IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent of:	Zhang	
U.S. Patent No.:	6,432,586	Attorney Docket No.: 39531-0002IP1
Issue Date:	August 13, 2002	
Appl. Serial No.:	09/546,266	
Filing Date:	April 10, 2000	
Title:	SEPARATOR FOR A HIGH ENERGY RECHARGEABLE LITHIUM BATTERY	

SUPPLEMENTAL DECLARATION OF

DR. KUZHIKALAIL M. ABRAHAM

1. This is a supplemental to the previous declaration (Exhibit 1003; "Original Declaration") that I provided in conjunction with these *Inter Partes* Review ("IPR") proceedings. Because Patent Owner has raised new issues regarding which I was not previously given an opportunity to offer testimony, I further provide the following opinion.

2. For purposes of this declaration, I have reviewed the following documents in connection with the IPR concerning the '586 patent (in addition to the documents listed in my Original Declaration and others incorporated into my declaration below):

   a. PTAB's Decision to Institute IPR (Paper 13);

   b. Celgard's Response to Petition for IPR [Redacted] (Paper 34) and references cited therein;

c. Declaration of Dr. Ralph White [Redacted] (Exhibit 2917) and references cited therein;

d. Declaration of Dr. C. Glen Wensley (Exhibit 2015) and references cited therein;

e. Deposition Transcript of Ralph E. White (Exhibit 1030); and

f. Deposition Transcript of Ralph E. White from IPR2014-00680 (SKI v. Celgard) (Exhibit 1033).

3. My findings, as explained below, are based on my education, experience, and background in the fields discussed in my Original Declaration.

I. Claim Construction

4. I understand that the terms of the claim should be interpreted according to their broadest reasonable construction in light of the specification, and that the words of the claims should be given their plain meaning unless that meaning is consistent with the specification. I further understand that the clam terms should be construed from the perspective of a person of ordinary skill in the art (POSITA) at the time of the filing of the ’586 patent.

5. Based on my review of the documents listed above, it is my understanding that Celgard has proposed for the first time during these IPR proceedings that the proper construction of “ceramic composite layer . . . adapted to at least block dendrite growth” should be “ceramic composite layer . . . capable

of preventing dendrites from growing all the way through the ceramic composite layer during the specified, stated or intended number of repetitive charge-discharge cycles of a rechargeable battery." Celgard Response at page 15. I also understand that Celgard's experts, in particular Dr. White, has not provided any testimony in support of this construction.

6. I have also reviewed the PTAB's construction for the same term and understand that the Board has interpreted such ceramic composite layer as one that is "capable of blocking dendrite growth with any degree of effectiveness." Institution Decision at page 7.

7. I disagree with Celgard's argument that this language in the '586 patent, when read by a POSITA, would be interpreted according to their proposed alternative construction. For example, the '586 patent mentions nothing about preventing dendrite growth "during the specified, stated or intended number of repetitive charge-discharge cycles of a rechargeable battery." The background section of the '586 patent merely acknowledges that dendrite growth can occur "after repetitive charge-discharge cycling," which a POSITA would understand as meaning that dendrites can form after one or more charge-discharge cycles, and in no way associates the occurrence of dendrite growth to a specific number of charge-discharge cycles. '586 patent at 1:20-23.

8. I further disagree with Celgard's argument that the claim language above only covers blocking any dendrite from "growing all the way through." Based on my understanding of how, in the IPR context, the terms of the claim should be interpreted according to their broadest reasonable construction in light of the specification, a POSITA would understand that any ceramic layer that is "adapted to at least block dendrite growth and to prevent electronic shorting" would be one that is designed and intended to mitigate the growth of dendrites—and the potential of shorting that can arise therefrom—with any degree of effectiveness.

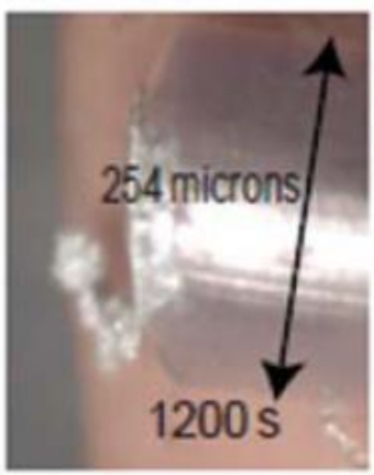
9. Accordingly, for at least the reasons above, I believe that a POSITA would find Celgard's proposed alternative to be not only unsupported by the '586 patent but also unduly narrow.

II. Dendrites

10. Regarding the mechanism of dendrite growth in a lithium battery, Celgard argues that "it is likely to follow the path of ionic flow toward the holes of Tojo." Celgard Response at page 19; *see also* White Declaration at ¶¶ 104 and 107. Dr. White further contends that dendrites can grow "in the form of needles" with "a diameter of less than 0.1 µm" so as to be able to grow through the 0.1 µm opening in Tojo. White Declaration at ¶ 102. However, while it *may* be possible for some portions of the dendrites to grow into holes as needle-like formations,

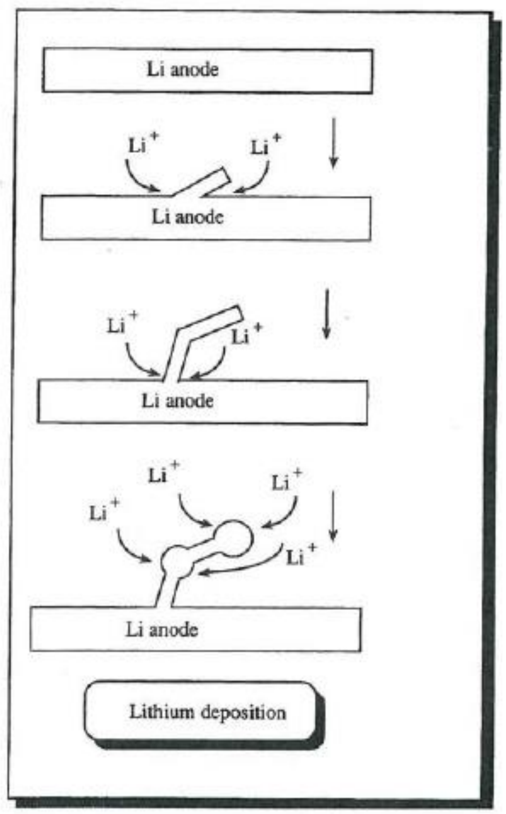
which is all that both Celgard and Dr. White appear to be suggesting, lithium dendrites are in no way limited to this size, shape, and kind of growth.

11. Indeed, as I stated before, lithium dendrites tend to form as "large clumps of granular, mossy type collections of lithium." Abraham Dep. Tr. at 81:6-22. Sample images of lithium dendrites that Dr. White himself provides support for my description. White Declaration at p. 16 (reproduced below). Dr. White's own example is far from the growth formation that he elsewhere calls "needle-like spikes." *Id*. at ¶ 48. And while certain portions of the lithium dendrite may include what Dr. White refers to as "needles [that] can form to a diameter of less than 0.1 μm," the overall dendrite structure is often much larger, often reaching tens or hundreds of microns in width. *Id*. at ¶ 102. Dr. White's example below shows a dendrite having a width of approximately 50 microns.



12. I further disagree with Dr. White's characterization of lithium dendrite growth as one in which the tip simply "grows into the pore following the path of

least resistance." White Dep. Tr. at 54:19-22. While such growth may be theoretically possible, it is unlikely to occur with the pore structure in Tojo's separator. On the other hand, it is well established that dendrites more likely than not grow from the base. In fact, Exhibit 2007, which was submitted by Celgard, shows a lithium dendrite that grows from the base as more lithium is deposited in the base region. *See* Exhibit 2007 at page 344, Fig. 2 (partially reproduced below).



13. A large number of other research works prior to the invention of the '586 patent, as well as my own understanding of dendrite growth based on years of industry experience studying dendrite formation in lithium batteries, reflect this view of dendrite growth. Just as one example, Yamaki et al. have explained that,

for lithium dendrites, "[t]heir tip morphology remains unchanged during their growth, which means they grow from the base...." Ex. 1032 at Abstract.

14. I also disagree with Dr. White's characterization that "[b]ecause the current density is higher at the pore/electrode interface, it is more likely that a dendrite would form at this location on the electrode." White Dec. at ¶ 47. While Dr. White contends that I "confirmed" this during my deposition, I disagree. *Id*. To be clear, pore location in the separator has nothing to do with determining where the dendrite forms because ion flow coming out of the separator is essentially uniform. Indeed, uniform distribution of Li-ions is a basic requirement of separators for use in Li-ion batteries. Rather, the dendrite will tend to form at points of irregularity on the anode surface that provide for increased conductivity and thus ion flux—which is something even Dr. White seems to agree with. *See* White Depo Tr. at 57:12-58:3. Occurrence of trapped gas in the anode during the manufacturing process, not to mention normal repetitive cycling or storage conditions, that creates a region of relatively low resistivity and subsequent increase in Li-ion flow around the gas bubble, when compared to the gas bubble region itself, is one example of anode irregularity that can lead to dendrite formation.

15. I also disagree with Dr. White's characterization that a "dead lithium particle with an SEI layer cannot cause a short" because it is somehow rendered

completely nonconductive. White Declaration at ¶ 49. Rather, "dead lithium" can still remain conductive and lead to a short circuit since, for instance, the SEI layer may not be fully formed around a detached lithium particle. Generally, the SEI layer is very thin and isn't fully formed around the detached lithium particle; consequently, such particle can easily become electrically re-connected to the anode due to expansion/contraction that occurs within the cell during repetitive cycling. It is also possible that any SEI layer that has formed around lithium particles may be removed due to the expansion/contraction. Not surprisingly, one of the papers Celgard cites explains that even dead lithium can cause shorts. *See* Exhibit 2005 at page A806. This is because even "dead" lithium can transmit a charge, contrary to Dr. White's statements otherwise.

16. I further disagree with Dr. White's statements that "it is unlikely that lithium dendrites would be formed in the cell during formation" because "the SEI layer occurs almost immediately." White Declaration at ¶ 64. This is because even 2 to 3 cycles, which is on the low range of number of cycles performed during formation, are sufficient to cause dendrite growth. While Dr. White has admitted that he knows nothing about the details of such formation processes due to his lack of industry experience, I have performed it many times, as I stated previously during my deposition, in the context of lithium batteries and have

observed evidence of dendrite growth during this time. *See* Abraham Dep. Tr. at 137:6-17.

III. Tojo

A. Tojo Blocks Dendrites

17. Dr. White contends that Tojo has only "straight-through holes" in its protective surface layer. *See* White Depo Tr. at 63:14-24. I disagree for at least the reason that Tojo clearly describes various pore structures that even Dr. White agrees would be tortuous and not straight-through.

18. Specifically, while Dr. White appears to rely on paragraph 25 of Tojo to support his contention that Tojo is only directed to "straight-through holes"—a term by the way that is mentioned nowhere in Tojo—Tojo clearly states that the description in paragraph 25 is just "[a]n example of a method" for avoiding the type of problems described earlier in paragraph 24 of Tojo. Tojo at ¶ 25. In fact, paragraph 27 lists other example methods of solving "[t]he problems previously described." *Id.* at ¶ 27. For example, paragraph 27 mentions the use of "extracting, stretching, [or] adding a foaming agent" among others. *Id.*

19. Extracting is a well-established method of creating a porous structure by evaporating the solvent from a mixture comprised of the solvent and other materials, such as binders and inorganic particles. Evaporating just the solvent

from this mixture leaves behind a porous and <u>tortuous</u> structure—not "straight-through holes" as Celgard may contend.

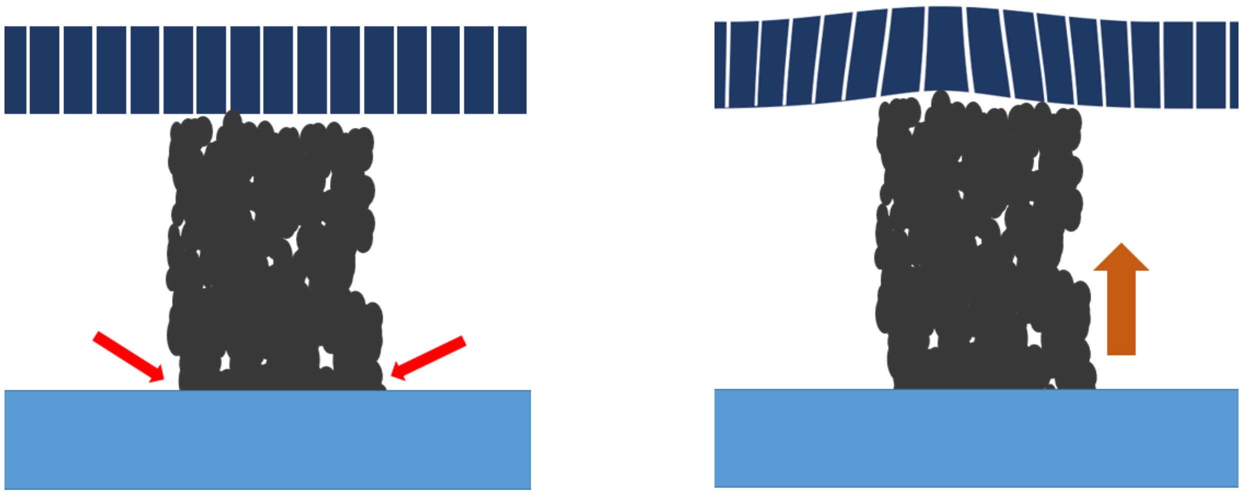
20. Even Dr. White, Celgard's expert, seems to agree. For example, looking through the publicly available deposition transcript of Dr. White from the parallel IPR proceedings involving Celgard and SKI, I see that Dr. White points to the extraction process I described above as a way of making tortuous pores. *See* White Dep. Tr. from SKI IPR proceedings, 81:12-21 ("**Q.** ...How would one create tortuous pores in a ceramic composite layer? . . . **A.** ...I mentioned that it was possible to form such pores by evaporation of a solvent that had been used to form the paste that was used to form the ceramic composite layer on the micro pore separator."). In the same deposition, Dr. White further characterized the pores in Tojo by saying that "the range of porosity of the layers referenced in the Tojo reference go all the way from straight through pores to **<u>non-straight through pores</u>**." *Id.* at 96:20-23, emphasis added).

21. Another point I'd like to reiterate, based on Dr. White's assertion that "other adverse events" as mentioned in Tojo are "unclear" and "vague," is that dendrite growth and penetration are precisely the type of "adverse events" that Tojo is designed to prevent. *See* White Dec. at ¶ 83. Indeed, as I noted above, dendrite growth is something that can occur during the formation process taking place during manufacturing—something even Dr. White doesn't deny. *Id.* at ¶ 64.

Therefore, the problem that Tojo is designed to address—which Dr. White characterizes as "penetration of the membrane during assembly and storage"—clearly encompasses prevention of dendrite growth and penetration. *Id*. at ¶ 83.

22. As I explained above, I disagree with Dr. White's characterization of Tojo as being limited to straight-through pores. However, even if Tojo had "open straight-through holes," which Celgard and Dr. White falsely contend Tojo is limited to, Tojo would *still* block dendrites. *Id*. at ¶ 92.

23. To illustrate, I provide two drawings below. In each, a dendrite is shown stretching from the bottom surface (anode) to the top surface (separator). Further, the separator is illustrated, for sake of argument only, with what Dr. White appears to refer to as "straight-through holes." The dendrite is shown to be approximately 50 µm in width, based on the picture provided by Dr. White in page 16 of his declaration, and the holes are shown to be approximately 1 µm wide, which is much larger than the example low range of 0.1 µm mentioned in paragraph 25 of Tojo.



24. As illustrated above on the left, lithium metal can deposit at the base portion of the dendrite and contribute to the base-growth mechanism I described above. As a result, as shown above on the right, the dendrite can grow from the base and further push into the separator. Here, regardless of any atomic scale growth that may be occurring at the tip of the dendrites, as Dr. White indicated, the base-growth of the dendrite can exert pressure on the separator, potentially causing it to tear. The presence of ceramic particles, as taught by Tojo, will help minimize any such deleterious effects. The straight-through nature of the holes, which Celgard and Dr. White contend, does not take away the penetration-prevention effect of Tojo's separator.

B. Tojo Informs a POSITA to Select the Matrix Materials Claimed in the '586 Patent

25. In response to Dr. White's assertion that Tojo fails to inform a POSITA to select the matrix materials being claimed in the '586 patent, I further provide the following opinion. *See* White Dec. at ¶ 116.

26. To be clear, the types of matrix materials used in the '586 patent were well known by a POSITA at the time of the filing of the '586 patent. In fact, the '586 patent itself recognizes this and simply states that "any gel forming polymer suggested for use" can be used. '586 patent at 3:31-33. In other words, the inventors of the '586 patent are simply listing materials that were commonly known for use in lithium battery separators at the time of filing. Even Dr. White acknowledges that the '586 patent simply lists known gel-forming polymers that were in use at this time. *See* White Dep. Tr. at 100:3-101:7.

27. Additionally, I further respond to Celgard's and Dr. White's assertions by identifying the following five exemplary references showing that PEO and PVDF, two of the materials claimed in the '586 patent, were conventionally used as gel polymer electrolyte separators during and before the filing of the '586 patent.

- Exhibit 1034 is a paper entitled "Trends in Polymer Electrolytes for Secondary Lithium Batteries," and includes in Section 3.1 a description of extensive use prior to 2000 of PEO in dry solid polymer electrolyte separators and in sections 4.1 and 4.2 on gel polymer electrolyte separators.

- Exhibit 1035 is a paper entitled "Studies on PVdF-based gel polymer electrolytes," and describes extensive use prior to 2000 of PVDF in gel polymer electrolyte separators.
- Exhibit 1036 is a paper that I co-authored in 1996 entitled "Studies of some poly(vinylidene fluoride) electrolytes," also describing extensive prior use of PVDF in gel polymer electrolyte separators.
- Exhibit 1037 is a paper that I co-authored in 1996 entitled "Characterization of Some Polyacrylonitrile-Based Electrolytes" that describes polyacrylonitrile-based gel polymer electrolyte separators.
- Exhibit 1038 is a 1998 paper entitled "Review of gel-type polymer electrolytes for lithium-ion batteries" that describes polyethylene oxide (PEO), polyacrylonitrile (PAN) poly_methyl methacrylate (PMMA) and polyvinylidene fluoride (PVdF)-based gel polymer electrolyte separators.

28. Dr. White's assertion that I relied on the '586 patent "[w]ithout any reasoning" to invalidate the claimed matrix materials is nonsensical for at least the reason that even the '586 patent admits that there was nothing special about the listed matrix materials and further because such materials were already in wide use at the time, as evidenced by the example publications noted above. Moreover, U.S. Patent No. 5,631,103 to Eschbach (Exhibit 1039), which I understand was relied on by the Patent Office during prosecution of the '586 patent and also

predates it by several years, teaches that "the matrix material is polyethylene oxide, polyvinylidene fluoride, polytetrafluoroethylene, polyurethane," thus further proving my point that these were commonly known materials. Eschbach at 4:1-19.

29. And while Dr. White refers to the declaration of Dr. Wensley to somehow suggest that the specific matrix materials claimed involved a complex evaluation process, the steps that Dr. Wensley describes are routine activities for a person of ordinary skill in the art at the time of the invention of the '586 patent and involve no inventive work whatsoever. *See* White Dec. at 121, citing to Wensley Dec. at 14-24.

IV. Tobishima in View of Tojo

30. Celgard argues that "adding the inorganic particles of Tojo would decrease the already low conductivity of Tobishima, resulting in a battery does not work." Celgard Response at page 42. Celgard further contends that "[a] POSITA who considered the particles of Tojo would not be motivated to add them because doing so would further reduce conductivity of Tobishima's electrolyte impregnated polymer layer." *Id*. at page 43. I disagree with this rationale for several reasons.

31. First, a battery with reduced conductivity is not one that "does not work." In fact, as Celgard acknowledges, even Tobishima describes that its battery still functions at "about 1/10 of the electric conductivity." Celgard Response at page 42 (citing from Exhibit 1005 at paragraph 11). Tobishima also contemplates

reducing this conductivity even further by using an optional cross-linking process, a fact which Celgard acknowledges. *See* Celgard Response at 43; *see also* Exhibit 1005 at paragraph 11.

32. Second, while certain types of ceramic particles may decrease the conductivity of a layer into which they are impregnated, a POSITA would certainly know to only add enough particles to achieve the optimal balance of strength and conductivity performance. This would actually provide a way to improve the mechanical strength of Tobishima without going through the <u>optional</u> cross-linking step, which Celgard admits would lead to undesirable loss of conductivity.

33. Third, depending on the type of ceramic particles used, the overall conductivity of the polymer layer may not even be decreased by the addition of such ceramic particles. In fact, the '586 patent itself mentions that while "[t]he inorganic particles 28 are normally considered nonconductive, however, these particles, when in contact with the electrolyte, appear...to develop a superconductive surface which improves the conductivity (reduces resistance) of the separator 16." '586 patent at 3:47-52. This would provide an additional motivation for a POSITA looking to improve the mechanical strength of Tobishima without going through the <u>optional</u> step of cross-linking, which Celgard admits leads to undesirable reduction in conductivity.

V. Secondary Considerations

34. I understand that Celgard's expert, Dr. White, has provided an analysis of the secondary considerations, which he refers to as the "objective signs of non-obviousness," alleging that "there is some link between the claimed invention and the secondary factors considered." White Declaration at ¶ 145. To this end, Dr. White opines that "the wide ranging industry acceptance, and promotion of the inventions claimed in the '586 patent is strong evidence of non-obviousness." *Id.* at ¶ 163 (emphasis added). Similarly, Dr. White states that "there is substantial evidence of commercial success of the claimed inventions." *Id.* at ¶ 165 (emphasis added). I disagree with such assessment.

35. To be clear, the "claimed inventions" that Dr. White refers to is not coextensive with all ceramic coated separators in general. In other words, the '586 patent does not in any way represent the invention of ceramic coated separators generally. Rather, the "claimed inventions" of the '586 patent, as reflected in its claims, are directed to a separator that possesses each of the various claimed features. *See, e.g.*, White Dep. Tr. at 28:19-29:7 (Dr. White explaining that "it is the Claim 1 **in its entirety** that is the invention of the '586 patent) (emphasis added); *see also id.* at 26:7-23, 27:8-25, 30:8-18. Thus, any analysis that points to, for example, merely the success of ceramic coated separators in general does not in any way inform a POSITA whether the *claimed* subject matter of the '586 patent would enjoy similar success.

VI. Closing

36. I currently hold the opinions expressed in this declaration. But my analysis may continue. If and as my study of the investigation continues, I may acquire additional information and/or attain supplemental insights that result in added observations.

37. I hereby 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.

Signature: Kuzhikalail M. Abraham Date: 5-4-2015

Kuzhikalail M. Abraham, Ph. D.