

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent of: Clive P. Bosnyak, et al.
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Title: DISPERSIONS COMPRISING DISCRETE CARBON
NANOTUBE FIBERS

SUPPLEMENTAL DECLARATION OF SAMUEL P. GIDO

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I, Samuel P. Gido, declare the following:

1. I am submitting this declaration (my “Second Declaration”) in support of the Petitioner’s Reply to Patent Owner’s Response (“POR”) to the Petition for *inter partes* review of the ’649 Patent. This Second Declaration supplements certain portions of my prior declaration in support of the Petition for this IPR Proceeding (LGES-1003, “my First Declaration”). In my First Declaration, I address many topics, including (but not limited to) my background and qualifications, the level of skill in art, an overview of the ’649 Patent, a POSITA’s background knowledge, certain legal standards explained to me by counsel, legal principles applied for the purposes of my analysis and this declaration, and a detailed analysis of the prior art against the claim elements of the ’649 Patent.
2. I maintain all of my earlier analysis and continue to hold the opinions expressed in my First Declaration. This Second Declaration does not replace my First Declaration. These declarations are complementary and should be read in tandem. Accordingly, this Second Declaration does not cover every topic addressed in my First Declaration.
3. In forming this Second Declaration, I have considered, among various documents, the following:

- MRD's Patent Owner Response ("POR") and documents cited therein
- MRD-2006 Declaration of Deborah D. L. Chung. Ph. D.
- LGES-1062 "Percolation and gel-like behavior of multiwalled carbon nanotube/polypropylene composites influenced by nanotube aspect ratio." Yongzheng Pan, Lin Li. *Polymer*, 54 (2013) 1218-1226. ("Pan")
- LGES-1063 "Correlations between Percolation Threshold, Dispersion State, and Aspect Ratio of Carbon Nanotubes." Jing Li, Peng-Cheng Ma, Wing Sze Chow, Chi Kai To, Ben Zhong Tang, J.-K. Kim. *Advanced Functional Materials* 2007, 17, 3207–3215. ("Li")
- LGES-1064 "The effect of the aspect ratio of carbon nanotubes on their effective reinforcement modulus in an epoxy matrix." Alfonso Martone, Gabriella Faiella, Vincenza Antonucci, Michele Giordano, Mauro Zarrelli. *Composites Science and Technology* 71 (2011) 1117–1123. ("Martone")
- LGES-1065 "Discrete carbon nanotubes increase lead acid battery charge acceptance and performance." Steven W. Swogger,

Paul Everill, D.P. Dubey, Nanjan Sugumaran. *Journal of Power Sources* 261 (2014) 55-63. (“Swogger”)

- LGES-1066 “Effects of the Dispersion State and Aspect Ratio of Carbon Nanotubes on Their Electrical Percolation Threshold in a Polymer” H.D. Bao et al., *J. of Applied Polymer Science* 128 (1), 735-740, 2013. (“Bao”)
- LGES-1067 “Effect of multi-walled carbon nanotube aspect ratio on mechanical and electrical properties of epoxy-based nanocomposites” M.R. Ayatollahi et al., *Polymer Testing* 30 (2011) 548–556. (“Ayatollahi”)

4. 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.

I. “Discrete carbon nanotubes”

5. PO’s construction of “discrete carbon nanotubes” should not be adopted because its narrowness is simply inconsistent with not only the ’649 Patent itself, but also with abundant other evidence indicative of the usage of the term in the art.

6. PO defines the term “discrete carbon nanotubes,” as “carbon nanotubes that are separate and distinct from other nanotubes; not attached to other nanotubes.” POR, 8. The ’649 Patent describes “discrete individual fibers” as “not entangled as a mass.” LGES-1001, 5:39-40. It says nothing prohibiting discrete nanotubes from being attached to one another, or from touching one another, and as such it provides no support for PO’s assertion that discrete nanotubes are “not attached to other nanotubes” POR, 8.
7. Even at low concentrations, it is not possible for carbon nanotubes, due to their high aspect ratio (length divided by diameter), to avoid crossing and contacting one another at least at a few points. This can be illustrated by a children’s game called Pick-Up Sticks, shown below. The randomly oriented sticks only occupy a few percent of the volume of the space they inhabit, yet due to their high aspect ratio, it is impossible for them to not cross one another at localized points. Similarly, a POSITA referring to carbon nanotubes would understand that the carbon nanotubes are dispersed and discrete, even if a limited number of isolated contacts are present. Also, a POSITA would recognize that the likelihood of isolated crossings and contacts between carbon nanotubes increases as the aspect ratio increases.



8. To ensure that carbon nanotubes are completely isolated and never touch one another, the concentration of nanotubes in the dispersion would need to be so low that the dispersion or composite would not achieve the properties for which carbon nanotubes are added. As an example, transmission electron microscopy (TEM) images of carbon nanotubes with aspect ratios of 51, 84, and 167 from Pan are shown below. LGES-1062, 1221. It is clear from these images that even at the lowest aspect ratio, 51, carbon nanotubes are crossing one another. As the aspect ratio increases, the number of contacts necessarily increases and by the time an aspect ratio of 167 is reached, virtually all of the nanotubes are crossing other nanotubes at multiple contact points. This is an unavoidable consequence of the great length of the nanotubes. If one were to move the nanotubes in Fig. 2C far enough apart that they never touched one another, then the concentration of nanotubes would need to be about 0.005%. At that percentage, which corresponds to one part in 20,000, the concentration of nanotubes would be so low that they

would not produce any appreciable influence on properties of interest, such as mechanical or electrical properties.

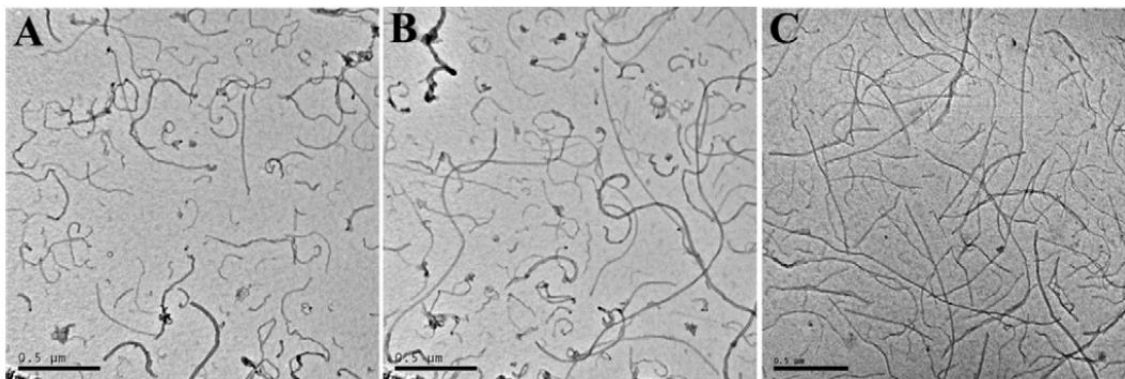
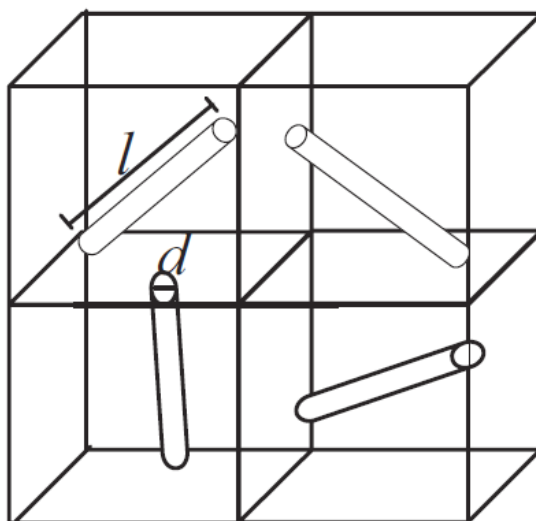


Fig. 2. TEM images of (A) MWCNT51, (B) MWCNT84 and (C) MWCNT167.

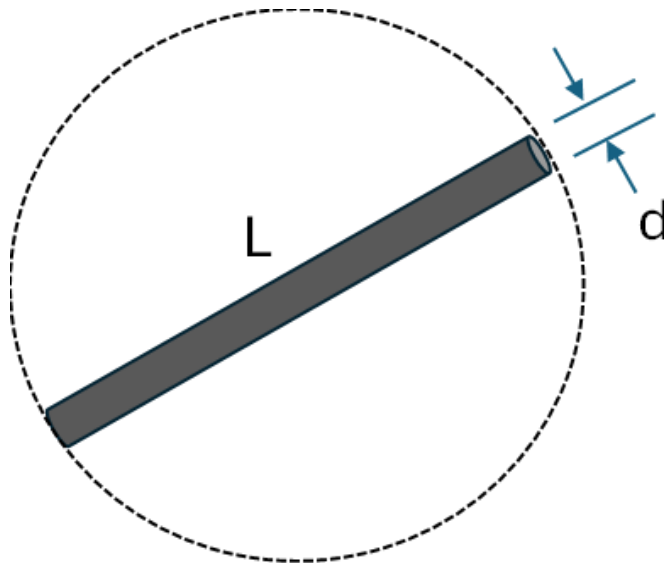
Excerpt from Pan, Fig. 2

9. As shown in the figure below, for carbon nanotubes to be completely isolated, each nanotube must reside in its own volume of space from which other nanotubes are excluded. LGES-1063, 3212.



Excerpt from Li, Fig. 7a

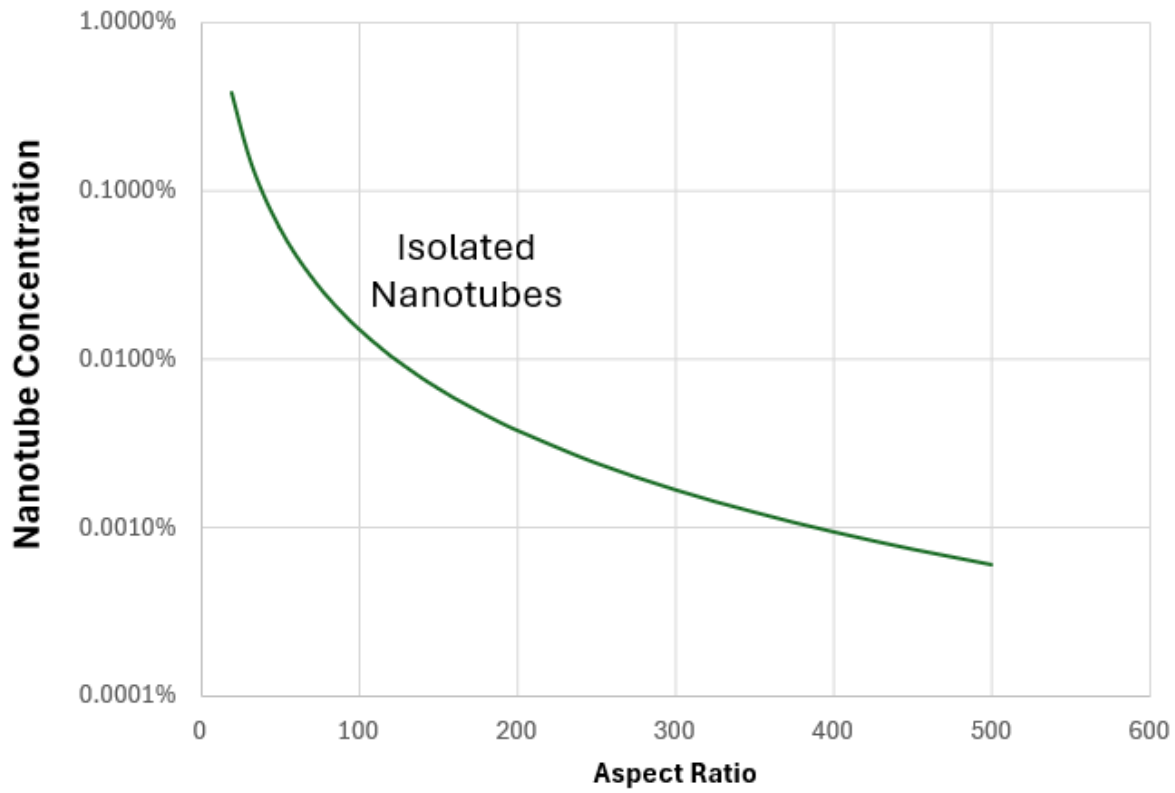
10. A POSITA can calculate the critical concentration of nanotubes, above which they begin to touch, by taking the ratio of the volume of a nanotube to the volume of the space around it (boxes in the illustration above, or a sphere around a nanotube as in the calculation illustrated below) from which other nanotubes must be excluded to prevent contacts.
11. The upper limit of nanotube concentration (expressed as a volume fraction of nanotubes) for which the nanotubes are completely isolated (*e.g.*, never touching each other, even at a single point) can be calculated by taking the ratio of the volume of a single nanotube (with length (L) and diameter (d)) to the volume of a sphere. The sphere has a diameter equal to L and represents the space from which all other nanotubes must be excluded to ensure they never touch.



12. The volume of the nanotube is $V_{NT} = \pi (d/2)^2 L$ while the volume of the sphere it occupies is $V_S = (4/3) \pi (L/2)^3$. The volume fraction (concentration) of nanotubes is given by the ratio of these volumes, as shown in the equation below. In this equation, AR is the aspect ratio (L/d).

$$\phi = \frac{\pi \left(d/2\right)^2 L}{(4/3)\pi(L/2)^3} = \frac{3}{2} \left(\frac{d}{L}\right)^2 = \frac{3}{2} \left(\frac{1}{AR}\right)^2$$

13. I have plotted an “Isolated Nanotubes” curve in the graph below. For each aspect ratio, a dispersion must have a concentration lower than the “Isolated Nanotubes” curve for the nanotubes to not touch one another. At even modest aspect ratios, these concentrations are so low that the carbon nanotubes would produce negligible influence on the material properties. For instance, at an aspect ratio of 60, the nanotube concentration would need to be less than about 0.04% (one part in 2,500) for the nanotubes to not touch one another. At this extremely low nanotube concentration, a POSITA would not consider this material to be a dispersion or a composite. Instead, the POSITA would view this material as essentially a pure matrix material (such as a polymer) with a small amount of an impurity, which in this case is carbon nanotubes.



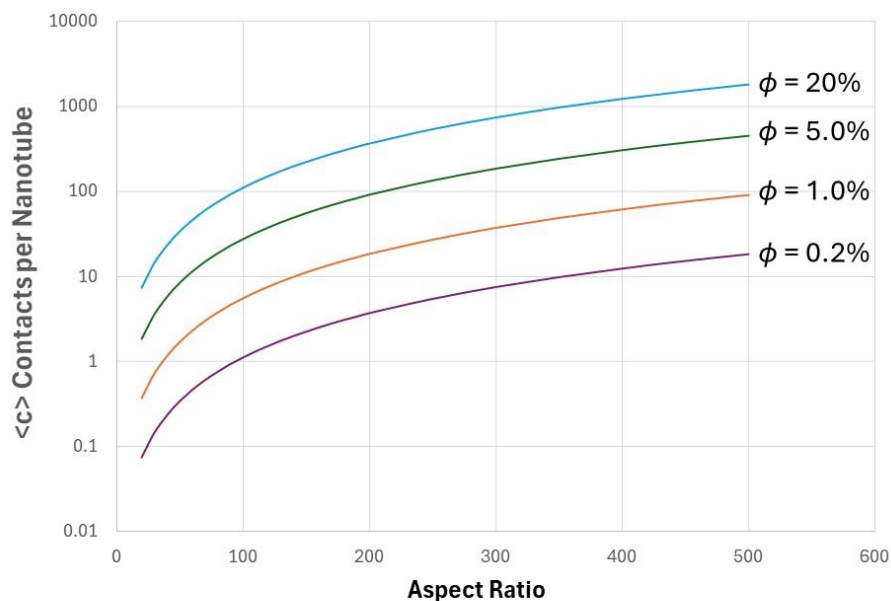
14. The concentration of nanotubes in the '649 Patent's compositions (*e.g.*, 0-30% by weight *see* LGES-1001, claim 1) is high enough that a POSITA would expect contact between the nanotubes in these compositions. Yet, the '649 Patent refers to its nanotubes as "discrete." This indicates that PO's construction of "discrete," that prohibits physical contact between nanotubes, is misaligned with the morphology that a POSITA would understand to be present in the '649 Patent's compositions.
15. To further demonstrate that the PO's definition of "discrete" should not be adopted, one can consider a mathematical relationship between the average

number of contacts each nanotube has with other nanotubes, $\langle c \rangle$, the aspect ratio (AR), and the concentration of nanotubes ϕ . LGES-1064, 1120.

$$\langle c \rangle = w \phi \left(4 + \frac{3AR^2}{3AR + 2} \right)$$

Excerpt from Martone, Equation 7

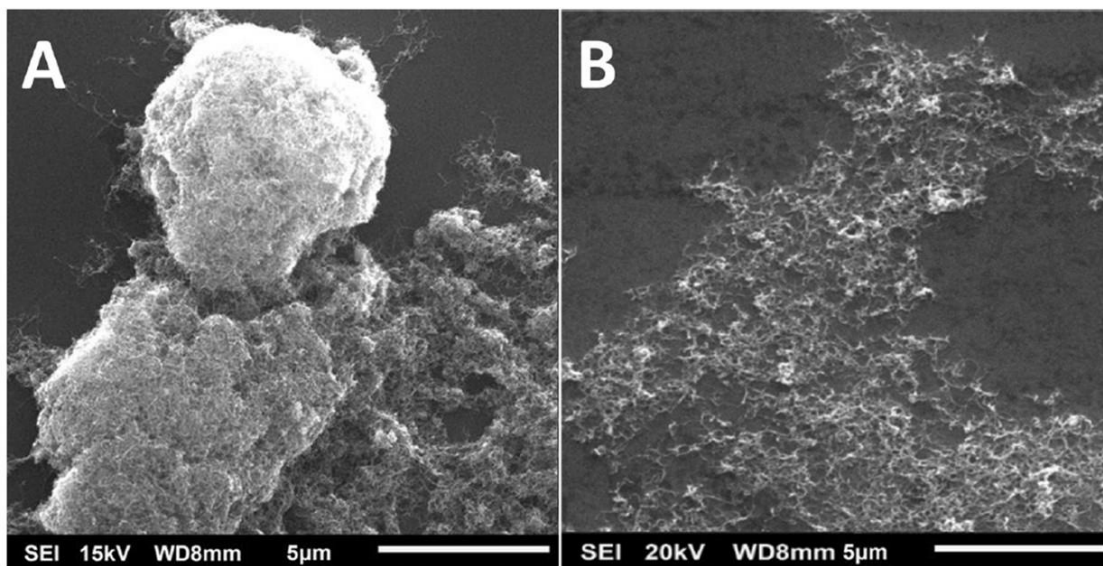
Here, Martone includes w , a “waviness” parameter, that considers the fact that carbon nanotubes are not straight throughout the entirety of their length. Martone provides values of this waviness parameter for carbon nanotubes across a range of aspect ratios. With this information and the equation above, I have plotted the average number of contacts between nanotubes per nanotube, $\langle c \rangle$, as a function of aspect ratio for nanotube concentrations ranging from 0.2% to 20%.



16. This graph illustrates how difficult it would be to prevent carbon nanotubes of even modest aspect ratios from contacting other nanotubes at localized points. For example, with a 1% concentration of nanotubes and an aspect ratio of about 35, there will be, on average, one contact per nanotube. At the same 1% concentration of nanotubes, when the aspect ratio reaches 500 (upper end of the range claimed in the '649 Patent), there are, on average, about 92 contacts per nanotube. The '649 patent claims nanotube concentrations ranging up to 30% (claim 1), 15% to 25% (claim 6), and 18% to 22% (claim 7). To provide perspective of the impact of the claimed nanotube concentration ranges on the number of contacts between nanotubes, I have plotted the average number of contacts per nanotube for a 20% concentration in the graph above. At a 20% concentration of nanotubes and an aspect ratio of 25 (lowest aspect ratio value claimed in the '649 Patent), there are, on average, about 10 contact points with other nanotubes for each nanotube. When the aspect ratio reaches the upper end of the claimed aspect ratio range, 500, there are well over 1000 contacts per nanotube. These observations demonstrate that it is impossible to have the combinations of aspect ratios and nanotube loadings (concentrations) claimed in the '649 patent without having numerous points of contact

between the nanotubes. PO's construction of discrete nanotubes as completely isolated nanotubes is unjustified.

17. PO's construction for discrete is also at odds with the usage of the term "discrete" in the art. For instance, Fig.1 of Swogger, reproduced below, compares scanning electron microscope (SEM) images of bundled carbon nanotubes shown in Fig. 1A, with discrete, untangled carbon nanotubes with aspect ratios approaching 70 in Fig. 1B. LGES-1065, 57. Swogger's description of the nanotubes in Fig. 1B as "discrete" is a clear indication that the common usage of this term encompasses non-bundled nanotubes that nevertheless can still cross and contact one another.

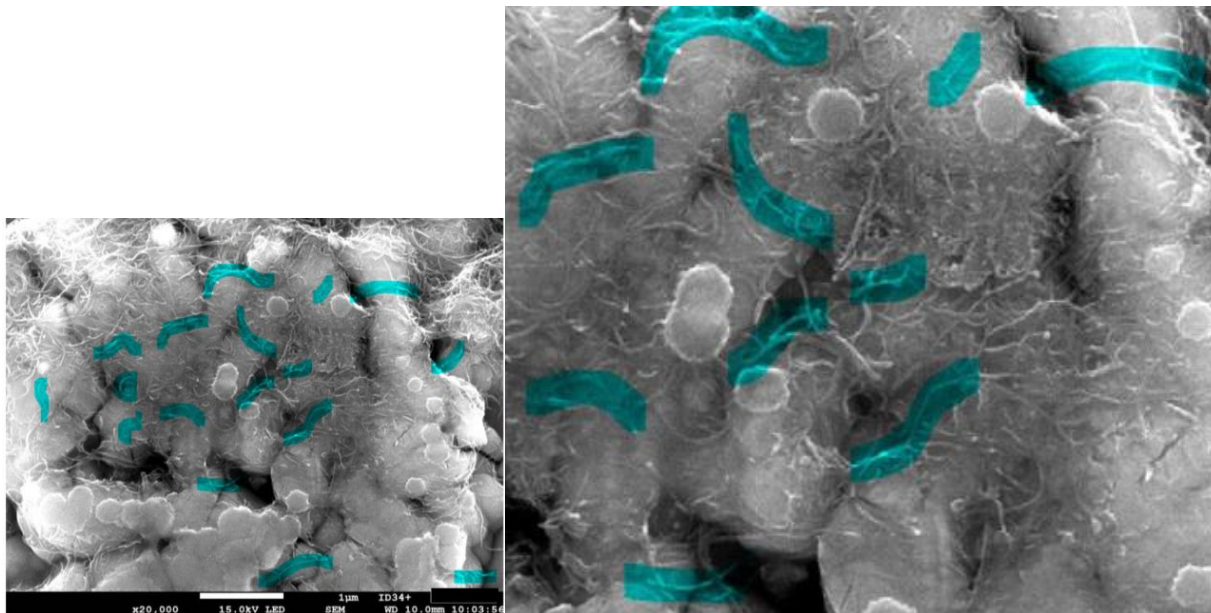


Excerpt from Swogger, Fig. 1

18. I also understand that PO asserted infringement of its "discrete" carbon nanotubes in a related proceeding in district court. In particular, I have

reviewed the SEM images below that PO provided as alleged evidence of such assertion, and I understand that PO provided the blue highlighting to indicate what it regards as discrete nanotubes in the accused products.

However, it can be clearly seen that most if not all of the nanotubes that PO highlighted are touching another nanotube at some point along its length, confirming my explanations above concerning how difficult it would be to prevent carbon nanotubes of even modest aspect ratios from contacting other nanotubes at localized points.



LGES-1050, 33 (original)

Id. (cropped, magnified)

19. To be sure, a fully isolated carbon nanotube that is not touching any other nanotube is likely “discrete.” However, this is a description, not a definition. Just as a carbon nanotube is undeniably a nanotube without all

nanotubes needing to be carbon nanotubes, a fully isolated nanotube can be discrete without all discrete nanotubes needing to be fully isolated.

II. Sun – dispersions of MWNTs

20. Regarding Sun's alleged failure to achieve discrete MWNTs, PO states that "a POSITA would not be surprised that a procedure leveraging specific surface area, such as one counting on the mechanical forces of nanoplatelets attached to the outside surface of CNTs for disentangling the CNTs, would not be as successful with CNTs having low specific surface area (MWCNTs) as it might be for CNTs having high specific surface area (SWCNTs)." (POR, 27). But this is simply backwards: a POSITA would recognize that high specific surface area materials are in fact often more difficult to disperse than lower surface area materials, due at least in part to stronger interactions (*e.g.*, increased van der Waals forces between high specific surface area CNTs) which promote aggregation or bundling of the CNTs.
21. But even if Sun were understood not to have actually produced dispersions of discrete MWNTs (which it does), that lack of experimental results does not render Sun a non-enabling disclosure.
22. Beyond its experimental results, Sun provides both a general process description (*see* LGES-1004, [0042]-[0052]) as well as a detailed discussion of the mechanism by which the MWNTs are rendered discrete in its

dispersions (*e.g.*, by interaction with nanoplatelets; *see id.*, [0044]). Sun's disclosure provides sufficient detail to inform a POSITA regarding MWNT dispersions.

23. Regarding PO's demand that Sun disclose "what alterations would need to be made to achieve the same SWCNT results with the fundamentally different MWCNTs" (POR, 35), Sun provides a detailed experimental protocol for creation of dispersions of MWNTs. *See* LGES-1004, [0054]-[0058]. To the extent that slight adjustments may be necessary to improve nanotube dispersion, this would have been well within the skill of a POSITA. For instance, with Sun's disclosed methods, a POSITA would recognize that experimental parameters, such as surface oxidation levels, sonication treatment times, length of the reaction, or reaction temperature, can be adjusted to improve nanotube dispersion. PO's assertion that Sun's disclosure does not "give a POSITA a place to even begin" is simply wrong. POR, 36.

III. Aspect ratios

24. PO argues that since Sun's aspect ratio ranges extend beyond the claimed ranges of the '649 Patent, it cannot be relied upon to disclose the claimed aspect ratio ranges. POR, 28. PO focuses on how Sun's carbon nanotube aspect ratio range may be as large 10,000 to argue that Sun discloses "at

least 9,500 more potential aspect ratios compared to the claimed aspect ratio (i.e., $10,000 - 500 = 9,500$).” POR, 30. PO asserts that “a POSITA would have to undertake undue experimentation,” (POR, 39) “[h]aving to test over potentially thousands of different aspect ratios where structure and behavior of the MWCNTs vary widely.” *Id.*

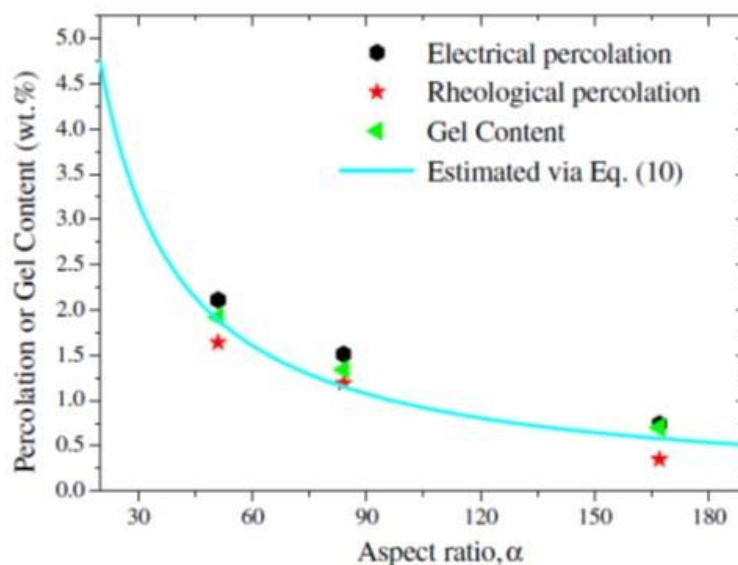
25. While focusing on the largest aspect ratio value in Sun’s disclosure, PO fails to establish that there is any meaningful difference between nanotubes having an aspect ratio in the claimed 25-500 range and Sun’s nanotubes, which have an aspect ratio of “at least about 10” or “at least about 20.” LGES-1004, [0038]. PO has offered generic statements alleging “differences relating to the structure and/or behavior of the oxidized CNTs of Sun versus the oxidized CNTs in the claimed invention” (POR, 31), without providing any evidence that these effects (to the extent they exist at all) are specific to the particular claimed range. The ’649 Patent itself does not describe any unexpected results stemming from nanotubes having an aspect ratio of 25-500, as compared to those with aspect ratios of at least about 10 or at least about 20.
26. Moreover, PO’s assertions that “[h]aving to test over potentially thousands of different aspect ratios where structure and behavior of the MWCNTs vary

widely ... is undue experimentation” (POR, 39) is inconsistent with the reality of nanotube experimentation.

27. No POSITA, when investigating the effect of aspect ratios on material properties, would ever prepare samples and assess properties for every single integer value of the aspect ratio range. This is not practical and not necessary for multiple reasons.
28. First, there is always a distribution of aspect ratios in any given sample of nanotubes, and a POSITA generally works with an average value obtained by averaging over this distribution. This means that any sample of nanotubes, with a given average aspect ratio, inherently contains many nanotubes that do not have the stated average value of the aspect ratio. For instance, two samples of nanotubes, one with an average aspect ratio of 100 and another with an average aspect ratio of 101, will both include nanotubes with the same aspect ratios in roughly the same amounts.
29. Second, a single digit difference between two aspect ratios has negligible impact on the properties of the dispersion, composite, or composition. A comparison between two composite samples, one with a nanotube aspect ratio of 100 and another with an aspect ratio of 101, is unlikely to reveal differences in relevant properties, due to this small variation in aspect ratio.

30. Third, a POSITA would understand that in order to investigate the influence of a parameter on a property (*e.g.*, mechanical, electrical, percolation threshold), it is not necessary to test every single possible value of the parameter. Rather, a POSITA would recognize that they can test the property at several values of the parameter distributed across the range. A POSITA can thereby develop an understanding of how the property varies as a function of the parameter.
31. The technical literature contains numerous published studies investigating the influence of carbon nanotube aspect ratio on material properties. Researchers of these studies do not prepare and test samples at every single possible value of the aspect ratio. Rather, they test a few values of the aspect ratio and develop a useful correlation between aspect ratio and the property of interest, which can then be used to find the value of the property at other values of the aspect ratio for which no experiments were conducted.
32. For instance, Pan studied the influence of nanotube aspect ratio on the mechanical (rheological) and electrical properties of MWNT-polymer composites at three average aspect ratios: 51, 84, and 167. LGES-1062, 1226. Pan produced the following graph, where the percolation threshold is plotted for the three tested aspect ratio values. Pan then developed a mathematical formula (Equation 10) based on the experimental data, which a

POSITA can use to estimate percolation threshold values at aspect ratios that were not directly tested. The blue curve is a plot of mathematical formula, which can be used to determine values of the percolation threshold at aspect ratios for which no experiments were conducted. LGES-1062, 1225. For instance, from the graph below, a POSITA would understand that the percolation threshold at an aspect ratio of 120 is about 1.0%.



Excerpt from Pan, Fig. 11

33. Similarly, Bao studied the effect of carbon nanotube aspect ratio on the electrical percolation threshold, which is a determinant of electrical properties. Bao considered a limited number of experimental samples with different nanotube aspect ratios given in Table 1, reproduced below. LGES-1066, 739.

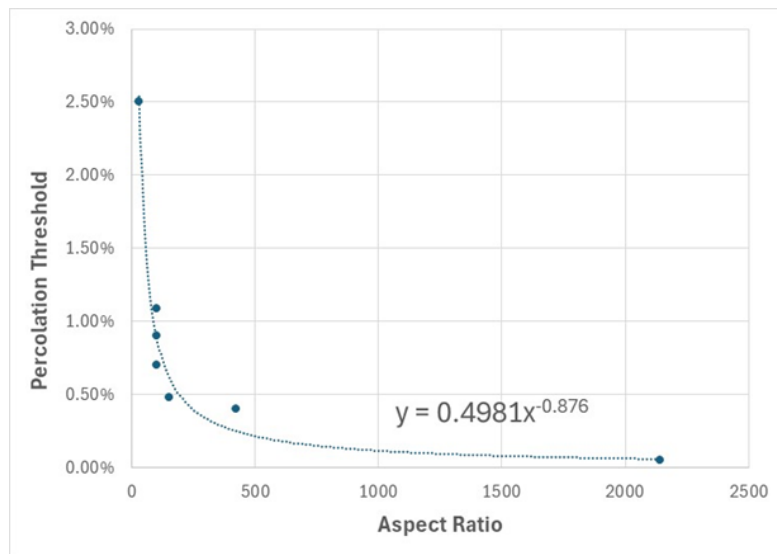
Table 1. The References, Matrix Types, and Processing Conditions for all the Samples

Matrix type	Aspect ratio	Processing method	Percolation threshold (vol %)	Dispersion state (P)	Reference	Symbol
Polyimide/CNT	2143 ^a	Solution compounding	0.05	0.29	16	F
POM/CNT	100 ^b	Melt mixing	0.70	0.85	3	C
PA6/CNT	100 ^b	Melt mixing	1.09	0.43	3	A
PP/CNT	100 ^b	Melt mixing	0.90	0.60	3	B
Epoxy resin/CNT	420 ^a	Solution compounding	0.40	0.03	4	E
Polycarbonate/CNT	150 ^a	Melt mixing	0.48	0.79	5	D
PP/CNT	30 ^a	Melt mixing	2.5	0.89	29	G

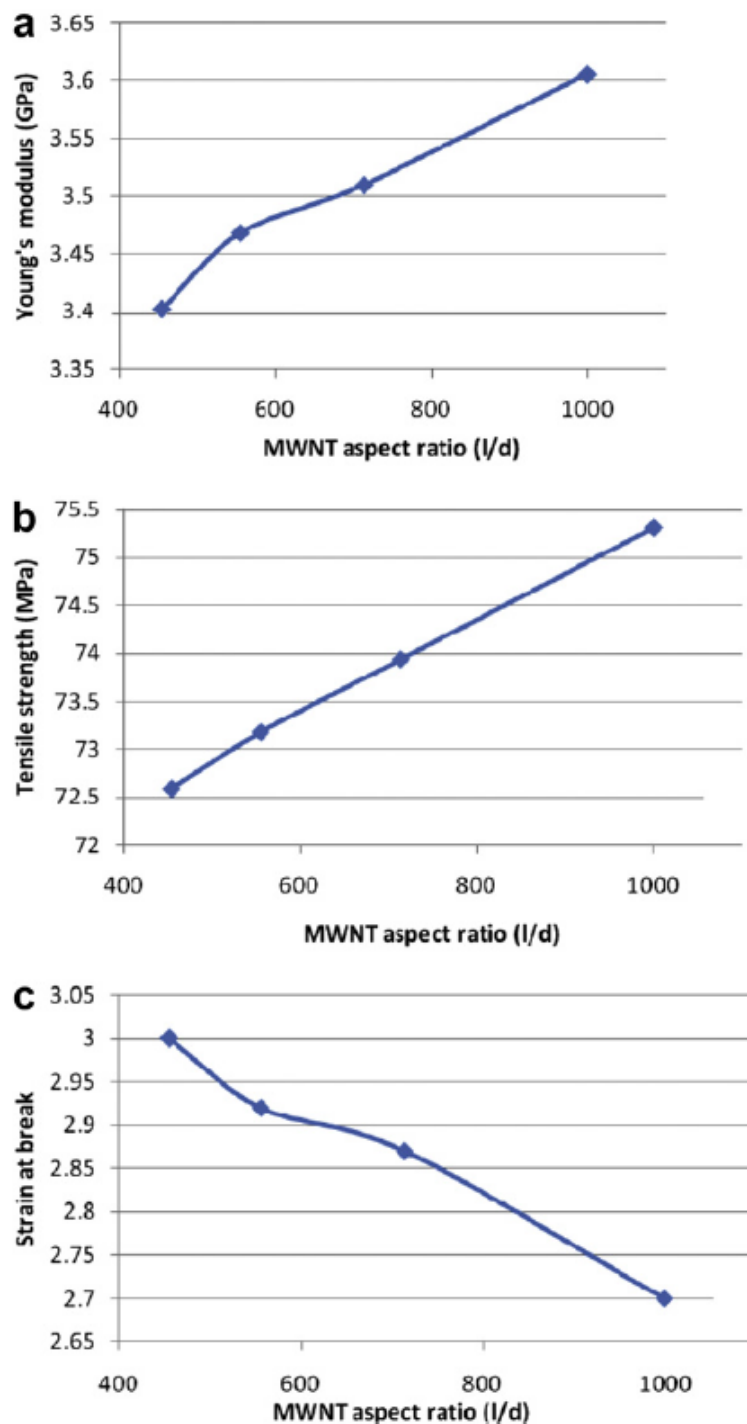
^aOriginal data., ^bMeasured data after compounding.

Excerpt from Bao, Table 1

34. Using the data from Table 1 of Bao, I plotted the percolation threshold versus aspect ratio and fitted a power-law curve to it (see below). From this analysis, a POSITA can easily tell that the percolation threshold is less than 0.25% across the whole range of aspect ratios between 500 and 2000. I know this without having to test any samples in that aspect ratio range. In fact, I can tell that the percolation threshold is less than 0.25% for any aspect ratio above 500, including all the way up to 10,000.



35. In another example, Ayatollahi studied the effect of aspect ratio of MWNTs on mechanical properties of nanotube-polymer composites. Ayatollahi prepared and tested only four samples with average aspect ratios ranging from 455 to 1000, and generated the following graphs for Young's modulus (stiffness), tensile strength, and strain at break as a function of aspect ratio. The diamond shaped symbols on these graphs represent experimental data from actual samples. LGES-1067, 551. The curves between these symbols can be reliably used by a POSITA to understand what the value of the property will be at other aspect ratios, say 900 for instance, at which no testing was done.



Excerpt from Ayatollahi, Fig. 4.

36. PO's argument that it is necessary to conduct tests at every single possible aspect ratio in order to understand their suitability for a particular property

or application, is flawed. POR, 39. No POSITA would conduct investigations in this manner. Instead, as demonstrated above, a POSITA would test a limited number of samples spanning the range of aspect ratios of interest and then draw curves and correlations from which conclusions may be reliably drawn for other values of the aspect ratio that were not directly tested.

IV. ADDITIONAL REMARKS

37. 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.
38. 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: 7/14/2025 By: 