stability Room Temperature Sodium-Sulfur Battery" Carbon Conference 2016, PA, July 10-15, 2016, presentation
6) Kewei Liu, Yu Zhu, "Three-Dimensional Bi-Continuous Graphene Monolith from Polymer Templates" Carbon Conference 2016, PA, July 10-15, 2016, presentation
7) Kun Yang, Yu Zhu, "Diketopyrrolopyrrole-based oligophenylenethiophenes with latent hydrogen bonding for solution-processed organic field-effect transistors", 252nd ACS National Meeting, Philadelphia PA, August 20-25, 2016, poster
8) Yu-Ming Chen and Yu Zhu, "Utilization of nitrogen doped carbonized metal organic framework for high stability room temperature sodium-sulfur battery", 252nd ACS National Meeting, Philadelphia PA, August 20-25, 2016, presentation
9) Yu Zhu "Porous Graphene from Polymer Templates and its Use in Energy Storage Applications" Gordon Research Conferences "Nanomaterials for Applications in Energy Technology", Feb. 22, 2015, poster
10) Yu Zhu "Improving the cyclability of lithium-air battery with small molecule solvent and free-standing carbon nanotube electrode" Gordon Research Conferences "Batteries", Mar. 9, 2014, poster
11) Yu Zhu "Advanced Polymer Binder for Silicon Anodes Based High Energy Density Lithium Ion Battery" Gordon Research Conferences "Batteries", Feb. 11-16 2016, poster
12) Yu Zhu "Photo-patterning of Conjugated Polymers with Latent Hydrogen Bonding" Gordon Research Conferences "Polymer", July 13-18, 2014, poster
13) Yu Zhu "Patternable Conjugated Polymers with Latent Hydrogen–bonding on the Main Chain" Gordon Research Conferences "Macromolecular Materials", Jan. 11, 2015, poster
14) Yu Zhu "Conjugated Polymers and Small Molecules with Hydrogen-bonding on the Main-Chain" Gordon Research Conferences "Polymer", June. 14, 2015, poster

15) Yu Zhu "Conjugated Polymers and Small Molecules with Hydrogen-bonding on the Main-Chain" Gordon Research Conferences "Polymer", July. 24-29, 2016, poster
16) Kewei Liu and Yu Zhu, "Bi-continuous Graphene Monolith via Polymer Template as High-performance Electrode for Lithium-oxygen Battery", Beyond Lithium Ion Conference VII, June 3-4, 2014, poster
17) Kewei Liu, Yu-Ming Chen and Yu Zhu, "Investigation of Lithium-Air Batteries with Bi-Continuous Graphene Cathode", Beyond Lithium Ion Conference VIII, June 2-4, 2015, poster
18) Feng Zou and Yu Zhu, "Advanced Polymer Binder for Silicon Anodes Based High Energy Density Lithium Ion Battery", Beyond Lithium Ion Conference IX, May 24-26, 2016, poster

INTELLECTUAL PROPERTY

Patents

1) Yu Zhu, Xiang Li, Asymmetric Metal Complex as Active Material of Flow Battery (USPTO: 63/080,374)
2) Yu Zhu, Kewei Liu, Impact Resistant Electrolyte Enabling Safe Li-Ion Batteries (USPTO 62/717,292)
3) Yu Zhu, High Ionic Conductivity Solid Polymer Electrolyte Composite (US20190379034)
4) Hongyou Fan, Yu Zhu, Self-Assembly Method for Core/Shell Nanoparticles with Enhanced Emission (USPTO 62/509,389)
5) Yu Zhu, Feng Zou, Crosslinked Polymer Binders for Electrochemical Energy Storage Devices (US 20200350588 A1)
6) Yu Zhu, Tianda He Conductive Film and Method of Making Same (WO 2015109137 A1)
7) James M. Tour, Yu Zhu Transparent electrodes based on graphene and grid hybrid structures, WO 2011112589 (2011)
8) James M. Tour, Yu Zhu Graphene-based thin films in heat circuits and methods of making the same, WO 2012100178 and US 20120208008 (2012) (Licensed)
9) James M. Tour, Yu Zhu Graphene nanoribbon composites and methods of making the same, WO2012112435 (2012) (Licensed)
10) Mathieu G. R. Turbiez, René Albert Johan Janssen, Martinus Maria Wienk, Hans Jürg Kirner, Mathias Düggeli, Bernd Tieke, Yu Zhu Diketopyrrolopyrrole polymers as organic semiconductors. WO2008000664 (2008)

PROFESSIONAL AND SCIENTIFIC SERVICE

Editorial Service

- Editor-in-Chief, Colloidal and Polymer Science, Springer-Nature Publisher (since 2021)

Proposal Reviewer Service

- Department of Energy (BES, 2023)
- National Science Foundation Division of Civil, Mechanical, and Manufacturing Innovation (review panel 2015)
- National Science Foundation Division of Chemical, Bioengineering, Environmental, and Transport Systems (ad hoc 2018, 2019,2023,2024, review panel 2018, Career panel 2019)
- National Science Foundation Division of Materials Research (review panel 2015, ad hoc 2015,2016,2017, 2018, 2019, 2020, 2021, 2022)
- Army Research Office (2024)
- Petroleum Research Fund (ad hoc 2015, 2018, 2019, 2021)
- American Association for the Advancement of Science (ad hoc 2016, remote panel 2019)
- Deutsche Forschungsgemeinschaft, DFG (ad hoc 2015, 2017, 2019, 2024)
- Israel Science Foundation (ad hoc 2015)
- New University Researchers Start-Up Program of the Fonds de recherche du Québec (ad hoc 2013)

National Technical Service

- Chair "Polymer Processing Society" Conference 2014.
- Co-Chair "2D Materials for Energy Storage" at ACS National Meeting, 2014 Spring Meeting
- Co-Chair "2D Materials for Energy Storage" at ACS National Meeting, 2014 Fall Meeting,
- Co-Chair "2D Materials for Energy Storage" at ACS National Meeting, 2015 Fall Meeting,
- Symposium Organizer "2D Materials: Graphene and Beyond, and Their Device Applications" at ACS National Meeting, 2016 Fall Meeting.
- Symposium Organizer "2D Materials for Energy and Fuel" at ACS National Meeting, 2017 Fall Meeting.
- Co-Chair "Li-ion Battery and Beyond" at ECS National Meeting, 2017 Spring Meeting
- Symposium Organizer "2D Materials Symposium: Innovative Materials and Devices for Energy and Fuel" at ACS National Meeting, 2018 Fall Meeting.

- Symposium Organizer "2D Materials and Beyond: Innovative Materials, Assemblies, and Devices for Energy and Fuel" at ACS National Meeting, 2019 Fall Meeting.

Journal Referee Service

Science, Nature Energy, Nature Communications, Science Advnaces, Chemical Society Review, Journal of the American Chemical Society, ACS Nano, Nano Letters, Nano Energy, Advanced Materials, Advanced Functional Materials, Advanced Energy Materials, Advanced Materials Technologies, Advanced Electronic Materials, Journal of Material Chemistry A, Journal of Material Chemistry C, Nano Research, Nanoscale, Polymer Chemistry, Applied Surface Science , Chemistry of Materials, Carbon, Macromolecules, Macromolecular Chemistry and Physics, Chemical Communications, Chemistry A European Journal, Tetrahedron, ACS Applied Materials and Interfaces, Polymer, Journal of Applied Polymer Science, Scientific Reports, Journal of Physical Chemistry, Journal of Applied Physics, Chemelectrochem , Journal of Power Sources, ACS Applied Energy Materials, Energy Storage Materials, ACS Applied Nano Materials, ACS Applied Polymer Materials.

APPENDIX B

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CALCULATION 1: Based on Wepasnick's Fig. 6

HNO3 %	Atomic % O	Atomic % C	**Weight % oxygen**
		(= 100 - at.% O)	(16 * at.%O / (16 * at.%O + 12 * at.% C)) *100
70%	9.5	90.5	**12.3**
50%	8.1	91.9	**10.5**
30%	5.3	94.7	**6.9**
20%	4.3	95.7	**5.7**
Pristine	1	99	**1.3**

Atomic mass O: 16 g/mol
Atomic mass C: 12 g/mol

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CALCULATION 2: Based on Wepasnick's Fig. 7 (and Fig. 6, for total %O values)

Nitric Acid Concentration	%O[Total]	% O[C-OH]	%O[COOH]	%O[C=O]	Atomic % C	Atomic % functionalized carbon atoms	Atomic % unfunctionalized carbon atoms
					(= 100 - at.% O)	%O[C-OH] + 0.5 * %O[COOH] + %O[C=O]	100 - at.% functionalized carbon atoms
0							
20%	4.3	0.8	1.9	0.9	95.7	2.65	93.05
30%	5.3	0.6	2.4	0.8	94.7	2.6	92.1
50%	8.1	1.2	4.3	1.4	91.9	4.75	87.15
70%	9.5	1.6	3.5	1.9	90.5	5.25	85.25

Atomic mass O: 16 g/mol
Atomic mass C: 12 g/mol

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CALCULATION 2 (continued)

Nitric Acid Concentration	Unfunctionalized C in 100 mol sample	OH weight in 100 mol sample	COOH weight in 100 mol sample	C=O weight in 100 mol sample	**Weight % functional groups**
	12 * % unfunct. C	(12 + 16) * %O[C-OH]	(0.5*12 + 16) * %O[COOH]	(12 + 16) * %O[C=O]	100*(OH, COOH, C=O) / (OH, COOH, C=O + unfunctionalized C)
0					
20%	1116.6	22.4	41.8	25.2	**7.4**
30%	1105.2	16.8	52.8	22.4	**7.7**
50%	1045.8	33.6	94.6	39.2	**13.8**
70%	1023	44.8	77	53.2	**14.6**

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CALCULATION 3: Based on Wepasnick's Fig. 9(a)

Oxidant	Atomic % O	Atomic % C	**Weight % oxygen**
		(= 100 - at.% O)	(16 * at.%O / (16 * at.%O + 12 * at.% C)) *100
KMnO4	5.3	94.7	**6.9**
H2SO4/HNO3	5.1	94.9	**6.7**
HNO3	5.5	94.5	**7.2**
O3	4.7	95.3	**6.2**
H2O2	4.5	95.5	**5.9**
(NH4)2S2O8	4.5	95.5	**5.9**

Atomic mass O: 16 g/mol
Atomic mass C: 12 g/mol

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CALCULATION 4: Based on Wepasnick's Fig. 9(a)

Oxidant	%O[Total]	% O[C-OH]	%O[COOH]	%O[C=O]	Atomic % C	Atomic % functionalized carbon atoms	Atomic % unfunctionalized carbon atoms
					(= 100 - at.% O)	%O[C-OH] + 0.5 * %O[COOH] + %O[C=O]	100 - at.% functionalized carbon atoms
KMnO4	4.5	0.5	1.6	1	95.5	2.3	93.2
H2SO4/HNO3	4.5	0.8	1.7	2.1	95.5	3.75	91.75
HNO3	4.7	0.9	1.8	2	95.3	3.8	91.5
O3	5.5	0.5	2.6	0.9	94.5	2.7	91.8
H2O2	5.4	0.3	2.6	0.9	94.6	2.5	92.1
(NH4)2S2O8	5.3	0.5	3.7	1	94.7	3.35	91.35

Atomic mass O: 16 g/mol
Atomic mass C: 12 g/mol

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CALCULATION 4 (continued)

Oxidant	Unfunctionalized C in 100 mol sample	OH weight in 100 mol sample	COOH weight in 100 mol sample	C=O weight in 100 mol sample	**Weight % functional groups**
	12 * % unfunct. C	(12 + 16) * %O[C-OH]	(0.5*12 + 16) * %O[COOH]	(12 + 16) * %O[C=O]	100*(OH, COOH, C=O) / (OH, COOH, C=O + unfunctionalized C)
KMnO4	1118.4	14	35.2	28	**6.5**
H2SO4/HNO3	1101	22.4	37.4	58.8	**9.7**
HNO3	1098	25.2	39.6	56	**9.9**
O3	1101.6	14	57.2	25.2	**8.0**
H2O2	1105.2	8.4	57.2	25.2	**7.6**
(NH4)2S2O8	1096.2	14	81.4	28	**10.1**

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CALCULATION 5: Based on Wepasnick's Fig. 9(a)

Oxidant	At. %O	% O(C-OH)	%O(COOH)	%O(C=O)	%O(Residual)	Atomic % C	Atomic % functionalized carbon atoms	Atomic % unfunctionalized carbon atoms
						(= 100 - at.% O)	%O[C-OH] + 0.5 * %O[COOH] + %O[C=O]	100 - at.% functionalized carbon atoms
KMnO4	4.5	0.5	1.6	1	1.2	95.5	2.3	93.2
H2SO4/HNO3	4.5	0.8	1.7	2.1	0.1	95.5	3.75	91.75
HNO3	4.7	0.9	1.8	2	0.1	95.3	3.8	91.5
O3	5.5	0.5	2.6	0.9	1.3	94.5	2.7	91.8
H2O2	5.4	0.3	2.6	0.9	1.1	94.6	2.5	92.1
(NH4)2S2O8	5.3	0.5	3.7	1	0	94.7	3.35	91.35

Atomic mass O: 16 g/mol
Atomic mass C: 12 g/mol

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CALCULATION 5 (continued)

Oxidant	Unfunctionalized C in 100 mol sample	OH weight in 100 mol sample	COOH weight in 100 mol sample	C=O weight in 100 mol sample	Residual O weight in 100 mol sample	**Weight % functional groups**
	12 * % unfunct. C	(12 + 16) * %O(C-OH)	(0.5*12 + 16) * %O(COOH)	(12 + 16) * %O(C=O)	16 * %O(Residual)	100*(OH, COOH, C=O, Residual) / (OH, COOH, C=O, Residual + unfunctionalized C)
KMnO4	1118.4	14	35.2	28	19.2	**7.9**
H2SO4/HNO3	1101	22.4	37.4	58.8	1.6	**9.8**
HNO3	1098	25.2	39.6	56	1.6	**10.0**
O3	1101.6	14	57.2	25.2	20.8	**9.6**
H2O2	1105.2	8.4	57.2	25.2	17.6	**8.9**
(NH4)2S2O8	1096.2	14	81.4	28	0	**10.1**

Atomic mass O: 16 g/mol
Atomic mass C: 12 g/mol

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CALCULATION 6: Based on Wepasnick's Fig. 5

	Atomic % O	Atomic % C	Weight % oxygen
		(= 100 - at.% O)	(16 * at.%O / (16 * at.%O + 12 * at.% C)) *100
	4	96	**5.3**
	6	94	**7.8**
	7	93	**9.1**
	10	90	**12.9**
	12	88	**15.4**

Atomic mass O: 16 g/mol
Atomic mass C: 12 g/mol

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