

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent of: Clive P. Bosnyak, et al.
U.S. Patent No.: 10,608,282 Attorney Docket No. 18768-0210IP8
Issue Date: Mar. 31, 2020
Appl. Serial No.: 15/881,071
Filing Date: Jan. 26, 2018
Title: BINDERS, ELECTROLYTES AND SEPARATOR FILMS FOR
ENERGY STORAGE AND COLLECTION DEVICES USING
DISCRETE CARBON NANOTUBES

SUPPLEMENTAL DECLARATION OF SAMUEL P. GIDO

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 ’282 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 ’282 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 ’282 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-2004 Declaration of Deborah D. L. Chung. Ph. D.
- LGES-1036 "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-1037 "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-1038 "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-1039 "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”)

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 ’282 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, 13-14. The ’282 Patent describes “discrete” carbon nanotubes as “alternatively termed exfoliated carbon nanotubes” that can be “obtained from as-made bundled carbon nanotubes[.]” LGES-1001, 6:50-54. 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, 13-14. Rather, because the ’282 Patent is interested in,

among other things, achieving “low electrical resistivity” as one would when making a conductive binder as per the '282 Patent, a POSITA would readily understand that the “discrete” carbon nanotubes in the '282 Patent are touching one another to form a conductive electrical network.

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-1036, 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.

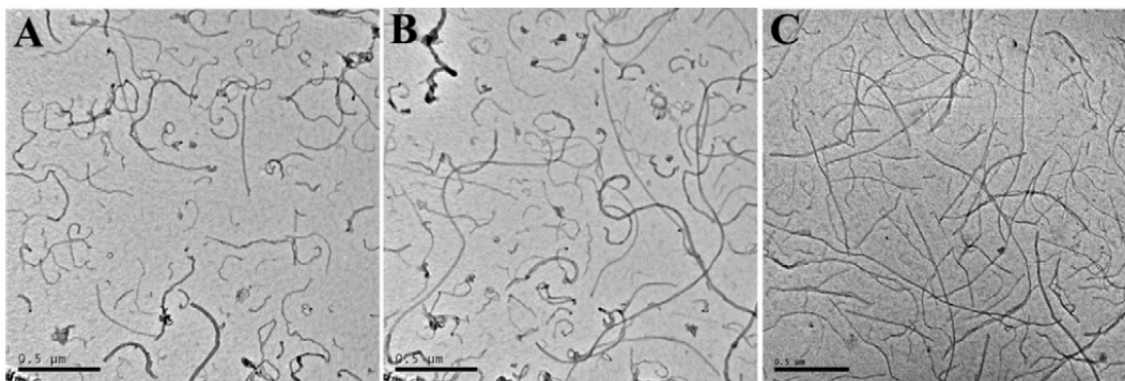
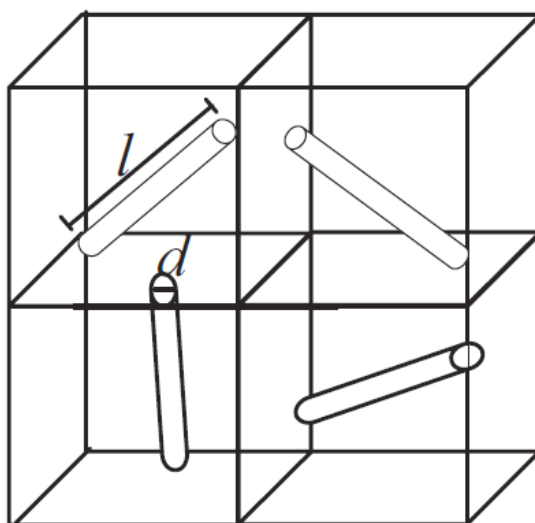


Fig. 2. TEM images of (A) MWCNT51, (B) MWCNT84 and (C) MWCNT167.

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-1037, 3212.

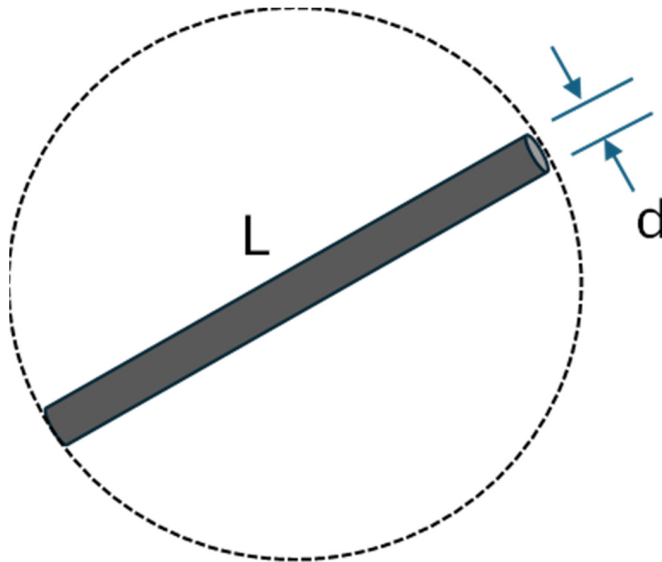


Excerpt from Li, Fig. 7a

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.

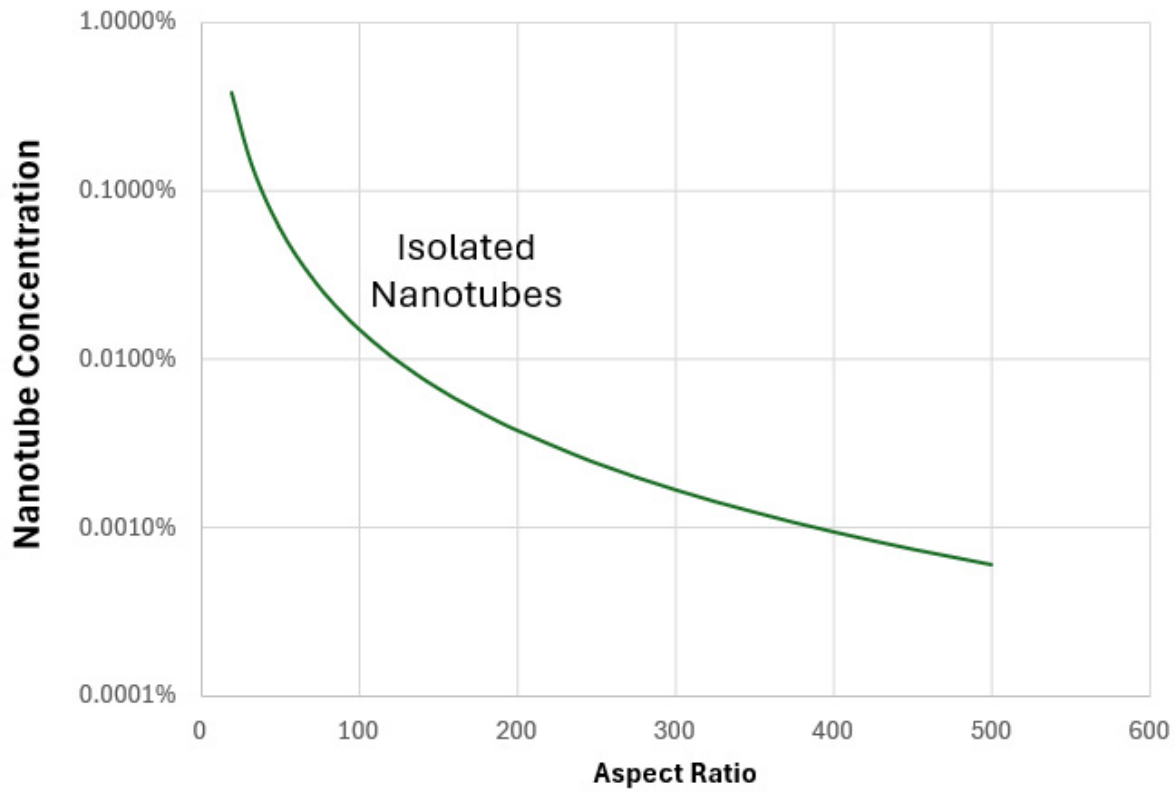


12. The volume of the nanotube is $V_{NT} = \pi (d/2)^2 L$ while the volume of the sphere it occupies is $V_S = (4/3) \pi (L/2)^3$. The volume fraction

(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.



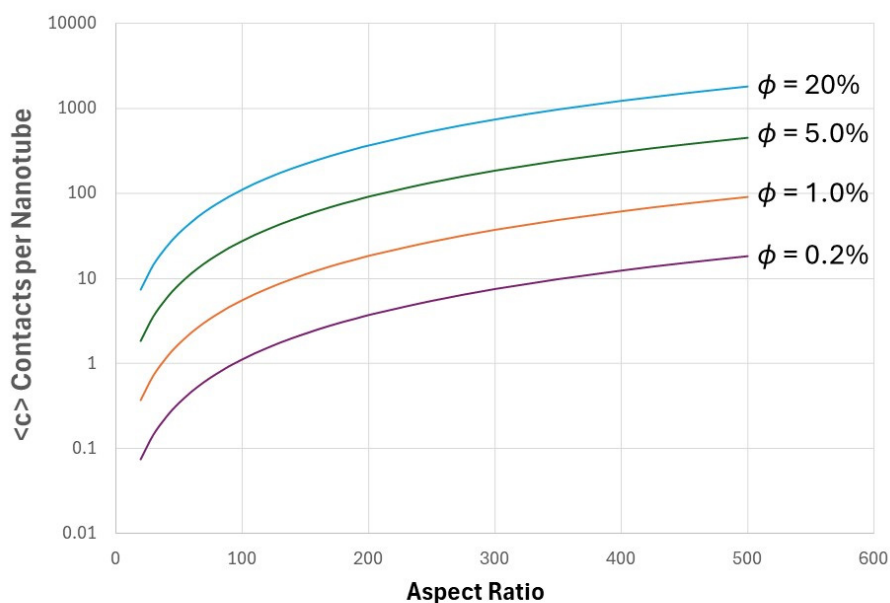
14. The concentration of nanotubes in the '282 Patent's compositions (*e.g.*, 10-50% by weight *see* LGES-1001, 9:21-24) is high enough that a POSITA would expect contact between the nanotubes in these compositions. Yet, the '282 Patent refers to its nanotubes as "discrete." This indicates that PO's construction of "discrete," that prohibits physical contact between nanotubes, is misaligned with the morphology that a POSITA would understand to be present in the '282 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-1038, 1120.

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

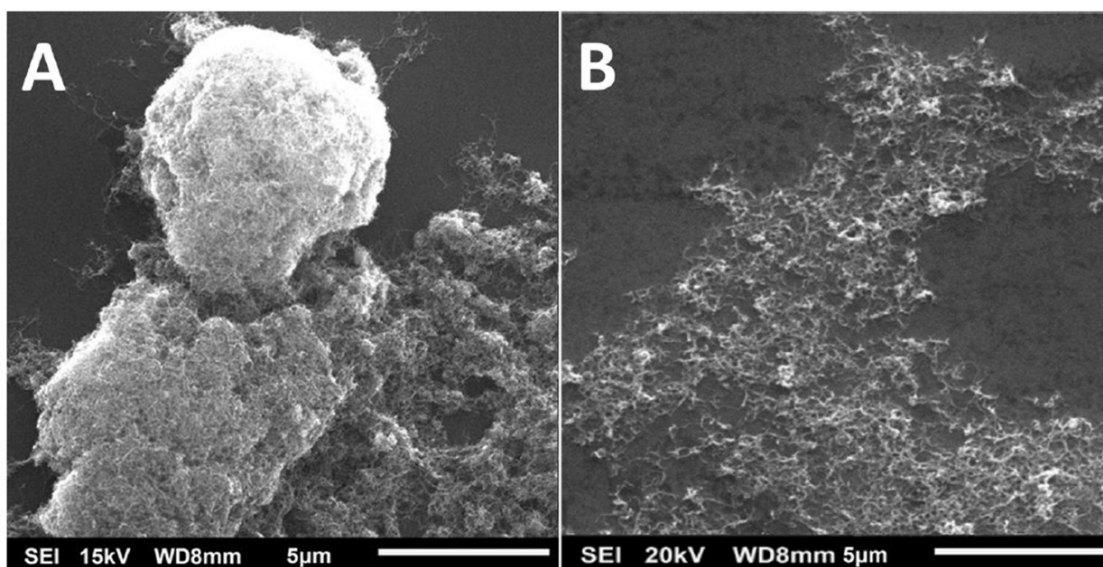
Excerpt from Martone, Equation 7

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 '282 Patent), there are, on average, about 92 contacts per nanotube. 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 (both well within contemplated ranges in the '282 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 '282 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-1039, 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.

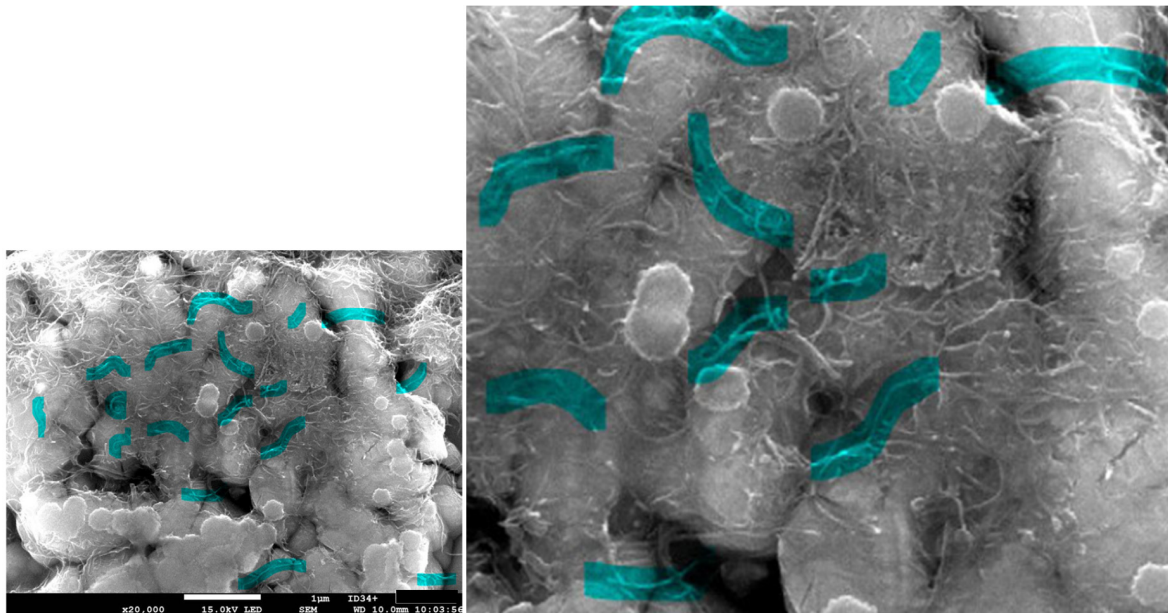


Excerpt from Swogger, Fig. 1

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 SEMs 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-1027, 40 (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. ADDITIONAL REMARKS

20. 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.
21. 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: 