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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 multiple 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 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. 8,968,483 ("the '483 Patent") and prior art references relating to its subject matter. I have reviewed the '483 Patent, and relevant excerpts of the prosecution history of the '483 Patent. I have also reviewed the following references:

LGES-1001	U.S. Patent No. 10,153,483 ("the '483 Patent")
LGES-1002	Excerpts from Prosecution History of the '483 Patent ("the Prosecution History")
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 Chinese Patent Pub. No. CN 101364643A

LGES-1007 Certified English Translation of CN 101364643 (LGES-1006) and Translator's Declaration ("Gong")

LGES-1008 Chinese Patent Pub. No. CN 102456881A

LGES-1009 Certified English Translation of CN 102456881A (LGES-1008) and Translator's Declaration ("Liu")

LGES-1010 Chinese Patent Pub. No. CN 101714627A

LGES-1011 Certified English Translation of CN 101714627A (LGES-1010) and Translator's Declaration ("Cheng")

LGES-1012 Int'l Patent Pub. No. WO 2011056290 ("Peng")

LGES-1013 U.S. Patent No. 10,608,282 ("the '282 Patent")

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-1016 U.S. Patent Pub. No. 2010/0124713 ("Farrugia")

LGES-1017	"Chemical oxidation of multiwalled carbon nanotubes," V. Datsyuk et al. Carbon 46 (2008) 833-840 ("Datsyuk")
LGES-1024	U.S. Patent Pub No. 2009/0035469 ("Sue")
LGES-1025	U.S. Patent No. 8,241,794 ("Sheem")
LGES-1026	U.S. Patent Pub. No. 2011/0159366A1 ("Nakura")
LGES-1027	"A Lithium-Ion Cell Based on a Sodium Insertion Material." J. Barker et al., Electrochemical and Solid-State Letters, 9 (4) A190-A192 (2006) ("Barker")
LGES-1028	"A multifunctional 3.5 V iron-based phosphate cathode for rechargeable batteries," Nature Materials 6 (2007), 749-753 ("Nazar 2007")
LGES-1029	"Crystal Structure and Electrochemical Properties of A_2MPO_4F Fluorophsphates (A = Na, Li; M = Fe, Mn, Co, Ni)," Brian L. Ellis et al., Chem. Mater, 2010, 22, 1059-1070 ("Nazar 2010")
LGES-1030	"Attachment of Metal Oxide Nanoparticles on Carbon Nanotubes with Different Oxidation Levels," C.T. Hsieh et al. Advanced Materials Research, Vols. 55-57, pp. 545-548 ("Hsieh")
LGES-1031	"Incorporation of MWCNTs into leaf-like CuO nanoplates for superior reversible Li-ion storage," J.Y. Xiang et al., Electrochemistry Communications 12 (2010) 1103-1107 ("Xiang")
LGES-1032	"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-1033 Wikipedia: The Free Encyclopedia, "Chemical bond" at https://en.wikipedia.org/wiki/Chemical_bond, last accessed 6/7/2024

LGES-1034 U.S. Patent Pub. No. 2006/0269842 ("Ichinose")

LGES-1035 "Functionalization of Single-Walled Carbon Nanotubes," A. Hirsch, *Angew. Chem. Int. Ed.* 2002, 41, No. 11, 1853-1859 ("Hirsch")

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-1050 *Molecular Rebar Design, LLC, and Black Diamond Structures, LLC v. LG Energy Solution, Ltd.*, Plaintiffs' Disclosure of Asserted Claims and Infringement, 2:24-cv-00107-JRG

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 a person of ordinary skill in the art related to the '483 Patent at the time of the priority date of the '483 Patent, and I have done so during my review of these materials. At the outset, I noted that the earliest *possible* priority date for the '483 Patent would be June 23, 2011, based on the filing date of the

provisional application that the ’483 Patent claims priority to. However, Counsel has informed me that the *actual* priority date to which the ’483 Patent is entitled priority (“Critical Date”) can be no earlier than June 21, 2012, because that’s the first time that applicant added disclosure regarding the specifically claimed ratios, for instance, limitations [1c] and [1f]. I have therefore used that Critical Date in my analysis below. However, applying the June 23, 2011 date would not have altered my analysis.

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 ’483 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 ’483 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 1-4 and 7-18 are obvious over Ohata in view of Kavan, in further view of Gong or Liu; and
- Claims 1, 2, 7-18 are obvious over Ohata in view of Kavan, in further view of Ichinose.

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 ’483 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 '483 PATENT

13. The '483 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 '483 Patent. *See*, *e.g.*, LGES-1004 (Ohata), [0004]; LGES-1005 (Kavan), Abstract. 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). As further detailed below, claim 1's feature of "wherein a ratio of lithium ion to other total ions in the ion active materials is at least 2:1 to 25:1" is rendered obvious by the Ohata-Kavan-Gong/Ohata-Kavan-Liu combinations, among others.

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-1008 (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-1017 (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-1017 (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. INTERPRETATIONS OF THE '483 PATENT CLAIMS AT ISSUE

18. I understand that, for purposes of my analysis in this *inter partes* review proceeding, the terms appearing in the patent claims should be interpreted "using the same claim construction standard that would be used to construe the claim in a civil action under 35 U.S.C. 283(b)...." 37 C.F.R. § 42.100(b). In that regard, I understand that the best indicator of claim meaning is its usage in the context of the patent specification as understood by a POSITA. I further understand that the words of the claims should be given their plain meaning unless that meaning is inconsistent with the patent specification or the patent's history of examination before the Patent Office. I also understand that the words of the claims should be interpreted as they would have been interpreted by a POSITA at the time of the invention was made

A. "ratio of lithium ion to other total ions in the ion active materials"

19. It is my opinion that the claim term "a ratio of lithium ion to other total ions in the ion active materials," which appears in claims 1, 7-9, and 11-18, should be interpreted as a ratio of lithium ions to other migratable ions in the ion active materials. I believe this to be the proper interpretation of this term for several reasons.

20. For instance, I note that the ***only*** place in the specification of the ’483 Patent where the specifically recited ratio values, namely 2:1, 3:1, 10:1, and 25:1, appear is the following passage: “When more than one ion is used, then the ratio of Li ion to other total ions is preferably at least 2:1, or 3:1 or 10:1 and can be as high as 25:1 (mole ratios).” LGES-1001, 4:57-59. Together with the preceding sentence in the specification where the patent talks about using a “migratable ion mixture” where multiple migratable ions (including lithium and other ions) are used as the migratable ion, these passages make clear to a POSITA that the “ratio” associated with the “2:1, 3:1, 10:1, and 25:1” ratio values is one of lithium ions to ***other migratable ions***. *See id.*, 4:53-57.

21. Apart from this interpretation, I do not see how the ion active material as contemplated in the ’483 Patent can provide “other total ions” that, when compared with the amount of lithium ions present in the ion active material, would result in the claimed ratio values of 2:1, 3:1, 10:1, and 25:1. For example, looking at the primary examples of ion active materials provided in the ’483 Patent, namely $LiFePO_4$ and $LiMnPO_4$, where Fe, Mn, and/or PO_4 may *conceivably* be viewed as “other total ions,” it is not possible to get ratios of 2:1, 3:1, 10:1, and 25:1 because Fe, Mn, and/or PO_4 within a lithium ion active material will be present at a higher mole ratio compared to Li, not the other way around as required by the claims. LGES-1001,1:31-35.

VII. PRIOR ART ANALYSIS

A. [GROUND 1A/1B] – CLAIMS 1-4 AND 7-18 ARE OBVIOUS OVER OHATA-KAVAN-GONG OR OHATA-KAVAN-LIU

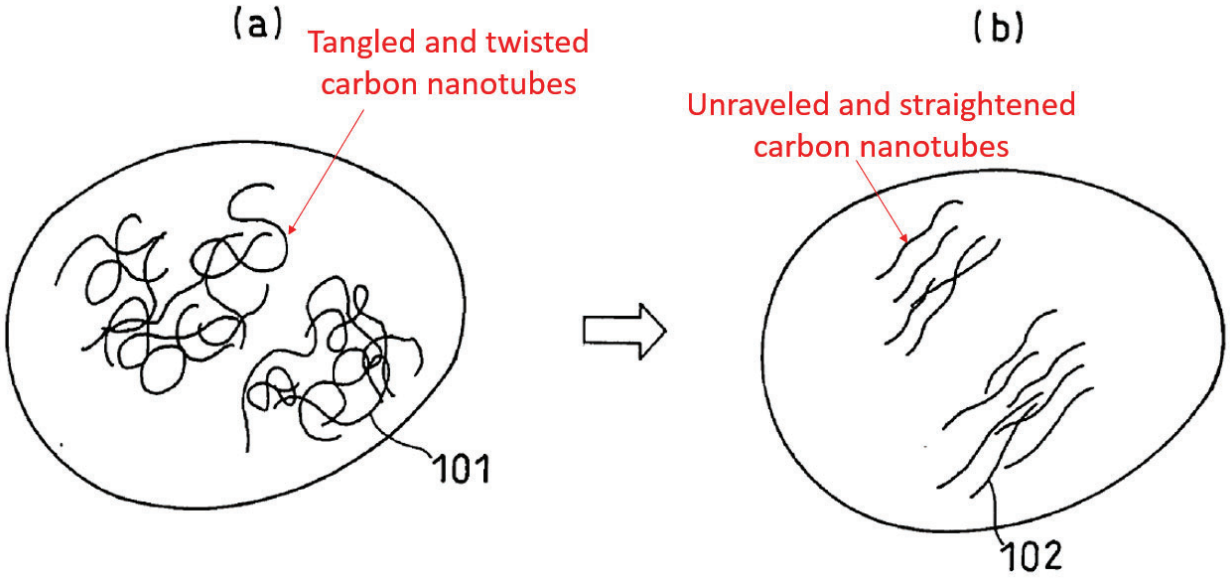
22. It is my opinion that a POSITA would have found claims 1-4 and 7-18 to be obvious over the combination of Ohata and Kavan further in view of Gong or Liu, as I discuss in the following sections.

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

23. 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].

24. 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)

25. 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].

26. 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

27. 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.

28. 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.

29. In Kavan, Oxidized nanotubes are 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 are prepared using the resulting material. LGES-1005 (Kavan), 2. The structure, composition, and electrochemical behavior of the resulting composites were then characterized.

30. 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 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

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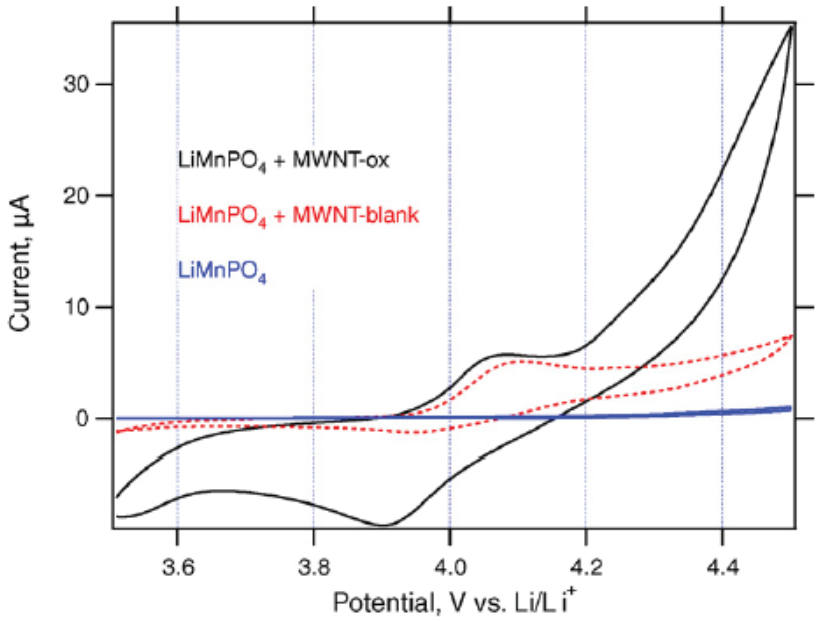
[1] "MWNT-bl" refers to a control sample of "non-functionalized blank material." LGES-1005 (Kavan), 2.

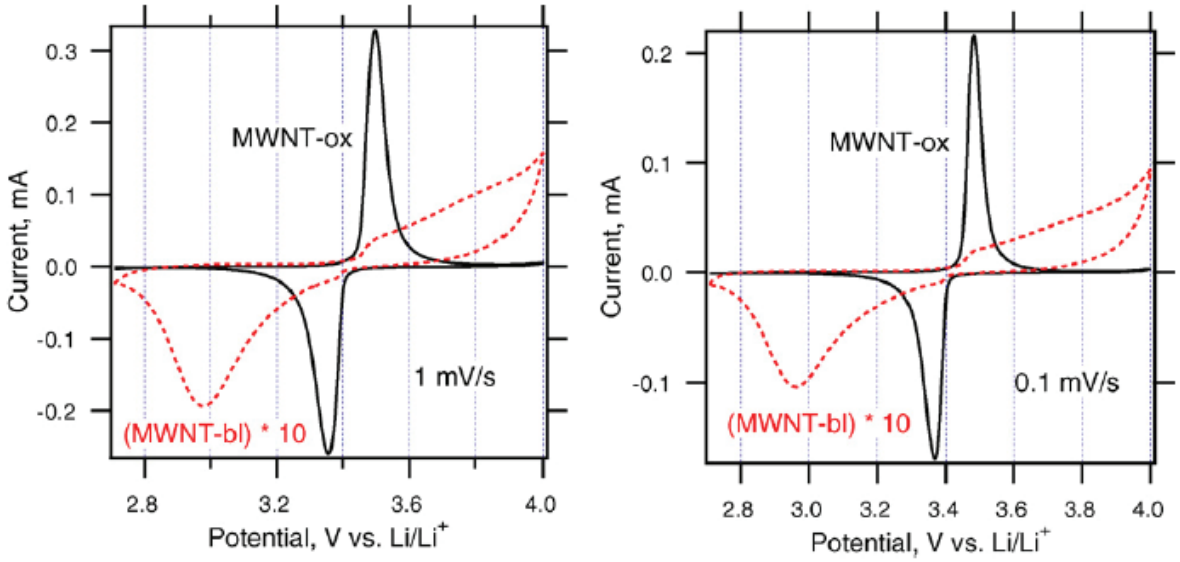
crucial for proper anchoring of nanotubes towards TiO_2." *Id.*, 4. Moreover, Kavan states that "[t]here is a clear analogy with the strong interaction of carboxyl-containing molecules … with the TiO_2 surface," which implies that strong binding can be formed between MWNTs' carboxyls and TiO_2. LGES-1005 (Kavan), 2.

31. As 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

32. 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

33. It is worth noting that the preparation of composites of MWNT-ox and $LiFePO_4$ in Kavan and Example 3 in the '483 Patent are similar. *See* LGES-1005 (Kavan), 2; LGES-1001 ('483 Patent), 5:55-67, 6:1-4. 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. Gong and Liu describe lithium position doping in olivine crystal structure for improved performance

34. Gong is generally directed to a lithium iron phosphate/carbon composite for use in a lithium-ion battery and, in particular, describes the doping of a lithium site of the olivine structure (*e.g.,* lithium iron phosphate) with a metal ion. *See* LGES-1007 (Gong), pp. 8, 26, 27, Claim 1. Among other things, Gong

describes a boron-containing lithium iron phosphate/carbon composite having a chemical formula of $Li_{1-x}M_xFe_{1-y}N_yP_{1-z}Si_zO_4 \cdot \xi BO_2/C$, where a metal element M can be doped at the lithium site, and where M is at least one of Mg, Ca, Zn, Al, Cr, V, Ti, Zr and Nb, and $0 \leq x \leq 0.1$. *Id.*, p. 8. Here, Gong is describing a lithium iron phosphate crystal structure where doping is used to replace a portion of the lithium ions with another metal ion.

35. Gong also describes various advantages for lithium-ion batteries based on such doping. For example, Gong also recognizes that, for lithium iron phosphate doped at lithium sites (*e.g.*, $Li_{0.98}Zr_{0.005}FePO_4$), the "changing trends of tapped density and average particle size are similar to those without doping" and further that, compared to undoped lithium iron phosphate, lithium iron phosphate doped at lithium sites has discharge capacity that is "generally higher." *Id.*, pp. 26-27. Gong also recognizes that "the composite material of the present invention [with lithium iron phosphate doped at lithium sites] has advantages such as low preparation temperature, small average particle size, high tapped density, and good rate charge and discharge characteristics." *Id.*, p. 27. Regarding the carbon source for the composite material, Gong discloses that "a carbon nanotube, acetylene black, or a carbon black conductive additive" can be used. *Id.*, Claim 7, p. 11.

36. Similar to Gong, Liu is also generally directed to "a positive electrode material for a secondary lithium battery ... and specifically relates to an olivine

phosphate positive electrode material." LGES-1009 (Liu), [0001], Abstract. With regard to "[t]ransition metal lithium phosphates ($LiMPO_4$, M = Fe, Co, Mn, or Ni) having an olivine structure," Liu recognizes common sources of poor performance, such as "low electron conductivity and low ion conductivity." *Id.*, [0002]. Liu also describes that "doping Li sites ... with aliovalent ions [can] improve the electron conductivity and ion conductivity of the material." *Id.*, [0003]. Thus, Liu also recognizes that lithium iron phosphate doped at lithium sites can lead to various performance improvements for lithium-ion battery systems.

37. In one example, Liu discloses an olivine phosphate positive electrode material mixed with carbon nanotubes, where the positive electrode materials are particles having an olivine crystal structure and having the following chemical composition: $Li_xA_aM_mB_bPO_zN_n$, where "doping element A is selected from at least one of Na, Mg, Ti, V, Cr, Cu, Mn, Co, Ni, Zn, Ga, In, Ge, W, Hg, Au, Ag, Nb, and Zr." LGES-1009, [0010]-[0013], Claim 1. Here, Liu is describing lithium iron phosphate olivine crystal particles that are doped at lithium sites with various types of metals. As to its carbon nanotubes, which are mixed with the olivine phosphates, Liu discloses that they can be "single-walled or multi-walled" and "hav[e] a diameter of 0.5 to 200 nm and an aspect ratio of 20 to 10000." LGES-1009, [0025], Claim 6.

4. The Ohata-Kavan combination

38. 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].

39. 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 a phosphate olivine such as $LiMnPO_4$ or $LiFePO_4$ (for a positive electrode composition). *See id.*, Abstract, 2.

40. 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").

41. 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].

42. 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 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].

43. 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 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, these

implementations render obvious the claims—a positive electrode material mixture paste including a phosphate olivine.

5. The Ohata-Kavan-Gong and Ohata-Kavan-Liu combinations

44. The Ohata-Kavan-Gong (Ground 1A) and Ohata-Kavan-Liu (Ground 1B) combinations each build on the Ohata-Kavan combination above in the following way. Specifically, in the Ohata-Kavan combination I described above, a POSITA would have found it obvious to form the positive electrode material mixture paste using a phosphate olivine, *e.g.*, $LiFePO_4$ or $LiMnPO_4$, for example as taught by Kavan. LGES-1005, 2. The resulting "electrode material mixture paste" can be used to form "[a]n electrode plate," *e.g.*, for a lithium ion battery. LGES-1004, [0025]. When a phosphate olivine is used as the electrode active material, the resulting electrode is a positive electrode for a lithium ion battery. Similarly, when TiO_2 is used as the electrode active material, the resulting electrode is a negative electrode for a lithium ion battery. Because both Ground 1A and 1B modify the underlying Ohata-Kavan combination in a similar manner, I may refer to them collectively as the Ohata-Kavan-Gong/Liu ground.

45. As I described above, Gong discloses that the performance of such phosphate olivine electrode materials, such as those in Ohata-Kavan, can be improved through the above-mentioned doping technique by replacing a portion of

the lithium ions in the phosphate olivine electrode active material with another, non-lithium metal ion. LGES-1007 (Gong), 8, 26-27. Liu similarly discloses that the phosphate olivine electrode materials as in Ohata-Kavan can be improved by doping at lithium sites with various types of metals. LGES-1009, [0002]-[0003], [0010]-[0013], Claim 1.

46. As I explain further below, a POSITA would have been motivated to apply doping to the positive electrode material of Ohata-Kavan (*i.e.*, a phosphate olivine such as $LiFePO_4$ or $LiMnPO_4$) as taught by either of Gong or Liu to obtain battery electrodes with improved performance.

6. Reasons for the combination

Reasons for incorporation of oxidized nanotubes, and expectation of success

47. 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.

48. The first reason for using oxidized nanotubes, for the specific case when the electrode material mixture paste is 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.

49. <u>The second reason</u> 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].

50. 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-1035 (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 prevent 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.

51. 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-1008 (Farrugia), [0027].

52. 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.

53. <u>The third reason</u> 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.

54. <u>The fourth reason</u> 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).

55. 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.

56. 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.

57. 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-1025 (Sheem), 3:28-52.

58. 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 a 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 a phosphate olivine as the electrode active material

59. It is 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 in this implementation, the combination electrode material mixture paste can be used to form a positive electrode of a lithium ion battery.

60. 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.

61. 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-1026 (Nakura), [0004].

62. 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-1026 (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.

63. <u>The fourth reason</u> 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.

64. 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.

65. 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).

66. It is my opinion that a POSITA would have expected success in incorporating phosphate olivine toward cathode material into the Ohata-Kavan electrode material mixture paste for at least the following reasons.

67. First, a POSITA would have expected to be successful in implementing phosphate olivine (for a positive electrode) in the combination because use phosphate olivines in compositions for lithium ion battery electrodes was conventional, and phosphate olivines 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 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-1026 (Nakura), [0004]) and Chen ("$LiFePO_4$ is a potential cathode candidate for the next generation of secondary lithium batteries." LGES-1041 (Chen), Abstract).

68. Second, a POSITA would have expected to be successful in implementing phosphate olivine in the combination as Ohata indicates that various electrode active material compositions can be used with its electrode material mixture paste. 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 phosphate olivines as electrode active materials as I indicated above, a POSITA would have expected phosphate olivines to be compatible with Ohata's electrode material mixture paste.

69. Third, a POSITA would have expected to be successful in implementing 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 phosphate olivines, as taught in the context of Kavan's slurry, could be used with Ohata's electrode material paste.

Reasons for doping the phosphate olivine in the electrode active material to replace lithium with another ion

70. It is my opinion that a POSITA would have been motivated to dope the phosphate olivine in the electrode active material of Ohata-Kavan, according to the teachings of Gong and/or Liu, to replace lithium with another ion.

71. The first reason is that a POSITA would have recognized that phosphate olivines such as $LiFePO_4$, which is used as an electrode material in Ohata-Kavan, can be doped as per the teachings of Gong and/or Liu to obtain

improved performance characteristics. For instance, Ohata notes that its object is "to provide an electrode superior in conductivity with a small amount of conductive material, yet with high capacity" and further provide one that is "excellent in cycle characteristics." LGES-1004, [0004], [0017]. Kavan similarly touts the importance of such improved performance characteristics. LGES-1005, 7, Abstract. Thus, a POSITA would have been motivated to apply lithium site doping to Ohata-Kavan's phosphate olivine electrode material to attain the performance benefits of such doping that each of Gong and Liu provides. As I described above, Gong describes that lithium iron phosphate doped at lithium sites can have higher discharge capacity as well as other advantages such as "low preparation temperature, small average particle size, high tapped density, and good rate charge and discharge characteristics." LGES-1007, pp. 26-27. Similarly, Liu describes that such doping can lead to improved "electron conductivity and ion conductivity." LGES-1009, [0002]-[0003]. A POSITA would have been motivated by such benefits to perform lithium position doping to the Ohata-Kavan material.

72. <u>The second reason</u> is that a POSITA would have found it obvious to pursue such lithium position doping implementation because doing so is merely the application of known techniques (lithium position doping a phosphate olivine of an electrode active material in lithium ion battery electrode composition) to a known

structure (Ohata/Kavan's positive electrode material) to yield predictable results (a positive electrode material with improved performance characteristics, such as improved conductivity and cyclability).

73. It is my opinion that a POSITA would have expected success in in incorporating lithium position doping, such as in Gong and/or Liu, into the Ohata-Kavan's phosphate olivine electrode material mixture paste for at least the following reasons.

74. First, use of lithium position doping when synthesizing phosphate olivines in compositions for lithium ion battery positive electrodes was conventional, with such doped phosphate olivines being well known to be used as positive electrode active materials. For example, Cheng (LGES-1011) describes that a "carbon nanotube doped-lithium iron phosphate composite cathode material [that] was synthesized by using a technique of doping in Li sites ... during synthesis of a lithium iron phosphate cathode material." LGES-1011, [0050]-[0051]. Peng (LGES-1012) is another prior art reference that describes the use of such dopant techniques in an iron phosphate electrode material. LGES-1012, Claims 1, 81 82 110, 111, 132. It was further well known to use such doped phosphate olivines together with *carbon nanotubes* to form composite materials as in Ohata-Kavan-Gong/Liu. LGES-1011, [0050]-[0051]; LGES-1012, Claims 127,

132, 133, 145, 153; LGES-1007, p. 11, Claim 7; LGES-1009, [0025], Claim 6; LGES-1011.

75. Second, composition of the Li-ion battery electrode material in both Gong and Liu is similar to that of Ohata-Kavan in that it, for example, is a lithium iron phosphate/carbon nanotube composite. *See* LGES-1007, pp. 8, 26, 27, Claim 1; LGES-1009, [0025], Claim 6. In view of such similarities in terms of both composition and application, a POSITA would have a reasonable expectation that doped phosphate olivines, as taught in the context of Gong and/or Liu, could be successfully implemented in Ohata-Kavan's positive electrode material.

7. Claim-by-Claim Analysis

(i) Claim 1

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

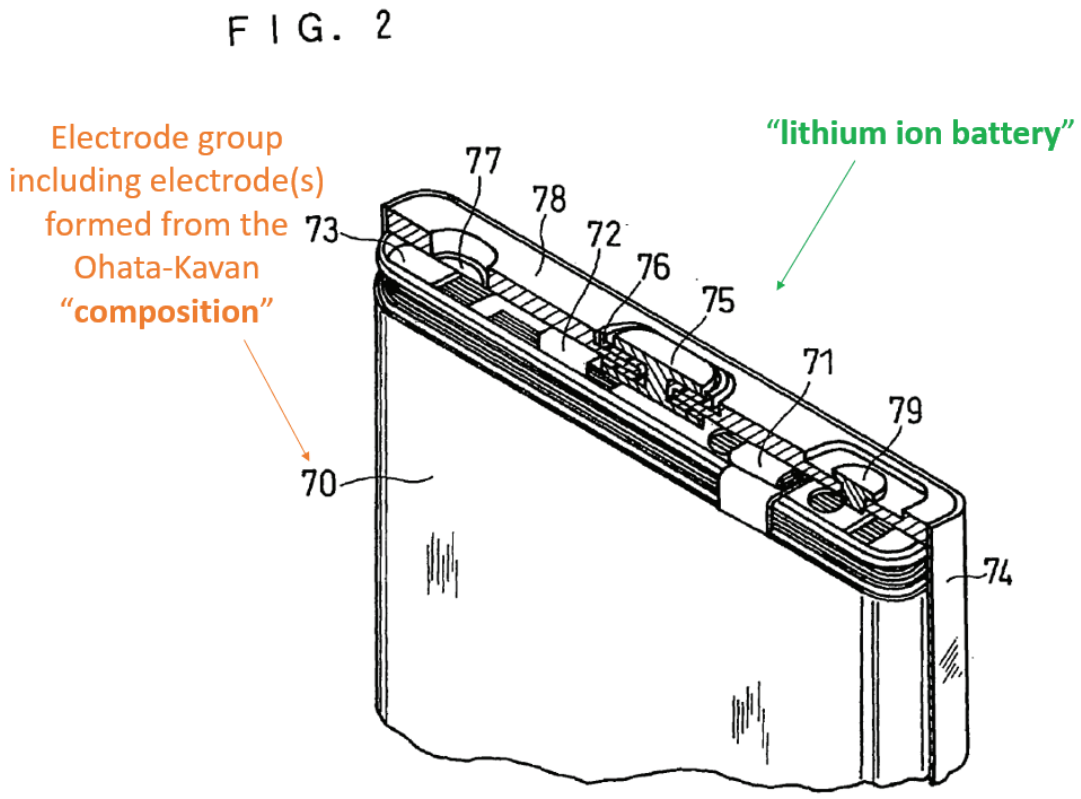
76. To the extent the preamble is deemed to be limiting, Ohata-Kavan-Gong/Liu renders it obvious.

77. First, the 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 positive electrode active material." *Id.*, [0007]; *see* [0026], claims 4. Specifically, in the combination **composition**, the positive electrode active material is implemented as phosphate olivine, based on the active material compositions described by Kavan. *See* LGES-1005 (Kavan), Abstract, 2; *see* my discussion of element [1b], *infra*.

78. 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").



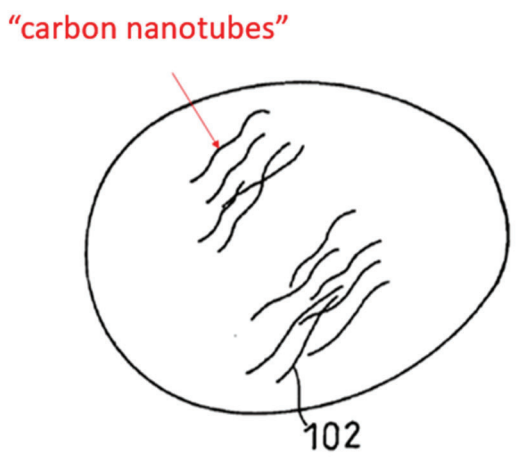
Excerpt from Ohata, FIG. 2 (annotated)

79. Importantly, even with the implementation of phosphate olivine as the electrode active material, based on Kavan, the combination composition remains **useful for lithium ion batteries**. Phosphate olivines were conventional electrode active materials for lithium ion batteries. *See* LGES-1005 (Kavan), 2 ("Since . . . $LiMPO_4$ 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-1026 (Nakura), [0004] ("As an alternative positive electrode active material to lithium cobaltate, lithium phosphates with an olivine structure are receiving attention").

[1a] discrete, non-agglomerated, and exfoliated carbon nanotubes having a surface, and

80. As I discussed above, Ohata's "masterbatch" includes "***carbon nanotubes*** and a resin," as shown in Fig. 1(b) below. LGES-1004, Abstract; *see also id.,* [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 100 parts by weight of the resin"), [0017] ("The resin with the unraveled and straightened ***carbon nanotubes*** dispersed therein").



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

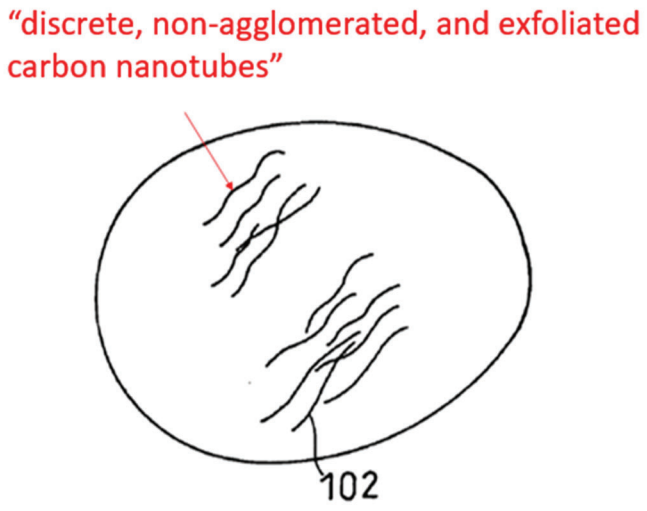
81. In the combination, which is based on Ohata's electrode material mixture paste as I discussed for [1pre], the composition is formed by "blending an electrode material mixture containing at least the masterbatch and an electrode active material with a dispersion medium to prepare an electrode material mixture paste." LGES-1004, Abstract; *see* [0005], [0023], [0032], claim 1. And I just discussed immediately above, the masterbatch includes **carbon nanotubes**. Thus, the **composition** of the Ohata-Kavan-Gong/Liu combination, which is based on Ohata's electrode material mixture paste, thus likewise include **carbon nanotubes**. *See* LGES-1004, [0024] ("The amount of the carbon nanotubes contained in the electrode material mixture"), [0007]-[0008], [0060]-[0061]. Indeed, Ohata 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 ($LiCoO_2$) as an active material and 45 parts by weight of N-methyl-2-pyrrolidone." LGES-1004, [0030], [0032]; *see* [0040]-[0042].

82. The **composition** of the Ohata-Kavan-Gong/Liu combinations further includes nanotubes that are **discrete/non-agglomerated/exfoliated** nanotubes, as taught by Ohata and as required by the claim. As an initial matter, I note that in the '282 Patent (US 10,608,282), which is another MRD patent from the inventors of this '483 Patent, "***discrete*** carbon nanotubes" are noted as being "***alternatively termed exfoliated*** carbon nanotubes." LGES-1012, 6:50-57. That is, MRD equates discrete with exfoliated. As for Ohata's nanotubes, during 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). The result is a "resin with the unraveled and straightened carbon nanotubes ***dispersed therein***." LGES-1004, [0017]; *see also id.,* [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). So to the extent **discrete** is interpreted as referring to nanotubes that are generally uniformly dispersed or individually distinct, a POSITA would have found it obvious that at least some these "unraveled and straightened carbon nanotubes" are **discrete** because they are generally uniformly "dispersed" as individual, distinct nanotubes (separated into individual tubes) in the resin, as described and illustrated by Ohata. LGES-1004, [0017].

83. Thus, in view of Ohata's description of its nanotubes as being "unraveled and straightened carbon nanotubes" and generally "dispersed" as individual nanotubes in the resin, A POSITA would have understood Ohata's nanotubes as being discrete, non-agglomerated, and exfoliated. And looking again to Ohata's illustrative example, provided below, it can be seen that most of the nanotubes are depicted as being discrete, non-agglomerated, and exfoliated. Indeed, a POSITA would have understood a carbon nanotube that is, as shown below, fully isolated from each other to be discrete, non-agglomerated, and exfoliated. The carbon nanotubes in the combination also necessarily have a surface because, among other things, Kavan discloses functionalizing the ***surface*** of its nanotubes. *See* LGES-1005, 2, 4.



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

84. A POSITA also would have understood or found obvious that the nanotubes in the combination composition are **discrete/non-agglomerated/exfoliated** because, at least in some instances, oxidation of nanotubes can help promote non-agglomeration of the nanotubes. *See* my analysis for [1e] below. The sidewalls of oxidized nanotubes are functionalized by species such as carboxylic acid or hydroxyl groups. *See* LGES-1010, Abstract; LGES-1014, 821-822; LGES-1015, 4186-4187; LGES-1016, [0027]-[0028]. In some cases, the presence of these functionalities can contribute to physically and chemically inhibiting aggregation of nanotubes into clumps; instead, oxidized nanotubes are generally present as individually distinct nanotubes. 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, non-agglomerated, and exfoliated**.

[1b] mixtures of ion active materials comprising migratable ions said materials comprising crystals or layers of lithium ion active material,

85. The combination composition in Ohata-Kavan-Gong/Liu (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, Abstract; *see* [0005], [0023], [0032], claim 1, claims 8-10. Here, the electrode active material, *e.g.*, a "positive electrode active material," including a doped phosphate olivine crystal that is based on the teachings of Kavan/Gong/Liu as I discussed, corresponds to the claimed mixture of ion active materials. LGES-1004, [0026]-[0027].

86. Moreover, a POSITA would have known that such phosphate olivine electrode materials 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, p.4; *see id.* ("the ***olivine crystals*** are typically hundreds of nm in size"), p.1 ("Carbon nanotubes form conductive composites with inorganic ***nanocrystals***"). Indeed, olivine electrode materials contemplated in the combination, such as $LiMnPO_4$ and

$LiFePO_4$, are mentioned in the ’483 Patent itself as being “[e]xamples of ion active materials.” LGES-1001, 4:35-38.

87. A POSITA would have also known that Li+ ions present within either $LiMnPO_4$ or $LiFePO_4$ are ***migratable ions***. *See* LGES-1001, 4:27-38 (“The migrating ion is preferably lithium ion....”). A POSITA would have further recognized that the ions used for lithium position doping, as in Gong and Liu, to take the place of the migratable lithium ions within the phosphate olivine material—including but not limited to Mg (magnesium), Na (sodium), Al (aluminum), Ni (nickel), Hg (mercury), Au (gold), and Ag (silver)—can be considered to be additional examples of migratable ions because they are taking the place of migratable lithium ions, as opposed to taking the place of other ions such as manganese and iron that are fixed within the olivine crystal structure (and thus not migratable). Indeed, it is well recognized in literature that such ions, including Na+ ions, can migrate and intercalate as Li+ ions do within battery systems. *See, e.g.*, LGES-1027, 3; LGES-1028, 1-4; LGES-1029, 5-6. Moreover, the above-mentioned ions (*i.e.*, Mg, Na, Al, Ni, HG, Au, Ag), along with others that may be “like” them, are listed in the ’483 Patent as being examples of migratable ions in this context. LGES-1001, 4:32-34.

[1c] wherein a ratio of lithium ion to other total ions in the ion active materials is at least 2:1 to 25:1, and

88. As I discussed above in Section VI.A, a POSITA would have understood the term "ratio of lithium ion to other total ions in the ion active materials" in the context of the '483 Patent to refer to the ratio of lithium ions to other (*i.e.*, non-lithium) migratable ions. Under such an interpretation of this term, Ohata-Kavan-Gong/Liu renders obvious limitation [1c].

89. For example, the ion active material contemplated in the Ohata-Kavan base combination included a positive electrode material mixture paste formed using a phosphate olivine, *e.g.*, $LiFePO_4$ or $LiMnPO_4$, as taught by Kavan. LGES-1005, 2.

90. Referring now to the Ohata-Kavan-Gong combination (Ground 1A), Gong discloses replacing Li within the phosphate olivine structure of Ohata-Kavan with other migratable ions M (*i.e.*, $Li_{1-x}M_x$) such that ***x is between 0 and 0.1***. LGES-1007, Claim 1, p. 8. When applied to the phosphate olivine electrode material of Ohata-Kavan, the resulting composition of the ion active material would then become, for example, $Li_{1-x}M_xFePO_4$ or $Li_{1-x}M_xMnPO_4$, where M_x corresponds to the doped, migratable ion that takes the place of some of the lithium ions. In one example, by setting the value of x to 0.1 (which is within the contemplated range of 0 to 0.1), the ratio of lithium ions to other migratable ions in the resulting $Li_{0.9}M_{0.1}FePO_4$ or $Li_{0.9}M_{0.1}MnPO_4$ is **9:1**. Further, by setting the value

of x to 0.04 (which is again within the contemplated range of 0 to 0.1), the resulting ratio of lithium ions to other migratable ions in $Li_{0.96}M_{0.04}FePO_4$ or $Li_{0.96}M_{0.04}MnPO_4$ is **24:1**. Thus, the range of the claimed ratio as disclosed by Ohata-Kavan-Gong falls well within "at least 2:1 to 25:1" requirement of limitation [1c]. As I explained above, M_x corresponds to non-lithium, migratable ions for use in the ratio calculation.

91. Referring now to the Ohata-Kavan-Liu combination (Ground 1B), Liu discloses replacing Li within the phosphate olivine structure of Ohata-Kavan with other migratable ions A (*i.e.*, Li_xA_a) such that ***x is between 0.9 and 1.8 and a is between 0 and 0.1***. LGES-1009, [0010]-[0013], Claim 1. When applied to the phosphate olivine electrode material of Ohata-Kavan, the resulting composition of the ion active material would then become, for example, $Li_xA_aFePO_4$ or $Li_xA_aMnPO_4$, where A_a corresponds to the doped, migratable ion that takes the place of some of the lithium ions. In one example, assuming the metal A has the same positive charge as lithium, by setting the value of x to 0.909 (which is within the contemplated range of between 0.9 and 1.8) and the value of a to 0.091 (which is within the contemplated range of between 0 and 0.1), the ratio of lithium ions to other migratable ions in the resulting $Li_{0.909}A_{0.091}FePO_4$ or $Li_{0.909}A_{0.091}MnPO_4$ is **10:1**. Further, by setting the value of x to 0.9615 (which is between 0.9 and 1.8) and the value of a to 0.0385 (which is between 0 and 0.1), the ratio of lithium ions

to other migratable ions in the resulting $Li_{0.9615}A_{0.0385}FePO_4$ or $Li_{0.9615}A_{0.0385}MnPO_4$ is **25:1**. Thus, the range of the claimed ratio as disclosed by Ohata-Kavan-Liu also falls squarely within "at least 2:1 to 25:1" as claimed in [1c]. As I explained above, A_a in Liu corresponds to non-lithium, migratable ions for use in the ratio calculation.

92. Here, it is worth noting that the '483 Patent itself provides no indication of criticality or unexpected results associated with the claimed range of ion ratios. That is, the patent simply appears to have listed example ratio ranges without showing, or even attempting to show, why such ratio ranges are in any way critical or lead to unexpected results.

[1d] wherein the discrete carbon nanotubes have crystals or layers of lithium ion active material ionically attached to their surface and

93. As I discussed above for limitation [1b], the positive electrode active material (the **ion active material**) in the combination's electrode material 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, 2; *see also* LGES-1004, [0026] ("The positive electrode active material … varies depending on the battery type"); LGES-1005, 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 $LiFePO_4$").

94. Moreover, according to Kavan (*see* [1e], *infra*), the phosphate olivine crystals of the combination are attached to the nanotube surface due at least in part to the surface oxidation of the nanotubes: "The surface oxidation of carbon nanotubes … was sometimes mentioned to improve ***the anchoring of … $LiMPO_4$***." LGES-1005, 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"). Indeed, as I described above, the carbon nanotubes used in preparation of the combination's masterbatch (*see* LGES-1004, [0015]) are functionalized, *e.g.*, "by carboxylic groups," as taught by Kavan, and are thus capable of becoming attached to the lithium ion active material (*e.g.*, lithium position doped phosphate olivine crystal). LGES-1005, Abstract. Kavan does not specify the type of bond that can be formed between the carboxyl group

and the olivine material (*i.e.*, lithium ion active material), but suggests to a POSITA that various types may be possible. LGES-1005, 2.

95. As a consequence, a POSITA would have recognized or otherwise found it obvious that one way by which the ion active materials of Kavan (e.g., $LiFePO_4$) become attached to the surface of Ohata-Kavan's nanotubes would be through ionic bonding/attachment. Indeed, it was well-known that the oxidized nanotubes, as in Ohata-Kavan, "have a negative electrostatic charge on the outer surface of the nanotube wall" and, thus, a POSITA would have recognized that such "negative electrostatic charge on the outer surface of the nanotube wall" would be used to enhance attachment of positively charged metal ions in the ion active material (e.g., $LiFePO_4$) to the nanotube surface via an ionic/electrostatic interaction. LGES-1024, [0057]; *see* LGES-1030 ("By oxidizing the nanotubes for various periods, different oxidation levels of the treated nanotubes can be easily prepared. This procedure is able to introduce carboxyl, carbonyl, and hydroxyl groups grown at any defects in the sidewalls of nanotubes. These oxygen groups ***show an affinity to couple with metal ions*** ... via the ***interaction of ionic exchange or surface polarity***."); *see also* LGES-1031, 2, FIG. 2g. In view of this, a POSITA would have recognized or otherwise found obvious that the negatively charged carboxyl group on the surface of the carbon nanotubes in Ohata-Kavan-Gong/Liu would be electrostatically attracted to and become capable of ionically

attaching to the positively charged metal ions (*e.g.*, Fe++, Mn++, or the like) in the phosphate olivines such as $LiMnPO_4$ or $LiFePO_4$.

[1e] wherein the discrete carbon nanotubes comprise oxidized species selected from the group consisting of carboxylic, ketone, or hydroxyl functionalities, and

96. As I described above in Section VII.A.4, it would have been obvious to have carbon nanotubes that are oxidized in the combination based on the teachings of Kavan. For instance, Kavan teaches that its nanotubes are "***functionalized by oxidation*** with HNO_3." LGES-1005, 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 (*see* LGES-1004, Abstract, [0015]-[0016],) and thus are present in the combination composition.

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

97. In the Ohata-Kavan-Gong/Liu 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

98. 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**.

99. The range of aspect ratios ("***not less than 100***" and "***not greater than 1000***") for the nanotubes of the Ohata-Kavan combination further 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-Gong/Liu combination renders obvious the claimed range.

100. I note that the nanotubes in the combination are oxidized, as I discuss in [1e]. 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.

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

101. 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.

102. 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.

103. As I explained above in my technical overview section, the **oxidation level by weight of the nanotubes** can be determined 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 moles (*i.e.*, 7 at.%) oxygen atoms, at 16 g/mol atomic weight; and 93 moles (*i.e.*, 93 at.%) carbon atoms, at 12 g/mol atomic weight. The oxidation level of Kavan's carbon nanotubes thus can be calculated as follows:

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

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

[2] The '483 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, 3:65-4:1.

$$= 9.1 \text{ wt.\% oxygen.}$$

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.

104. I note that Kavan's titration results indicate that "[t]he amount of acid groups in the nanotubes was found to be 2.54×10^{-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 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.

105. 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 a lithium metal salt and an element selected from the group consisting of: iron, manganese, cobalt, copper, nickel, vanadium, titanium, and mixtures thereof.

106. As I described above for limitation [1d], the positive electrode active material (the ion active material) in the Ohata-Kavan-Gong/Liu combination 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, 2; *see also* LGES-1004, [0026]. Both $LiMnPO_4$ and $LiFePO_4$ are well-known lithium metal salts, as acknowledged by the '483 Patent. LGES-1001, Claim 3.

107. Moreover, based on Gong and Liu, the phosphate olivine crystals can further be doped, for various performance benefits, to replace not only Li but alsoFe with various types of metals including, but not limited to, ***titanium, copper, cobalt, vanadium, and nickel***. *See* LGES-1007, 8, Claim 1; LGES-1009, [0010]-[0013], Claim 1. Accordingly, the doped phosphate olivine crystals of Ohata-Kavan-Gong/Liu includes crystals of lithium ion active material comprising both a "lithium metal salt" and "an element selected from the group consisting of: iron, manganese, cobalt, copper, nickel, vanadium, titanium, and mixtures thereof,"

namely titanium, copper, or nickel. Moreover, phosphate olivines such as $LiMnPO_4$ and $LiFePO_4$ each provides a lithium metal salt having an element selected from the claimed group, namely manganese and iron.

(iii) Claim 3

[3] The composition of claim 2, wherein the lithium metal salt has an olivine crystal structure and wherein the lithium metal salt is $LiFePO_4$ or $LiMnPO_4$.

108. As I described above for limitations [1d] and [2], the Ohata-Kavan-Gong/Liu combination renders obvious a positive electrode active material (*e.g.*, lithium metal salt) that is a phosphate olivine having an olivine crystal structure such as $LiMnPO_4$ or $LiFePO_4$ based on Kavan's teachings of forming an electrode from "nanotubes" and "***$LiMnPO_4$ or $LiFePO_4$***." LGES-1005, 2; *see also* LGES-1004, [0026]. Moreover, both $LiMnPO_4$ and $LiFePO_4$ are well known to have a crystal structure. LGES-1005, 4 ("crystals of $LiMnPO_4$" and "$LiFePO_4$ crystals").

(iv) Claim 4

[4] 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.

109. As I described above for limitations [1b]-[1d], in the Ohata-Kavan-Gong/Liu combination, the lithium position of the lithium ion active material (*e.g.*, phosphate olivine crystals) can be doped to replace lithium with various types of metals including, but not limited to, ***copper and titanium***. *See* LGES-1007, 8,

Claim 1; LGES-1009, [0010]-[0013], Claim 1. 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"). As such, the crystals or layers of lithium ion active material comprise tin, silicon, ***copper***, antimony, aluminum, germanium, ***titanium***, or mixtures thereof.

(v) Claim 7

[7pre] A composition useful for lithium ion batteries, comprising

110. I believe that [7pre] is obvious for the reasons discussed above in [1pre]. *See* [1pre] (incorporated here).

[7a] discrete, non-agglomerated, and exfoliated carbon nanotubes having a surface, and

111. I believe that [7a] is obvious for the reasons discussed above in [1a]. *See* [1a] (incorporated here).

[7b] mixtures of ion active materials comprising migratable ions said materials comprising crystals or layers of lithium ion active material,

112. I believe that [7b] is obvious for the reasons discussed above in [1b]. *See* [1b] (incorporated here).

[7c] wherein a ratio of lithium ion to other total ions in the ion active materials is at least 2:1 to 25:1,

113. I believe that [7c] is obvious for the reasons discussed above in [1c]. *See* [1c] (incorporated here).

[7d] wherein the discrete carbon nanotubes have crystals or layers of lithium ion active materials ionically attached to their surface, and

114. I believe that [7d] is obvious for the reasons discussed above in [1d]. *See* [1d] (incorporated here).

[7e] wherein the carbon nanotubes are in a weight ratio of carbon nanotubes to lithium salts of 0.1% to 10% and

115. In the Ohata-Kavan-Gong/Liu 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 positive electrode active materials (*see id.*, [0007], [0009], claim 4) and negative electrode active materials (*see id.*, [0008], [0010], claim 5).

116. 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

carbon nanotubes being present in a weight percentage of 0.2% (0.2 parts nanotubes per 100 parts ion active material) to 3.0% (3:100) by weight of the ion active material. Alternatively/additionally, to the extent the weight percentage is based on the total weight of both nanotubes and ion active material,[3] 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. Because the ion active material of the Ohata-Kavan-Gong/Liu combination can be in the form of lithium (metal) salts such as $LiMnPO_4$ and $LiFePO_4$, the above weight percentage range corresponds to a weight ratio of carbon nanotubes to lithium salts.

117. 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 4. 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-Gong/Liu combination

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[3] The '483 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, 3:65-4:1.

thus renders obvious the claimed range of weight percentages. And even if fewer than all of the proposed combination's nanotubes are discrete nanotubes, a POSITA still would have found it obvious that the weight percentage of discrete nanotubes would be generally 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.

118. 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 4.

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

119. Specifically, I have mapped the CNT parts by weight for each of Examples 1-8 to a percentage by weight of the 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 ion active material (%)	Weight of CNTs to weight of CNTs and 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)

[7f] wherein the discrete carbon nanotubes comprise oxidized species selected from the group consisting of carboxylic, ketone, or hydroxyl functionalities, and

120. I believe that [7f] is obvious for the reasons discussed above in [1e]. *See* [1e] (incorporated here).

[7g] wherein the discrete carbon nanotubes have an aspect ratio of 10 to 500 and

121. I believe that [7g] is obvious for the reasons discussed above in [1f]. *See* [1f] (incorporated here).

[7h] oxidation levels from 1% to 15% by weight of the carbon nanotube.

122. I believe that [7h] is obvious for the reasons discussed above in [1g]. *See* [1g] (incorporated here).

(vi) Claim 8

[8pre] A composition useful for lithium ion batteries, comprising

123. I believe that [8pre] is obvious for the reasons discussed above in [1pre]. *See* [1pre] (incorporated here).

[8a] oxidized discrete, non-agglomerated, and exfoliated carbon nanotubes having a surface, and

124. I believe that [8a] is obvious for the reasons discussed above in [1a] and [1e]. *See* [1a], [1e] (incorporated here). In particular, as I discussed for [1e], the nanotubes of Ohata-Kavan-Gong/Liu are oxidized.

[8b] mixtures of ion active materials comprising migratable ions said materials comprising crystals or layers of lithium ion active material,

125. I believe that [8b] is obvious for the reasons discussed above in [1b]. *See* [1b] (incorporated here).

[8c] wherein a ratio of lithium ion to other total ions in the ion active materials is at least 2:1 to 25:1,

126. I believe that [8c] is obvious for the reasons discussed above in [1c]. *See* [1c] (incorporated here).

[8d] wherein the discrete carbon nanotubes have crystals or layers of lithium ion active materials attached to their surface, and

127. I believe that [8d] is obvious for the reasons discussed above in [1d]. *See* [1d] (incorporated here).

[8e] wherein the carbon nanotubes are uniformly dispersed without clustering on a dimension scale of less than about 1 micrometer and

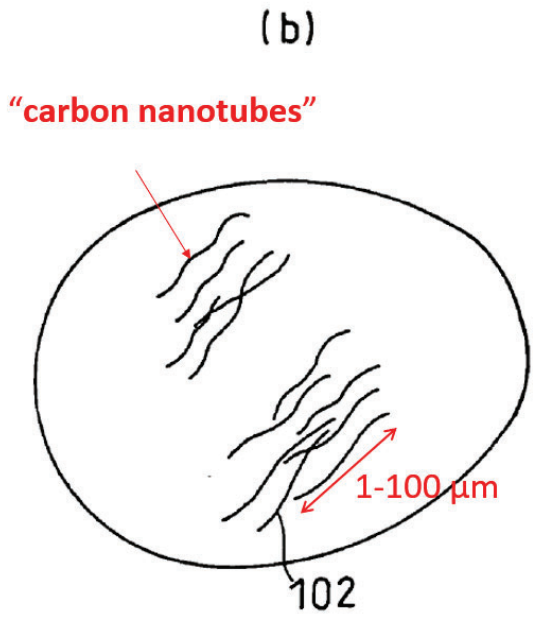
128. Ohata describes that in a masterbatch process, "a conductive material is ***uniformly predispersed*** in a binder." LGES-1004 (Ohata), [0002]. In the combination, this "conductive material" is the oxidized carbon nanotubes, and via

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].

129. It is my opinion that a POSITA would have understood, or at least found obvious, that the nanotubes in the Ohata-Kavan-Gong/Liu 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.

130. 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**. Moreover, 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)

[8f] wherein the discrete carbon nanotubes comprise oxidized species selected from the group consisting of carboxylic, ketone, or hydroxyl functionalities, and

131. I believe that [8f] is obvious for the reasons discussed above in [1e].

See [1e] (incorporated here).

[8g] wherein the discrete carbon nanotubes have an aspect ratio of 10 to 500 and

132. I believe that [8g] is obvious for the reasons discussed above in [1f].

See [1f] (incorporated here).

[8h] oxidation levels from 1% to 15% by weight of the carbon nanotube.

133. I believe that [8h] is obvious for the reasons discussed above in [1g]. *See* [1g] (incorporated here).

(vii) Claim 9

[9pre] A composition useful for lithium ion batteries, comprising

134. I believe that [9pre] is obvious for the reasons discussed above in [1pre]. *See* [1pre] (incorporated here).

[9a] uniformly dispersible oxidized, non-agglomerated carbon nanotubes having a surface, and

135. As I discussed above for limitations [1a] and [1e], the Ohata-Kavan-Gong/Liu combination provides oxidized and non-agglomerated carbon nanotubes having a surface. Regarding the "uniformly dispersible" limitation, 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].

136. 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.

137. Accordingly, the nanotubes of the combination composition are **uniformly** dispersed. 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**.

138. 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.

[9b] mixtures of ion active materials comprising migratable ions said materials comprising crystals or layers of lithium ion active material,

139. I believe that [9b] is obvious for the reasons discussed above in [1b]. *See* [1b] (incorporated here).

[9c] wherein a ratio of lithium ion to other total ions in the ion active materials is at least 2:1 to 25:1,

140. I believe that [9c] is obvious for the reasons discussed above in [1c]. *See* [1c] (incorporated here).

[9d] wherein the discrete carbon nanotubes have crystals or layers of lithium ion active materials ionically attached to their surface and

141. I believe that [9d] is obvious for the reasons discussed above in [1d]. *See* [1d] (incorporated here).

[9e] wherein the discrete carbon nanotubes comprise oxidized species selected from the group consisting of carboxylic, ketone, or hydroxyl functionalities, and

142. I believe that [9e] is obvious for the reasons discussed above in [1e]. *See* [1e] (incorporated here).

[9f] wherein the discrete carbon nanotubes have an aspect ratio of 10 to 500 and

143. I believe that [9f] is obvious for the reasons discussed above in [1f]. *See* [1f] (incorporated here).

[9g] oxidation levels from 1% to 15% by weight of the carbon nanotube.

144. I believe that [9g] is obvious for the reasons discussed above in [1g]. *See* [1f] (incorporated here).

(viii) Claim 10

[10] The composition of claim 9, wherein the crystals or layers of lithium ion active materials are chemically attached to the surface of the dispersible oxidized carbon nanotubes.

145. As I explained above for limitations [1d] and [9d], a POSITA would have recognized or otherwise found it obvious that the ion active materials of Kavan (e.g., $LiFePO_4$) would be ionically attached to the surface of Ohata-Kavan's nanotubes via an electrostatic interaction. Here, it is worth noting that materials that are "chemically attached" are well understood to be those that are attached covalently, ionically, or by Van der Waals, among others. That is, ionic bonding is merely a type of chemical bonding. *See, e.g.,* LGES-1033. Moreover, when oxygen groups become attached to the nanotube surface as per Kavan, "oxygen is chemically bonded with the ends or defects of carbon networks during the

oxidation process." LGES-1030, 2. Thus, the phosphate olivine can be considered to be chemically attached to the nanotubes via the oxygen groups.

(ix) Claims 11-18

[11] The composition of claim 1 where the ratio of lithium ion to other total ions in the ion active materials is from 3:1 to 25:1.

[12] The composition of claim 1 where the ratio of lithium ion to other total ions in the ion active materials is from 10:1 to 25:1.

[13] The composition of claim 7 where the ratio of lithium ion to other total ions in the ion active materials is from 3:1 to 25:1.

[14] The composition of claim 7 where the ratio of lithium ion to other total ions in the ion active materials is from 10:1 to 25:1.

[15] The composition of claim 8 where the ratio of lithium ion to other total ions in the ion active materials is from 3:1 to 25:1.

[16] The composition of claim 8 where the ratio of lithium ion to other total ions in the ion active materials is from 10:1 to 25:1.

[17] The composition of claim 9 where the ratio of lithium ion to other total ions in the ion active materials is from 3:1 to 25:1.

[18] The composition of claim 9 where the ratio of lithium ion to other total ions in the ion active materials is from 10:1 to 25:1.

146. The Ohata-Kavan-Gong/Liu combination renders obvious Claims 11-18 for the same reasons I provided above with respect to limitation [1c]. Indeed, as I explained for [1c], the explanations for which is incorporated herein, the Ohata-Kavan-Gong combination (Ground 1A) provides a ratio of lithium ion to other total ions in the ion active materials that ranges, in one instance, from **9:1 to 24:1**. Similarly, the Ohata-Kavan-Liu combination (Ground 1B) provides the claimed

ratio in the range of **10:1 to 25:1**. Both of these ranges fall within the ratio ranges recited in Claims 11-18.

B. [GROUND 2] – CLAIMS 1, 2, AND 7-18 ARE OBVIOUS OVER OHATA, KAVAN, AND ICHINOSE

147. It is my opinion that a POSITA would have additionally found claims 1, 2, and 7-18 to be obvious over the combination of Ohata, Kavan, and Ichinose, as I discuss in the following sections.

148. As I described above in Section VI.A, a POSITA would have understood "a ratio of lithium ion to other total ions in the ion active materials" to refer to the ratio of lithium ions to other (*i.e.*, non-lithium) migratable ions. It is my opinion that this construction is most consistent with the '483 Patent itself. *See, e.g.*, LGES-1001, 4:53-59.

149. However, Counsel has informed me that MRD appears to have advanced an apparent construction of this ratio term as meaning a ratio of Li+ ions to "PF6$^-$ or similar Li$^+$ counter-ion[s]." LGES-1050, 85. I agree with this assessment concerning MRD's apparent construction. Here, PF6$^-$ (or PF_6^- to be more technically correct) refers to the anion component of $LiPF_6$, which is a commonly used ***electrolyte*** in lithium-ion battery systems. It should be noted that there is no mention of $LiPF_6$ or "PF6$^-$ or similar Li$^+$ counter-ion" anywhere in the '483 Patent. And because the electrolyte is not an "ion active material," it is

unclear how Patent Owner can point to anions within this liquid electrolyte as somehow corresponding to "other total ions in the ion active materials" as required by the claim language. *See* LGES-1001, ("The ion active material which can be attached to the discrete carbon nanotubes is defined here as meaning an ion can migrate into or out of the ion active material thereby exchanging electrons."). An anion of liquid electrolyte can neither be attached to the carbon nanotubes nor support ion migration into and out of itself to exchange electrons. Not surprisingly, and consistent with this understanding, the only examples of ion active materials provided in the '483 Patent are well-known cathode/anode particles such as "lithium iron phosphate, lithium manganese phosphate, lithium cobalt oxide, silica, manganese dioxide, cadmium, titanium dioxide and carbon." LGES-1001, 4:34-37. It is also worth pointing out that the claims of the '483 Patent are merely directed to a "composition useful for lithium ion batteries" and not a full lithium-ion battery ("LIB") with electrolyte, separator, and other battery components.

150. MRD contends in the infringement contentions that "[g]iven the migratable nature of the Li+, its concentration within the cathode varies during charge and discharge processes but is configured to be within a 2:1 to 25:1 ratio at

least when at approximately a 44% to 98% state of charge."[4] LGES-1050, 85. But if this apparent construction for the ratio term were to be adopted, basically all lithium-ion batteries will at some point of its charging/discharging satisfy the claimed ratio as a varying number of lithium ions move back and forth between the cathode and the anode. In other words, to the extent Patent Owner argues that the claimed ratio refers to the ratio of Li+ to PF_6^- or similar Li+ counter-ions, which again I do not believe is proper in view of the specification and the claim language, such interpretation would result in the claimed ratio being satisfied by basically all lithium-ion batteries.

151. Nevertheless, I provides below an alternative analysis based on Ohata, Kavan, ***and Ichinose*** in view of Patent Owner's apparent construction of the ratio term.

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[4] Since a "98% state of charge" refers to a state in which most of the migratable Li+ ions have intercalated into the anode (such that there are fewer migratable Li+ ions in cathode, rendering the ratio of migratable Li+ in cathode to other ions (counter ions like $PF6^-$ in electrolyte based on the MRD's statement) lower), PO appears to have meant to say "98% state of ***discharge***" (rendering the ratio of migratable Li+ in the cathode to other ions higher).

152. Ichinose is generally directed to a "non-aqueous electrolyte secondary battery including an electrode assembly [and] a non-aqueous electrolyte." LGES-1034, Abstract, [0001]. Ichinose describes a LIB system that uses electrolyte made from "lithium salt such as $LiPF_6$," which is quite common for lithium-ion batteries. *Id.*, [0043], [0002]. Specifically, Ichinose discloses an example electrolyte volume to battery capacities as 0.18 to 1.117 ml/Ah, where the electrolyte used in this patent is 1 M $LiPF_6$ in a mixture of ethylene carbonate and ethyl methyl carbonate (volume ratio 1:3), and where the cathode used in the study is $LiCoO_2$, with a theoretical specific capacity of 142 mAh/g. *Id.*, [0049], [0111], [0112]

1. Analysis

[1c], [7c], [8c], [9c], and [11]-[19]

153. The Ohata-Kavan base combination, which I described above in Section VII.A.4, renders obvious most of the limitations of Claims 1, 2, and 7-19. The only exception, which is why the further combination with Gong/Liu was proposed, concern limitations [1c], [7c], [8c], [9c], and [11]-[19], which recite a specific "ratio of lithium ion to other total ions in the ion active materials," Gong and Liu were not relied upon to reach the claim limitations addressed in Grounds 1A/1B. In other words, the Ohata-Kavan combination renders obvious all limitations of Claims 1, 2, and 7-19 ***except for*** limitations [1c], [7c], [8c], [9c], and

[11]-[19] that require a specific "ratio of lithium ion to other total ions in the ion active materials."

154. Below, based on example parameters provided in the LIB of Ichinose, I provide the following exemplary steps to calculate the claimed ratio based on Patent Owner's apparent construction of this ratio term. Indeed, Ichinose's patent provided detailed information that would allow a POSITA to calculate the ratio of Li+ and PF_6^- at different states of discharge. To obtain the ratio, the following steps can be used.

1) Firstly, consider a battery with a capacity of 1 Ah, indicating that the cathode's capacity is 1 Ah. The number of moles of electrons corresponding to this capacity can be calculated.

1 Ah is equivalent to 3600 Coulombs. Coulomb is the unit of electric charge, defined as the amount of charge transported by a current of 1 ampere in 1 second. This relationship is expressed in the equation below:

1 Ah = 3600 C = 3600 s * 1 A

The electric charge can also be expressed as the charge carried by a certain number of electrons. One mole of electrons carries 96485 Coulombs of electric charge. The number of moles of electrons that carry a charge of 1 Ah can be calculated as follows:

Number of moles of electrons = 3600 C / (96485 C/mol) ≈ 0.0373 moles

2) Secondly, the lithium utilization should be calculated. It is well known that $LiCoO_2$ cannot utilize all of its lithium for intercalation. Typically, only about 50% of the lithium in $LiCoO_2$ can be used without affecting long-term cycling performance. Considering all the lithium in $LiCoO_2$, the true theoretical specific capacity is 274 mAh/g. In Ichinose's patent, the "theoretical" specific capacity of the cathode is specified to be 142 mAh/g, implying that only a portion of the lithium can be utilized. The percentage of utilizable lithium can be calculated as follows:

142 mAh/g / 274 mAh/g *100% ≈ 52%

This means that 48% of the lithium in $LiCoO_2$ cannot be removed from the cathode. This portion of lithium is considered immovable as it must remain in the cathode to support the structure of $LiCoO_2$. It is worth noting that in LGES-1050, the PO claims the state of charge (which should be state of discharge) to be between 44% and 98%. The 44% value likely refers to the amount of lithium that must be retained in the cathode.

3) By following the first and second steps, the total amount of lithium ions in the fully discharged cathode (state of discharge = 100%) can be calculated for a 1 Ah battery. In step 3, considering a 1 Ah battery, it can store charges equivalent

to 0.0373 moles of electrons. Each lithium atom can carry the same amount of charge as one electron. Therefore, the amount of utilizable lithium is also 0.0373 moles. The total lithium in $LiCoO_2$ is then calculated as follows:

0.0373 moles / 0.52 ≈ 0.0717 moles

Therefore, the total amount of lithium ions in the fully discharged cathode is 0.0717 moles.

4) By knowing the total amount of lithium ions in the fully discharged cathode, the movable lithium in the cathode at any state of discharge (SoD) can be calculated. The two extreme states are SoD = 100% and SoD = 48%. At SoD = 100%, all lithium ions are in the cathode. Since 52% of the lithium can be utilized in Ichinose's cathode, at SoD = 48%, all removable lithium has been extracted from the cathode.

When SoD = x, the total lithium in the cathode is 0.0717 * x moles. Among these, the movable lithium in the cathode is calculated as follows:

0.0717 * x - 0.0717 * (1 - 0.52)

5) Since the PO of LGES-1050 considers the ions in the electrolyte as migratable ions, this means that both the lithium ions in the electrolyte and the counter ions in the electrolyte are included in the calculation. We assume the ratio

mentioned by the PO refers to the ratio of movable lithium ions in the cathode at a specific SoD plus lithium ions in the electrolyte versus counter ions in the electrolyte. To calculate this ratio, the amount of electrolyte must be known. In Ichinose's patent, the electrolyte quantity is provided within a range, with the lower limit being 0.18 ml/Ah and the upper limit being 1.117 ml/Ah.

The amount of Li ions in electrolyte is:

At lower limit: 0.18 ml * 1 mol/L / 1000 = 0.00018 moles

At upper limit: 1.117 ml * 1 mol/L / 1000 = 0.001117 moles.

The amount of counter ion PF_6^- in electrolyte is:

At lower limit: 0.18 ml * 1 mol/L / 1000 = 0.00018 moles

At upper limit: 1.117 ml * 1 mol/L / 1000 = 0.001117 moles.

At SoD = x ($0.48 < x < 1$), the total movable lithium ions in the cathode plus the lithium ions in the electrolyte are:

At lower limit:

0.00018 + 0.0717 * x - 0.0717 * (1 - 0.52) mole

At upper limit:

0.001117 + 0.0717 * x - 0.0717 * (1 - 0.52) mole

Therefore, the ratio of movable lithium ions (including the movable lithium ions in the cathode at SoD = x and the lithium ions in the electrolyte) to PF_6^- is:

At lower limit:

(0.00018 + 0.0717 * x - 0.0717 * (1 - 0.52)) / 0.00018

At upper limit :

(0.001117 + 0.0717 * x - 0.0717 * (1 - 0.52)) / 0.001117

6) The calculation results are summarized in the table below, reporting two SoD states: SoD = 98% and SoD = 50%.

At the lower limit of electrolyte loading, the ratio changes from 200.166667 at SoD = 98% to 33.094897 at SoD = 50%. At the upper limit of electrolyte loading, the ratio changes from 8.9666667 at SoD = 98% to 2.283796 at SoD = 50%.

SoD	**Total Movable Li (Lower Limit)**	**Total Movable Li (Upper Limit)**	**Ratio (Lower Limit)**	**Ratio (Upper Limit)**
0.98	0.036030	0.036967	200.166667	33.094897
0.5	0.001614	0.002551	8.966667	2.283796

155. In view of the sample calculations above, a POSITA would have recognized that the ratio of **(active Li in cathode + Li in electrolyte) : PF_6^-** will, in essentially all standard lithium-ion batteries that far pre-date the ’483 Patent, eventually fall within the claimed ratio depending on the state of charge (or

discharge). Specifically, the resulting ratio range of ~2.3:1 to ~200:1 overlaps with the claimed ranges of 2:1 to 25:1, 3:1 to 25:1, and 10:1 to 25:1.

VIII. LEGAL PRINCIPLES

A. Anticipation

156. 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.

157. 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

158. 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.

159. 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.

IX. CONCLUSION

160. 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.

161. 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: 6/12/2024 By: Gu Zhu

APPENDIX A

Yu Zhu
Associate Director and Professor of
School of Polymer Science and Polymer Engineering
The University of Akron
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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

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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[36] Kewei Liu, Yu-Ming Chen, Gina M. Policastro, Matthew L. Becker, Yu Zhu* "Three-dimensional Bi-continuous Graphene Monolith from Polymer Templates" *ACS Nano* 2015, 9(6), 6041-6049 (impact factor: 15.881, citations=55)

[35] Yu-Ming Chen, Aozhen Xie, Yu Zhu* "Cyclability of a Lithium–Oxygen Battery Containing N-Methyl-2-Pyrrolidone and a Vertically Aligned Carbon Nanotube Cathode" *ChemElectroChem* 2015, 2(2), 208-212 (impact factor: 4.590, citations=7)

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Journal of Materials Chemistry A 2014, 2, 20902-20907. (Yu Zhu as corresponding author at U. Akron) (impact factor: 12.732, citations=40)

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[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

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.