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In re Patent of:	Clive P. Bosnyak, et al.	
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Title:	LITHIUM ION BATTERIES USING DISCRETE CARBON NANOTUBES, METHODS FOR PRODUCTION THEREOF AND PRODUCTS OBTAINED THEREFROM	

DECLARATION OF DR. YU ZHU

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I. QUALIFICATIONS AND BACKGROUND INFORMATION

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

2. I am currently the Associate Director and Professor of the School of Polymer Science and Polymer Engineering at The University of Akron. I am specializing in nanomaterials, polymers, energy storage devices and organic electronics. I received my PhD from the University of Cologne in Germany in 2007, where I worked on polymer synthesis, electrochemistry and organic electronics. From 2008 to 2012, I joined Rice University as a postdoctoral research associate, where my research included synthesis of graphene, carbon nanotube, low-dimensional nanomaterials and their applications in energy storage devices. I became an assistant professor at the University of Akron from August 2012, and was promoted to associate professor in 2018 and full professor in 2021. I have published over 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. 8,968,924 ("the '924 Patent") and prior art references relating to its subject matter. I have reviewed the '924 Patent, and relevant excerpts of the prosecution history of the '924 Patent. I have also reviewed the following references:

LGES-1001	U.S. Patent No. 8,968,924 ("the '924 Patent")
LGES-1004	U.S. Patent Pub. No. 2004/0160156 ("Ohata")
LGES-1005	"Multi-walled carbon nanotubes functionalized by carboxylic groups: Activation of TiO_2 (anatase) and phosphate olivines ($LiMnPO_4$; $LiFePO_4$) for electrochemical Li-storage." Ladislav

	Kavan et al. Journal of Power Sources 195 (2010) 5360-5369 ("Kavan")
LGES-1006	Chinese Patent Pub. No. 101714627 ("Cheng")
LGES-1007	Certified English Translation of CN 101714627 and Translator's Declaration ("Cheng")
LGES-1008	U.S. Patent Pub. No. 2010/0124713 ("Farrugia")
LGES-1009	"Chemical oxidation of multiwalled carbon nanotubes." V. Datsyuk et al. Carbon 46 (2008) 833-840 ("Datsyuk")
LGES-1010	"Surface and structural characterization of multi-walled carbon nanotubes following different oxidative treatments." Kevin A. Wepasnick, et al. Carbon 49 (2011) 24-36 ("Wepasnick")
LGES-1011	"Carbon Nanotubes Properties and Applications." https://www.cheaptubes.com/carbon-nanotubes-properties-and-applications/ ("Cheaptubes")
LGES-1012-1013	Reserved
LGES-1014	"Surface modified multi-walled carbon nanotubes in CNT/epoxy-composites." Florian H. Gojny, Jacek Nastalczyk, Zbigniew Roslaniec, Karl Schulte. Chemical Physics Letters 370 (2003) 820–824 ("Gojny")
LGES-1015	"Sonochemical Oxidation of Multiwalled Carbon Nanotubes." Yangchuan Xing, Liang Li, Charles C. Chusuei, and Robert V. Hull. Langmuir 2005, 21, 4185-4190 ("Xing")
LGES-1016	Reserved

LGES-1017 Chinese Patent Pub. No. 102136576 (“Zhao”)

LGES-1018 Certified English Translation of CN 102136576 and Translator’s Declaration (“Zhao”)

LGES-1019-1020 Reserved

LGES-1021 (Gan) U.S. Patent Pub. No. 2012/0058255 (“Gan”)

LGES-1022 Reserved

LGES-1023 Int’l Patent Pub. No. WO 2010/117392 (“Bosnyak”)

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 “Enhanced photocatalytic activity of mesoporous TiO_2 aggregates by embedding carbon nanotubes as electron-transfer channel.” J. Yu, T. Ma, S. Liu. Phys. Chem. Chem. Phys. 13 (2011) 3491–3501 (“Yu”)

LGES-1028 “Hydrothermal synthesis of lithium iron phosphate.” J. Chen, M. Whittingham. Electrochemistry Communications 8 (2006) 855-858 (“Chen”)

LGES-1029 “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-1030 “Incorporation of MWCNTs into leaf-life CuO nanoplates for superior reversible Li-ion storage.” J.Y. Xiang, J.P. Tu, J. Zhang, J.

Zhong, D. Zhang, J.P. Cheng. Electrochemistry Communications 12 (2010) 1103-1107 ("Xiang")

LGES-1031 A. Hirsch. "Functionalization of Single-Walled Carbon Nanotubes" *Angew. Chem. Int. Ed.* 2002, 41, No. 11, 1853-1859 ("Hirsch")

LGES-1032 "A porous $LiFePO_4$ and carbon nanotube composite." Y. Zhou, J. Wang, Y. Hu, R. O'Hayre, Z. Shao, Chemical Communications (2010), 46, 7151-7153 ("Zhou")

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

6. Counsel has informed me that I should consider these materials through the lens of one of ordinary skill in the art related to the '924 Patent at the time of the earliest possible priority date of the '924 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 '924 Patent would be June 23, 2011, based on the filing date of the provisional application that the '924 Patent claims priority to. However, Counsel has informed me that the *actual* priority date to which the '924 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 [3]. 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 ’924 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 ’924 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 5 are obvious over Ohata and Kavan;
- Claims 1, 4, and 5 are obvious over Cheng and Gan; and
- Claims 2 and 3 are obvious over Cheng, Gan, and Zhao.

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 '924 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 '924 PATENT

13. The '924 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 '924 Patent. *See*, *e.g.*, LGES-1004 (Ohata), [0004]; LGES-1005 (Kavan), Abstract; LGES-1007 (Cheng), 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), 4; LGES-1007 (Cheng), [0021]; LGES-1021 (Gan), [0024].

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-1009 (Datsyuk), 2 ("Oxidised [sic] MWCNTs were tested by X-Ray photoelectron spectroscopy in order to quantify and qualify the nature of oxygen present on the MWCNT surface"), 4 ("With reference to functionality grafting, XPS is one of the surface analytical techniques, which can provide useful information on the nature of the functional groups and also on the presence of structural defects on the nanotube surface"). A second surface-sensitive technique is energy-dispersive X-ray spectroscopy (EDX or EDS). *See* LGES-1010 (Wepasnick), Abstract ("energy dispersive spectroscopy (EDX) was used to characterize the extent of surface oxidation"); 6 ("XPS and EDX were both used to quantify the atomic oxygen concentration following different oxidative treatments"). Thermogravimetric analysis (TGA) is yet another technique that can be used to measure the composition throughout the thickness of a sample. *See* LGES-1009 (Datsyuk), 2 ("Oxidised [sic] MWCNTs were tested by X-Ray photoelectron spectroscopy in order to quantify and qualify the nature of oxygen present on the MWCNT surface. The characterization [sic] of the oxidised [sic] MWCNTs was completed by thermogravimetric analysis"). Titration techniques can be used to determine select

information about nanotube functionalization, such as the presence of certain types of functional groups on the nanotubes. *See* LGES-1005 (Kavan), 2 ("The extent of functionalization was determined by titration … The amount of acid groups in the nanotubes was found to be 2.54×10^{-4} mol g^{-1}"); LGES-1009 (Datsyuk), 6 ("A direct acid-base titration technique using NaOH was performed to determine the concentration of surface groups").

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

VI. PRIOR ART ANALYSIS

A. [GROUND 1] – CLAIMS 1, 4, AND 5 ARE OBVIOUS OVER OHATA-KAVAN

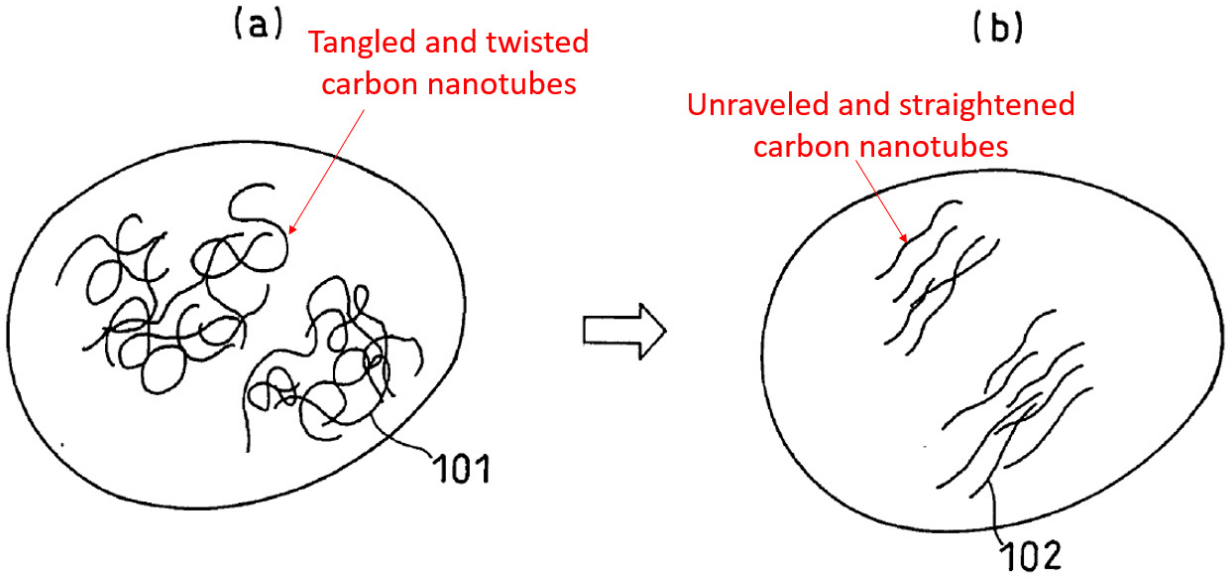
18. It is my opinion that a POSITA would have found claims 1, 4, and 5 to be obvious over the combination of Ohata and Kavan, as I discuss in the following sections.

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

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

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

FIG. 1



Excerpt from Ohata, FIG. 1 (annotated)

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

22. Ohata indicates that the size of the nanotubes impacts the conductivity of the electrode, and provides suitable size ranges for the diameter and length of

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

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

23. Kavan describes multi-walled carbon nanotubes (MWNTs) functionalized by carboxylic groups, used in composites with either negative electrode active materials or positive electrode active materials, for use as electrodes in lithium ion batteries. *See* LGES-1005 (Kavan), Introduction. Kavan sets out its "central motivation" as a desire to gain an "understanding … of the MWNT functionalization (by surface oxidation forming carboxyls) for optimum attachment of MWNT towards titania or olivine surfaces." *Id.*, 2.

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

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

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

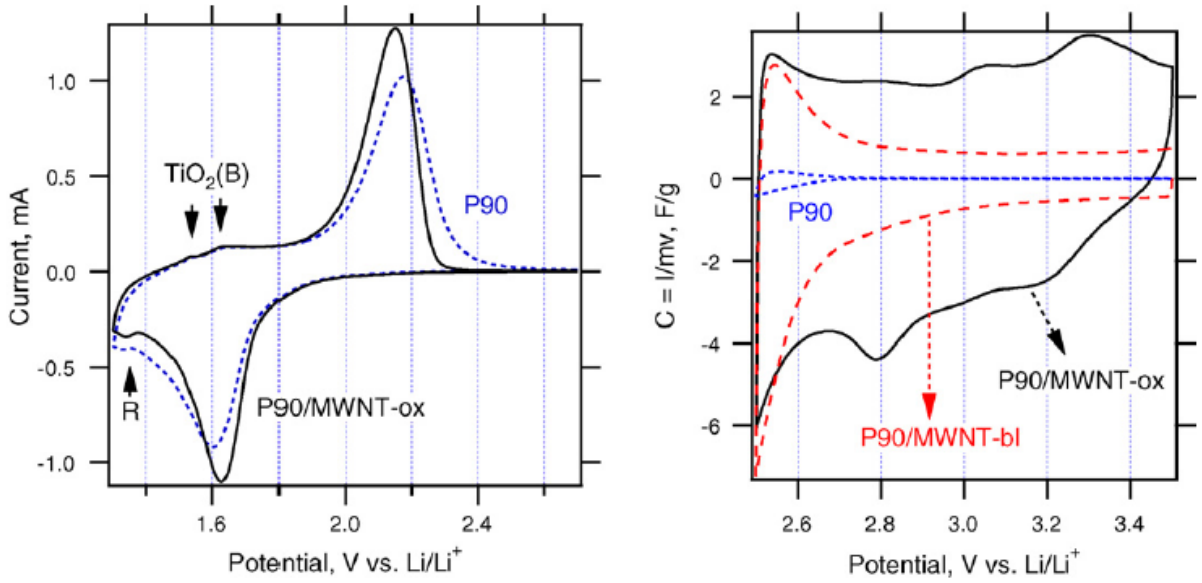
[1] "MWNT-bl" refers to a control sample of "non-functionalized blank material." LGES-1005 (Kavan), 2.

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.

27. In addition to directly mixing the lithium ion active materials particles with oxidized MWNTs, the '924 Patent also disclosed procedures of synthesized lithium active materials on the surface of oxidized MWNTs using soluble precursors (example 1,2,4,5). However, such procedure has also been reported in the literature. For example, synthesizing TiO_2 on oxidized MWNTs was reported by J. Yu. *See* LGES-1027 (Yu), Abstract, 2. This procedure was generally considered to form chemical bonds between CNT and lithium ion active materials. As stated in J. Yu's report, "Such surface functional groups provide active sites for directed deposition of anatase [TiO_2] nanocrystals around the surface of MWCNTs. For example, the -OH groups on the Ti precursor possibly react with the -OH and -COOH groups on the MWCNTs surface through esterification to form C-O-Ti or O=C-O-Ti bonds…" *Id.*, 6.

28. The attachment between TiO_2 and nanotubes enhances the electrochemical performance of the combination material. For instance, Kavan provides cyclic voltammetry data in Fig. 5, *infra*, for both TiO_2 electrodes and electrodes composed of a composite of TiO_2 and functionalized nanotubes. These

data show that while "the Li-storage capacities of both materials are identical," there are nevertheless electrochemical advantages associated with the TiO_2-nanotube composite. LGES-1005 (Kavan), 5. Specifically, "[t]he beneficial effect of nanotube networking" in the composites of TiO_2 and oxidized nanotubes can be seen in "the shape of cyclic voltammograms of Li-insertion," where for the TiO_2-nanotube composite, "[t]he peak-to-peak separation gets smaller" and "the slopes of insertion onset … and extraction offset … are steeper." *Id.* That is, "the rate of electrochemical charge transfer is higher due to nanotube networking." *Id.*, 9.

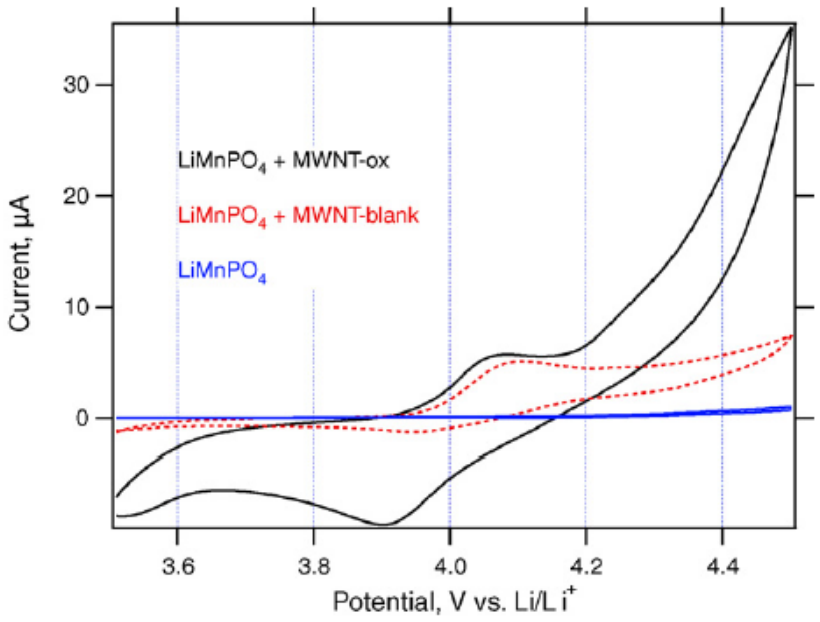


Excerpt from Kavan, Fig. 5

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

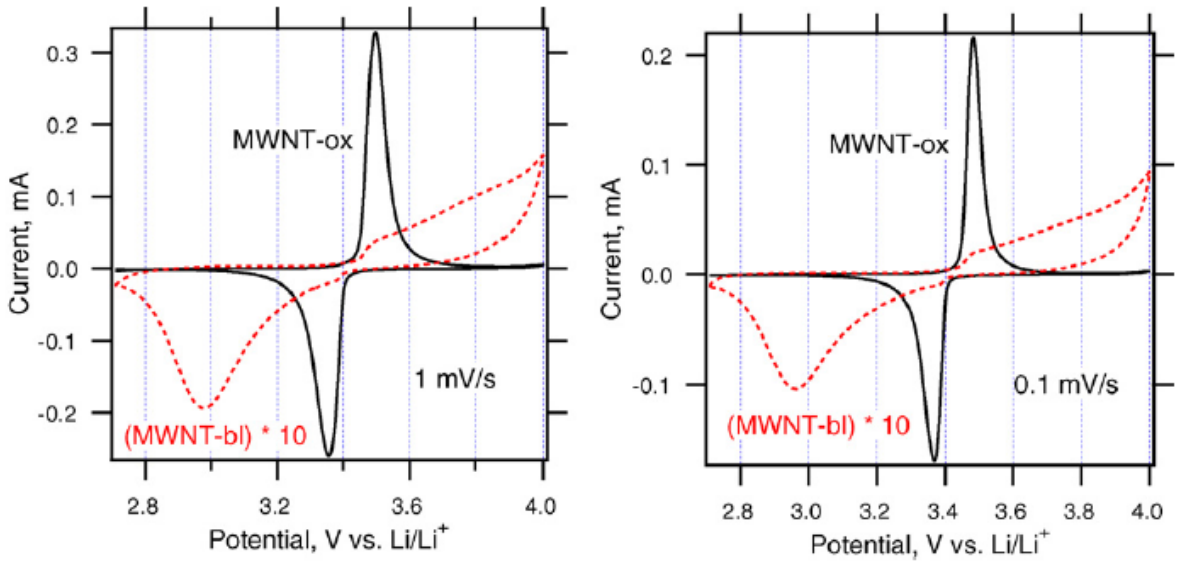
cyclic voltammogram of the composite to the creation of "new sites for pseudocapacitive interfacial redox chemistry." *Id.*

30. For the phosphate olivines, Kavan describes that structurally, "naked nanotubes are easily recognized between the individual crystals" of $LiMnPO_4$ or $LiFePO_4$. LGES-1005 (Kavan), 4; *see* Fig. 3. Overall, Kavan concludes that "the functionalization of nanotubes is … crucial for enhancing the electrochemical activity of the composite materials with $LiFePO_4$." *Id.*, 8. For instance, Fig. 7, *infra*, shows cyclic voltammetry results of $LiMnPO_4$ electrodes (blue curve), electrodes composed of a composite of $LiMnPO_4$ and non-oxidized nanotubes ("$LiMnPO_4$ + MWNT-blank," red dashed curve) and electrodes composed of a composite of $LiMnPO_4$ and oxidized nanotubes ("$LiMnPO_4$ + MWNT-ox," black curve). These results demonstrate that while "oxidation of $LiMnPO_4$ to MnPO4 is roughly identical for both electrodes" (*i.e.*, both nanotube-containing electrodes), "the oxidative treatment of nanotubes improves the reversibility of $LiMnPO_4$ charge/discharge." *Id.*, 7.



Excerpt from Kavan, Fig. 7

31. Regarding $LiFePO_4$, Kavan states that "the $LiFePO_4$ olivine exhibits excellent performance in a composite with MWNT-ox" and that "the improvement" in electrochemical activation of $LiFePO_4$ "caused by surface oxidation of nanotubes is dramatic." *Id.*, 7. Cyclic voltammograms in Fig. 8, *infra*, show results for electrodes composed of a composite of $LiFePO_4$ and non-oxidized nanotubes ("MWNT-blank," red dashed curve) and electrodes composed of a composite of $LiMnPO_4$ and oxidized nanotubes ("MWNT-ox, black solid curves"). *Id.* Kavan characterizes the results for the oxidized nanotube based composite as "impressive" in terms of both "capacity" and "charge/discharge reversibility." *Id.*



Excerpt from Kavan, Fig. 8

32. It is worth noting that the preparation of composites of MWNT-ox and $LiFePO_4$ in Kavan and Example 3 in the '924 Patent are similar. *See* LGES-1005 (Kavan), 2; LGES-1001 ('924 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. The Ohata-Kavan combination

33. The Ohata-Kavan combination composition is based initially on the teachings of Ohata. Specifically, "an electrode material mixture containing the masterbatch and an electrode active material" is blended with "a dispersion medium" to form "[a]n electrode material mixture paste." LGES-1004 (Ohata), [0023]. The masterbatch contains carbon nanotubes dispersed in a resin, where the

aspect ratio of the nanotubes is between 100 and 1000, as taught by Ohata. *See id.*, [0015], [0020]. The ratio of carbon nanotubes to electrode active material is also consistent with Ohata's teachings, *e.g.*, "[t]he amount of the carbon nanotubes contained in the electrode material mixture is, for example, 0.2 to 3 parts by weight per 100 parts by weight of the electrode active material," and "is preferably not greater than 0.8 parts by weight," which Ohata indicates is sufficient "to obtain a high-capacity electrode." *Id.*, [0024].

34. Based on the teachings of Kavan, the carbon nanotubes used in the masterbatch of the combination are oxidized, as discussed below. *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.

35. In the Ohata-Kavan combination, the carbon nanotubes used in preparation of the masterbatch (*see* LGES-1004 (Ohata), [0015]) are functionalized, *e.g.*, "by carboxylic groups," as taught by Kavan. LGES-1005 (Kavan), Abstract. For instance, the nanotubes are exposed to the "oxidative functionalization" process described by Kavan, in which the nanotubes are "functionalized by oxidation with HNO_3" such that "[t]he amount of atomic oxygen present" on the nanotube surface is "7%." *Id.*, 2,4; *see* 2 ("For oxidative functionalization, 1 g of purified nanotubes was mixed with 50 mL of 65% HNO_3

and refluxed for 3 h at 120 ºC under stirring. The nanotubes were filtered on a glass frit, washed with water until the filtrate reached a pH of 6, and dried at 120 ºC for 48 h").

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

37. As taught by Ohata, "an electrode material mixture containing the masterbatch and an electrode active material" is blended with "a dispersion medium" to form "[a]n electrode material mixture paste." LGES-1004 (Ohata), [0023]. As taught by Ohata, the dispersion medium is "selected according to the resin to be used." *Id.*, [0024]. Ohata indicates that "[t]he positive electrode active material" or "[t]he negative electrode active material to be used" in the electrode material mixture "varies depending on the battery type." LGES-1004 (Ohata), [0026]-[0027]. Consistent with this flexibility, in the Ohata-Kavan combination, a positive electrode material mixture paste is formed using a phosphate olivine, *e.g.*,

$LiFePO_4$ or $LiMnPO_4$, as also taught by Kavan. *Id.*, 2. In this combination electrode material mixture paste, the nanotubes and electrode active material are provided, as taught by Ohata, in amounts such that "[t]he amount of the carbon nanotubes … is, for example, 0.2 to 3 parts by weight per 100 parts by weight of the electrode active material," and "preferably not greater than 0.8 parts by weight." LGES-1004 (Ohata), [0024].

38. The resulting "electrode material mixture paste" can be used to form "[a]n electrode plate," *e.g.*, for a lithium ion battery. LGES-1004 (Ohata), [0025]. For instance, the combination electrode material mixture paste is applied "onto an electrode substrate, which is then dried and rolled" and "cut into a predetermined shape to give an electrode intended." *Id.*, [0025]. Specifically, when 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.

4. Reasons for the combination

Reasons for incorporation of oxidized nanotubes, and expectation of success

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

40. The first reason for using oxidized nanotubes, 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.

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

42. 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-1031 (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.

43. 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].

44. 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.

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

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

47. I note that oxidized nanotubes may have a lower conductivity than their unoxidized counterparts. However, use of oxidized nanotubes could also bring 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, e.g., as taught in Kavan, 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.

48. 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.

49. 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.

50. Second, a POSITA would have expected to be successful in using oxidized nanotubes in the context of Ohata's electrode material mixture paste because the composition of Kavan's slurry is generally similar to the composition of Ohata's electrode material paste. For instance, both use a fluorocarbon resin

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

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

51. 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.

52. 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-1029 (Feng), Abstract.

53. 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].

54. 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-1028 (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-1029 (Feng), 1.

55. The fourth reason for using phosphate olivines as the positive electrode active material is that they are environmentally friendly and relatively non-toxic: "the phosphate olivines … are promising cathode materials for Li-ion batteries due to … environmental tolerability." LGES-1005 (Kavan), 1. For instance, Chen specifically addresses $LiFePO_4$, stating that "[a] good cathode material has to meet the following criteria: … low toxicity. $LiFePO_4$, with the Olivine structure, … appears to meet many of these requirements." LGES-1028 (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-1029 (Feng), 1.

56. 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-1028 (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-1029 (Feng), 1.

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

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

58. 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.

59. 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-1028 (Chen), Abstract).

60. 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.

61. 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.

5. Claim-by-Claim Analysis

(i) Claim 1

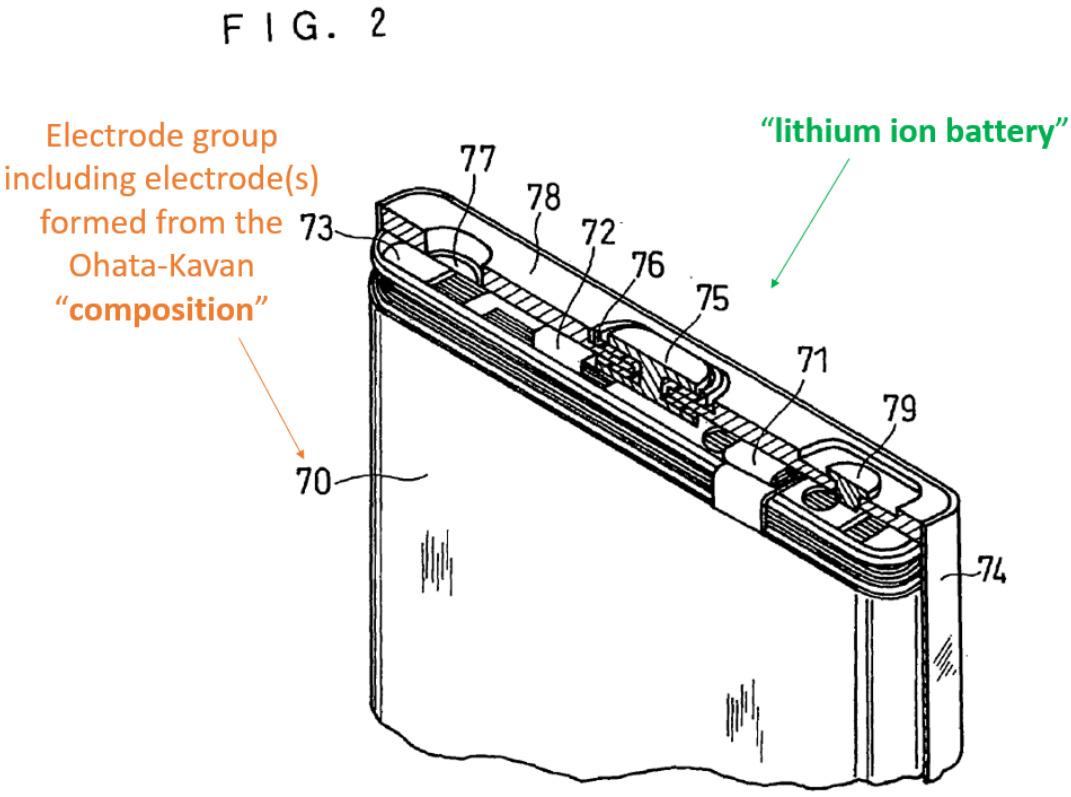
[1pre] A composition used in lithium ion batteries comprising:

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

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

64. 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)

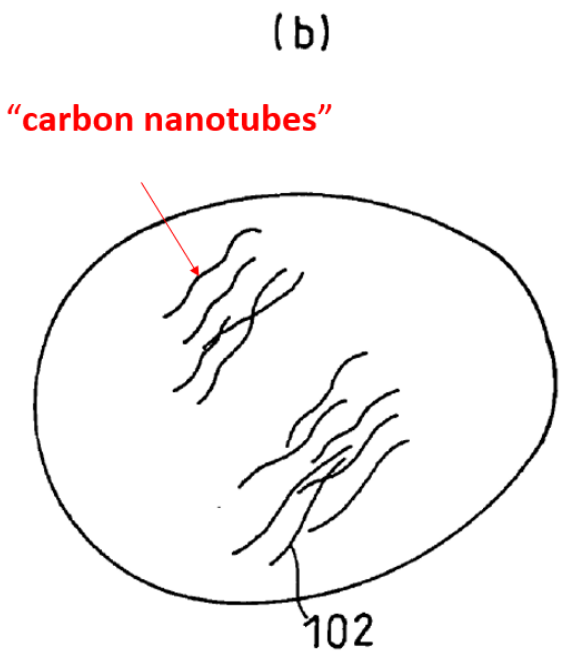
65. Importantly, even with the implementation of phosphate olivine as the electrode active material, based on Kavan, the combination composition remains **useful for lithium ion batteries**. Phosphate olivines were conventional electrode active materials for lithium ion batteries. *See* LGES-1005 (Kavan), 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 carbon nanotubes

The combination composition includes carbon nanotubes

66. Ohata's "masterbatch" includes "***carbon nanotubes*** and a resin," as shown in Fig. 1(b), *infra*. LGES-1004 (Ohata), Abstract; *see* [0015] ("producing a masterbatch by blending at least ***carbon nanotubes*** with a resin"), [0016] ("The ***carbon nanotubes*** are unraveled and straightened by blending them with a resin"), [0006] ("The proper amount of the ***carbon nanotubes*** contained in the masterbatch is 5 to 20 parts by weight per 10 parts by weight of the resin"), [0017] ("The resin with the unraveled and straightened ***carbon nanotubes*** dispersed therein"), [0005], [0009]-[0010], [0018]-[0022], [0030], claim 1, claim 3.



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

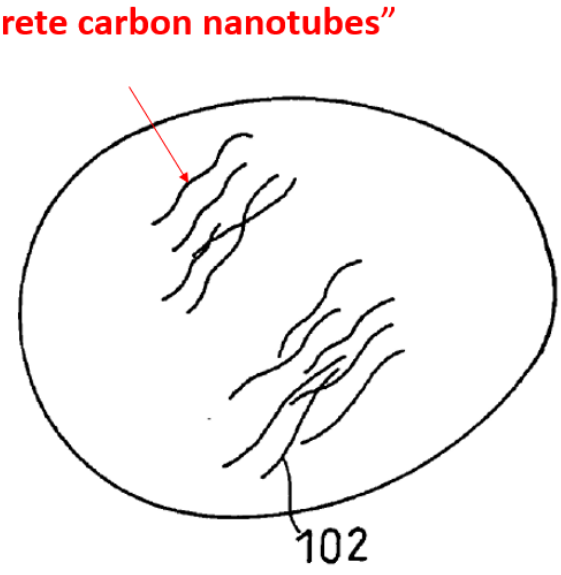
67. The combination composition (based on Ohata's electrode material mixture paste; *see* [1pre]) is formed by "blending an electrode material mixture containing at least the masterbatch and an electrode active material with a dispersion medium to prepare an electrode material mixture paste." LGES-1004 (Ohata), Abstract; *see* [0005], [0023], [0032], claim 1. As discussed in the preceding paragraph, the masterbatch includes **carbon nanotubes**. Thus, the **composition** of the Ohata-Kavan combination (based on Ohata's electrode material mixture paste), which includes the masterbatch, thus likewise includes **carbon nanotubes**. *See* LGES-1004 (Ohata), [0024] ("The amount of the carbon nanotubes contained in the electrode material mixture"), [0007]-[0008], [0060]-[0061].

68. 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 (LiCoO2) as an active material and 45 parts by weight of N-methyl-2-pyrrolidone." LGES-1004 (Ohata), [0030], [0032]; *see* [0040]-[0042].

The carbon nanotubes in the combination composition are discrete

69. The **composition** of the Ohata-Kavan combination (based on Ohata's electrode material mixture paste) includes a masterbatch, which contains **discrete** nanotubes, as taught by Ohata. Specifically, Ohata describes "producing a masterbatch by blending at least carbon nanotubes with a resin." LGES-1004 (Ohata), [0015]. Prior to this blending, the nanotubes "are tangled and twisted with one another," as illustrated in Ohata's FIG. 1(a). *Id.*, [0016]. In the process of "blending them with a resin" to form the masterbatch, "[t]he carbon nanotubes are ***unraveled and straightened***," as shown in FIG. 1(b), *infra*. *Id.* The result is a "resin with the unraveled and straightened carbon nanotubes ***dispersed therein***," *e.g.*, a masterbatch containing **discrete carbon nanotubes**. *Id.*, [0017]; *see* [0016]

("The unraveled and straightened carbon nanotubes 102 are expected to form an excellent conductive path in an electrode"), [0022] ("As the resin in which ***the carbon nanotubes are dispersed***, a fluorocarbon resin can be used"), [0002] ("in order to reduce the amount of conductive material used, it is proposed to use a masterbatch process, whereby ***a conductive material is uniformly predispersed in a binder***, in the production of a positive electrode"). For instance, these "unraveled and straightened carbon nanotubes" are **discrete** because they are generally "dispersed" as individual nanotubes in the resin, as described and illustrated by Ohata. LGES-1004 (Ohata), [0017].



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

70. Alternatively/additionally, a POSITA would have understood or found obvious that the nanotubes in the combination composition are **discrete**. As I discuss further for element [1d], *infra*, the nanotubes in the Ohata-Kavan

combination are oxidized. At least in some instances, oxidization can help promote non-agglomeration of nanotubes, *e.g.*, oxidized nanotubes can be discrete.

71. The sidewalls and/or ends of oxidized nanotubes are functionalized by species such as carboxyl, carbonyl, or hydroxyl groups. *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"). As I explained above, by the presence of these functionalities, oxidized nanotubes are physically and chemically inhibited from aggregation into clumps, and instead are generally present as individually distinct nanotubes. *See* LGES-1014 (Gojny), 821-822 ("[t]he presence of

carboxylic acid groups on the surface" "enables a better dispersion of CNTs" showing images of "oxidised [sic] MWCNTs … [that] are completely dispersed in the [epoxy resin] matrix due to the appearance of carboxylic groups on the surface"); LGES-1015 (Xing), 4186-4187 ("treated [oxidized] CNTs had better dispersion"); LGES-1008 (Farrugia), [0027] (describing that functionalization of carbon nanotubes "results in one or more chemical moieties attached to the carbon nanotubes," which "results in better dispersion of the nanotubes throughout the matrix").

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

[1b] having ion active material attached to their surface . . . wherein the 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

73. The combination composition (based on Ohata's electrode material mixture paste; *see* [1pre]) is formed by "blending an electrode material mixture containing at least the masterbatch and ***an electrode active material*** with a dispersion medium to prepare an electrode material mixture paste." LGES-1004 (Ohata), Abstract; *see* [0005], [0023], [0032], claim 1, claims 8-10. The electrode

active material, *e.g.*, either a "positive electrode active material" or a "negative electrode active material" corresponds to **ion active material**. LGES-1004 (Ohata), [0026]-[0027]. The specific composition of this electrode active material as phosphate olivine is based on the teachings of Kavan, as discussed in the following paragraphs.

Ion active material for cathodes

74. The positive electrode active material (the **ion active material**) in the Ohata-Kavan electrode material mixture paste is a phosphate olivine such as $LiMnPO_4$ or $LiFePO_4$ based on Kavan's teachings of forming a "paste" including "nanotubes" and "***$LiMnPO_4$ or $LiFePO_4$***." LGES-1005 (Kavan), 2; *see* LGES-1004 (Ohata), [0026] ("The positive electrode active material … varies depending on the battery type"); LGES-1005 (Kavan), 2 (describing use of "carbon-free olivines, $LiFePO_4$ or $LiMnPO_4$ in fabricating samples), 4 ("Fig. 3 shows the morphological features of our olivine/MWNT-ox composites"), 6 ("Fig. 6 shows a series of cyclic voltammograms of $LiMnPO_4$/MWNT-ox composite"), 7 ("Fig. 8 shows cyclic voltammograms" for "[t]he composite $LiFePO_4$/MWNT-ox"), 7 ("Fig. 9 shows a series of cyclic voltammograms for a series of composite materials $LiFePO_4$/MWNT-ox"), 8 ("Fig. 11 shows the initial discharge/charge of $LiMnPO_4$/MWNT-ox at varying charge rates from C/10 to C. A similar plot for

$LiFePO_4$/MWNT-ox is shown in Fig. 12"), 9 ("composite materials in which multi-walled carbon nanotubes … were interfaced to … $LiMnPO_4$ and $LiFePO_4$").

75. Phosphate olivines such as $LiMnPO_4$ and $LiFePO_4$ are each an **ion active material**, as confirmed by Kavan: 2 ("***$LiMPO_4$ olivines are promising cathode materials***"), 1 ("From the viewpoint of electrochemical lithium storage, the ***phosphate olivines*** … have been extensively studied in the past. They ***are promising cathode materials*** for Li-ion batteries"). Indeed, that these materials are ion active materials was well-known. *See* LGES-1026 (Nakura), [0004] ("As an alternative positive electrode active material to lithium cobaltate, lithium phosphates with an olivine structure are receiving attention"). This is also consistent with the disclosure of the '924 Patent, which identifies a "lithium metal salt with an olivine crystal structure" as an example of an "ion active material." LGES-1001, 2:40-45, 4:34-37**.** Further, I notice the claim states the ion active material includes both a lithium metal salt and an element selected from the specified group. I understand this element can either be a constituent of the lithium metal salt or another component of the ion active material. For instance, none of the relevant experimental examples in the '924 Patent disclose extra "element" in addition to the lithium metal salt, and it merely states that "[e]xamples of ion active materials are lithium iron phosphate, lithium manganese phosphate, lithium cobalt oxide, silica, manganese dioxide, cadmium, titanium dioxide and carbon." LGES-

1001 (the ’924 Patent), 2:40-45, 4:34-37. Therefore, because the ion active material in the Ohata-Kavan electrode material includes **phosphate olivines** such as **LiMnPO4** and **LiFePO4**, which are well known examples of lithium metal salts, and further because such **phosphate olivines** further contain **an element** selected from the group consisting of: **iron**, **manganese**, cobalt, copper, nickel, vanadium, titanium, and mixtures thereof, Ohata-Kavan renders obvious this limitation.

76. Moreover, based on Kavan, the nanotubes in the combination electrode material paste have phosphate olivine crystals (**crystals of ion active material**) **attached to their surface**. For instance, the phosphate olivine crystals are attached to the nanotube surface due at least in part to the surface oxidation of the nanotubes (*see* [1d], *infra*): “The surface oxidation of carbon nanotubes … was sometimes mentioned to improve ***the anchoring of … $LiMPO_4$***.” LGES-1005 (Kavan), 2; *see id.* (“detailed understanding is desirable of the MWNT functionalization (by surface oxidation forming carboxyls) for ***optimum attachment of MWNT towards … olivine surfaces***”), 4 (“the interaction of -COOH with phosphate surface”). For instance, the attachment between the phosphate olivine (*e.g.*, $LiFePO_4$ or $LiMnPO_4$) and the surface of the oxidized nanotubes can be a covalent bond, *e.g.*, between the carboxyl groups on the surface

of the nanotubes and the transition metals (*e.g.*, Fe or Mn) in the $LiFePO_4$ or $LiMnPO_4$ particles.

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

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

TABLE 2

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

TABLE 2-continued

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

CNT: Carbon nanotube

"aspect ratio of 10 to 500"

Excerpt from Ohata, Table 2

78. 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**.

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

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

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

81. 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.

82. 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.

83. 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 mol (*i.e.*, 7 at.%) oxygen atoms, at 16 g/mol atomic weight; and 93 mol (*i.e.*, 93 at.%) carbon atoms, at 12 g/mol atomic weight. The oxidation level of Kavan's carbon nanotubes thus can be calculated as follows:

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

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

= 9.1 wt.% oxygen.

[2] The '924 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, 3:64-67.

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.

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

85. 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 4

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

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

87. 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

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

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

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

[4] The '924 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, 3:64-67.

89. 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

90. 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)

(iii) Claim 5

[5] The composition of claim 1, comprising uniformly dispersible oxidized carbon nanotubes having ion active materials attached to their surface.

91. The combination composition includes oxidized (as I discussed for element [1d], *supra*) carbon nanotubes (as I discussed for element [1a], *supra*)

having ion active materials attached to their surface (as I discussed for element [1b], *supra*).

92. 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].

93. 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.

94. As described above, the combination composition includes uniformly dispersed, oxidized carbon nanotubes having ion active materials attached to their surface.

95. 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**.

96. 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.

B. [GROUND 2A] – CLAIMS 1, 4, AND 5 ARE OBVIOUS OVER CHENG-GAN

97. It is my opinion that a POSITA would have found claims 1, 4, and 5 to be obvious over the combination of Cheng and Gan, as I discuss in the following sections.

1. Cheng describes obtaining carbon nanotube composite based on conducting agent and lithium iron phosphate

98. Cheng describes issues regarding [1] poor conductivity of electrode material of a lithium-ion battery and [2] dispersion of carbon nanotubes themselves and dispersion of the carbon nanotubes at a precursor. *See* LGES-1007 (Cheng), [0003], [0008], [0010], [0012]. For instance, Cheng states that "it is difficult to further increase the power density because of the poor conductivity of the electrode material," and it was recognized in the prior art that "the conductivity of the composite electrode is the most important factor in designing a high performance battery." *See id.*, [0003], [0008].

99. More specifically, Cheng describes that the issues include [1] carbon nanotubes not being "fully dispersed" [2] carbon nanotubes "being distributed only on the surface of the electrode material," [3] carbon nanotubes not "form[ing] a continuously connected three-dimensional electrically conductive network structure both in the interior and on the surface of the material." *See id.*, [0010], [0012].

100. Cheng focuses on addressing the above issues (poor conductivity of electrode material of a lithium-ion battery and dispersion of carbon nanotubes) by focusing on improving carbon nanotube dispersion (dispersion of carbon nanotubes and dispersion in a precursor), such that carbon nanotubes can "form a continuously connected three-dimensional electrically conductive network structure both in the interior and on the surface of the material," thereby improving electrical conductivity of the positive electrode composite material and the lithium ion batteries. *See id.*, [0005], [0007], [0008], [0010], [0012], [0021], [0031]-[0033]. For instance, when the carbon nanotubes "form a continuously connected three-dimensional electrically conductive network structure," it improves the electrical conductivity of the composite material, as well as the electrical conductivity of the electrode and the utilization rate of the active material. *Id.,* [0005] ("Carbon nanotubes have good electrical conductivity, and the fibrous structure thereof can better connect electrode active materials into a continuous "network" that is electrically conductive, thereby improving the electrical conductivity of the electrode and the utilization rate of the active material), [0007] ("Uniform dispersion of carbon nanotubes can also significantly improve infiltration of the electrolyte in the electrode material"), [0008] ("introducing an appropriate quantity of carbon nanotubes into the electrode material and dispersing the same uniformly can greatly improve both the power density and the cycle

performance of the lithium ion battery, thereby achieving high use value"), [0021] ("Only when the effective conductive network is formed, the conductive network and the acetylene black can act on each other to achieve a synergistic effect"), Abstract ("When the obtained composite cathode material is used as a positive electrode of a lithium ion battery, the power output and the cycle characteristics can be significantly improved").

101. Cheng focuses on improving carbon nanotube dispersion by providing a carbon nanotube/lithium iron phosphate composite material for producing a positive electrode of the lithium ion battery. *Id.,* [0010] ("[u]niform dispersion of carbon nanotubes in the bulk phase and on surfaces of lithium iron phosphate is achieved by means of an in situ solid phase reaction process, thereby solving the problem in the prior art in which carbon nanotubes are distributed only on the surface of the electrode material and cannot form a continuously connected three-dimensional electrically conductive network structure both in the interior and on the surface of the material, and also solving the problem in which the properties of carbon nanotubes cannot be fully utilized because the carbon nanotubes cannot be fully dispersed"). The composite material is prepared by at least making the carbon nanotubes, acetylene black (conducting agent), and dispersant mixture "in water or an organic solvent" based on ultrasonic dispersion, mixing the mixture with "a lithium source, an iron source and a phosphate salt," and performing

further mixing and processing steps. *See id.,* Abstract, [0012]–[0020], [0040], [0041], [0044], [0048], [0049], [0063], [0064], [0071], [0072], [0076], [0077], claim 2, claim 8. Cheng describes that the obtained carbon nanotube composite material [1] provides "carbon nanotube dispersion (dispersion of carbon nanotubes and dispersion in a precursor)," [2] exhibits a uniform distribution of carbon nanotubes both "on the surface of and in the lithium iron phosphate material," [3] forms "effective conductive network" or "a uniformly distributed network structure," thereby enhancing the electrical conductivity of the carbon nanotube composite material. *See id.,* Abstract, [0012], [0021], [0023], [0030]–[0033]

2. Gan describes obtaining carbon nanotube composite based on conducting agent and functionalization of the carbon nanotube

102. The "subject matter" of Gan "relates to composites of carbon nanotubes and conductive polymer materials [corresponding to conducting agents]." LGES-1021 (Gan), [0002]. More specifically, Gan describes "functionalized carbon nanotubes" that are mixed with conducting agents to obtain "composites that exhibit increased electrical conductivity." *Id.,* Abstract. "Generally such mixing can be achieved by adding the functionalized CNTs [carbon nanotubes] to the conductive polymer material directly and sonicating the mixture to uniformly disperse the functionalized CNTs in the conductive polymer material." *Id.*, [0026].

103. Gan attributes this "increased electrical conductivity" of the functionalized carbon nanotube / conductive polymer composite primarily to improved dispersibility of nanotubes in water or polymer in Gan, resulting in a better constructed electrically conductive network (or "percolated network") within the composite. *See id.,* Abstract, [0018]-[0024], [0041] ("percolated network refers to the network of CNTs and conductive polymer that form a continuous conductive path extending through the composite. The presence of such continuous conductive path through the composite is believed to be a reason why the electrical conductivity of the conductive polymer [conducting agent] material increases when CNTs are present."). For instance, Gan describes that "[a]gglomeration of the CNTs within the polymer material matrix is undesirable because it results in formation of voids and aggregates in the composite which adversely affects the electrical conductivity properties of the composite as well as the physical properties of the composite" and Gan focuses on improving the dispersibility of the carbon nanotubes. *See id.,* [0019], [0022], [0023], [0024], [0027]. To achieve improved dispersibility of the carbon nanotubes, Gan controls the functionalization level (e.g., oxidation level) of the carbon nanotubes. *See id.,* [0018]-[0024].

104. Gan further attributes the "increased electrical conductivity" of the composite to the reduced contact resistance between the carbon nanotubes at or within the composite. *See id.,* [0020]-[0023]

105. For instance, Gan describes that "[t]he functionalization of the CNTs achieves at least one or more of the following objectives," which include the "reduce[d] contact resistance between CNTs" and improvement in "solubility and dispersibility of the CNTs in water and conductive polymer materials." *See id.* For instance, Gan explains that the improved dispersibility and the reduced contact resistance are due to the presence of carboxyl and/or hydroxyl groups that are "attached to the sidewalls of the CNTs." *Id.,* [0022] ("carboxyl groups (—COOH) are a preferred functional group that can be chemically attached to the CNT's because they render the CNT more soluble in water and conductive polymer materials . . . In addition, carboxyl groups can form hydrogen bonds between carbonyl and hydroxyl groups of two different CNTs that contain carboxyl groups . . . effectively reducing electrical contact resistance between the CNTs . . . Hydroxyl groups [are] effective at increasing the solubility and dispersibility of the CNTs").

106. Further, Gan emphasizes controlling the degree of functionalization of the carbon nanotubes with carboxyl and/or hydroxyl groups to obtain [1] improved dispersibility of nanotubes in water or polymer, [2] reduced contact resistance of

the carbon nanotubes, and [3] preservation of the intrinsic properties of the carbon nanotubes, including the electrical conductivity of pristine carbon nanotubes. *See id.,* [0019]-[0024]. For instance, Gan discloses that the "suitable functionalization with carboxyl or hydroxyl groups ranged from about 2.5 to about 10 wt%" "2.5 to about 10 wt%," which indicates a safe oxidation level range that preserves the intrinsic properties of the carbon nanotubes while maximizing their dispersibility in water or polymer. *See id.,* [0024]. Moreover, intrinsic properties include the electrical conductivity of pristine carbon nanotubes. For instance, Gan explicitly states that:

> The degree to which the CNTs are functionalized is a balance between introducing the desired functional groups into the CNTs to achieve one or more of the three objectives, while minimizing the effect on the desirable intrinsic properties of the CNTs. For example, while a greater degree of functionalization may improve the dispersibility of the CNTs, such improvement may come at a decrease in intrinsic properties of the pristine CNTs.

Id., [0024].

3. The Cheng-Gan combination

107. In the Cheng-Gan combination, the carbon nanotubes in Cheng's carbon nanotube/lithium iron phosphate composite would have carbon nanotubes that are functionalized with carboxyl or hydroxyl groups at "about 2.5 to 10 wt%"

or have oxidation levels at "about 2.5 to 10 wt%," based on Gan's disclosure.[5] LGES 1001, 3:60-64; LGES-1021 (Gan), [0024]. I note that the '924 Patent defines the term "oxidized carbon nanotubes" as carbon nanotubes oxidized to have "carboxylic, ketone or hydroxyl functionalities." LGES-1001 (the '924 Patent), 3:60-63. Gan's description regarding functionalization of nanotubes with carboxyl groups and hydroxyl groups equates to the oxidation of nanotubes. LGES-1021 (Gan), [0018]-[0023]. The term "functionalization" can be interchangeably used with "oxidation," at least in the instances relating to functionalizing the carbon nanotubes with functional groups having carboxylic, ketone or hydroxyl functionalities.

108. As shown below, the composition of the Challenged Claims is rendered obvious by [1] functionalizing the carbon nanotubes of Cheng using well-known methods, as described in Gan, or [2] obtaining the functionalized carbon nanotubes from commercial sources, as described in Gan, and applying such functionalized nanotubes in preparing the composite described in Cheng. *See* LGES-1021 (Gan), [0018]-[0024].

[5] The '924 Patent defines the term "oxidized carbon nanotubes" as carbon nanotubes oxidized to have "carboxylic, ketone or hydroxyl functionalities." LGES-1001, 3:60-63

4. Reasons for the combination

109. It is my opinion that a POSITA would have been motivated to use oxidized nanotubes (or oxidized nanotubes having oxidation levels such as those described in Gan) in the Cheng's nanotube composite for at least the following reasons.

110. The first reason for using oxidized nanotubes (or oxidized nanotubes having the functionalization levels (oxidation levels) such as those described in Gan) is that a POSITA would have recognized the importance of "about 2.5 to 10 wt%" oxidation levels in **obtaining a better constructed electrically conductive network of carbon nanotubes within the nanotube / lithium iron phosphate composite**, which is a design objective in Cheng. *See* LGES-1007 (Cheng), Abstract, [0003]-[0010], [0021], [0030]–[0033]. Bearing in mind the well-known role of **functionalization** of the carbon nanotubes in improving their dispersibility in an aqueous solution and the well-known fact that the **functionalized carbon nanotubes** can retain their dispersion and incorporate directly into the porous $LiFePO_4$ particulates, resulting in an intimately interlaced 3D composite network and promising electrode performance of the lithium ion batteries, a POSITA would have looked to Gan for guidance as to the functionalization level or oxidation level appropriate for this objective. LGES-1032 (Zhou), 1 ("**The porous LiFePO4–CNT composites were synthesized via a facile in situ sol–gel method** (see

details in the experimental section of ESI), where **the carbon nanotubes were functionalized** using a mixed acid method **to ensure uniform dispersion** into the aqueous sol. The nanotubes **retained their dispersion** and were **incorporated directly into the porous LiFePO4 particulates during the subsequent gelatinization and calcination steps resulting in an intimately interlaced 3D composite network**"). For instance, Zhou describes that the nanotubes were incorporated throughout the $LiFePO_4$ particulates during the calcination steps and the functionalized nanotubes have "retained their dispersion" throughout the process and achieved the "**highly dispersed conductive networks**," where the "uniform dispersion of **CNT networks** into the $LiFePO_4$ is crucial for performance." *Id.*

111. For instance, a POSITA would have recognized Gan's teachings about using carbon nanotubes that are functionalized with carboxyl or hydroxyl groups at "about 2.5 to 10 wt%," and would have applied this knowledge to the nanotubes in Cheng according to Gan's guidance. *See* LGES-1021 (Gan), [0018]-[0024]. As described above, Gan's guidance regarding "2.5 to 10 wt%" oxidation level is highly relevant to obtaining a better constructed electrically conductive network of the carbon nanotubes within the composite. *See id.*, Abstract, [0018]-[0024], [0041]. For instance, "2.5 to 10 wt%" oxidation level is highly relevant in [1] improving dispersibility of nanotubes in water or polymer, and [2] reducing

contact resistance of the carbon nanotubes among the carbon nanotubes themselves or carbon nanotube composite formed based on the conducting agent. *See id.,* [0018]-[0024].

112. The second reason for using oxidized nanotubes (or oxidized nanotubes having the functionalization levels (oxidation levels) such as those described in Gan) is that a POSITA would have been motivated to enhance **dispersion of carbon nanotubes in water** and/or conducting agent in Cheng. For instance, enhancing dispersion of carbon nanotubes in water and/or conducting agent in Cheng would have facilitated the steps that contribute to a design objective in Cheng, which relates to "carbon nanotube dispersion (dispersion of carbon nanotubes and dispersion in a precursor)". *See* LGES-1007 (Cheng), Abstract, [0010]-[0014]. For instance, Cheng makes mixture of carbon nanotubes, acetylene black (conducting agent), and dispersant by "ultrasonically dispersing the same in **water,"** and after dispersion, mixes the mixture with "a lithium source, an iron source and a phosphate salt" and "organic carbon source" using the mixing medium such as "**deionized water**." *Id.* [0013]-[0017], claim 2. With regards to this context, Gan notes that the functional groups such as carboxyl groups or hydroxyl groups "are effective at increasing the solubility and dispersibility of the CNTs in **water,**" as well as in conducting agent such as conductive polymers. *See* LGES-1021 (Gan), [0018]-[0021]. Moreover, it was well-known to a POSITA that

the carboxyl or hydroxyl groups are "hydrophilic substituents [that are incorporated] into the exterior CNT sidewalls," which improve the hydrophilicity and "create CNTs that disperse in water." *See* LGES-1010 (Wepasnick), 1 ("To improve the CNT's hydrophilicity, their surfaces are often tailored by covalent or non-covalent modification strategies. Covalent surface modification involves the incorporation of hydrophilic substituents into the exterior CNT sidewalls"), 2 ("Among the various types of covalent surface modification strategies, deliberately grafting oxygen-containing functional groups at the open ends and sidewalls of CNTs is a popular and versatile approach [5]. Indeed, this strategy is often used to create CNTs that disperse in water").

113. The third reason for using oxidized nanotubes having the functionalization levels (or oxidation levels) such as those described by Gan is that, a POSITA would have recognized Gan's guidance of oxidation levels at **"about 2.5 to 10 wt%" remain within a safe threshold that preserves "the desirable intrinsic properties of the CNTs."** *Id.,* [0024]. For instance, Gan emphasizes that, when finding a suitable level for functionalization, the goal is to "**minimiz[e] the effect on the desirable intrinsic properties** of the CNTs." For instance, Gan teaches the **suitable level of oxidation at "about 2.5 to 10 wt%" and that "while a greater degree of functionalization may improve the dispersibility of the CNTs, such improvement may come at a decrease in intrinsic properties of the**

pristine CNTs." *Id*. This indicates a safe oxidation level range that preserves the intrinsic properties of the carbon nanotubes, including electrical conductivity of pristine nanotubes, while maximizing their dispersibility in water or polymer.

114. The fourth reason of using oxidized carbon nanotubes in Cheng's design, as described in Gan, is that extensive applications of oxidized or functionalized carbon nanotubes were well-known in various fields including lithium ion battery industry. *See* LGES-1023 (Bosnyak), Abstract, [0005], [0014], [0032], [0037]; LGES-1024 (Sue), [0006] [0056] [0101]; *see supra* Part IV.A.2. & A.5.(i).(1d). For instance, Gan identifies "commercial sources such as CheapTubes, Inc . . . and Carbon Solutions, Inc.," where oxidized nanotubes having specific oxidation levels can be obtained. These sources highlight their broad applications, particularly in "[e]nergy [s]torage" components, and more particularly, in "lithium-ion batteries." LGES-1021 (Gan), [0018]; LGES-1011 (stating "CNTs have the intrinsic characteristics desired in material used as electrodes in batteries and capacitors"). Further, for instance, the Patent Owner of the '924 patent describe, in a patent application that is a prior art herein, use of exfoliated oxidized carbon nanotube in electrodes for lithium ion batteries. *See* LGES-1023 (Bosnyak), Abstract, [0005], [0014], [0032], [0037], [0074], [0077], [0078].

115. The fifth reason of using oxidized carbon nanotubes in Cheng's design, as described in Gan, is that a POSITA would have recognized well-known facts that the [1] "role of oxidation is to create 'foothold' functional groups, such as carboxyl groups, which then serve as attachment points to anchor . . . nanoparticles" such as $LiFePO_4$ and [2] oxidized carbon nanotubes improve the "colloidal stability of CNTs," ensuring their uniform dispersion. *See* LGES-1010 (Wepasnick), 2 ("In functionalization strategies, the role of oxidation is to create ''foothold'' functional groups, such as carboxyl groups, which then serve as attachment points to anchor either larger biomolecular structures [31,32] or nanoparticles [19,28–32]"), 2 ("surface oxidation has been shown to exert a pronounced effect on both the sorption properties and colloidal stability of CNTs [12,13,29,30]"). Based on this knowledge, along with teachings of Cheng and Gan, a POSITA would have understood that the functional groups on oxidized nanotubes [1] facilitate the bonding between carbon nanotubes and nanoparticles (such as lithium iron phosphate) and [2] improve the "colloidal stability of CNTs." *Id.* This supports Cheng's design objective of achieving better dispersion of both the carbon nanotubes themselves and throughout the lithium iron phosphate. *See* LGES-1007 (Cheng), [0010]–[0014]. Further, as described above, Gan specifies a suitable degree of oxidation level (e.g., "2.5–10 wt%"), which would have motivated a POSITA, as they would have understood that this level of oxidation

[1] is within the safe threshold that preserves "the desirable intrinsic properties of the CNTs," [2] improves the dispersibility of the carbon nanotubes in water, and [3] aids in reducing contact resistance between the carbon nanotubes to boost conductivity.

116. The sixth reason of using oxidized carbon nanotubes in Cheng's design, as described in Gan, is that a POSITA would have been motivated to pursue this implementation due to its ease of implementation, easy access to functionalized nanotubes, and predictable results. For instance, implementing carbon nanotubes would have involved either [1] obtaining "functionalized SWNTs and MWNTs . . . from commercial sources" or [2] the application of "known techniques such as by sonicating pristine CNTs in an acid bath of sulfuric acid and nitric acid" to a known structure (carbon nanotube) to yield predictable results. LGES-1021 (Gan), [0018]. For instance, the predictable results based on functionalization of the carbon nanotubes include at least the following:

[a] improving dispersibility of the carbon nanotubes;

[b] retaining carbon nanotubes dispersion and incorporation into $LiFePO_4$ particulates, which in turn result in a better constructed electrically conductive network of the carbon nanotubes within the composite and "yields promising electrode performance" of lithium ion batteries;

[c] reducing contact resistance between the carbon nanotubes to thereby enhance electrical conductivity of the nanotube composite;

[d] improving the "colloidal stability of CNTs" to further ensure uniform dispersion of carbon nanotubes; and

[e] facilitating the bonding between carbon nanotubes and nanoparticles (such as lithium iron phosphate) while not harming **"**the desirable intrinsic properties of the CNTs."

LGES-1010 (Wepasnick), 1, 2; LGES-1021 (Gan), [0018]-[0024], [0029], [0030]-[0033]; LGES-1032 (Zhou), 1.

It would have been a very simple and easy implementation as POSITA can easily obtain the functionalized carbon nanotube from commercial sources prior to performing initial steps of making the carbon nanotube, acetylene black, and dispersant mixture at water based on ultrasonic dispersion in Cheng.

117. Moreover, it is my opinion that a POSITA would have expected success in incorporating nanotubes having functionalization levels (or oxidation levels) such as those described by Gan in the Cheng-Gan combination for at least the following reasons.

118. First, as described above, the oxidized carbon nanotubes or the oxidation levels taught in Gan: [1] remain within the safe threshold that preserves (or does not harm) the desirable intrinsic properties of the carbon nanotubes; [2]

help producing better constructed electrically conductive network of carbon nanotubes within the composite; [3] help reduce contact resistance between carbon nanotubes to thereby enhance conductivity of the nanotube composite; [4] improve the dispersibility of carbon nanotubes; [5] improve the "colloidal stability of CNTs" to further ensure uniform dispersion of carbon nanotubes; and [6] provide enhanced "sorption properties," as they would offer further "attachment points to anchor" lithium iron phosphate. LGES-1010 (Wepasnick), 1, 2; LGES-1021 (Gan), [0018]-[0024], [0029], [0030]-[0033]; LGES-1032 (Zhou), 1.

119. Second, oxidation (or functionalization) of carbon nanotube with carboxyl or hydroxyl groups, or effect of such oxidation in enhancing electrical conductivity of the electrode or nanotube composite and improving dispersibility of the carbon nanotubes were well-known in lithium ion battery industry. *See* discussion *supra* Part IV.A.2. As described above, 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" and discloses cyclic voltammograms with results for electrodes composed of a composite of $LiFePO_4$ and non-oxidized nanotubes and electrodes composed of a composite of $LiMnPO_4$ and oxidized nanotubes. *Id.* Again, Kavan characterizes the results for the oxidized nanotube

based composite as "impressive" in terms of both "capacity" and "charge/discharge reversibility." *Id.*

120. Third, enhancing "sorption properties" between carbon nanotubes and ion active materials (or transition metal oxide) based on functionalization of carbon nanotubes was well-known in the lithium ion battery industry. *See* LGES-1010 (Wepasnick), 2; LGES-1030 (Xiang), 2; LGES-1027 (Yu), 6. For instance, it was well-known that carboxyl or hydroxyl groups on functionalized nanotubes offer further "attachment points to anchor either larger biomolecular structures or nanoparticles." *See* LGES-1010 (Wepasnick), 2. Further, it was demonstrated that ion active materials or transition metal oxide, such as copper oxide or titanium dioxide precursors or nanoparticles that are formed on the carbon nanotubes, interact or react with the carboxyl or hydroxyl groups of the functionalized carbon nanotubes to form attachment or chemical bonding. LGES-1027 (Yu), 6 ("Such surface functional groups provide active sites for directed deposition of anatase nanocrystals around the surface of MWCNTs. For example, the –OH groups on the Ti precursor possibly react with the –OH and –COOH groups on the MWCNTs surface through esterification to form C–O–Ti or O=C–O–Ti bonds"); LGES-1030 (Xiang), 2 ("Due to the electrostatic effects, some Cu (II) ions added in the solution are absorbed in the outward end of the micelles while some are still dispersed in the solution (Step B). Once NaOH solution is dropped in, $Cu(OH)_2$

nanowires are immediately formed, some lie on MWCNTs while some are around MWCNTs (Step C). Then, $Cu(OH)_2$ nanowires dispersed in the solution self-assemble parallel to those lying on MWCNTs and become leaf-shaped nanoplates (Step D)"; LGES-1030 (Xiang), 3 "The enhanced capability and reversibility of CuO/MWCNT is attributed to the MWCNT network in the composite. Firstly, MWCNTs dispersed in the composite build up a network to connect CuO … ensure good electrical contact of active materials. Secondly, MWCNTs incorporated into CuO … provide fast diffusion channels for Li+ and facilitate the charge–transfer on CuO/electrolyte interfaces. Thirdly, MWCNTs with excellent mechanical property act as a soft matrix to buffer the volume changes and decrease the pulverization and deterioration of CuO during cycling").

121. Fourth, oxidation levels that fall within the range disclosed in Gan (2.5–10 wt%) were well-known to be used extensively in various technical fields, including lithium ion battery. *See supra* Part IV.A.5.(i).(1d); *see also* LGES-1011.

122. Fifth, oxidation levels that fall within the range disclosed in Gan (2.5–10 wt%) were well-known to enhance electrochemical activity of the nanotube composite materials with LiFePO4." *See supra* Part IV.A.2 & A.5.(i).(1d). For instance, Kavan demonstrates an **oxidation level** of **9.1%** and states that in the $LiFePO_4$/MWNT-ox composite "[t]he improvement caused by surface oxidation of nanotubes is dramatic" and "[i]t is obvious that the functionalization of nanotubes

is, indeed, crucial for enhancing the electrochemical activity of the composite materials with $LiFePO_4$." LGES-1005 (Kavan), 7, 8. For instance, Kavan 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.

123. Therefore, a POSITA would have expected success in applying nanotubes having functionalization level (or oxidation level) such as those described by Gan in the Cheng's carbon nanotube/lithium iron phosphate composite.

5. Claim-by-Claim Analysis

(i) Claim 1

[1pre] A composition used in lithium ion batteries comprising:

124. To the extent the preamble is deemed to be limiting, Cheng-Gan renders it obvious.

125. In the Cheng-Gan combination, Cheng's carbon nanotube/lithium iron phosphate composite is a composition used to produce the "cathode material" for positive electrode of the lithium ion battery. *See* LGES-1007 (Cheng), Abstract. When Cheng's carbon nanotube/lithium iron phosphate composite is used as the "cathode material" for positive electrode of the lithium ion battery, "the power output and the cycle characteristics can be significantly improved," "the capability

of binding with an aluminum foil current collector is strong," "electrode sheet has high compacted density," and "[t]he flexibility and winding property of the electrode sheet are good." *Id.*

[1a] discrete carbon nanotubes

126. At least for the reasons described below, carbon nanotubes in Cheng-Gan combination are discrete.

127. First, in the Cheng-Gan combination, Cheng describes and claims methods of "carbon nanotube dispersion," in which the carbon nanotube disperses **[1]** themselves and **[2]** in a "precursor" that includes lithium iron phosphate. *See* LGES-1007 (Cheng), Abstract, [0012]-[0015], claim 2. Cheng states that these methods address issues such as **[1]** the inability of the "carbon nanotubes" to disperse fully, which had led to the "problem in which the properties of carbon nanotubes cannot be fully utilized," **[2]** the carbon nanotubes being "distributed only on the surface of the electrode material," and **[3]** the carbon nanotubes being unable to "form a continuously connected three-dimensional electrically conductive network structure both in the interior and on the surface of the material." *See id.*, [0010]. Therefore, Cheng focuses on solving the issues of nanotube dispersion and states that resulting composite material contains nanotubes that are "**uniformly dispersed**." *See id.*, Abstract, [0010], [0012], [0041], claim 1.

128. For instance, Cheng describes that the steps for "carbon nanotube dispersion" include at least ultrasonically dispersing the carbon nano-tube, acetylene black, and dispersant (e.g., polyvinylpyrrolidone (PVP), polyvinyl alcohol (PVA)) "in water or an organic solvent to obtain a mixture." *Id.*, [0013], claim 2. Cheng describes that the steps for dispersing the carbon nanotube in "the precursor" include at least **[1]** mixing the above "mixture" with "a lithium source, an iron source, and a phosphate salt at an atom ratio of Li : Fe : P = 1 : 1 : 1" and "organic carbon source" (e.g., sucrose, glucose or phenolic resins) using "mixing medium" (e.g., least one of "deionized water, industrial alcohol, or the absolute ethyl alcohol") and **[2]** ball milling and drying.[6] *See id.,* [0013]-[0018], claim 2. After ball milling, the precursor powder is calcined and sieved. *See id.,* [0019], claim 2. As a result of the carbon nanotube dispersion (the [1] "dispersion of carbon nanotubes" and [2] "dispersion in a precursor" that includes lithium iron

[6] Cheng fabricates its cathode active material using a "ball-milling" process. LGES-1007, Abstract, [0015]. I understand that the accused products are fabricated using a ball milling process. To the extent ball milling is compatible with discrete carbon nanotubes, it is my opinion that the Cheng's process is compatible with discrete nanotubes.

phosphate), the carbon nanotubes are "**uniformly dispersed**." *Id.,* Abstract, [0010], [0012], [0041], claim 1.

129. Second, in the Cheng-Gan combination, Cheng emphasizes that the carbon nanotube composite material obtained based on the disclosed methods result in **non-aggregated** and **uniform dispersion** of nanotubes. For instance, Cheng emphasizes that the "carbon nanotubes or carbon nanofibers are uniformly dispersed" in both "the surface of and in the lithium iron phosphate material" and that "the infiltration of the electrolyte in the electrode material" can be "significantly improve[d]" as a result of "the uniform dispersion of carbon nanotubes." *See* LGES-1007 (Cheng), Abstract, [0007], [0010], [0021]. Further, for instance, Cheng emphasizes that the carbon nanotubes of the composite "form a uniformly distributed network structure" internally within the composite material, as a result of the uniform dispersion of the carbon nanotube. *See id.*, [0023]. In addition to emphasizing that the nanotubes of composite are uniformly dispersed and form uniformly distributed network structure, Cheng emphasizes that the nanotubes are in **non-aggregated state**. For instance, Cheng describes that when there is an "agglomerat[ion]," it is difficult to form an "effective conductive network" or an "uniformly distributed network structure." *Id.*, [0023], [0073]. Accordingly, the carbon nanotube composite material of Cheng result in **non-aggregated** and **uniform dispersion** of nanotubes.

130. Third, in the Cheng-Gan combination, Cheng describes use of diameter and length that contribute to lesser aggregation and better dispersion of carbon nanotubes, thereby contributing to the **uniform dispersion** and **non-aggregated state** of **nanotubes**. For instance, Cheng describes the use of carbon nanotubes having "tube diameter of 10 to 200 nm and the length of 5 to 20 μm," and "tube diameter of 50 to 150 nm and the length of 10to 15 μm" for the "optimal effect," and states that the described ranges of diameter and length contribute to forming of "effective conductive network," where "the carbon nanotubes are **less entangled** with each other and are **more easily dispersed**." *Id.*, [0021].

131. Fourth, in the Cheng-Gan combination, when Cheng is modified to use the **oxidized nanotubes**, as described in Gan, the method of carbon nanotube dispersion (dispersion of carbon nanotubes and dispersion in a precursor) is similar to a dispersion method disclosed in the '924 patent. For instance, the '924 patent has two examples (Examples 2 & 3), which allegedly describe "Discrete Carbon Nanotubes with Lithium Iron Phosphate Coated on the Surface of the Tubes Suitable for a Lithium Ion Battery Cathode Material." LGES-1001, 5:30-67, 6:1-4. Here, the Example 2 of the '924 patent describes sequential steps of [1] forming oxidized carbon nanotube / solvent mixture based on ultrasonic dispersion, [2] mixing the mixture with lithium source, iron source and phosphate source with mixing medium (e.g. deionized water), and [3] heat treatment. Similarly, Cheng,

in the Cheng-Gan combination, when utilizing the oxidized nanotube as taught by Gan, discloses each and all of the above-noted steps of Example 2 in sequential order. *See* LGES-1007 (Cheng), Abstract, [0013]-[0019], claim 2. This suggests that similar steps in forming the composite would have likely yielded similar dispersion results. To add more, as described above with respect to the "Reasons for the combination" section, Zhou describes a similar method where the **functionalized carbon nanotubes** / $LiFePO_4$ composite were produced via "in-situ" method, where the nanotubes were incorporated throughout the $LiFePO_4$ particulates during the calcination steps. *See* ¶110. In Zhou, the functionalized nanotubes have "**retained their dispersion**" throughout the process and achieved the "**uniform dispersion**" and "**highly dispersed** conductive networks." *See id*.

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

chemically prevent the nanotubes from aggregating into clumps and instead allow the nanotubes to be generally present as individually distinct nanotubes. *See* LGES-1005 (Kavan), Abstract; LGES-1010 (Wepasnick), Abstract; LGES-1014 (Gojny), 821-822; LGES-1015 (Xing), 4186-4187; LGES-1008 (Farrugia), [0027]-[0028]; LGES-1032 (Zhou), 1. In light of this well-known behavior of oxidized nanotubes, a POSITA would have understood or found obvious that oxidized nanotubes of the combination composition can be present as generally individually distinct nanotubes, *e.g.*, that the nanotubes are **discrete**.

[1b] having ion active material attached to their surface . . . wherein the 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

133. In the Cheng-Gan combination, Cheng's carbon nanotube/lithium iron phosphate composite is a composition used to produce the "cathode material" for positive electrode of the lithium ion battery and the lithium iron phosphate corresponds to an ion active material.[7] LGES-1007 (Cheng), Abstract. The

[7] As described above with respect to paragraph 75, I notice the following: claim states that the ion active material includes both a lithium metal salt and an element selected from the specified group. I understand this element can either be a constituent of the lithium metal salt or another component of the ion active

lithium iron phosphate includes a lithium metal salt having an element selected from the group consisting of: **iron**, manganese, cobalt, copper, nickel, vanadium, titanium, and mixtures thereof. Therefore, because the ion active material in the Cheng-Gan electrode material includes lithium iron phosphate, which is a well-known example of lithium metal salts, and further because such lithium iron phosphate further contains **an element** selected from the group consisting of: **iron**, manganese, cobalt, copper, nickel, vanadium, titanium, and mixtures thereof, Cheng-Gan renders obvious this limitation. Further, lithium iron phosphate, as active ion materials, are attached to the nanotubes surfaces at least for the following reasons.

134. First, in the Cheng-Gan combination, Cheng's methods of forming the composite, including **[1]** mixing carbon nanotube, acetylene black, and dispersant mixture with lithium source, iron source, phosphate salt, and organic carbon source

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material. For instance, none of the relevant experimental examples in the '924 Patent disclose extra "element" in addition to the lithium metal salt and it merely states that "[e]xamples of ion active materials are lithium iron phosphate, lithium manganese phosphate, lithium cobalt oxide, silica, manganese dioxide, cadmium, titanium dioxide and carbon." LGES-1001 (the '924 Patent), 2:40-45, 4:34-37; *see supra* ¶75.

using mixing medium, **[2]** ball milling, **[3]** drying, **[4]** calcination, and **[5]** sieving indicate that the ion active material [lithium iron phosphate] is attached to the surfaces of carbon nanotubes. *See* LGES-1007 (Cheng), [0013]-[0018], claim 2. For instance, Cheng claims and describes "a carbon nanotube/iron phosphate composite," in which the carbon nanotubes are "uniformly dispersed in a bulk phase and on surfaces of lithium iron phosphate," and describes that "[t]he the high-performance carbon nanotube lithium iron phosphate composite material obtained" has features of the "carbon nanotubes or carbon nanofibers" being "**uniformly distributed on the surface of and in the lithium iron phosphate material** by means of an **in situ solid phase reaction**." LGES-1007 (Cheng), Abstract, [0012], [0021], claim 1, 2. For instance, Cheng claims and describes that the "**in situ solid phase reaction**" includes certain steps including [1] calcining the dried precursor powder at 600-850 ℃ for 4-49 hours, [2] pulverizing the burned material, and [3] then sieving the same through a sieve of 150-400 meshes (here, the dried precursor powder refers to a powder that is obtained by [a] mixing carbon nanotube, acetylene black, and dispersant mixture with lithium source, iron source, phosphate, and organic carbon source using the mixing medium, [b] ball milling the mixture, and [c] drying the mixture). *See id.*, [0019], claim 2. As a consequence of above-noted steps, Cheng obtains "the particulate homogeneous composite of carbon nanotubes and the lithium iron phosphate cathode material,"

where the carbon nanotubes "not only are present on the surface of the material, but also form a uniformly distributed network structure inside the material." *Id.*, [0022], [0023].

135. Accordingly, the above-noted steps in Cheng indicate that the **ion active material [lithium iron phosphate] are attached to the surface of the carbon nanotubes**.

136. Second, in the Cheng-Gan combination, Cheng can incorporate oxidized nanotubes of Gan, and steps of attaching or coating the "**ion active material [lithium iron phosphate]" to the surface of the carbon nanotubes** are similar to those described in the '924 patent. For instance, the '924 patent has two examples (Examples 2 & 3), which allegedly describe "Discrete Carbon Nanotubes with Lithium Iron Phosphate ***Coated on the Surface of the Tubes*** Suitable for a Lithium Ion Battery Cathode Material." LGES-1001, 5:30-67, 6:1-4. Here, the Example 2 of the '924 patent describes sequential steps of [1] forming oxidized carbon nanotubes / solvent mixture based on ultrasonic dispersion, [2] mixing the mixture with lithium source, iron source and phosphate source with mixing medium (e.g. deionized water), and [3] heat treatment. *See id.,* 5:30-53.

137. Similarly, Cheng, when utilizing the oxidized nanotube as taught by Gan, discloses each and all of the above-noted three steps of Example 2 in sequential order. *See* LGES-1007 (Cheng), Abstract, [0013]-[0019], claim 2. For

instance, Cheng describes the steps for [1] ultrasonically dispersing the carbon nano-tube, acetylene black, and dispersant (e.g., polyvinylpyrrolidone (PVP), polyvinyl alcohol (PVA)) "in water or an organic solvent to obtain a mixture" and [2] mixing the "mixture" with "a lithium source, an iron source, and a phosphate salt at an atom ratio of Li : Fe : P = 1 : 1 : 1" and "organic carbon source" (e.g., sucrose, glucose or phenolic resins) using "mixing medium" (e.g., deionized water), ball milling the overall mixed precursor powder or composite, and [3] calcining the overall mixed precursor powder or composite. *See id.,* [0013]-[0018], claim 2. This suggests that **similar steps in "coat[ing]" or attaching the lithium iron phosphate would have likely yielded similar attachment results**.

138. Third, in the Cheng-Gan combination, Cheng can incorporate oxidized nanotubes of Gan, and it was well-known in the lithium ion battery industry that oxidized nanotube would improve the attachment of the ion active material (lithium iron phosphate or the like) to the carbon nanotube surface.

139. For instance, it was well-known that "[t]he surface oxidation of carbon nanotubes by HNO_3, H_2SO_4/HNO_3 or H_2O_2, producing among other groups also carboxyls, was sometimes mentioned to improve the anchoring of … $LiMPO_4$" [e.g., M = transition metal including Fe]). LGES-1005 (Kavan), 2. For instance, it was well-known that [1] "the role of oxidation is to create 'foothold' functional groups, such as carboxyl groups, which then serve as attachment points

to anchor either larger biomolecular structures or nanoparticles," and [2] the "surface oxidation has been shown to **exert a pronounced effect** on both the sorption properties . . . of CNTs." LGES-1010 (Wepasnick), 2. Again, referring to paragraph 76 and other discussions relating to Kavan, it was demonstrated that oxidation of nanotubes improves the attachment between the phosphate olivine (e.g., $LiFePO_4$) and the functional groups on the surface of oxidized nanotubes and such attachment. *See* ¶76.

140. Further, it was well-known, with regards to the battery electrode materials development, that ion active materials or transition metal oxide, such as copper oxide or titanium dioxide precursors or nanoparticles that are formed on the carbon nanotubes, interact or react with the carboxyl or hydroxyl groups of the functionalized carbon nanotubes to form attachment or chemical bonding. LGES-1027 (Yu), 6 ("Such surface functional groups provide active sites for directed deposition of anatase nanocrystals around the surface of MWCNTs. For example, the –OH groups on the Ti precursor possibly react with the –OH and –COOH groups on the MWCNTs surface through esterification to form C–O–Ti or O=C–O–Ti bonds"); LGES-1030 (Xiang), 2 ("Due to the electrostatic effects, some Cu (II) ions added in the solution are absorbed in the outward end of the micelles while some are still dispersed in the solution (Step B). Once NaOH solution is dropped in, $Cu(OH)_2$ nanowires are immediately formed, some lie on MWCNTs

while some are around MWCNTs (Step C). Then, $Cu(OH)_2$ nanowires dispersed in the solution self-assemble parallel to those lying on MWCNTs and become leaf-shaped nanoplates (Step D)"; LGES-1030 (Xiang), 3 " The enhanced capability and reversibility of CuO/MWCNT is attributed to the MWCNT network in the composite. Firstly, MWCNTs dispersed in the composite build up a network to connect CuO … ensure good electrical contact of active materials. Secondly, MWCNTs incorporated into CuO … provide fast diffusion channels for Li+ and facilitate the charge–transfer on CuO/electrolyte interfaces. Thirdly, MWCNTs with excellent mechanical property act as a soft matrix to buffer the volume changes and decrease the pulverization and deterioration of CuO during cycling").

141. Further, for instance, it was demonstrated that the oxidized nanotubes "have a negative electrostatic charge on the outer surface of the nanotube wall" and a POSITA would have easily recognized that such "negative electrostatic charge on the outer surface of the nanotube wall" would be used to enhance attachment of the positive ion active material (e.g., $LiFePO_4$) to the nanotubes' surfaces. *See* LGES-1024 (Sue), [0057] ("The oxidized nanotubes . . . have a negative electrostatic charge on the outer surface of the nanotube wall").

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

142. In the Cheng-Gan combination, Cheng describes nanotubes that have an aspect ratio in the preferred range of 67-300, which falls within the claimed range ("10 to 500"). For instance, Cheng describes that "the carbon nanotubes used in the present invention need to have the tube diameter of 10 to 200 nm and the length of 5 to 20 μm, and the optimal effect is achieved when the carbon nanotubes have the tube diameter of 50 to 150 nm and the length of 10 to 15 μm. Compared with carbon nanotubes having a smaller tube diameter and a greater length, the carbon nanotubes are less entangled with each other and are more easily dispersed. Therefore, formation of an effective conductive network is facilitated." LGES-1007 (Cheng), [0021]. Based on Cheng's disclosure, Cheng's carbon nanotube exhibits the aspect ratio in the preferred range of 67-300 (e.g., length 10-15 μm is equal to 10,000 to 15,000 nm, and when length 10,000-15,000nm is divided by diameter 50-150nm, result is 66.67 to 300). As such Cheng-Gan combination's aspect ratio falls within the claimed range ("10 to 500").

143. Alternatively/additionally, taking into consideration the full range of aspect ratio taught in Cheng, with "the tube diameter of 10 to 200 nm and the length of 5 to 20 μm", the aspect ratio in Cheng corresponds to 25-2000 (e.g., length 5-20 μm is equal to 5,000 to 20,000 nm, and when length 5,000-20,000 nm is divided by diameter 10-200 nm, result is 25 to 2000). LGES-1007 (Cheng),

[0021]. Thus, the aspect ratio of 25-2000 overlaps with claimed range of 10 to 500. Further, the '924 Patent does not provide any discussion of aspect ratio, let alone any evidence, of unexpected results or criticality of the claimed range of aspect ratio.

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

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

145. In the Cheng-Gan combination, the nanotubes (e.g., oxidized nanotubes) has an oxidation level of 2.5-10 wt%. For instance, Gan discloses that the "suitable functionalization with carboxyl or hydroxyl groups ranged from about 2.5 to about 10 wt," which equates to oxidation levels at about 2.5 to 10 wt%. LGES-1021 (Gan), [0024].

146. As discussed above, Gan emphasizes controlling the degree of functionalization of the carbon nanotubes with carboxyl and/or hydroxyl groups "at about 2.5 to 10 wt%" to obtain [1] improved dispersibility of nanotubes in water or polymer, [2] reduced contact resistance of the carbon nanotubes, and [3]

preservation of the intrinsic properties of the carbon nanotubes, including the electrical conductivity of pristine carbon nanotubes. *See* LGES-1021 (Gan), [0019]-[0024].

147. For instance, Gan describes that "the degree to which the CNTs are functionalized" achieves "one or more of the three objectives" including "improv[ing] the solubility and dispersibility of the CNTs in water and conductive polymer materials." For instance, Gan attributes this solubility and dispersibility of the carbon nanotubes to the "carboxyl groups (—COOH) [, which] are a preferred functional group that can be chemically attached to the CNT's because they render the CNT more soluble in water and conductive polymer materials" and the "[h]ydroxyl groups," which "are effective at increasing the solubility and dispersibility of the CNTs." *Id.*, [0022].

148. For instance, Gan further describes another objective that can be achieved based on the functionalization corresponds to "reduc[ing] contact resistance between CNTs" that are incorporated into conducting agent (e.g., conductive polymer), thereby enhancing the conductivity of the network formed by carbon nanotubes. LGES-1021 (Gan), [0020], [0021]. For instance, Gan attributes this enhanced conductivity at least to the ability of carboxyl groups to "form hydrogen bonds between carbonyl and hydroxyl groups of two different CNTs that

contain carboxyl groups," thereby "effectively reducing electrical contact resistance between the CNTs." LGES-1021 (Gan), [0022].

149. Moreover, Gan emphasizes that "while a greater degree of functionalization may improve the dispersibility of the CNTs, such improvement may come at a decrease in intrinsic properties of the pristine CNTs." *Id.,* [0024]. This explains or implies that Gan's guidance of "at about 2.5 to 10 wt%" oxidation level, intrinsic properties of the carbon nanotubes would be preserved.

150. As described above with respect to the "Reasons for the combination" section (Part IV.B.4.), there would have been sufficient motivation for a POSITA to apply nanotubes having functionalization level (or oxidation level) such as those described by Gan to the Cheng's carbon nanotube/conducting agent/lithium iron phosphate composite, and, at the same time, would have expected success. *See* discussion *supra* Part IV.B.4.

151. Therefore, this range of oxidation level recited in Gan ("2.5 to 10%"), as applied to the nanotubes of the combination, renders obvious a composition in which the nanotubes have oxidation levels from 1% to 15% by the weight of the carbon nanotube.

152. Alternatively/additionally, oxidized nanotubes with oxidation levels that fall within the claimed range ("1% to 15%") were well-known and extensively used various applications, including lithium ion batteries and further, the effect of

oxidized nanotubes with oxidation level such as 9.1% was well-known to improve dispersibility of the carbon nanotubes and thus to enhance electrical conductivity of conductive network in the electrode composite in lithium ion battery industry. *See* discussion *supra* Part IV.A.5.(i).(1d).

153. First, oxidation levels that fall within the claimed range in [1d] ("1% to 15%") were well-known to be used extensively in various technical fields, including battery. *See* LGES-1024 (Sue), [0006], [0056], [0101] (stating "**[o]xidation of the nanotubes is at least about 1%, and more preferable at least at about 4%,"** and "[n]anotubes are a novel class of nanostructures that exhibit remarkable mechanical, electrical, and thermal properties, thus having potential applications such as nanoscale probe devices, **energy storage components**, sensors, flame retardant materials, and electrical conductors in the aerospace, automotive, micro-electric, photovoltaic, and energy transmission industries"); *see also* discussion *supra* Part IV. A.5.(i).(1d).

154. Second, oxidation levels that fall within the claimed range in [1d] ("1% to 15%") were well-known to enhance an electrochemical activity of the carbon nanotube / lithium iron phosphate composite materials, for use toward positive electrodes of lithium ion batteries. For instance, it was well-known that "[t]he improvement caused by surface oxidation of CNTs" with **oxidation level that fall within the claimed range in element [1d] ("1% to 15%") was known**

to be "dramatic." *See* discussion *supra* Part IV. A.5.(i).(1d) (explaining that **Kavan shows oxidation level of 9.1%);** *see also* LGES-1005, 8 ("the functionalization of nanotubes [was known to be], indeed, crucial for enhancing the electrochemical activity of the composite materials with $LiFePO_4$").

155. Therefore, the oxidized nanotubes with oxidation levels that fall within the claimed range in element [1d] ("1% to 15%") were well-known and extensively used various applications, including lithium ion battery, and more particularly, toward forming a carbon nanotube/lithium iron phosphate composite for use toward lithium ion battery positive electrode materials.

(ii) Claim 4

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

156. In the combination, Cheng describes that the "composite cathode material obtained in the present invention is the particulate homogeneous composite of the carbon nanotubes and the lithium iron phosphate cathode material," where the "carbon nanotubes account for 0.5-15% by weight, and the lithium iron phosphate accounts for 85-99.5% by weight." LGES-1007 (Cheng), Abstract, [0023], claim 1. In particular, Cheng describes that the preferable range is where the "carbon nanotubes accounts for 0.5-3% by weight, and the lithium iron phosphate accounts for 92-97% by weight, the coating carbon accounting for 1-5% by weight." *Id.*, [0023].

157. Cheng's preferred weight percentage range (0.5-3 % of carbon nanotube and 92-97 % of lithium iron phosphate) corresponds to the carbon nanotubes being present in a weight percentage ("wt%") of 0.52 % (0.5 wt % nanotubes / 97 wt % lithium iron phosphate X 100%) to 3.26% (3 wt % nanotubes / 92 wt % lithium iron phosphate X 100%) by weight of the ion active material.

158. Alternatively/additionally, to the extent the weight percentage is based on the total weight of both nanotubes and ion active material,[8] Cheng's weight percentage range corresponds to the carbon nanotubes being present in a weight percentage of 0.51% (0.5 wt % nanotubes / (0.5 wt % nanotubes + 97 wt % lithium iron phosphate) X 100%) to 3.16% (3 wt % nanotubes / (3 wt% nanotubes + 92 wt % lithium iron phosphate) X 100%) by weight. LGES-1007 (Cheng), Abstract, [0023], claim 1.

159. The range of weight percentages disclosed by Cheng (***0.52-3.26%***) falls within the claimed **0.1% to 10% by weight** range recited in claim 4. Therefore, this specific range of weight percentages disclosed by Cheng is a

[8] The '924 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, 3:64-67.

relevant disclosure for the recited weight percentage range, and the Cheng-Gan combination thus renders obvious the claimed range of weight percentages.

(iii) Claim 5

[5] The composition of claim 1, comprising uniformly dispersible oxidized carbon nanotubes having ion active materials attached to their surface.

160. As discussed above with respect to discussion of [1a], [1b], and [1d], the Cheng-Gan combination uses oxidized carbon nanotubes (that are either obtained commercially or by oxidizing the nanotubes using well-known technique), in which the oxidized carbon nanotube disperses and is dispersible throughout both the [1] "carbon nanotubes" themselves and [2] "the precursor" that includes lithium iron phosphate. *See* discussion *supra* Part IV. B.4. & B.5.(i).[1a], [1b], [1d]. Again, as discussed above, such oxidized carbon nanotubes dispersion results in non-aggregated and uniform dispersion of CNTs. *See id.* As a result of, or evidenced by, "uniform dispersion," "uniformly distributed network structure," or non-aggregated state, the oxidized carbon nanotubes are uniformly dispersible tubes. *See id*. Moreover, the oxidized nanotubes are **dispersible** because the combination composition is capable of being mixed or blended.

C. [GROUND 2B] – CLAIMS 2 AND 3 ARE OBVIOUS OVER CHENG-GAN-ZHAO

(i) Claim 2

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

161. Zhao focuses on "provid[ing] a conductive agent for a lithium-iron-phosphate battery, the conductive agent having high electron conductivity and uniform heat conductivity." LGES-1018 (Zhao), [0004]. "In order to achieve the above purpose," Zhao describes addition of graphene to carbon nanotube/carbon black (e.g., acetylene black) composite material for positive electrodes of lithium ion batteries. *Id.*, Abstract, [0006], [0009], [0011]. For instance, here, Zhao refers to the "conductive agent" (hereinafter "composition") as "carbon nanotube/carbon black composite material, graphene, and a binder." *See id.,* Abstract. Zhao describes that such use of the composition "effectively utilizes respective advantages of fibrous carbon nanotubes, spherical carbon black, and sheet-like graphene, and cause the same to achieve a synergistic effect, thereby significantly improving electrochemical performance of the battery. In addition, the addition of the binder not only can reduce the falling speed of the conductive agent and improve the storage stability of the prepared conductive agent, but also can improve the compatibility between carbon nanotubes and the lithium iron phosphate as the active material in the slurry preparation process." *See id.*, [0012].

162. For instance, Zhao explains that adding graphene to a carbon nanotube/carbon black composite material enhances electron transport, reduces internal resistance, and improves heat dissipation. *Id.,* [0011] ("graphene has a greater electron transport rate and better heat dissipation performance than carbon nanotubes, so that further addition of graphene to the carbon nanotube/carbon black composite material can not only reduce the internal resistance of the carbon nanotube/carbon black composite material, but also improve heat dissipation performance of a unit cell"). Moreover, for instance, Zhao describes that combining graphene with a carbon nanotube/carbon black composite material enables multi-dimensional electron conduction between lithium iron phosphate and achieves synergistic effects due to their fibrous, spherical, and sheet-like characteristics. *Id.* ("[a]fter combination of the graphene and the carbon nanotube/carbon black composite material, **multi-dimensional conduction of electrons between lithium iron phosphate as the active material can also be achieved"** and **synergistic effects** "**can be achieved by means of fibrous, spherical, and sheet-like characteristics achieved by the combination of the carbon nanotube/carbon black composite material and the graphene**, thereby improving physical and chemistry properties of the active material, and playing an **important role in the consistency [or uniformity] of the battery**").

163. Given the above-noted advantages of graphene toward carbon nanotube / carbon black composite material, a POSITA would have been motivated to add graphene toward the carbon nanotube / acetylene black (specific form of carbon black) / lithium iron phosphate composite material in Cheng-Gan combination and would have expected success.

(ii) Claim 3

[3] The composition of claim 2, wherein the weight ratio of graphene to carbon nanotube is in the range of 0.1:99.9 to 99.9:0.1.

164. First, in Cheng-Gan-Zhao combination, Zhao discloses a weight ratio of graphene to carbon nanotube that falls within the broad range of claimed weight ratio of graphene to carbon nanotube. For instance, the claimed weight ratio of graphene to carbon nanotube in [3], which ranges from 0.1:99.9 to 99.9:0.1, indicates a broad range. This ratio indicates that the weight percentage of either graphene or carbon nanotube in sum of graphene and carbon nanotube corresponds to 0.1 wt% - 99.9 wt%.

165. Second, Zhao describes that the preferred weight percentage of graphene in sum of graphene and carbon nanotube is about 9 wt%., based on weight ratio of graphene to carbon nanotube (with carbon black) being 0.1 : 1. For instance, Zhao describes the following:

> A conductive agent for a lithium-iron-phosphate battery, consisting of a carbon nanotube/carbon black composite material, graphene, and a binder, the weight ratio of the carbon nanotube/carbon black

> composite material, the graphene, and the binder being: carbon nanotube/carbon black composite material : graphene : binder = 1 : (0.001 to 0.1) : (0.01 to 1). Preferably, the weight ratio of the carbon nanotube/carbon black composite material, the graphene, and the binder is: carbon nanotube/carbon black composite material : graphene : binder = 1 : 0.1 : 1.

LGES-1018 (Zhao), [0006], [0007].

This indicates that the weight percentage of graphene in the sum of graphene and carbon nanotube/carbon black composite corresponds to about 9 wt% (0.1 graphene / (0.1 graphene + 1 carbon nanotube/carbon black composite) x 100% = 9 wt %).

166. Third, 9 wt% of graphene in the sum of graphene and carbon nanotube / carbon black composite should fall within the 0.1 wt% - 99.9 wt% of graphene in the sum of graphene and carbon nanotube, even excluding the carbon black. For instance, even if the carbon black is not considered when determining the weight percentage of graphene in the sum of graphene and carbon nanotube, it would have been obvious that the teaching of using a specific weight percentage of graphene would still be relevant and applicable within the broad claimed range.

167. For instance, a POSITA would have recognized that the disclosed range for the weight ratio of graphene to carbon nanotube / carbon black composite in Zhao inherently encompasses various composite formulations, including those without carbon black. First, a POSITA would have known that graphene and/or carbon nanotubes can replace or partially replace carbon black without reducing

the electrode's conductivity. Considering the similarity between graphene and carbon nanotubes as alternatives to conventional carbon fillers (e.g., carbon black) used in lithium-ion batteries, both graphene and carbon nanotubes are highly conductive, low-dimensional materials with high aspect ratios. In contrast, carbon black consists of round particles with an aspect ratio of approximately 1 and medium conductivity. Thus, graphene and/or carbon nanotubes can replace or partially replace conventional carbon fillers (e.g., carbon black) without reducing the electrode's conductivity. Second, the ratio of low-dimensional materials in composites is determined by their intrinsic conductivity and aspect ratio (which affects capability to form a conductive network). Therefore, graphene and carbon nanotube can be interchangeable in this regard due to their similar aspect ratio and conductivity range. As such, based on [1] the replaceability of carbon black with graphene or nanotubes, and [2] the interchangeability of carbon black and graphene, a POSITA would have thus considered a wide range of ratios (e.g., weight ratio of graphene to carbon nanotube is in the range of 0.1:99.9 to 99.9:0.1) to be acceptable.

168. Therefore, a weight ratio of graphene to carbon nanotube in the range of 0.1:99.9 to 99.9:0.1 is rendered obvious based on the Cheng-Gan-Zhao combination.

VII. LEGAL PRINCIPLES

A. Anticipation

169. 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.

170. 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

171. 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.

172. I have been informed that a claim can be obvious in light of a single prior art reference or multiple prior art references. To be obvious in light of a single prior art reference or multiple prior art references, there must be a reason to modify the single prior art reference, or combine two or more references, in order to achieve the claimed invention. This reason may come from a teaching, suggestion, or motivation to combine, or may come from the reference or references themselves, the knowledge or "common sense" of one skilled in the art, or from the nature of the problem to be solved, and may be explicit or implicit from the prior art as a whole. I have been informed that the combination of familiar elements according to known methods is likely to be obvious when it does no more than yield predictable results. I also understand it is improper to rely on hindsight in making the obviousness determination.

VIII. CONCLUSION

173. 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.

174. 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

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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)
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[47] Sarang M. Bhaway, Zhe Qiang, Yanfeng Xia, Xuhui Xia, Byeongdu Lee, Kevin G. Yager, Lihua Zhang, Kim Kisslinger, Yu-Ming Chen, Kewei Liu, Yu Zhu, and Bryan D. Vogt* "Operando Grazing Incidence Small Angle X-Ray Scattering/X-Ray Diffraction of Model Ordered Mesoporous Lithium Ion Batteries Anodes" *ACS Nano, 2017*, 11, 1443-1454(impact factor: 15.881, citations=30)

[46] Zhe Qiang, Yu-Ming Chen, Burcu Gurkan, Yuanhao Guo, Miko Cakmak, Kevin A. Cavicchi, Yu Zhu, Bryan D. Vogt* "Cooperatively Assembled Nitrogen-doped Ordered Mesoporous Carbon/Iron Oxide Nanocomposites For Low-cost, Long Cycle Life Sodium-ion Batteries" *Carbon, 2017*, 116, 286-293 (impact factor: 9.594, citations=36)

[45] Zhe Qiang‖, Yu-Ming Chen‖, Yanfeng Xia, Wenfeng Liang, Kevin A. Cavicchi, Yu Zhu* and Bryan D. Vogt* "Ultra-Long Cycle Life Room Temperature Sodium-Sulfur Batteries Enabled by Highly Doped (N, S) Nanoporous Carbons for Stationary Energy Storage" *Nano Energy 2017*, 32, 59-66. (**Yu Zhu and Bryan Vogt as co-corresponding author and Students from Zhu's group and Vogt's group as first authors with equal contribution**) (impact factor: 17.881, citations=138)

[44] Kewei Liu, Zitian Yu, Xiaowen Zhu, Shuo Zhang, Feng Zou, Yu Zhu* "A Universal Surface Enhanced Raman Spectroscopy (SERS)-Active Graphene Cathode for Lithium-Air Battery Discharge Product Investigation", *RSC Advances*, 2016, 6, 102272-102279. (impact factor: 3.361, citations=6)

[43] Feng Zou, Yu-Ming Chen, Kewei Liu, Zitian Yu, Wenfeng Liang, Min Gao, Sarang M. Bhaway, Yu Zhu* "Metal Organic Frameworks Derived Hierarchical Hollow NiO/Ni/Graphene Composites for Lithium and Sodium Storage" *ACS Nano*, 2016, 10, 377-386 (impact factor: 15.881, citations=417)

[42] Yu-Ming Chen, Wenfeng Liang, Si Li, Feng Zou, Sarang M. Bhaway, Zhe Qiang, Min Gao, Bryan D. Vogt, Yu Zhu* "A Nitrogen Doped Carbonized Metal–organic Framework for High Stability Room Temperature Sodium–sulfur Batteries" *Journal of Materials Chemistry A* 2016, 4, 12471-12478(impact factor: 12.732, citations=117)

[41] Ruixuan He, Mauricio Echeverri, Daniel Ward, Yu Zhu, and Thein Kyu* "Highly Conductive Solvent-free Polymer Electrolyte Membrane for Lithium-ion Batteries: Effect of Prepolymer Molecular Weight" *Journal of Membrane Science* 2016, 498, 208-217(impact factor: 8.742, citations=81)

[40] Jeongwoo Lee, Yu-Ming Chen, Yu Zhu, Bryan D. Vogt* "Tuning SEI Formation on Nanoporous Carbon-titania Composite Sodium Ion Batteries Anodes and Performance with Subtle Processing Changes" *RSC Advances*, 2015, 5, 99329-99338 (impact factor: 3.361, citations=20)

[39] Tianyu Meng, Chang Liu, Kai Wang, Tianda He, Yu Zhu, Abdullah Al-Enizi, Ahmed Elzatahry, Xiong Gong* "High Performance Perovskite Hybrid Solar Cells with E-beam-Processed TiOx Electron Extraction Layer" *ACS Applied Materials & Interfaces* 2016, 8, 1876-1883 (impact factor: 9.229, citations=32)

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

[37] Abdul-Rahman O. Raji, Tanvi Varadhachary, Kewang Nan Tuo Wang, Jian Lin, Yongsung Ji, Bostjan Genorio, Yu Zhu, Carter Kittrell, James M. Tour* "Composites of Graphene Nanoribbon Stacks and Epoxy for Joule Heating and Deicing of Surfaces" *ACS Applied Materials & Interfaces* 2016, 8 (5), 3551–3556 (impact factor: 9.229, citations=75)

[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)

[34] Yu-Ming Chen, Yu Zhu* "CVD of Carbon Nanotubes in Porous Nickel for Anodes in Lithium Ion Battery" *Current Opinion in Chemical Engineering* 2015, 7, 32-39 (impact factor: 5.164, citations=8)

[33] Kun Yang, Tianda He, Xiaoyi Chen, Stephen Z. D. Cheng, Yu Zhu* "Patternable Conjugated Polymers with Latent Hydrogen–bonding on the Main Chain" *Macromolecules* 2014, 47 (24), 8479–8486 (impact factor: 5.985, citations=45)

[32] Tianda He, Aozhen Xie, Darrell H. Reneker, Yu Zhu* "A Tough and High-Performance Transparent Electrode from a Scalable and Transfer-Free Method" *ACS Nano* 2014, 8 (5), 4782–4789 (impact factor: 15.881, citations=98)

[31] Jeongwoo Lee, Yu-Ming Chen, Yu Zhu, Bryan D. Vogt* "Fabrication of Porous Carbon/TiO2 Composites through Polymerization-Induced Phase Separation and Use as An Anode for Na-Ion Batteries" *ACS Applied Materials & Interfaces* 2014, 6 (23), 21011–21018. (impact factor: 9.229, citations=79)

[30] Pei Dong, Yu Zhu*, Jing Zhang, Feng Hao, Jingjie Wu, Sidong Lei, Hong Lin, Robert H Hauge, James M Tour*, Jun Lou* "Vertically Aligned Carbon Nanotubes/Graphene Hybrid Electrode as a TCO- and Pt- free Flexible Cathode for Solar Cell Applications"

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

[29] Pei Dong, Yu Zhu*, Jing Zhang, Cheng Peng, Zheng Yan, Lei Li , Zhiwei Peng , Gedeng Ruan , Wanyao Xiao , Hong Lin , James M. Tour*, Jun Lou* "Graphene on Metal Grids as the Transparent Conductive Material for Dye Sensitized Solar Cell" *Journal of Physical Chemistry* C 2014, 118 (45), 25863–25868. (Yu Zhu as corresponding author at U. Akron, Yu Zhu's student Wanyao Xiao as co-author) (impact factor: 4.309, citations=40)

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[27] Vladimir Volman‖, Yu Zhu‖, Abdul-Rahman O Raji, Bostjan Genorio , Wei Lu , Changsheng Xiang , Carter Kittrell , James M. Tour* "Radiofrequency Transparent, Electrically Conductive Graphene Nanoribbon Thin Films as De-icing Heating Layers" *ACS Applied Materials & Interfaces* 2014, 6 (1), 298–304 (‖with equal contribution) (impact factor: 9.229, citations=43)

[26] Chenguang Zhang, Zhiwei Peng, Jian Lin, Yu Zhu, Gedeng Ruan, Chih-Chau Hwang, Wei Lu, Robert H. Hauge, and James M. Tour* "Splitting of a Vertical Multiwalled Carbon Nanotube Carpet to a Graphene Nanoribbon Carpet and Its Use in Supercapacitors" *ACS Nano* 2013, 7(6), 5151–5159 (impact factor: 15.881, citations=77)

[25] Jian Lin, Chenguang Zhang, Zheng Yan, Yu Zhu, Zhiwei Peng, Robert H Hauge, Douglas Natelson, James M Tour* "3-Dimensional Graphene Carbon Nanotube Carpet-Based Microsupercapacitors with High Electrochemical Performance" *Nano Letters* 2013, 13, 72-78(impact factor: 11.189, citations=652)

[24] Wei Lu, Gedeng Ruan, Bostjan Genorio, Yu Zhu, Barbara Novosel, Zhiwei Peng, James M Tour* "Functionalized Graphene Nanoribbons via Anionic Polymerization Initiated by Alkali Metal-Intercalated Carbon Nanotubes" *ACS Nano* 2013, 7, 2669-2675 (impact factor: 15.881, citations=35)

[23] Yu Zhu, Lei Li, Chenguang Zhang, Gilberto Casillas, Zhengzong Sun, Zheng Yan, Gedeng Ruan, Zhiwei Peng, Abdul-Rahman O Raji, Carter Kittrell, Robert H Hauge, James M Tour* "A Seamless Three-Dimensional Carbon Nanotube Graphene Hybrid Material" *Nature Communications* 2012, 3, 1225 (impact factor: 14.919, citations=515)

[22] Yu Zhu, Dustin K James, James M Tour* "New Routes to Graphene, Graphene Oxide and Their Related Applications" *Advanced Materials* 2012, 24, 4924-4955 (impact factor: 30.849, citations=334)

[23] Yu Zhu, Xianyu Li, Qinjia Cai, Zhengzong Sun, Gilberto Casillas, Miguel Jose-Yacaman, Rafael Verduzco, James M Tour* "Quantitative Analysis of Structure and Bandgap Changes in Graphene Oxide Nanoribbons during Thermal Annealing" *Journal of the American Chemical Society* 2012, 134, 11774-11780 (impact factor: 15.419, citations=47)

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

[21] Zheng Yan‖, Lulu Ma‖, Yu Zhu‖, Indranil Lahiri, Myung Gwan Hahm, Zheng Liu, Shubin Yang, Changsheng Xiang, Wei Lu, Zhiwei Peng, Zhengzong Sun, Carter Kittrell, Jun Lou, Wonbong Choi, Pulickel M Ajayan, James M Tour* "Three-Dimensional Metal–Graphene–Nanotube Multifunctional Hybrid Materials" *ACS Nano* 2012, 7, 58-64 (‖with equal contribution) (impact factor: 15.881, citations=216)

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

PRESENTATIONS

Invited Seminars and invited presentation in conferences

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

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.