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17 **UNITED STATES DISTRICT COURT**
18 **CENTRAL DISTRICT OF CALIFORNIA**

19 RJ TECHNOLOGY LLC,) CASE NO. 8:22-CV-1874-JVS-JDE
20)
21 Plaintiff,) **DEFENDANT APPLE INC.'S**
22) **RESPONSIVE CLAIM**
23 v.) **CONSTRUCTION BRIEF**
24)

23 APPLE INC.,) Date: July 31, 2023
24) Time: 3:00 p.m.
25 Defendant.) Ctrm: 10C
26) Judge: Hon. James V. Selna
27)

27) Second Amended Complaint Filed:
28) April 13, 2023

DEFENDANT APPLE INC.'S
RESPONSIVE CLAIM CONSTRUCTION
BRIEF

Case No. 8:22-CV-1874-JVS-JDE

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I. INTRODUCTION

In its Opening Brief, Plaintiff’s shifting positions make clear that its arguments change to whatever is convenient to its needs at the time. Dkt. 70 (“Pl.’s Br.”). Before the United States Patent and Trademark Office (“USPTO”), Plaintiff made representations on which the examiner relied in order to issue the ’641 Patent.¹ And in other forums around the world, Plaintiff repeatedly defined the claim terms at issue here while seeking to avoid invalidation. Despite narrowing the scope of its invention to gain and defend its patent, Plaintiff now seeks to impermissibly re-broaden the scope of the ’641 Patent. This it cannot do. Though Plaintiff broadly asserts the plain and ordinary meaning applies to the terms for which construction is requested, tellingly, Plaintiff avoids its own statements made during the prosecution of both the ’641 Patent and proceedings regarding the foreign counterpart patent. These statements are critically at odds with both the arguments it makes, as well its expert’s opinions. Confirming its made-for-litigation positions, Plaintiff’s own expert has offered different opinions—outside this litigation—that contradict Plaintiff’s arguments here, and support Apple’s positions. As a result, Plaintiff requests the Court: (1) find indefinite two terms, and (2) adopt Apple’s construction for the remaining three.

II. DISPUTED TERMS

Term / Claims	Apple’s Proposal	Plaintiff’s Proposal
“a charge cut-off voltage” (claims 5–7)	The voltage at which the charging changes from constant current charging to constant voltage charging.	No construction needed. Alternatively, plain and ordinary meaning: the maximum voltage to which the battery is configured to be charged.

¹ Apple disputes Plaintiff’s characterization that it has “profited from this groundbreaking invention,” (Pl.’s Br. 1) as well as allegations that Apple’s products infringe the asserted patent.

A. “a charge cut-off voltage”

In its Opening Brief, Plaintiff characterizes the dispute at issue as whether Apple’s construction properly limits the charge cut-off voltage to CC/CV charging.² Pl.’s Br. 8. But the Court need not decide at this stage whether: (1) Plaintiff’s characterizations of different charging methods are accurate,³ or (2) whether Apple’s proposed construction does or does not include certain of those methods. Indeed, many variables affect whether or not any given charging method would meet either party’s construction, and Plaintiff’s high-level characterizations of various ways that batteries have been historically charged is an irrelevant distraction from the issues before this Court. Horn Decl., ¶¶8–9, 11. Indeed, Plaintiff’s attempt to introduce the question of which types of charging infringe the patent are premature questions of infringement that need not be resolved at the *Markman* stage, and should thus be disregarded. *Eon Corp. IP Holdings v. Silver Spring Networks*, 815 F.3d 1314, 1319 (Fed. Cir. 2016) (“[A]t the claim construction stage ... courts should not resolve questions that do not go to claim scope, but instead go to infringement.”); *SRI Int’l v. Matsushita Elec. Corp. of Am.*, 775 F.2d 1107, 1118 (Fed. Cir. 1985) (“A claim is construed in the light of the claim language, the other claims, the prior art, the prosecution history, and the specification, *not* in light of the accused device.”) (emphasis in original). Therefore, whether or not a given charging method meets either party’s construction is a question of fact appropriate for fact and expert discovery.

² Plaintiff’s complaints regarding the production of Apple’s technical documents are unfounded. Plaintiff has produced a total of 140 public documents, despite its obligation to produce documents in connection with PLR 3-2, and multiple requests from Apple regarding purported testing performed by Plaintiff. To the contrary, Apple has produced over 6,000 documents to date, including technical documents.

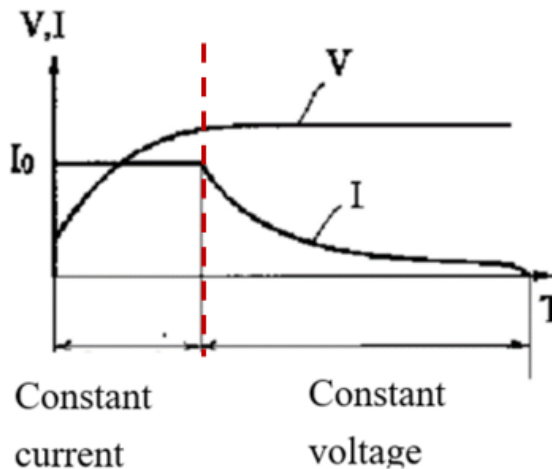
³ While Apple does not agree with all of Plaintiff’s statements regarding the technology overview, Apple disputes only the relevant portions for each term in the sections below.

1 Instead, the question for the Court is whether, in light of Patentees'⁴ explicit
 2 statements regarding what it represented as a "standard general term in the field" during
 3 litigation over a foreign counterpart, Patentees' statements should be adopted. In
 4 opposition to invalidation proceedings for the '641 Patent's Chinese counterpart, Apple
 5 contended that the term charge cut-off voltage was "not clear." Dkt. 68 at 5 ("Apple's
 6 Br."), Exs. C–F (CNIPA Case Nos. 4W107406 & 4W112700 (collectively, "CN
 7 Invalidation Proceedings")). Patentees responded that "[t]his technical term is a
 8 standard general term in the field of battery related technology, *and its meaning is*
 9 *determined*, which refers to *the voltage value of the battery when it transitions from*
 10 *charging at a constant current to charging at a constant voltage*. The meaning of 'the
 11 charging limit voltage' in the description of the present patent is the same as it usually
 12 has in this field."⁵ Apple's Br., Ex. D at 7. Moreover, when attempting to distinguish
 13 prior art during the CN invalidity proceedings, the Patentees doubled down, specifically
 14 referencing the below figure and stating, "[f]or the charging curve of the limited
 15 charging voltage, refer to Figure 35 of D10 (Page 395/431). That is, *the limited*
 16 *charging voltage is the voltage value when the battery is charged from the constant*
 17 *current to constant voltage*." Apple's Br., Ex. F at 27; *see also id.* at 30, 50.

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 26 ⁴ The '641 Patent inventors are Xiaoping Ren and Jie Sun, who are also "Plaintiff's
 founders and principals." Dkt. 51, ¶ 7.

27 ⁵ All emphases added unless otherwise noted.



Apple’s Br., Ex. F at 27 (annotation added). As explained in Apple’s Opening Brief, these statements are intrinsic evidence and clearly reflect that the charge cut-off voltage is voltage when the battery is charged from constant current to constant voltage. *See also* Apple’s Br., Ex. G at 5–6 (Plaintiff’s prior PLR 4-2 disclosure cited China National Standard, which states the battery charges “*switch[es] to constant voltage charging* when the voltage at the battery side reaches the limited charge voltage *until charging current* is less than or equal to 20mA.”); *see also* Pl.’s Br. 11 (“[T]he Linden handbook reaffirms that the then-industry standard 4.2V limit was typically implemented via CC-CV charging at that level.”); Horn Decl., ¶10. Moreover, the principle of judicial estoppel should apply. *See Rissetto v. Plumbers & Steamfitters Loc.*, 94 F.3d 597, 600 (9th Cir. 1996) (“Judicial estoppel ... precludes a party from gaining an advantage by taking one position, and then seeking a second advantage by taking an incompatible position.”); *Intel Corp. v. XMTT, Inc.*, No. 2021-2127, 2022 WL 1152312, at *1 (Fed. Cir. Apr. 19, 2022) (finding judicial estoppel when party “changes its position and advocates for a claim construction that is clearly inconsistent with its position before the [prior tribunal]”); *see also Guinness PLC v. Ward*, 955 F.2d 875, 899 (4th Cir. 1992) (finding judicial estoppel can arise from positions taken in a foreign proceeding); *Ajinomoto Co. v. Archer-Daniels-Midland Co.*, 228 F.3d 1338, 1349 (Fed. Cir. 2000) (relying on patentees’ own usage of term in submission to foreign government to

1 construe term).

2 Rather than address its own statements,⁶ Plaintiff identifies intrinsic evidence that
 3 does nothing to justify Plaintiff's construction over Apple's. Plaintiff first identifies
 4 three quotes from the '641 Patent, contending the quotes refer to the charging voltage
 5 "maximum." Pl.'s Br. 9. But these quotes merely restate the claim language, explaining
 6 that the "charge cut-off voltage" is set to 4.2V or 5.8V, but providing no explanation
 7 for *what* that charge cut-off voltage is. Nor does Plaintiff's recitation of the file history
 8 discussion regarding "charging voltage" support its construction: again, both only
 9 discuss the deleterious effects when the "charging voltage increases beyond 4.2V,"
 10 which does not lend support to Plaintiff's construction over Apple's. Indeed, Apple
 11 does not dispute Plaintiff's argument that the intrinsic record "repeatedly refers to 4.2V
 12 as the then-universal industry standard for charge cutoff voltage," but rather disputes
 13 Plaintiff's interpretation of what the industry standard *meant* by "charge cutoff
 14 voltage." Pl.'s Br. 10. Plaintiff's own statements during the CN invalidation
 15 proceedings provide that answer: "[t]his technical term is a standard general term in the
 16 field of battery related technology, *and its meaning is determined*, which refers to *the*
 17 *voltage value of the battery when it transitions from charging at a constant current*
 18 *to charging at a constant voltage*." Apple's Br., Ex. D at 7.

19 Plaintiff then attempts to redefine its construction, arguing that its proposed
 20 "maximum" cut-off voltage construction "does not mean that during normal use of the
 21 battery, the charging voltage can never exceed the charge cut-off voltage." Pl.'s Br. 9.
 22 This pivot—admitting Plaintiff's proposal of "maximum" means something other than
 23 "maximum"—is fatal, as it renders Plaintiff's proposed construction indefinite and
 24 confusing. Nor is Plaintiff's explanation for its adjustment appropriate: Plaintiff's
 25 construction further introduces the idea that the voltage must be "configured" (*i.e.*,

26
 27 ⁶ Despite Apple's identification of Patentees' statements in the CN Invalidation
 Proceedings, Plaintiff failed to address those statements in its Opening Brief.

designed and manufactured”) (*id.*), but identifies no support in the intrinsic record for why the “configuration” of the battery or the underlying design intent is relevant to, or within the scope of, the asserted claims. *See Tempo Lighting, Inc. v. Tivoli, LLC*, 742 F.3d 973, 977 (Fed. Cir. 2014) (rejecting proposed construction adding “limitations that lack support in any form of intrinsic evidence”); *see also Radiall S.A. v. Glenair, Inc.*, No. 2:14-CV-00822-ODW, 2015 WL 1321518, at *5 (C.D. Cal. Mar. 20, 2015) (same).

Moreover, Plaintiff’s added limitation that the maximum voltage must be “designed and manufactured” (Pl.’s Br. 9) improperly introduces a requirement as to the ***subjective intent*** of a designer or manufacturer regarding the cut-off voltage. In other words, Plaintiff’s proposed construction introduces the new requirement that a battery be “***configured***” (“*i.e., designed and manufactured*”) (*id.*) to be charged at a charge cut-off voltage of 4.2 V, not just that it have the physical property of a charge cut-off voltage set at 4.2 V, as claim 5 requires. *See* Horn Decl., ¶12. Plaintiff’s proposed construction is thus improper. *See Cochlear Bone Anchored Sols. AB v. Oticon Med. AB*, 958 F.3d 1348, 1356 (Fed. Cir. 2020) (“To the extent that Cochlear is suggesting that the language requires a particular intent or objective of a hearing-aid designer or manufacturer, we reject the suggestion.”); *VetStem BioPharma, Inc. v. Cal. Stem Cell Treatment Ctr., Inc.*, No. 2:19-CV-04728-AB-FFM, 2022 WL 1601387, at *4 (C.D. Cal. Mar. 17, 2022) (injecting intent “runs contrary to the well-established principle that intent is typically irrelevant to patent infringement”). Because Apple’s proposed construction is consistent with the intrinsic evidence, Patentees’ own statements to a patent authority, and Patentee’s initial proposal in this case, Apple’s proposed construction should be adopted.

B. “ratio ... as calculated by a theoretic capacity”

Term / Claims	Apple’s Proposal	Plaintiff’s Proposal
“ratio of positive electrode material to negative electrode material as calculated by a	Indefinite.	No construction needed.

1 2 3 4 5 6 7 8 9 10 11 12	theoretic capacity with a charge cut-off voltage set at 4.2 V” (claim 5)	Alternatively, the ratio calculated as the product of the theoretic capacity of the cathode active material per mass unit and the amount of the cathode active material in the same mass unit divided by the product of the capacity of the anode active material per mass unit and the amount of the anode active material in the same mass unit, when the cell charge cutoff voltage is set at 4.2 V, wherein “theoretic capacity” is voltage independent and means “theoretical, rather than actual, capacity that assumes all of the active substances participate in the reaction of the battery.”	Alternatively, plain and ordinary meaning: the ratio calculated as the product of the capacity of the cathode active material per mass unit and the amount of the cathode active material in the same mass unit divided by the product of the capacity of the anode active material per mass unit and the amount of the anode active material in the same mass unit, when the charge cutoff voltage is 4.2 V.
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13 Plaintiff’s arguments in its Opening Brief underscore that if the “ratio” term is
14 not determined to be indefinite, Apple’s construction should be adopted, as Plaintiff
15 seeks to impermissibly broaden the scope of “*theoretic* capacity” to mean *any* capacity,
16 including *specific* capacity. Moreover, the term is indefinite even under Plaintiff’s
17 proposed construction: Plaintiff both admits that the patent at least “sometimes” uses
18 “theoretical capacity” to refer to the “theoretical maximum capacity,” yet maintains that
19 theoretical capacity “must refer to the theoretical capacity at [4.2V] because it means
20 some of the lithium may not participate in the reactions”—*i.e.* something *less* than
21 “theoretical maximum capacity” and hence akin to “specific capacity.” Pl.’s Br. 15.
22 Plaintiff’s own positions thus underscore that this term is indefinite.

23 1. If Not Invalid, Apple’s Construction Should Be Adopted

24 As a preliminary matter, the Court need not construe the “ratio” term, as it is
25 indefinite for the reasons set forth in the next section. However, if the term is not found
26 indefinite, Plaintiff’s proposed construction should be rejected. As Plaintiff concedes,
27 both parties generally agree how the ratio of cathode to anode material should be

1 calculated. *See* Pl.’s Br. 13. However, there are two apparent disputes: (1) whether the
 2 “theoretic capacity” used to calculate the ratio should be expanded to capture all forms
 3 of “capacity” (Plaintiff’s proposal), or whether the theoretic capacity means
 4 “theoretical, rather than actual, capacity that assumes all of the active substances
 5 participate in the reaction of the battery” (Apple’s proposal); and (2) whether the charge
 6 cutoff voltage refers to the “cell” charge cutoff voltage.

7 **First**, with respect to theoretical capacity, as noted in Apple’s Opening Brief, the
 8 plain language of claim 5 reciting “theoretical capacity” weighs in favor of construing
 9 it as just that: a capacity theoretically assuming that all of the active materials participate
 10 in the reaction. Consistent with this plain meaning, claim 1 recites “specific capacity”
 11 (or actual capacity), as calculated at a charge cut-off voltage of 4.2 V. As different
 12 terms are presumed to have different meanings, “theoretic capacity” must mean
 13 something different from “specific capacity.” *See Bd. of Regents of the Univ. of Tex.*
 14 *Sys. v. BENQ Am. Corp.*, 533 F.3d 1362, 1371 (Fed. Cir. 2008) (different terms are
 15 “presumed to have different meanings”). In other words, since claim 1 recites specific
 16 capacity (*i.e.*, actual capacity), claim 5’s recitation confirms that “theoretic capacity”
 17 must mean something different, and only Apple’s proposal captures that distinction.

18 Plaintiff’s Opening Brief conflates the two distinct measures of capacity: Plaintiff
 19 claims that ***theoretic*** capacity refers only to a “portion of” the active material being
 20 engaged at “less than 100%” of the maximum capacity, *i.e.*, at a charge cut-off voltage
 21 of 4.2 V. Pl.’s Br. 14. But claim 1, reciting specific capacity, references the same
 22 capacity calculation “at a charge cut-off voltage of 4.2 V.” ’641 Patent, cl. 1. The
 23 specification further distinguishes between “theoretical capacity” and “actually used
 24 capacity,” (*id.*, 1:40–43), and Plaintiff’s expert confirms that actual capacity refers to
 25 specific capacity. Yao Decl., ¶50 (“Such specific capacity is also closer to the actually
 26 usable capacity, so it acts as a good reference point to determine how to balance the
 27 capacities of cathode and anode in a battery.”); *see also* Horn Decl., ¶¶ 14–15. Thus,

1 despite Plaintiff admitting theoretical and specific capacity are different, if theoretic
2 capacity is construed in the way Plaintiff requests, there will be the same.

3 Underscoring Plaintiff's attempt to transpose the two concepts, Plaintiff's expert
4 explicitly uses "specific capacity" in the ratio calculation. *See* Yao Decl., ¶38
5 ("[W]here C_p is the specific capacity of the cathode active material ... C_n is the specific
6 capacity of the anode active material ..."), *id.*, ¶39 ("The ratio refers to a ratio **based**
7 **on specific capacity**"). But specific capacity is recited in claim 1, which Plaintiff has
8 elected to withdraw from this case. Plaintiff cannot now seek to broaden asserted claim
9 5 to capture every measure of capacity that exists, including the dropped specific
10 capacity in claim 1. Plaintiff's attempt to manipulate the straightforward meaning of
11 "theoretical" into "actual" (or specific) capacity is both factually and legally incorrect:
12 the claims and specification confirm that theoretical and specific capacity refer to
13 different types of capacity as set forth in Apple's proposed construction, and Plaintiff
14 cites no authority stating that two different terms should be construed to mean the same
15 thing.

16 Rather than remaining consistent with the language of the claims and
17 specification, Plaintiff and its expert confusingly use a number of terms to reference
18 capacity: (1) theoretic capacity; (2) theoretic maximum capacity; (3) specific capacity;
19 (4) maximum specific capacity; (5) initial capacity; and (6) remaining capacity. *See*,
20 *e.g.*, Yao Decl., ¶¶ 38–40, 44–46, 72–74. But Plaintiff does not explain how these
21 concepts are different in the context of the recitations of claims 1 and 5. In fact,
22 Plaintiff's creation of the new "theoretic maximum capacity" term is referenced
23 nowhere in the specification. *See* Pl.'s Br. 16; Yao Decl., ¶¶ 43–44. To the contrary,
24 the specification clearly provides examples of "theoretical capacity"—which is when
25 all active materials participate in the reaction (*i.e.*, roughly double specific capacity for
26 LiCoO_2). *See, e.g.*, '641 Patent, 1:40–44, 1:65–2:1. Plaintiff's expert's explanation for
27 the so-called "theoretical maximum capacity" confirms he is in fact referring to what
28

the specification describes as “theoretical capacity.” *Compare* Yao Decl. ¶ 48 (“A POSA would readily appreciate that the theoretical maximum capacity is not achievable in real life.”), *with* ’641 Patent, 1:43–45 (“[A]bout half of said theoretic capacity is not utilized. This is mainly caused by the limitation of charge cut-off voltage commonly used in the art.”). And Plaintiff’s interchangeable use of its new capacity terms confuses, rather than clarifies the capacity calculation recited in claim 5. *See* Pl.’s Br. 17 (theoretical capacity “sometimes [refers] to the theoretical maximum”). Indeed, Plaintiff does not address (nor cite any case law) supporting its departure from the disclosures in the specification that contradict its contrived re-writing of “theoretical capacity” to mean “maximum theoretical capacity.” Plaintiff’s unfounded attempt to depart from the specification and claim language, to contend a POSITA would supposedly understand it to mean something different, should be rejected.

Indeed, Patentees themselves made contrary representations during the CN Invalidation Proceedings. Specifically, with respect to the claimed “ratio of positive and negative electrodes of the single lithium ion battery is between 1:1.0 and 1:2.5 based on the theoretical gram capacity when the charging limit voltage is 4.2V,”⁷ the Patentees explained that “theoretical gram capacity” (*i.e.*, theoretic capacity) assumes all active substances participate:

The amount of electricity (electricity capacity) given ***by assuming that all active substances participate in the reaction of the battery.*** Specifically, where the active substance is LiCoO_2 , the Faraday constant $F = 96485$ C/mol, and the molar mass of LiCoO_2 $M = 97.8698$ g/mol. If all lithium ions are de-intercalated, then $F/M = 985.8535$ C/g, and after unit conversion, ***it is 273.848 mAh/g, which is the theoretical gram capacity.***

⁷ The ’641 Patent claims priority to PCT/CN02/00696, which claims priority to Chinese Patent No. ZL 01141615.7. Plaintiff has conceded the ’641 Patent is practically identical to the Chinese counterpart. Dkt. 51 at 16 (“[I]ts specification is translated directly from the Chinese Patent’s specification,” and “[c]laims 5–10 of the Asserted Patent are also ***literal translations of claims 5–10 of the Chinese Patent.***”).

1 However, the actual capacity released by the battery is only a part of the
 2 theoretical values mentioned above.
 3 Apple's Opening Brief, Ex. D, APL-RJT_00785537; Horn Decl., ¶ 18. In other words,
 4 Patentees' statements reflect their understanding that "theoretical gram capacity" is the
 5 theoretical capacity, *i.e.*, when all of the active materials participate.

6 Plaintiff's expert, Dr. Yao, has similarly made statements consistent with the
 7 specification regarding theoretical capacity and specific capacity. In his own patents
 8 and publications—predating this litigation—Dr. Yao contradicts Plaintiff's position
 9 that "theoretical capacity" refers to a capacity where less than all of the active materials
 10 participate. Specifically, Dr. Yao's patents and publications describe "specific"
 11 capacity as a percentage of "theoretical capacity." *See* Horn Decl. ¶ 19 (citing Horn
 12 Decl., Ex. 3 (Heavily n-Dopable π -Conjugated Redox Polymers with Ultrafast Energy
 13 Storage Capability) at 4957 ("***[T]he specific capacity of P(NDI2OD-TET) is 47.4 mAh***
 14 ***g⁻¹, or 90% of its theoretical capacity (52.7 mAh g⁻¹).***");⁸ Horn Decl., Ex. 4 (U.S.
 15 Patent No. 10,522,875) at 7:22–25 ("A ***specific capacity of up to 350 mAh/g*** is
 16 obtained, ***which is 91% of the theoretical capacity***").⁹

17 Plaintiff alleges that the claim requires the theoretical capacity "refer to a value
 18 modified by the applicable x factor." Pl's Br. 17. However, the theoretical capacity
 19 that is modified by this applicable x factor refers to the ***specific capacity***—not

21 ⁸ For certain materials at certain charge cut-off voltages, all of the material may
 22 engage. *Id.* ("***[T]he specific capacity of P(NDI2OD-T2) is 54.2 mAh g⁻¹, or 100%***
 of its theoretical capacity.").

23 ⁹ Plaintiff's assertion that "theoretic capacity" refers to the "4.2V capacity" is likewise
 24 unavailing. Plaintiff cites Linden's Table 1.1, but Linden does not specify or recite
 25 a "theoretical capacity," nor does it state any cut-off voltage (and certainly not 4.2
 26 V) that is applied. *See* Plaintiff's Ex. 6 at RJ00001804_Apple-RJ00001805_Apple;
 27 Horn Decl., ¶ 20. Indeed, Linden later explicitly describes "[t]he reversible value of
 x for LiCoO₂ ... is approximately 0.5" and "[t]hus although the ***theoretical capacity***
 of LiCoO₂ and LiNiO₂ (275 mAh/g)" Plaintiff's Ex. 6 at RJ00002798-Apple;
 Horn Decl., ¶ 20.

1 theoretical capacity. Horn Decl., ¶ 20; '641 Patent, 1:40–44 (LiCoO₂ “has a theoretical
2 capacity of 248 mAh/g, while the actually used capacity of it is merely about 140
3 mAh/g, i.e., about half of said theoretic capacity is not utilized.”).

4 **Second**, although lacking from Plaintiff’s Opening Brief, as explained in
5 Apple’s Opening Brief, clarification that the voltage to be used is a “cell” voltage is
6 necessary, as the recitation of “charge cut-off voltage” can ambiguously refer to either
7 the half-cell voltage, or the full-cell voltage. Horn Decl., ¶ 21.

8 2. The Term Is Indefinite

9 Plaintiff’s arguments confirm that regardless which construction applies, the term
10 is indefinite. Plaintiff argues that “theoretic capacity” as recited in claim 5 is modified
11 by the limitation “a charge cut-off voltage set at 4.2 V.” But as explained in the
12 preceding section, it is *specific* capacity, not *theoretical* capacity, that measures the
13 amount of material participating in a reaction at a specific charge cut-off voltage.
14 See '641 Patent, 1:40–45 (explaining that for a positive electrode material, the “charge
15 cut-off voltage” caused the “actually used capacity” to be “about half” of “theoretical
16 capacity”). Theoretical capacity, on the other hand, is voltage independent because it
17 assumes that *all* of the active materials participate. Thus, a POSITA would not
18 understand how to calculate a theoretical capacity, at a charge cut-off voltage of 4.2V.
19 Horn Decl., ¶ 22. If a POSITA understood the claim to refer to theoretical capacity, a
20 POSITA would assume all materials participate and thus the charge voltage would be
21 irrelevant; if a POSITA understood the claim to refer to specific capacity at a charge
22 cut-off voltage of 4.2 V, the POSITA would not be applying the plain language
23 “theoretical.” *Nautilus, Inc. v. Biosig Instruments, Inc.*, 572 U.S. 898, 901 (2014) (claim
24 indefinite when it “fail[s] to inform, with reasonable certainty, those skilled in the art
25 about the scope of the invention”).

26 Although Plaintiff asserts this confusion is created by Apple’s proposed
27 construction, in fact claim 5 creates this ambiguity. Confronted by two contradictory

1 elements, Plaintiff seeks to rewrite “theoretical capacity” as “specific capacity,” as
 2 confirmed by its explanation that theoretical capacity refers to a “theoretical value
 3 modified by an ‘x’ factor.” Pl.’s Br. 17. But Plaintiff cannot rewrite the claim to avoid
 4 indefiniteness, particularly where Patentees drafted the claims using the distinct terms
 5 “specific capacity” (claim 1) and “theoretical capacity” (claim 5). *Tandon Corp. v. U.S.*
 6 *Int’l Trade Comm’n*, 831 F.2d 1017, 1023 (Fed. Cir. 1987) (“There is presumed to be a
 7 difference in meaning and scope when different words or phrases are used in separate
 8 claims.”).

9 Despite contradictory evidence in the claims and specification, Plaintiff instead
 10 relies heavily on a single citation to the specification to assert that the patent “expressly
 11 defines theoretical capacity to mean the capacity at 4.2 V.” Pl.’s Br. 15. Not so. As an
 12 initial matter, the specification does not define the actual claim term “ratio of positive
 13 electrode material to negative electrode material as calculated by a theoretic capacity
 14 with a charge cut-off voltage set at 4.2 V,” instead referring to “theoretical capacities
 15 of the positive and negative electrode.” ’641 Patent, 4:5–9. In light of the other
 16 disclosures in the specification, Plaintiff’s cite does not satisfy the standard for an
 17 express definition of the disputed claim term. *See Thorner v. Sony Computer Ent. Am.*
 18 *LLC*, 669 F.3d 1362, 1365 (Fed. Cir. 2012) (“To act as its own lexicographer, a patentee
 19 must ‘clearly set forth a definition of the **disputed claim term**’ other than its plain and
 20 ordinary meaning.”); *In re Paulsen*, 30 F.3d 1475, 1480 (Fed. Cir. 1994) (definition
 21 must be described with “reasonable clarity, deliberateness, and precision”); *Bradium*
 22 *Techs. LLC v. Iancu*, 923 F.3d 1032, 1044 (Fed. Cir. 2019) (“It is not enough for a
 23 patentee to simply disclose a single embodiment or use a word in the same manner in
 24 all embodiments, the patentee must ‘clearly express an intent’ to redefine the term.”).
 25 Even if the statement were an express definition (it is not), the term is still indefinite
 26 because a POSITA would not know which form of “capacity” should be measured: *i.e.*,
 27 theoretic capacity, specific capacity, or Plaintiff’s new “initial capacity” or “remaining

capacity.” See Pl.’s Br. 23–24; *Merck & Co. v. Teva Pharms. USA, Inc.*, 395 F.3d 1364, 1370 (Fed. Cir. 2005) (finding definitional statement did not apply because “the passage cited by the district court from the specification for Merck’s definition of ‘about’ is ambiguous”); *Abbott Lab’ys v. Syntron Bioresearch, Inc.*, 334 F.3d 1343, 1355 (Fed. Cir. 2003) (“Because the specification provides two alternative definitions for the term at issue, the specification does not define the claim term in the manner required under *Renishaw*.”); *Bushnell Hawthorne, LLC v. Cisco Sys., Inc.*, 813 F. App’x 522, 527 (Fed. Cir. 2020) (finding a claim indefinite when “[a] POSA, faced with the claims and the specification, could not, with reasonable certainty, discern the meaning of the claim term”). Nor would a POSITA understand how to measure theoretical capacity at a charge cut-off voltage, in light of the numerous other disclosures in the specification. Horn Decl., ¶ 22.

Indeed, even under Plaintiff’s proposed construction, the term is indefinite because the specification does not teach a POSITA how to calculate how much less “theoretic capacity” is than Plaintiff’s supposed “maximum theoretical capacity.” Pl.’s Br. 17 (“Theoretical capacity in that context *must* refer to a value modified by the applicable x factor”).¹⁰ Nor does Plaintiff explain how it differs from specific capacity.

For the foregoing reasons, the claim term is indefinite. However, to the extent the term has any meaning to a POSITA, the Court should adopt Apple’s proposed construction.

C. “A secondary lithium ion cell or battery”

Term / Claims	Apple’s Proposal	Plaintiff’s Proposal
“A secondary lithium ion cell or battery” (claim 5)	A lithium ion battery, wherein the battery electrode materials	No construction needed.

¹⁰ To the extent Plaintiff contends a POSITA would have known the applicable x factor for a material, Plaintiff’s expert confirms that it is uncertain which value would be used: he opines that certain values could be assumed, but also opines that “higher values may be obtained in practice.” Pl.’s Br. 16.

	must not be substantially changed from what was available in the art and made without substantially decomposing the electrolyte, which can be recharged to its original capacity, without any decreased capacity or cycle life at a charge cut-off voltage above 4.2 V.	Alternatively, see RJ Technology’s proposed construction for the term “as calculated by a theoretic capacity with a charge cut-off voltage set at 4.2 V.”
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Plaintiff challenges two aspects of Apple’s proposed construction: (1) that the lithium ion battery “can be recharged ... *without any decreased* capacity or cycle life ... above 4.2 V,” and (2) that the “electrode materials *must not be substantially changed* from what was available in the art.”¹¹ Plaintiff’s arguments are unpersuasive.

First, Plaintiff argues that Apple’s construction is contrary to the patent claims, specification and prosecution history, which show “the claimed secondary battery experiences *low* capacity loss over hundreds of charges—not *no* capacity loss.” Pl.’s Br. 19 (emphasis in original). Not so: during prosecution, the Patentees repeatedly emphasized that their purported invention *did not* diminish the cycle life—an allegedly novel aspect of their invention. *See, e.g.*, Apple’s Br., Ex. B at 83 (“In a manner wholly unforeseen by the art, Applicant’s novel methodology *did not diminish* the recycle property of the secondary lithium ion battery.”); *see also* ’641 Patent, 4:20–21 (describing cells with “excellent recycle property” as batteries that maintain “more than 95% after 50 cycles, and and more than 80% after 300 cycles”); *id.* at 4:24–26 (explaining a “cell exhibit[ing] inferior recycle property” where “the capacity maintains merely 83.9% after 6 cycles”).

Moreover, Plaintiff’s argument that “secondary lithium ion battery” need not be construed is contrary to Patentees’ positions before the USPTO. For example, to overcome § 103 rejections over a combination of Idota with Tanaka and Idota with

¹¹ Plaintiff does not dispute that the battery be made without substantially decomposing the electrolyte.

1 Kobayashi, Patentees argued that either of these combinations would have resulted in a
 2 “lithium ion battery with very poor cycleability.” Apple’s Br., Ex. B at 51. Patentees
 3 clarified that “[t]hose in the battery field would not categorize such a battery as a
 4 ‘secondary’ type cell, since *that appellation denotes a battery that can be recharged to*
 5 *capacities relatively close to the original capacity.*” *Id.* Though Patentees attempt to
 6 step back from their own statements by arguing their statements referenced “secondary
 7 batteries” and not “primary batteries” (Pl.’s Br. 20), Patentees cannot show why their
 8 statements regarding “secondary batteries” should not apply to claim 5’s recitation of
 9 just that: “A secondary lithium ion cell or battery.”

10 Lastly, Plaintiff argues this requirement is “scientifically impossible.” However,
 11 as Dr. Horn explains and was known to a POSITA, batteries *can* be recharged to 100%
 12 capacity (and actually increase in capacity) after the first few cycles. *See* Horn Decl.,
 13 ¶¶ 25–26. Regardless, the prosecution history makes clear that this is in fact what
 14 Patentees claimed. Indeed, Plaintiff repeatedly told the USPTO that its invention
 15 allowed for battery performance in a “manner wholly unforeseen by the art,” and they
 16 may not now retract statements made to obtain the Asserted Patent. *See* Apple’s Br.,
 17 Ex. B at 83 (“[I]n a manner wholly unforeseen by the art, Applicant’s novel
 18 methodology did not diminish the recycle property of the secondary lithium ion
 19 battery.”); *id.* at 48 (distinguishing the prior art by arguing “no one ... could have
 20 ‘readily anticipated’ ... a secondary lithium ion battery free of poor recyclability and
 21 electrolyte breakdown” and pointing to publications of record “which indicate[] that the
 22 anticipated result of the claimed invention would have been ... a battery with poor
 23 recyclability and/or significant electrolyte breakdown”). Patentees’ statements reflect
 24 a clear and effective disclaimer with respect to the improved cyclability over the prior
 25 art. *See Data Engine Techs. LLC v. Google LLC*, 10 F.4th 1375, 1383 (Fed. Cir. 2021)
 26 (limiting “patentees to distinguishing statements made during prosecution even if they
 27 said more than needed to overcome a prior art rejection”).

1 **Second**, Plaintiff’s argument that the term need not be limited to “electrode
 2 materials not [] substantially changed” is similarly unpersuasive. Plaintiff argues the
 3 patent specification “specifically contemplates after arising technology,” but Patentees’
 4 brief statement that “the inventors believe that the method can still be used to positive
 5 and negative electrode materials having higher specific energy developed in the future”
 6 contemplates Patentees did not possess such materials at the time of their purported
 7 invention, and is thus insufficient to justify broadening claim scope. ’641 Patent, 7:12–
 8 15. Moreover, during prosecution, Patentees argued for a narrowed construction of the
 9 term by repeatedly highlighting that they “discovered an approach for raising the cut-
 10 off voltage above 4.2 V ***without substantially changing the electrode materials.***”
 11 Apple’s Br., Ex. B at 83. Patentees further distinguished the claimed invention from
 12 the prior art by emphasizing that ***known*** electrode materials could be used at voltages
 13 above 4.2 V without negatively impacting cycle life. *See id.* at 47 (“[T]he claimed
 14 invention contravened the understanding and expectations of the prior art and, hence,
 15 would have been wholly surprising to one of ordinary skill.”); *id.* at 145 (not disputing
 16 that the “present application utilize the same anode active material and cathode active
 17 material”).

18 Further, the specification explains that the positive and negative electrode
 19 materials can be created using conventional methods. *See* ’641 Patent, 8:37–41 (“[T]he
 20 present invention can use ***conventional*** conducting agent and binder, and the mixture
 21 ratio for each component[] in the positive electrode active material can be those well
 22 known in the art.”); *id.* 9:5–7 (“The mesophane asphalt based carbon fiber or mesophane
 23 asphalt based spherical carbon can be obtained according to the conventional method.”).
 24 Patentees’ statements make clear that the alleged invention was limited to electrode
 25 materials that were substantially similar to those used at the time of the invention. *See*
 26 *Kopykake Enterprises, Inc. v. Lucks Co.*, 264 F.3d 1377, 1380, 1383 (Fed. Cir. 2001)
 27 (construing term based on conventional processes at the time); *Steuben Foods, Inc. v.*

Oystar USA, Inc., 520 F.Supp.3d 310, 323 (W.D.N.Y. 2021) (limiting definition to that at time of filing); *Fundamental Innovation Sys. Int’l LLC v. Samsung Elecs. Co.*, No. 2:17-CV-145-JRG-RSP, 2018 WL 647734, at *11 (E.D. Tex. Jan. 31, 2018) (limiting term to existing standards).

D. “overcharging protection [...] voltage” terms

Term / Claims	Apple’s Proposal	Plaintiff’s Proposal
“overcharging protection voltage” (claim 9) / “overcharging protection release voltage” (claim 9) / “overcharge protection release voltage” (claim 10)	Overcharging protection voltage, implemented in the secondary lithium ion cell or battery, and not in a protection circuit.	No construction needed. Alternatively, plain and ordinary meaning: the voltage level at which the battery’s protection mechanism(s) engages to prevent the battery from being charged to a higher voltage.

Apple’s proposed construction for “overcharging protection” is consistent with Patentees’ elections made during prosecution, in response to restriction requirements.

Plaintiff agrees that “[o]vercharge protection can be implemented in the battery itself, such as in the battery management system or related circuitry, but it may also be implemented in software or firmware that operates on the battery. It may also be implemented in a protection circuit.” Pl.’s Br. 22 (internal citations omitted); *see also* Horn Decl., ¶ 28. Plaintiff does not dispute that during prosecution, Patentees complied with a restriction requirement, and did not elect the invention claiming protection circuitry. Thus, the question to be decided is whether the disclaimed protection circuitry should be excluded from the scope of the claim. It should.

Plaintiff argues that Patentees’ restriction election “is not entitled to ‘much weight’ here because the examiner did not construe or limit the term.” Pl.’s Br. 22–23 (citing *Honeywell Int’l, Inc. v. ITT Indus., Inc.*, 452 F.3d 1312, 1319 (Fed. Cir. 2006)). However, it is black-letter law that “[c]laims to the non-elected invention(s) are withdrawn from consideration and must be canceled before the application is allowed to issue as a patent.” *Helifix Ltd. v. Blok-Lok, Ltd.*, 208 F.3d 1339, 1348 (Fed. Cir.

2000). Thus, it would be antithetical to the restriction requirement to permit a patentee to disclaim an invention, then later construe a term in the issued patent to recapture the disclaimed invention.

Plaintiff's cited cases are inapposite. **First**, Plaintiff cites *Dynamic* to argue that a restriction requirement election does not necessarily imply a waiver of claim scope. Pl.'s Br. 22 (citing *Dynamic Digital Depth Rsch. PTY LTD v. LG Elecs., Inc.*, No. CV 15-05578 GW(EX), 2016 WL 7444569, at *7 (C.D. Cal. Nov. 7, 2016)). But *Dynamic* noted that "a restriction requirement election does not necessarily imply a disclaimer of claim scope" **if the election is unrelated to the claim language**. *Dynamic*, 2016 WL 7444569, at *25, n. 3 (citing *Plantronics, Inc. v. Aliph, Inc.*, 724 F.3d 1343, 1350–51 (Fed. Cir. 2013) (holding the election of certain concha stabilizer species does not imply a disclaimer of structure)). To the contrary here, Plaintiff's election is directly tied to the overcharging protection voltage claim language, because Plaintiff did not elect coverage for protection circuitry. Thus, Plaintiff cannot backdoor into the scope of the claims the protection circuitry.

Second, in *Honeywell*, the court did "not assign much weight" to a restriction issued in a divisional's parent application, when determining the divisional's claim construction.¹² Here, the Patentees (who never filed any divisional) were issued a restriction requirement during the '641 Patent's prosecution, and their election expressly withdrew "a protection circuit."

Plaintiff's last argument is that there is a "strong presumption created by the

¹² In *Honeywell*, the examiner issued restriction requirement because application contained three distinct inventions: "(1) a method for preventing breakdown of a fuel filter, (2) the fuel filter itself, and (3) a moldable polymeric material." *Id.* at 1314. "[T]he patentee chose the first invention" and filed a divisional application on a "fuel injection system component." *Id.* at 1315. Since the "only fuel component disclosed and claim in the patent was a fuel filter," the court disregarded the examiners restriction requirement in the parent application to claims for a "fuel filter" and a "fuel system component." *Id.* at 1318–19.

specification’s express preservation of scope in the embodiments,” presumably to argue that because the specification references protection circuitry, Plaintiff can claim protection circuitry. But this argument completely disregards Plaintiff’s election of an invention that was *not* protection circuitry. And Plaintiff’s own authority confirms its argument is improper: in *Dynamic*, the court “agree[d] that portions of the specification that refer to claims that have been removed as a result of a restriction requirement election *cannot properly be relied upon to support an interpretation of claims.*” *Dynamic Digital Depth Rsch. PTY LTD*, 2016 WL 7444569, at *7. Here, Patentees filed the specification with their original claims, including those covering protection circuitry. After Patentees did not elect the protection circuitry invention during prosecution, Patentees did not remove those references from the specification. Thus, Plaintiff cannot now seek to rely on the specification, to broaden their claims to recapture the protection circuitry invention they did not elect.

E. “maintains at least ... capacity” terms

Term / Claims	Apple’s Proposal	Plaintiff’s Proposal
“maintains at least 7500 of capacity after 400 cycles” (claim 11) / “maintains at least 75.83% of capacity after 400 cycles” (claim 12) / “maintains at least 80% of capacity after 400 cycles” (claim 13)/ “maintains at least 80.72% of capacity after 400 cycles” (claim 14)	Indefinite.	No construction needed. Alternatively, plain and ordinary meaning: maintains at least [75% / 75.83% / 80% / 80.72%] of capacity after 400 complete charge and discharge cycles of the battery.

Plaintiff’s Opening Brief confirms the two reasons why these terms are indefinite: (1) a POSITA would not have understood what “capacity” is used to calculate the claimed percentage of retained capacity; and (2) the ’641 Patent fails to teach a POSITA what parameters are used to measure the claimed “capacity.”

First, the terms are indefinite because a POSITA would not understand which

“capacity” is claimed. Plaintiff’s Opening Brief misses the mark—the indefiniteness stems not from how to calculate a percentage, but rather what values are used in order to arrive at the claimed percentage. As discussed in Apple’s Opening Brief and above, claims 1 and 5 of the ’641 Patent recite “specific capacity” and “theoretic capacity,” respectively, and therefore are “presumed to have different meanings.” *Bd. of Regents of the Univ. of Tex. Sys.*, 533 F.3d at 1371. Plaintiff’s Opening Brief not only fails to address this ambiguity, but also introduces **additional** ambiguities by referring to additional types of “capacity.” Plaintiff and its expert allege that a POSITA would have understood the percentage of the claimed “capacity” is calculated by dividing the “remaining capacity” by the “initial capacity.” Pl.’s Br. 23; Yao Decl., ¶ 72. However, the ’641 Patent **does not once** mention “initial capacity” or “remaining capacity,” nor does Plaintiff define these terms or how to measure them. Horn Decl., ¶ 30. A POSITA would not understand whether the “initial capacity” used in Plaintiff’s calculation, is the theoretic capacity (such that all active materials participate in the reaction) or instead the actual (or specific) capacity. *Id.* ¶ 30. Moreover, as discussed in Apple’s Opening brief, even if these claims did refer to the theoretic capacity because they depend from claim 5, the requirement that the battery maintain at least a certain percentage of capacity is indefinite because “theoretic capacity” assumes all of the material is engaged. *Id.*

Second, these terms are likewise indefinite because a POSITA would not understand what parameters are used to measure the claimed percent capacity maintained. Horn Decl., ¶ 31. As discussed in Apple’s Opening Brief and the accompanying declaration of Dr. Horn, parameters such as the cathode and anode materials, testing conditions, cycling conditions, rest time between charge and discharge, and temperature all impact a given capacity measurement. In fact, Plaintiff’s Opening Brief similarly admits that “[t]o perform such balancing, the manufacturer will carefully design and measure the mass, area, volume, and/or capacity of the active

1 material of each electrode.” Pl.’s Br. 6.

2 Plaintiff’s reliance on the “examples” in the specification do not save its position.
3 Plaintiff’s first citation discusses the “capacity” of lithium cobalt oxides “*reach[ing]*
4 *159 mAh/g* where the charge cut-off voltage is 4.35V, but it drops to 135 mAh/g after
5 several cycles, *and the capacity attenuates quickly when the charge cut-off voltage is*
6 *4.25V.*” ’641 Patent, 2:22–25. As an initial matter, the specification describes that the
7 capacity changes considerably based on what charge cut-off voltage is applied,
8 however, it does not describe the testing conditions, such as temperature applied
9 throughout the “several cycles.” Horn Decl., ¶ 32. Plaintiff’s second citation states
10 that “[a]fter formation and test using a charge cut-off voltage of 4.2 V, these cells have
11 an *arithmetic mean capacity* of 1113 mAh, a weight specific energy of 102 Wh/kg, an
12 average operating voltage of 3.70 V, and maintains 85.72% of capacity after 400
13 recycles.” ’641 Patent, 10:34–37. The specification again does not refer to an initial or
14 remaining capacity, but rather “an arithmetic mean capacity,” and does not describe the
15 testing conditions, such as temperature applied throughout the “400 cycles.” Horn
16 Decl., ¶ 32.

Moreover, Plaintiff's reliance on the Handbook of Batteries by Linden for the "Capacity Fade" principle further exemplifies why these terms are indefinite. Indeed, Plaintiff's own citations *explicitly* show the differing capacities based on different parameters applied—including the temperature variance. Horn Decl., ¶ 33. For example, Linden expressly describes the difference in a "fade rate," which changes with temperature. Horn Decl., ¶ 33. Specifically, "[a]t 21°C, the batteries had a fade rate of 0.024%/cycle, while the battery at 45 °C had a fade rate of 0.028%/cycle. Thus after 500 cycles, the battery cycled at the 1C rate **at 21 °C delivered 1.42 Ah, 91% of the initial value**, and the one cycled **at 45 °C** at the 1C rate **delivered 1.35 Ah on the 500th cycle, or 87% of the initial value.**" Plaintiff's Ex. 6 (Handbook of Batteries), at 35.42.

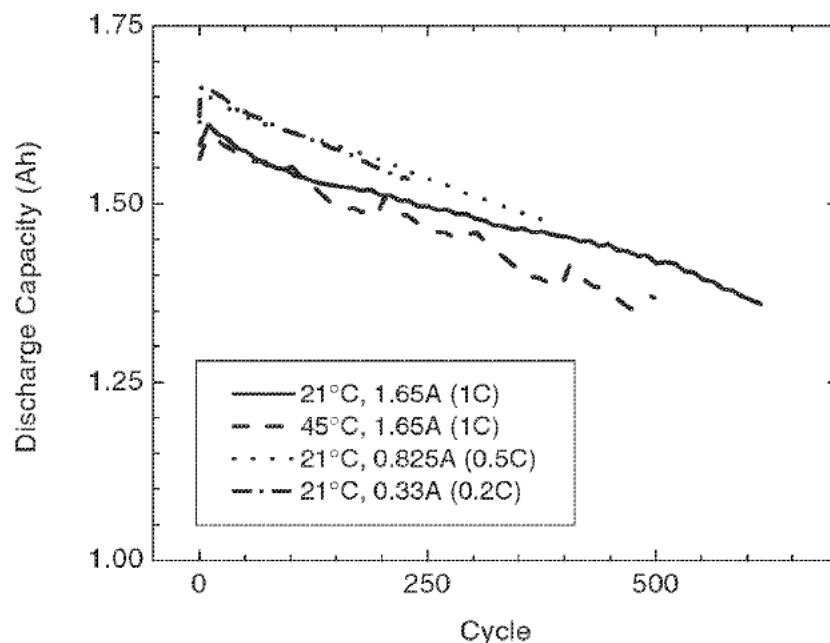


FIGURE 35.45 Capacity of 18650-type C/LiCoO₂ batteries when cycled at either 21°C or 45°C at rates from 0.2C (0.33 A) to 1.0C (1.65 A) between 4.2 V and 2.5 V. (Courtesy of NEC Moli Energy.)

Id. Thus, whether the battery is cycled at a given temperature would provide different results and could ultimately determine whether a given product meets the claimed percent capacity maintained. Horn Decl., ¶¶ 33–36.

Indeed, the specification discloses that if a battery is cycled at 45 °C, it may not “maintain[] at least 80.72% of capacity after 400 cycles,” but if that very same battery

1 is cycled at 0 °C, it may “maintain[] at least 80.72% of capacity after 400 cycles.”
 2 Because the specification provides no way for a POSITA to determine what parameters
 3 (including the cycling temperature) to apply, the terms are indefinite. *See Saso Golf,*
 4 *Inc. v. Nike, Inc.*, 843 F. App’x 291, 296 (Fed. Cir. 2021) (“[O]ur precedents indicate
 5 that ‘a claim may be invalid as indefinite when (1) different known methods exist for
 6 calculating a claimed parameter, (2) nothing in the record suggests using one method in
 7 particular, and (3) application of the different methods result in materially different
 8 outcomes for the claim’s scope.’”) (quoting *Ball Metal Beverage Container Corp. v.*
 9 *Crown Packaging Tech., Inc.*, 838 F. App’x 538, 542 (Fed. Cir. 2020)); *Pac. Coast*
 10 *Bldg. Prod., Inc. v. CertainTeed Gypsum, Inc.*, 816 F. App’x 454, 460 (Fed. Cir. 2020)
 11 (finding term indefinite where POSITA would have no reasonable certainty in
 12 calculating value based on availability of various choices).

13 Therefore, these terms are indefinite because (1) it is unclear what “capacity” is
 14 used, and (2) a POSITA would have no way to determine from the intrinsic record what
 15 combination of parameters to apply during the 400 cycles to determine whether the
 16 claimed percent capacity is maintained. Horn Decl., ¶ 36.

17 **III. CONCLUSION**

18 For the foregoing reasons, Apple respectfully requests that the Court find
 19 indefinite two terms, and adopt Apple’s proposed constructions for the remaining terms.
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1 DATED: July 14, 2023

Respectfully submitted,

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CERTIFICATION PER LOCAL RULE 11-6.2

The undersigned counsel of record for Defendant Apple, Inc. certifies that this brief contains 6,993 words, as calculated by the limitations in L.R. 11-6.1.

DATED: July 14, 2023

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**UNITED STATES DISTRICT COURT
 CENTRAL DISTRICT OF CALIFORNIA**

RJ TECHNOLOGY LLC,	)	CASE NO. 8:22-CV-1874-JVS-JDE
	)	
Plaintiff,	)	DECLARATION OF QUINN
	)	HORN, PH.D. IN SUPPORT OF
v.	)	DEFENDANT APPLE INC.'S
	)	RESPONSIVE CLAIM
APPLE INC.,	)	CONSTRUCTION BRIEF
	)	
Defendant.	)	Date: July 31, 2023
	)	Time: 3:00 p.m.
	)	Ctrm: 10C
	)	Judge: Hon. James V. Selna
	)	
	)	Second Amended Complaint Filed:
	)	April 13, 2023

1 I, Quinn Horn, do hereby declare as follows:

2 **I. INTRODUCTION**

3 1. I previously submitted a declaration in support of Apple Inc.’s (“Apple”) opening claim construction brief (“Opening Declaration”). See Dkt. 68-1. I hereby
4 incorporate by reference my previous declaration. *Id.*

6 **II. MATERIALS CONSIDERED**

7 2. In addition to the materials considered in my previous declaration, I have
8 also considered the following:

- 9 • The parties’ opening claim construction and related declarations
10 for certain terms in these patents, as well as the extrinsic evidence
11 identified by the parties therein;
- 12 • Dr. Yao’s declaration in support of Plaintiff’s opening brief; and
- 13 • Any other material cited or referred to in this declaration.

14 3. My opinions in this declaration are based upon my background knowledge
15 and experience relating to the topics discussed in my declaration, as well as the materials
16 that I have considered in making this declaration.

17 4. I reserve the right to supplement or amend this declaration in the event that
18 I hereafter become aware of additional relevant information, including applicable
19 rulings by the Court, and opinions expressed by the Plaintiff or its experts. I further
20 reserve the right to submit a declaration rebutting claim construction opinions that may
21 be expressed by others in this case.

22 **III. CLAIM CONSTRUCTION OPINIONS**

23 5. I understand that RJ Technology LLC asserts claims 5–14 of the ’641
24 Patent (the “Asserted Claims”).

25 6. I understand that the parties have identified certain claim terms for
26 construction and submitted opening claim construction briefs. Therefore, I have limited
27 my analysis to just these terms. I reserve the right to opine on other terms should the
28 need arise.

1 **A. “a charge cut-off voltage” (Claims 5–7)**

2 7. As I explained in my Opening Declaration, in my opinion a person of
3 ordinary skill in the art (“POSITA”) reading the ’641 Patent in light of the specification
4 and file history would have understood the claim term “a charge cut-off voltage” to
5 mean: “the voltage at which the charging changes from constant current charging to
6 constant voltage charging.”

7 8. I understand that Plaintiff frames the dispute as “whether the patent limits
8 the definition of charge cutoff voltage to this one specific algorithm through which it is
9 implemented (Apple’s construction, CC/CV), or whether it incorporates the full plain
10 and ordinary meaning of the term (RJ Technology’s construction).” Dkt. 70 (Pl. Brief)
11 at 8. However, many variables affect whether any given charging method would meet
12 either party’s construction. For example, various types of charging methods can be
13 used in combination with each other. Indeed, CC/CV charging is simply a combination
14 of constant current and constant voltage charging.

15 9. A POSITA would understand that any of the “charging algorithms”
16 identified by Plaintiff (*see* Dkt. 70 (Pl. Brief) at 6–7) could be used in any combination
17 with one another. Thus, a POSITA would understand that Apple’s construction (“the
18 voltage at which the charging changes from constant current charging to constant
19 voltage charging”) does not necessarily exclude the use of other charging methods.

20 10. Moreover, as I explained in my Opening Declaration, the intrinsic
21 evidence supports Apple’s proposed construction. Consistent with what was well-
22 understood in the art at the time of the invention, the Patentee in the related CN
23 Invalidity Proceedings confirmed this understanding that: “[t]his technical term is a
24 standard general term in the field of battery related technology, and its meaning is
25 determined, which refers to *the voltage value of the battery when it transitions from*
26 *charging at a constant current to charging at a constant voltage.*” Dkt. 69, Ex. D (The
27 Patentees’ Response to Invalidation Request re China National Intellectual Property
28 Administration, Case No. 4W107406, dated August 7, 2018) at 325.

11. Additionally, CC/CV charging was and continues to be a common and preferred method to charge a lithium-ion battery. CC/CV charging is very simple to implement and allows the battery to be charged to as high a capacity as possible without pushing the battery to damaging voltages. For example, CC/CV is preferred over constant-current (CC) alone because for a lithium-ion battery to be fully charged under CC, the current applied must remain a low value, and would take much longer to charge than CC/CV to an equivalent state of charge. Additionally, CC/CV is preferred over constant-voltage (CV) only because CV would not limit the amount of current that the battery will allow, which could damage the battery as it approaches unsafe temperatures, and also damage the cyclability. Finally, purely CV charging requires a power supply capable of generating extremely high currents when first starting to charge a fully-discharge battery. In practice, any charger configured as a purely CV charger would have an effective constant-current limit based on the physical limitations of the charger to deliver power.

12. Plaintiff's construction that "a charge cut-off voltage" means "the maximum voltage to which the battery is configured to be charged" introduces additional ambiguity as to the "maximum" and what "configured to be charged" means. Plaintiff and Dr. Yao's interpretation that the "maximum" that the battery is "configured to be charged" means the "maximum" that the battery is "designed and manufactured" to be charged introduces ambiguity. *See* Dkt. 70-1 ("Yao Decl.") ¶ 28. For example, while the cell designer may find the cell should not be charged past a certain voltage, the battery designer may find the battery should not be charged past a different voltage level. Moreover, Plaintiff's interpretation improperly implicates the intent of the designers rather than the physical properties of the battery itself.

B. "ratio ... as calculated by a theoretic capacity"

13. As I explained in my Opening Declaration, in my opinion, if the term is not found to be indefinite, a POSITA would understand that the term should mean "the ratio calculated as the product of the theoretic capacity of the cathode active material

1 per mass unit and the amount of the cathode active material in the same mass unit
2 divided by the product of the capacity of the anode active material per mass unit and
3 the amount of the anode active material in the same mass unit, when the cell charge
4 cutoff voltage is set at 4.2 V, wherein ‘theoretic capacity’ is voltage independent and
5 means ‘theoretical, rather than actual, capacity that assumes all of the active substances
6 participate in the reaction of the battery.’”

7 14. I understand that Plaintiff’s position is that the definition of “theoretic
8 capacity” used to calculate the ratio should be expanded to include all forms of
9 “capacity.” I disagree. In my opinion, a POSITA would understand that this claim term
10 (theoretic capacity) refers to the theoretical capacity, rather than actual capacity, and
11 assumes all of the active substantives participate in the reaction of the battery. Thus, a
12 POSITA would not expand this term to include, for example, a specific capacity
13 calculated from actively participating substances under specific conditions.

14 15. In my opinion, Plaintiff and Dr. Yao incorrectly conflate the two distinct
15 measures of capacity: specific capacity and theoretical capacity. For example, Dr. Yao
16 explicitly uses “specific capacity” when defining claim 5’s ratio calculation. *See* Yao
17 Decl., ¶ 38 (“where C_p is the specific capacity of the cathode active material... C_n is the
18 specific capacity of the anode active material....”), ¶ 39 (“The ratio refers to a ratio
19 ***based on specific capacity.***”) (emphasis added). A POSITA would understand that
20 claim 5 clearly references a distinct “theoretic capacity” separate from claim 1’s
21 “specific capacity,” and the ’641 Patent’s specification continuously distinguishes
22 between these two capacities. In my opinion, a POSITA would understand that
23 “specific capacity” refers to the capacity of the actively participating materials under
24 specific conditions (such as a specific charge cut-off voltage), whereas the theoretic
25 capacity is calculated assuming that all the active substances are utilized in the battery’s
26 reactions.

27 16. This understanding is confirmed by the ’641 Patent’s specification, which
28 distinguishes “theoretical capacity” from “actually used capacity.” *See* ’641 Patent,

1 1:40–43. Moreover, Dr. Yao appears to concede that actual capacity refers to specific
2 capacity. *See* Yao Decl., ¶ 50 (“Such specific capacity is also closer to the actually
3 usable capacity, so it acts as a good reference point to determine how to balance the
4 capacities of cathode and anode in a battery.”). Under Plaintiff’s construction, however,
5 there is no difference between theoretic capacity and specific capacity. Thus, a POSITA
6 would not understand Plaintiff’s and Dr. Yao’s substitution of “specific capacity” for
7 “theoretic capacity.”

8 17. Additionally, Plaintiff and Dr. Yao use many terms to reference capacity,
9 including (1) theoretic capacity; (2) theoretic maximum capacity; (3) specific capacity;
10 (4) maximum specific capacity; (5) initial capacity; and (6) remaining capacity. In my
11 opinion, a POSITA would not understand what these capacities mean in context of
12 the ’641 Patent, and specifically in context of claims 1 and 5. While Plaintiff and Dr.
13 Yao assert that Apple’s proposed construction of “theoretic capacity” requiring all
14 active materials participate in the reaction is allegedly a different concept, the “theoretic
15 maximum capacity,” I disagree. A POSITA reading the ’641 Patent specification would
16 not understand what “theoretic maximum capacity” is—and nowhere in the ’641 Patent
17 specification does the term “theoretic maximum capacity” once appear. Rather, a
18 POSITA would understand that “theoretic capacity”—consistent with the ’641 Patent
19 specification—refers to a capacity that assumes the active substances are fully utilized
20 in the battery’s reactions.

21 18. This understanding is further confirmed by Patentees’ statements made
22 during CNIPA Case Nos. 4W107406 & 4W112700 (collectively, “CN Invalidation
23 Proceedings”). For example, the Patentees explained that “theoretical gram capacity”
24 (*i.e.*, theoretic capacity) assumes all active substances participate:

25 The amount of electricity (electricity capacity) given ***by assuming that all***
26 ***active substances participate in the reaction of the battery***. Specifically,
27 where the active substance is LiCoO_2 , the Faraday constant $F = 96485$
28 C/mol , and the molar mass of LiCoO_2 $M = 97.8698 \text{ g/mol}$. If all lithium
ions are de-intercalated, then $F/M = 985.8535 \text{ C/g}$, and after unit

1 conversion, **it is 273.848 mAh/g, which is the theoretical gram capacity.**
2 However, the actual capacity released by the battery is only a part of the
3 theoretical values mentioned above.
4 Dkt. 69, Ex. D at pdf. pg. 10; Plaintiff's and Dr. Yao's attempt to interchange "specific
5 capacity" and "theoretic capacity" do not agree with Patentees' statements above.

6 19. Moreover, Dr. Yao has similarly made statements in his own patent and
7 publications that contradict Plaintiff's assertion that the "theoretic capacity" should be
8 conflated with "specific capacity." For example, Dr. Yao's patents and publications
9 predating this litigation describe "specific" capacity as a percentage of "theoretic
10 capacity":

- 11 • "[T]he **specific capacity** of P(NDI2OD-TET) is 47.4 mAh g⁻¹, or **90% of its**
12 **theoretical capacity** (52.7 mAh g⁻¹)." Ex. 3 (Heavily n-Dopable
 π -Conjugated Redox Polymers with Ultrafast Energy Storage Capability) at
13 4957 (emphasis added).
- 14 • "A **specific capacity** of up to 350 mAh/g is obtained, which is **91% of the**
15 **theoretical capacity**". Ex. 4 (U.S. Patent No. 10,522,875) at 7:22-25
16 (emphasis added).

17 Therefore, Plaintiff's and Dr. Yao's attempt to interchange "specific capacity" and
18 "theoretic capacity" do not agree with Dr. Yao's own statements above. A POSITA
19 would agree with Dr. Yao's statements above.

20 20. Plaintiff and Dr. Yao also argue that the claimed "[t]heoretical capacity in
21 that context *must* refer to a value modified by the applicable x factor." Dkt. 70 (Pl.
22 Brief) at 17. However, a theoretical capacity that is modified by a charge cut-off voltage
23 refers to the *specific capacity*, not the theoretical capacity. For example, the '641 Patent
24 states that LiCoO₂ "has a theoretical capacity of 248 mAh/g, while the actually used
25 capacity [or specific capacity] of it is merely about 140 mAh/g, i.e., about half of said
26 theoretic capacity is not utilized." '641 Patent, 1:40–44. Indeed, while Plaintiff and its
27 expert reference Linden for the assertion that x=0.5 for LiCoO₂ for the theoretic
28 capacity, Linden later explicitly describes that the theoretic capacity of LiCoO₂ is **275**
mAh/g—i.e., no applicable x factor. See Pl.'s Ex. 6, at RJ00002798-Apple ("[t]he
reversible value of x for LiCoO₂ ... is approximately 0.5" and "[t]hus although the

1 *theoretical capacity of LiCoO₂ and LiNiO₂ (275 mAh/g)*) (emphasis added). The
2 “applicable x factor” is the fraction of all available of lithium ions that participate in the
3 reaction. Linden’s use of the word “approximately” and the note that “higher values
4 [than $x = 0.5$] may be obtained in practice” undermine Plaintiff’s assertions that this
5 represents a theoretical capacity. These qualifying terms suggest that these values are
6 empirical or rule-of-thumb estimates, not based in theory. Therefore, in my opinion, a
7 POSITA would understand that a capacity “modified by the applicable x factor” refers
8 to a specific capacity, rather than the claimed “theoretic capacity.”

9 21. Additionally, in my opinion, the term “charge cut-off voltage” refers to the
10 cell charge cutoff voltage. As Plaintiff and Dr. Yao admit “[e]ach electrode has a
11 different capacity,” which is separate from “[t]he total capacity of the battery.” Yao
12 Decl. at 7. In the industry, it is common that specific capacities of active materials are
13 described in half-cell voltages (i.e., voltages relative to a known, fixed reference
14 potential, such as Li/Li⁺). However, the ’641 Patent’s specification describes cell-level
15 voltages. Thus, the construction of “cell charge cutoff voltage” should be adopted to
16 clarify the claim does not recite a half-cell voltage.

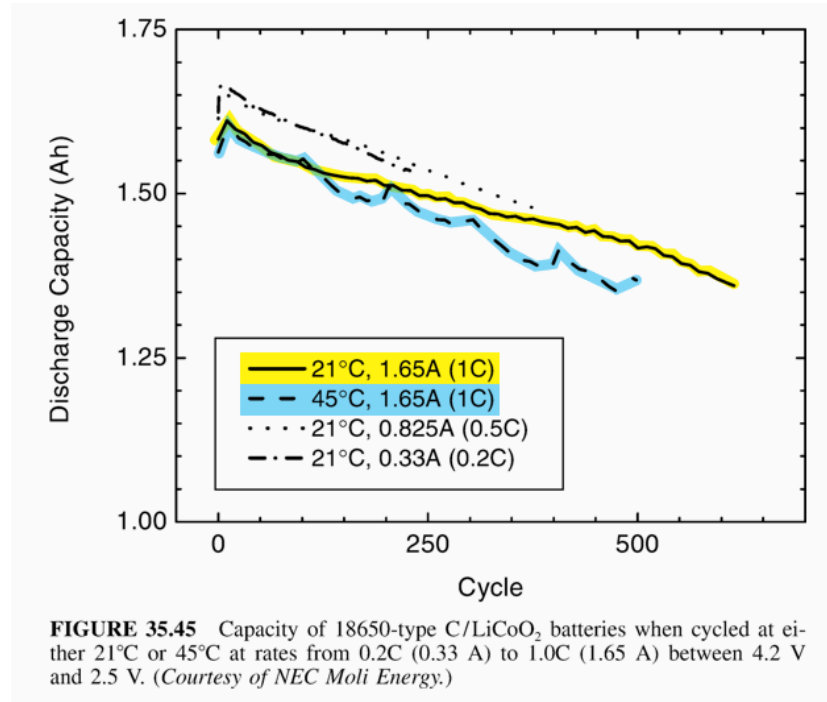
17 22. Moreover, while Apple’s proposed construction should be adopted if the
18 term is not found indefinite, in my opinion, the term is indefinite. As I explained in my
19 Opening Declaration, the term is indefinite because claim elements are rendered
20 meaningless by Plaintiff’s own proposed construction. If a POSITA understood the
21 claim to refer to theoretical capacity, a POSITA would assume all materials participate,
22 which would be independent of any charge cut-off voltage. Therefore, calculating a
23 theoretical capacity at a specific charge cut-off voltage would be nonsensical to a
24 POSITA. Additionally, as I previously discussed, the capacity at a particular charge
25 cut-off voltage refers to the *specific* capacity and not *theoretic* capacity. Therefore, in
26 my opinion, the claim fails to inform a POSITA with reasonable certainty about the
27 scope of the invention.

1 **C. “A secondary lithium ion cell or battery”**

2 23. As I explained in my Opening Declaration, in my opinion, “a secondary
3 lithium ion cell or battery” means “a lithium ion battery, wherein the battery electrode
4 materials must not be substantially changed from what was available in the art and made
5 without substantially decomposing the electrolyte, which can be recharged to its
6 original capacity, without any decreased capacity or cycle life at a charge cut-off voltage
7 above 4.2 V.”

8 24. I disagree with Plaintiff’s assertion that Apple’s proposed construction is
9 “scientifically impossible.” Dkt. 70 (Pl. Br.) at 19. Secondary lithium ion batteries *can*
10 be recharged to 100% capacity, but that recharge capacity may diminish after significant
11 use. Indeed, Plaintiff’s expert implicitly agrees that secondary lithium ion batteries can
12 be recharged to 100%: “[E]ven though secondary batteries may be recharged, they do
13 not *always* recharge to 100% of their original capacity. In fact, after enough use, no
14 battery can recharge to this ideal figure, and in many cases, there is irreversible capacity
15 loss after the very first charge.” Yao Decl., ¶ 59 (emphasis added). Thus, Dr. Yao
16 agrees that is is in fact scientifically possible that secondary batteries can be recharged,
17 for example after just one charge and discharge cycle, to 100% capacity.
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25. Indeed, this is consistent with the knowledge of a POSITA, as the below Figure 34.45 from Linden illustrates that the capacity can actually increase after the first few cycles before degrading:



Pl.'s Ex. 6, at RJ00002892_Apple (annotated).

26. Indeed, Dr. Yao's own patent shows the capacity of a secondary battery *increasing* after the first few cycles:

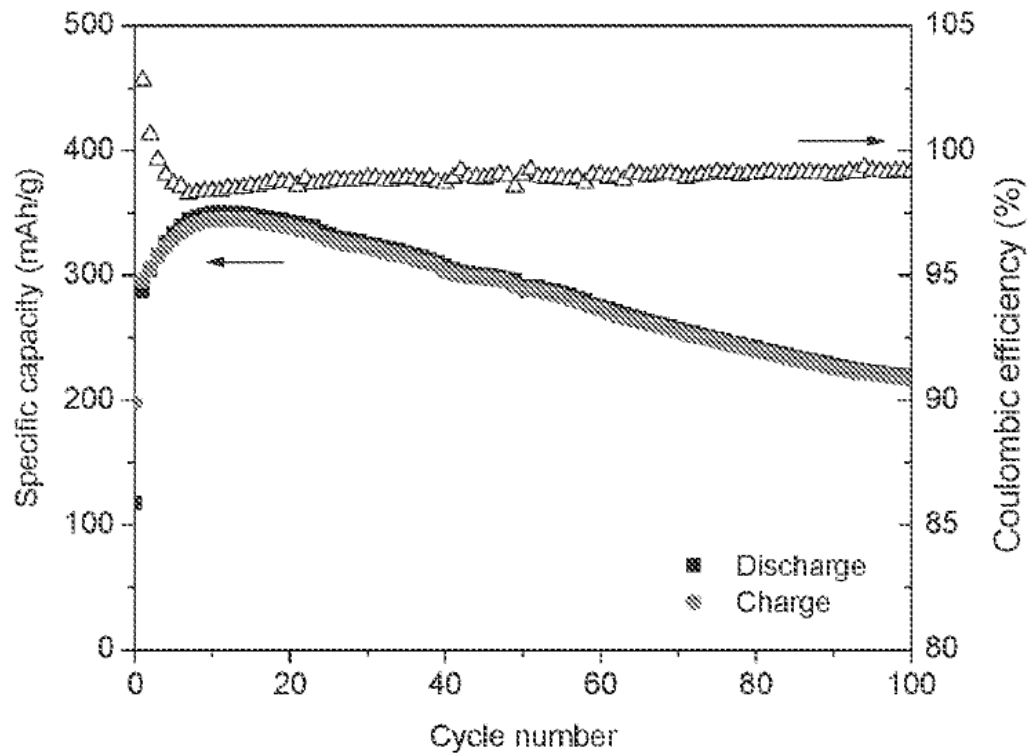


FIG. 7

Ex. 4 (U.S. Patent No. 10,522,875) at Fig. 7.

D. “overcharging protection [...] voltage” terms

27. As explained in my Opening Declaration, in my opinion, a POSITA reading the ’641 Patent in light of the specification and file history would have understood the claim term “overcharging protection voltage” to mean: “overcharging protection voltage, implemented in the secondary lithium ion cell or battery, and not in a protection circuit.”

28. Plaintiff and Dr. Yao concede that overcharge protection “can be implemented in the device (e.g., in circuitry or software) or in the battery itself (e.g., in the battery management system).” Yao Decl., ¶ 67; *see also* Dkt. 70 (Pl. Br.) at . 22. However, in my opinion, Plaintiff and Dr. Yao fail to account for Plaintiff’s restriction elections during prosecution such that the external protection circuitry cannot be within

1 the proper scope of the claim. Thus, there does not appear to be a dispute that the
2 overcharging protection voltage can be implemented within the secondary lithium ion
3 cell or battery, and Apple's proposed construction appropriately excludes
4 implementations of a separate protection circuit.

5 **E. "maintains at least ... capacity" terms**

6
7 29. As explained in my Opening Declaration, in my opinion, these terms are
8 indefinite. A POSITA would not understand these terms since (1) a POSITA would not
9 know what "capacity" is used to calculate the claimed percentage of retained capacity,
10 and (2) a POSITA reading the '641 Patent would not know what parameters to use to
11 measure the claimed "capacity."

12 30. As I explained above, Plaintiff and Dr. Yao add additional ambiguous
13 definitions of "capacity" beyond the claimed "specific" and "theoretic" capacity in
14 claims 1 and 5. For example, Plaintiff and Dr. Yao claim that a POSITA would have
15 understood the percentage of the claimed capacity is calculated by dividing the
16 "remaining capacity" by the "initial capacity." Dkt. 70 (Pl. Br.) at 23; Yao Decl. ¶ 74.
17 However, a POSITA would not understand what the "initial capacity" or remaining
18 capacity" used in Plaintiff's calculation is, as the '641 Patent does not mention these
19 terms and Plaintiff does not define these terms or how to measure them. In the context
20 of the '641 Patent, for example, the "initial capacity" could refer to either the theoretic
21 capacity (meaning all the active substances participate in the reaction) or instead the
22 actual (or specific) capacity. Assuming these terms referred to theoretic capacity, in my
23 opinion, a POSITA would not understand what a "remaining" theoretical capacity
24 means, since "theoretic capacity" assumes all of the material participates in the reaction.

25 31. Moreover, as I discussed in my Opening Declaration, a POSITA would not
26 understand what parameters are used to measure the claimed percent capacity
27 maintained. For example, parameters like the anode and cathode materials, cycling
28 conditions, testing conditions, rest time between charge and discharge, and temperature

1 would all impact a given capacity measurement. As Plaintiff recognized “[t]o perform
2 such balancing, the manufacturer will carefully design and measure the mass, area,
3 volume, and/or capacity of the active material of each electrode.” Pl.’s Br., at 6.

4 32. Plaintiff and Dr. Yao cite “examples” in the specification, which do not
5 clarify what the testing parameters were. For example, Plaintiff’s cite says “the capacity
6 of lithium cobalt oxides reaches 159 mAh/g when the charge cut-off voltage is 4.35 V,
7 but it drops to 135 mAh/g after several cycles; and the capacity attenuates quickly when
8 the charge cut-off voltage is 4.25 V.” ’641 Patent, 2:21–25. The ’641 Patent describes
9 that the capacity changes significantly based on the applied charge cut-off voltage, but
10 a POSITA would not understand what the testing conditions are. For example, a
11 POSITA would not know what temperature was applied throughout the “several
12 cycles,” which would significantly affect the capacity. Plaintiff’s other cite says that
13 “[a]fter formation and test using a charge cut-off voltage of 4.2 V, these cells have an
14 ***arithmetic mean capacity*** of 1113 mAh, a weight specific energy of 102 Wh/kg, an
15 average operating voltage of 3.70 V, and maintains 85.72% of capacity after 400
16 recycles.” ’641 Patent, 10:33–37. Again, the specification does not describe the testing
17 conditions, such as the temperature applied throughout the “400 cycles.” In both of these
18 citations, there is no reference to an initial or remaining capacity. *See e.g., id.* (referring
19 to “an arithmetic mean capacity”). Even if a POSITA understood what Plaintiff’s
20 “remaining capacity” or “initial capacity” meant (they wouldn’t), it would be unclear
21 what parameters were used to calculate these values.

22 33. Additionally, Plaintiff and Dr. Yao’s citations to Linden’s Handbook
23 further exemplifies why a POSITA would not understand what parameters are used to
24 calculate the remaining capacity after 400 cycles. As Plaintiff and Dr. Yao recognize,
25 Linden refers to “Capacity Fade,” describing loss of capacity after successive charging
26 cycles. *See* Dkt. 70 (Pl. Br.) at 24; Yao Decl., ¶ 76.

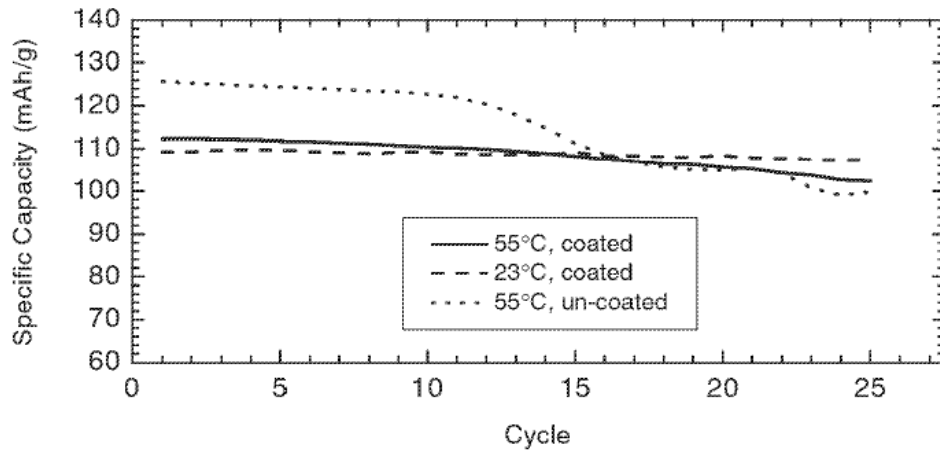


FIGURE 35.15 Specific capacity of LiCoO₂ coated and un-coated spinels cycled at the C/2 rate between 4.2 and 3.5 V at 23°C or 55°C. (Courtesy of Carus Chemical.)

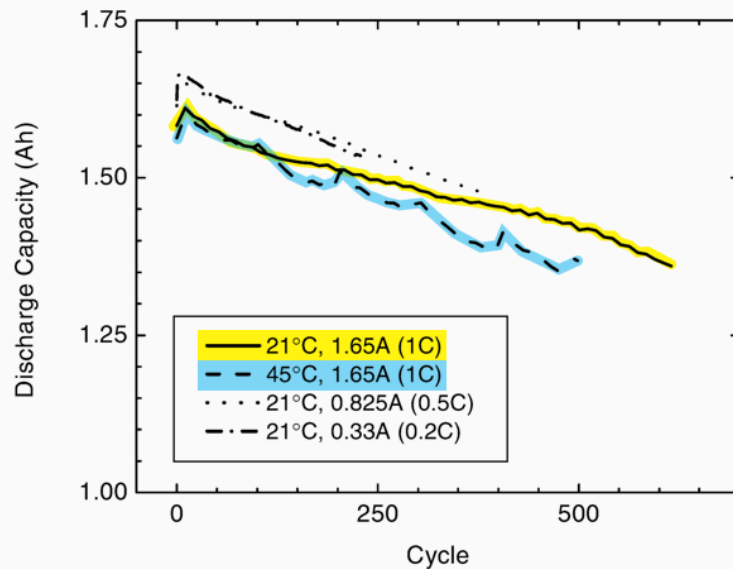


FIGURE 35.45 Capacity of 18650-type C/LiCoO₂ batteries when cycled at either 21°C or 45°C at rates from 0.2C (0.33 A) to 1.0C (1.65 A) between 4.2 V and 2.5 V. (Courtesy of NEC Moli Energy.)

Pl's Ex. 6, Figs. 35.15 (top) & 35.45 (bottom) (annotated).

34. However, Linden's Figs. 35.15 and 35.45 (including the key indicating different testing temperatures) show that the "Specific Capacity" and "Discharge Capacity" versus the "Cycle" change at *different temperatures*. For example, Linden explains "[a]t 21 °C, the batteries had a fade rate of 0.024%/cycle, while the battery at 45 °C had a fade rate of 0.028%/cycle. Thus after 500 cycles, the battery cycled at the 1C rate at 21 °C delivered 1.42 Ah, 91% of the initial value, and the one cycled at 45

1 °C at the 1C rate delivered 1.35 Ah on the 500th cycle, or 87% of the initial value.”
2 Plaintiff’s Ex. 6 (Handbook of Batteries), at 35.42 (emphasis added).

3 35. Therefore, these citations further exemplify why a POSITA would need to
4 know what parameters are applied in order to calculate how much capacity is retained
5 after discharge. Specifically, a POSITA would know that whether the battery is cycled
6 at a given temperature would provide different results, which could ultimately
7 determine whether a given battery meets the claimed percent capacity maintained.

8 36. Therefore, a POSITA would understand that any capacity calculations
9 after 400 cycles would have different results depending on parameters applied,
10 including the temperature. Because a POSITA would not know what temperature to
11 perform the tests at, how long to rest in between cycles, or whether the claimed
12 percentages of capacity retained are averages over a number of tests, or whether they
13 are the result of individual tests, a POSITA would not understand with reasonable
14 certainty the scope of the claimed invention.

15
16 I declare under penalty of perjury under the laws of the United States of America
17 that the foregoing is true and correct and that this declaration was executed on July 14,
18 2023.

19
20 

21 _____
22 Quinn Horn, Ph.D.
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EXHIBIT 3



Heavily n-Dopable π -Conjugated Redox Polymers with Ultrafast Energy Storage Capability

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Supporting Information

ABSTRACT: We report here the first successful demonstration of a “ π -conjugated redox polymer” simultaneously featuring a π -conjugated backbone and integrated redox sites, which can be stably and reversibly n-doped to a high doping level of 2.0 with significantly enhanced electronic conductivity. The properties of such a heavily n-dopable polymer, poly{[N,N'-bis(2-octyldodecyl)-1,4,5,8-naphthalenedicarboximide-2,6-diyl]-*alt*-5,5'-(2,2'-bithiophene)} (P(NDI2OD-T2)), were compared *vis-à-vis* to those of the corresponding backbone-insulated poly{[N,N'-bis(2-octyldodecyl)-1,4,5,8-naphthalenedicarboximide-2,6-diyl]-*alt*-5,5'-(2,2'-(1,2-ethanediyl)bithiophene)} (P(NDI2OD-TET)). When evaluated as a charge storage material for rechargeable Li batteries, P(NDI2OD-T2) delivers 95% of its theoretical capacity at a high rate of 100C (72 s per charge–discharge cycle) under practical measurement conditions as well as 96% capacity retention after 3000 cycles of deep discharge–charge. Electrochemical, impedance, and charge-transport measurements unambiguously demonstrate that the ultrafast electrode kinetics of P(NDI2OD-T2) are attributed to the high electronic conductivity of the polymer in the heavily n-doped state.

Organic π -conjugated polymers are emerging as a materials class for energy-related applications enabling a path to a more sustainable energy landscape without the need of energy-intensive, expensive, and sometimes toxic metal-based compounds.¹ Furthermore, the possibility to fabricate lightweight and mechanically flexible devices makes polymeric materials even more attractive. Hole-transporting (semi)conducting polymers with substantial redox activity and electronic conductivity have been long recognized as electrode materials for batteries, supercapacitors, and thermoelectrics.² However, all-polymer devices of this type have been difficult to realize due to the limitations of electron-transporting polymers.³ Two general classes of electron-transporting polymers are known: π -conjugated polymers and nonconjugated redox polymers.^{2a} Electron-transporting (or n-type) π -conjugated polymers typically need multiple repeating units in the backbone to stabilize injected electrons. Thus, the inability to stably and reversibly store these charges limits the n-doping level,⁴ which in turn reduces the amount of free electrons and therefore restricts the electronic conductivity. On the other hand, nonconjugated redox polymers have dedicated redox active

sites accepting one or more electrons per repeating unit⁵ but lack a π -conjugated backbone as found in π -conjugated polymers, which is crucial for efficient electron conduction. Since the advantages of the two classes of polymers perfectly complement each other, the weakness of both polymers could be addressed by a rational combination of the characteristics from each class, such as a “ π -conjugated redox polymer” (Figure 1a). Pioneering attempts to construct such a polymer

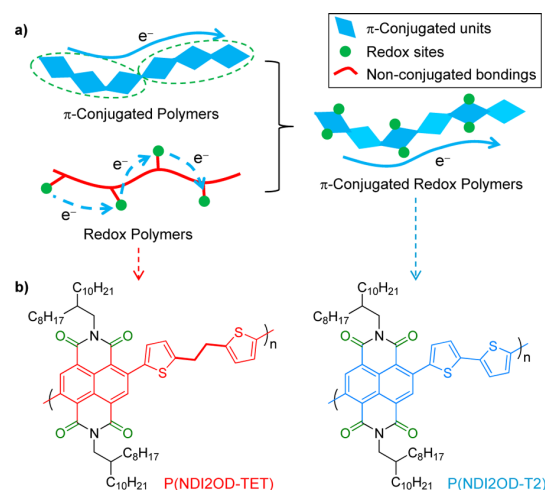


Figure 1. (a) Graphical illustration of the structural characteristics of π -conjugated polymers, redox polymers, and π -conjugated redox polymers. (b) Molecular structure of the nonconjugated P(NDI2OD-TET) and the π -conjugated P(NDI2OD-T2).

were limited by insufficient reversibility of the doping process,⁶ unknown doping level,^{6b,c,7} and/or uninvestigated/poor conductivity of the n-doped state.^{6,7} Thus, it remains a major challenge to develop heavily n-dopable π -conjugated polymers with high electronic conductivity.

Herein we report the first demonstration of a π -conjugated redox polymer in poly{[N,N'-bis(2-octyldodecyl)-1,4,5,8-naphthalenedicarboximide-2,6-diyl]-*alt*-5,5'-(2,2'-bithiophene)} (P(NDI2OD-T2)), which can be stably and reversibly n-doped to a high doping level (2.0) and exhibits significant electronic

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conductivity, enabling the realization of ultrafast rechargeable Li batteries (Figure 1b). P(NDI2OD-T2) is an electron-transporting π -conjugated polymer extensively studied in organic electronics, but only in its intrinsic and slightly n-doped states (doping level ≤ 0.1).⁸ Our results demonstrate that P(NDI2OD-T2) meets key criteria for an efficient π -conjugated redox polymer: a linear π -conjugated alternating naphthalene–bithiophene backbone providing an electron-transport pathway and a naphthalene dicarboximide (NDI) unit, which is a highly reversible two-electron acceptor,^{5b} ensuring a high n-doping level. The advantage of a π -conjugated redox polymer as an energy storage material over a nonconjugated one is highlighted by the design of a control polymer, poly{[N,N'-bis(2-octyldodecyl)-1,4,5,8-naphthalenedicarboximide-2,6-diyl]-alt-5,5'-[2,2'-(1,2-ethanyl)bithiophene]} (P(NDI2OD-TET), for synthetic details see SI), where a saturated $-\text{CH}_2-\text{CH}_2-$ group is inserted in between the two thienyl units to break polymer backbone π -conjugation, leaving all the other P(NDI2OD-T2) structural components unaltered (Figure 1b).

The *vis-à-vis* n-dopability of the two polymers was investigated using an *in situ* electrochemical doping method. This method enables precise control of the doping process and monitoring of the property change of the polymer. Li is an ideal doping reagent because the small size of Li^+ prevents phase segregation (*vide infra*). For this purpose, coin-type rechargeable Li cells were fabricated with a polymer/conductive carbon mixture as the working electrode and metallic Li as the counter and reference electrode (see SI). Polymer/carbon black composite films containing 60 wt % of polymer with an active mass of 0.3 mg cm^{-2} was galvanostatically cycled in a Li^+ -containing ethereal electrolyte solution to realize the redox process. Every NDI unit of our polymers contains four carbonyl groups, two of which are expected to be reversibly reduced via a two-step two-lithium addition reaction (Figure 2a; further

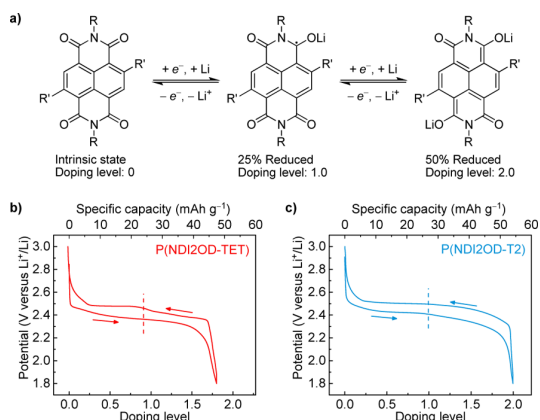


Figure 2. (a) Two-step n-doping/lithiation mechanism of the naphthalene diimide unit. (b,c) Potential profile of P(NDI2OD-TET) (b) and P(NDI2OD-T2) (c) during n-doping-undoping at 1C (2 h per cycle).

reduction of more carbonyl groups leads to irreversible transformation of the NDI structure).⁹ During the two-carbonyl reduction, two electrons will be delocalized in the polymer backbone repeating units, corresponding to an n-doping level of ~ 2.0 . Figure 2b,c shows the potential profile during the n-doping-undoping process, demonstrating that both polymers undergo two-step reduction. The two stages are

less discernible for P(NDI2OD-T2), reflecting the charge delocalization by the π -conjugated backbone. Judging from the charge injected during n-doping, both polymers roughly complete the two-electron reduction, leading to high n-doping levels of 1.8 and 2.0 for P(NDI2OD-TET) and P(NDI2OD-T2), respectively. If considered as a Li storage material, the specific capacity of P(NDI2OD-T2) is 54.2 mAh g^{-1} , or 100% of its theoretical capacity (54.2 mAh g^{-1} ; calculated for a 2-electron redox reaction), whereas the specific capacity of P(NDI2OD-TET) is 47.4 mAh g^{-1} , or 90% of its theoretical capacity (52.7 mAh g^{-1}). Thus, the π -conjugated backbone of P(NDI2OD-T2) does not decrease the n-dopability.

The electrode kinetics of P(NDI2OD-T2) and P(NDI2OD-TET) were compared via fast charge–discharge measurements. As shown in Figure 3a, the capacity retention at 10C (12 min

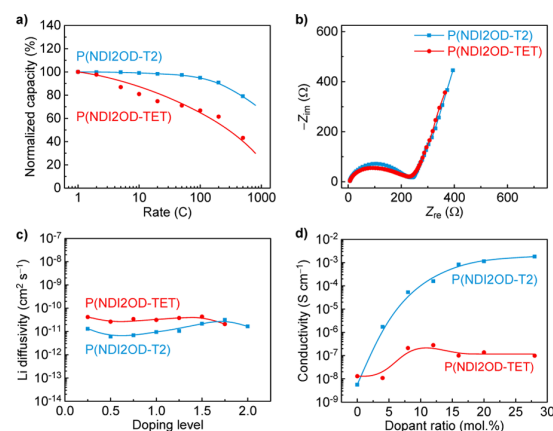


Figure 3. Electrochemical measurements for the indicated polymers. (a) Capacity retention of Li cells at increased charge–discharge rates. (b) Nyquist plot from EIS measurements of Li cells at a depth-of-discharge of 25%. (c) Solid-state Li diffusivity measured with GITT at different doping levels. (d) Electronic conductivity of polymer films without carbon additive in different chemically n-doped states.

per cycle) is 99% and 80% for P(NDI2OD-T2) and P(NDI2OD-TET), respectively, whereas at the ultrafast rate of 500C (14 s per cycle) the capacity difference between the two polymers is larger (79% and 43%, respectively). The potential difference between charge and discharge curves is also smaller for P(NDI2OD-T2) at all C-rates (Figure S1). Clearly, P(NDI2OD-T2) exhibits much faster electrode kinetics than that of P(NDI2OD-TET).

The kinetics of a solid-state battery electrode is determined by three major parameters: (1) reaction rate of the redox site, (2) solid-state ion (Li^+) transport, and (3) electronic conduction. Thus, we have measured these parameters for our polymers to understand the origin of the fast electrode kinetics of P(NDI2OD-T2). First, the redox activity was probed with electrochemical impedance spectroscopy (EIS). The obtained Nyquist plots (Figure 3b) for the two polymers at a depth-of-discharge of 25% (corresponding to a doping level of 0.5) show semicircles of about the same size in the low- to mid-frequency region, which can be fitted to yield almost identical charge transfer resistance ($R_{\text{ct}} = 206.7$ and 210.4Ω for P(NDI2OD-T2) and P(NDI2OD-TET), respectively). The R_{ct} values for the two polymers remain very similar (235.5 and 223.8Ω) at a depth-of-discharge of 75% (corresponding to a doping level of 1.5). Since R_{ct} is a measure of the

electrochemical reactivity of an electrode material at a given discharged state, the almost identical values indicate no obvious difference in the reaction rate for the two polymers in any n-doped state. Second, the solid-state Li diffusivity was assessed with galvanostatic intermittent titration technique (GITT), and the calculated diffusivity values are plotted against doping level (Figure 3c). The Li diffusivity–doping level dependence is similar for both polymers. The average Li diffusivities are 1.45×10^{-11} and 3.38×10^{-11} cm² s⁻¹ for P(NDI2OD-TET) and P(NDI2OD-T2), respectively, again very close to each other. These similarities are not unexpected given the largely identical functional groups in the two polymers. The slight advantage of P(NDI2OD-TET) over P(NDI2OD-T2) in Li diffusivity may be due to the less efficient stacking of P(NDI2OD-TET) compared with P(NDI2OD-T2) (Figure S2), which allows faster Li diffusion. Finally, the film electronic conductivity of our polymers was measured for chemically doped samples. Note, conductive carbon was not used in these measurements to extract the intrinsic conductivity of the polymers. Both pristine polymers exhibit a conductivity of $\sim 10^{-8}$ S cm⁻¹. However, when reduced with progressively larger amounts of the molecular dopant 4-(1,3-dimethyl-2,3-dihydro-1H-benzimidazol-2-yl)-N,N-diphenylaniline (N-DPBI),¹⁰ the conductivity of P(NDI2OD-T2) rapidly increases by 5 orders of magnitude to $\sim 10^{-3}$ S cm⁻¹, whereas that of P(NDI2OD-TET) only increases slightly ($\sim 10^{-7}$ S cm⁻¹). These data indicate that intrachain charge transport within the π -conjugated backbone of P(NDI2OD-T2) dramatically enhances electronic conductivity, whereas that of P(NDI2OD-TET) must rely only on the less efficient interchain charge hopping. In our lithium cells, although the addition of conductive carbon will increase the apparent conductivity of the electrodes based on both polymers, the electron transport within the polymer domains will be still dominated by the intrinsic conductivity of the doped polymers. The combined results demonstrate that the presence of the π -insulating ethanyl group in P(NDI2OD-TET) does not alter the redox activity nor the ionic conductivity and that the highly conducting nature of the heavily n-doped P(NDI2OD-T2) is unambiguously responsible for the ultrafast energy storage capability.

We believe that the electronic conductivity saturation observed in Figure 3d for P(NDI2OD-T2) at N-DPBI molar doping ratio >15 mol % is due to inefficiencies of the dopant rather than an intrinsic limitation of the polymer. Indeed, Figure 4 compares the morphology of the pristine P(NDI2OD-T2) film, the chemically doped polymer film with 28 mol % of N-DPBI, and the electrochemically doped polymer film with 200 mol % of Li ion. The pristine film shows fiber-like structure as previously reported.^{8a} N-DPBI-doped film at 28 mol % shows grainy aggregates, indicating significant phase segregation of the molecular dopant. This observation is in agreement

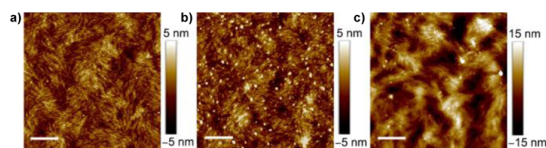


Figure 4. AFM height images of (a) pristine P(NDI2OD-T2) thin-film, (b) P(NDI2OD-T2) thin-film doped with 28 mol % of N-DPBI, and (c) P(NDI2OD-T2) thin-film doped with 200 mol % of Li ion. Scale bar: 500 nm. The rms roughness for three films is 1.07, 1.33, and 5.14 nm, respectively.

with previous studies showing similar phase segregation of molecular dopants in the polymer host films for N-DPBI dopant ratio >10 mol %, ^{8b} thus precluding further doping. In contrast, the electrochemically Li⁺-doped P(NDI2OD-T2) film exhibits negligible phase segregation and the increased surface roughness is due to the exceptionally large (200 mol %) dopant content (Figure 4c). Because of the much higher doping level, it is expected that Li-ion-doped films should exhibit similar trends but with far higher conductivities than those measured for the N-DPBI-doped films in Figure 3d.

The improved redox kinetics of P(NDI2OD-T2) encouraged us to further explore its fast charge–discharge capability. Electrodes with higher loading (up to 1.3 mg cm⁻²; see Figure S3) and higher active mass ratio (up to 80 wt % active mass; see Figure S4) were fabricated, and the results are compared to the state-of-the-art organic electrode materials (Table S1). For relatively “standard” high-rate conditions (~ 1 mg cm⁻², ~ 60 wt % active mass, ~ 10 C), very few materials approach the capacity retention level of P(NDI2OD-T2). Under more harsh testing conditions such as an ultrahigh-rate of 100C (72 s per cycle) and a high active mass ratio of ≥ 80 wt % (unusually high for organic electrode materials), the superiority of P(NDI2OD-T2) becomes even more evident (Table 1). Note that all these

Table 1. Comparison of the Rate Capability of P(NDI2OD-T2) with Those of State-of-the-Art Electrode Materials

compound	doping type	active ratio ^a	capacity retention ^b	ref
P(NDI2OD-T2)	n	60%	95% (100C)	this work
		80%	81% (10C)	
PTCDA-PI2	n	60%	28% (80C)	12
PVK	p	55%	78% (100C)	11a
PTMA	p	50%	71% (50C)	11b
Li ₄ C ₆ O ₆	n	80%	27% (6C)	13

^aOnly the results obtained with an active mass ratio of $\geq 50\%$ and non-thin-film devices (e.g., thickness > 1 μ m) are included. ^bRelative to the highest capacity recorded at low rates. Only the results obtained at ≥ 50 C (for active ratio ≥ 50 wt%) or ≥ 5 C (for active ratio ≥ 80 wt%) are included.

results were achieved without the need for the construction of porous polymer structures, incorporation of nanostructured 1-/2-/3-dimensional conducting reagents, or nanoscale morphology control, which are strategies usually necessary to achieve fast kinetics for organic electrode materials.^{5c,11} The fact that P(NDI2OD-T2) exhibits the best electrode kinetics despite the presence of significant amount ($\sim 57\%$ by mass; $> 65\%$ by volume from DFT computations) of redox-inactive/insulating alkyl chains in the molecular structure fully demonstrates the potential of this molecular design for energy storage purposes.

The stability of P(NDI2OD-T2) when heavily n-doped and the reversibility of the doping process were investigated through repeated doping/undoping cycles. Impressively, after 3000 cycles to a doping level of 2.0 and complete undoping to the intrinsic state, the dopability of P(NDI2OD-T2) remains intact (Figure 5), with a capacity retention of 96% and an average Coulombic efficiency $> 99.95\%$. These results demonstrate for the first time that π -conjugated redox polymers can be stably and reversibly n-doped to such high levels. As a charge storage material, the specific capacity of P(NDI2OD-T2) (~ 54 mAh g⁻¹) is lower than that of inorganic counterparts due to the unoptimized long solubilizing alkyl chains that do not

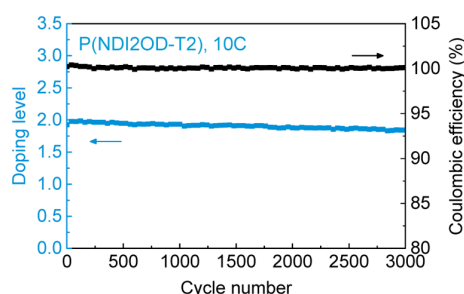


Figure 5. Reversibility of P(NDI2OD-T2) during repeated deep n-doping-undoping at 10C (12 min per cycle).

contribute to redox reaction. Thus, improved molecular design of π -conjugated redox polymers should result in significant improvement in energy density.

In summary, we have successfully demonstrated that π -conjugated redox polymers are a class of heavily n-dopable conducting polymers combining the high redox activity of redox polymers and the high electronic conductivity of π -conjugated polymers. A model polymer, P(NDI2OD-T2), was stably and reversibly n-doped to a high doping level of 2.0, a significant progress for electron-transporting π -conjugated polymers. The high electronic conductivity of the heavily n-doped P(NDI2OD-T2) leads to unprecedented electrode kinetics when evaluated as an organic electrode material for rechargeable Li batteries. With rational molecular design, π -conjugated redox polymers will establish new design space in polymer chemistry and see widespread applications especially in energy-related ones such as batteries, supercapacitors, and thermoelectrics.¹⁴

■ ASSOCIATED CONTENT

● Supporting Information

Details on material synthesis and electrochemical measurements, detailed electrochemical measurement results. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest.

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EXHIBIT 4

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- (51) **Int. Cl.**
H01M 10/06 (2006.01)
H01M 4/14 (2006.01)
H01M 10/08 (2006.01)
H01M 10/44 (2006.01)
B60L 53/10 (2019.01)
- (52) **U.S. Cl.**
 CPC *H01M 4/608* (2013.01); *H01M 10/06* (2013.01); *H01M 10/08* (2013.01); *H01M 10/44* (2013.01); *H01M 4/606* (2013.01); *H01M 2220/20* (2013.01); *Y02E 60/126* (2013.01); *Y02T 10/7016* (2013.01)
- (58) **Field of Classification Search**
 CPC .. H01M 4/608; H01M 2220/20; H01M 4/606; B60L 53/11; Y02E 60/126; Y02T 10/7016
 See application file for complete search history.
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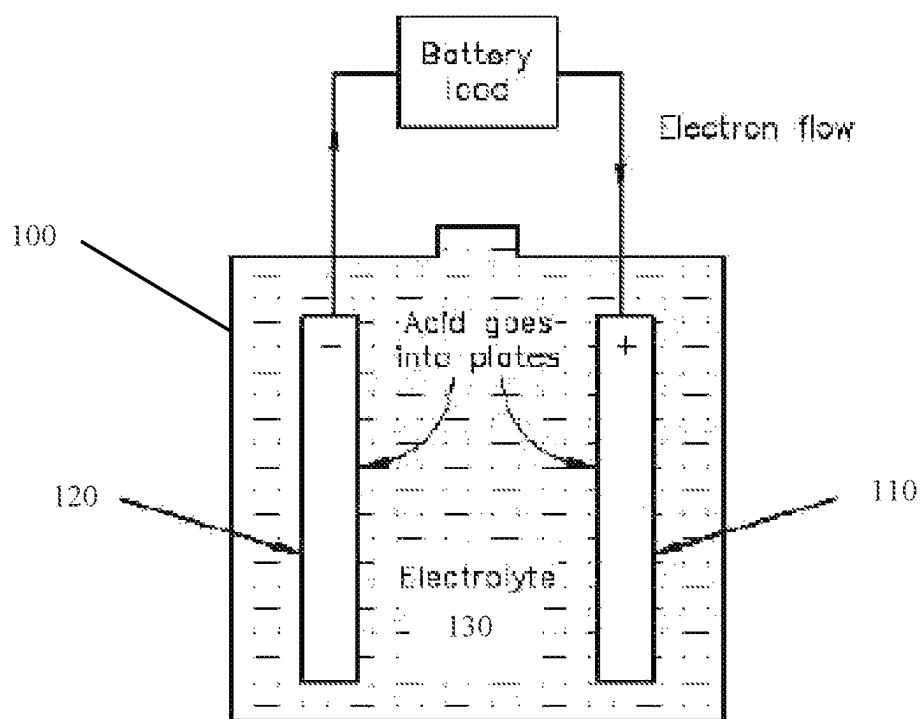


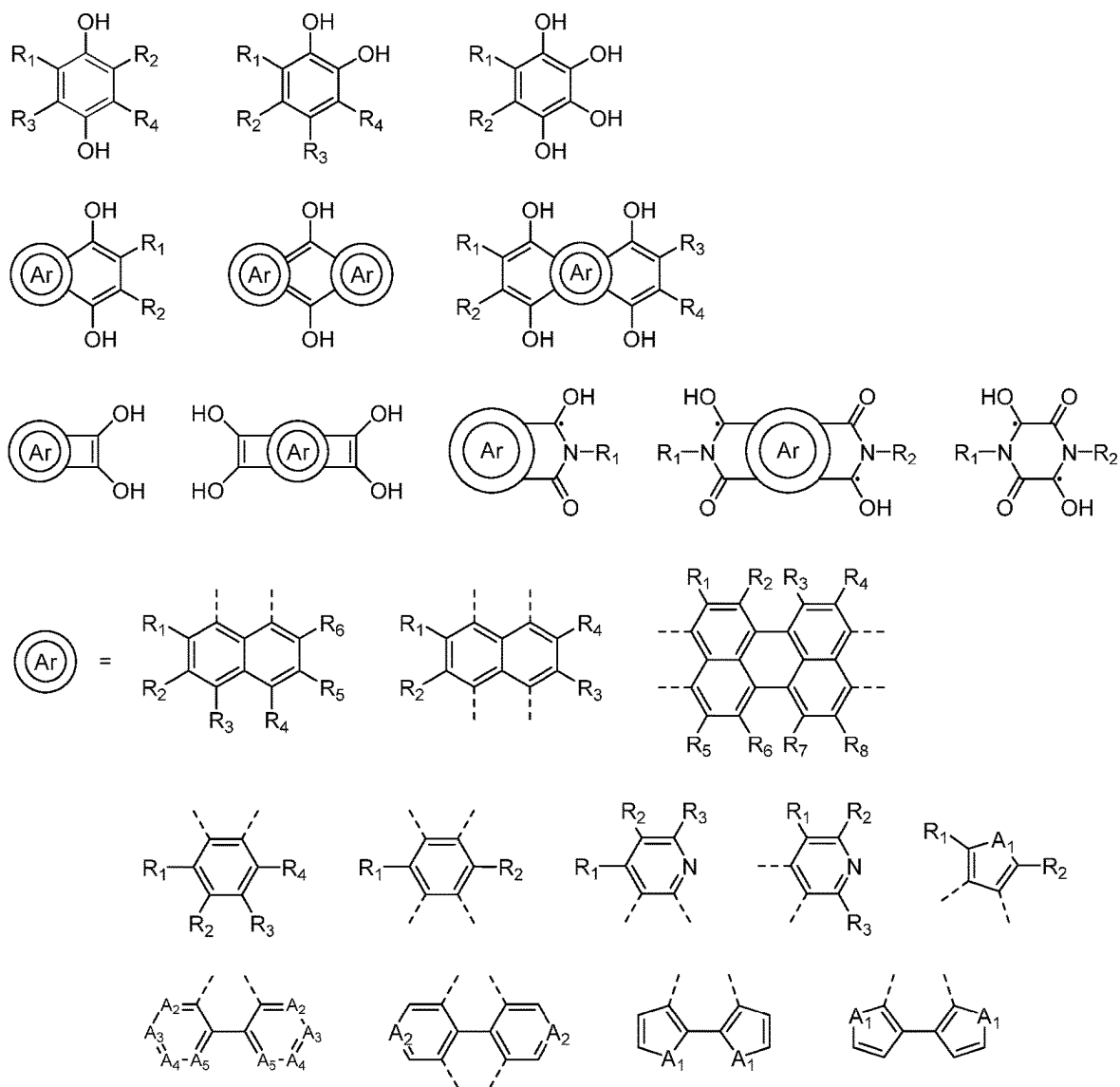
FIG. 1

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R_{1, 2, ..., 8} = M_x, CH₃, CH₂CH₃, CH₂CH₂CH₃, CH(CH₃)₂, C(CH₃)₃, CHCH₂, CCH, OM_x, OCH₃, OCH₂CH₃, OCH₂CH₂CH₃, OCH(CH₃)₂, OC(CH₃)₃, NH₂, NHCH₃, N(CH₃)₂, NHCH₂CH₂CH₃, NHCH(CH₃)₂, NHC(CH₃)₃, CN, Cl, Br, I, SO₃M_x

M = H, Li, Na, K, Mg, Ca, Al, or a mixture of these elements

x = 0.33~1

A₁ = NR₁, O, S

A_{2, 3, 4, 5} = C, N

FIG. 2

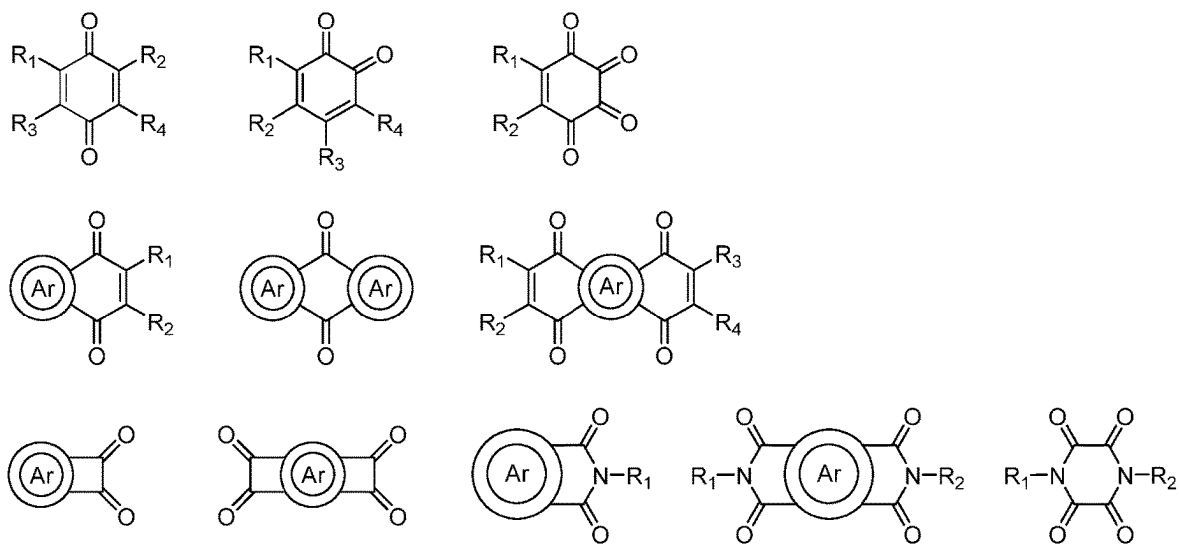
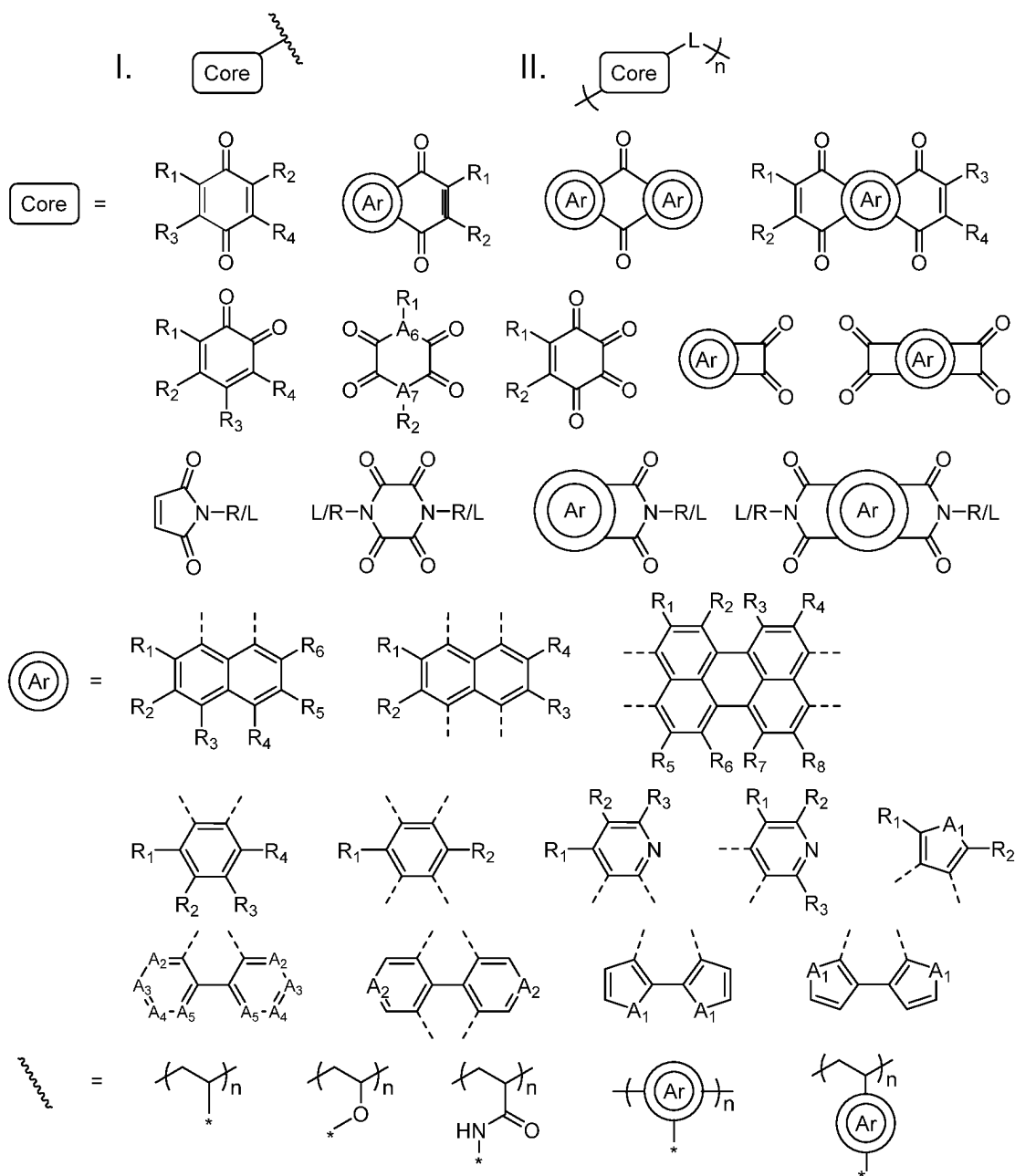


FIG. 3



$R_1, 2, \dots, 8$ = N/A, M_x , CH_3 , CH_2CH_3 , $CH_2CH_2CH_3$, $CH(CH_3)_2$, $C(CH_3)_3$, $CHCH_2$, CCH , OM_x , OCH_3 , OCH_2CH_3 , $OCH_2CH_2CH_3$, $OCH(CH_3)_2$, $OC(CH_3)_3$, NH_2 , $NHCH_3$, $N(CH_3)_2$, $NHCH_2CH_2CH_3$, $NHCH(CH_3)_2$, $NHC(CH_3)_3$, CN , Cl , Br , I , SO_3M_x

M = H, Li, Na, K, Mg, Ca, Al, or a mixture of these elements

x = 0.33~1

A_1 = NR_1 , O, S

$A_{2, 3, 4, 5}$ = C, N

$A_{6, 7}$ = CH_2 , CHR , $C(R)_2$, NH , NR , O, S

L = N/A, L' , $C(=O)$, $C(=O)-L'-C(=O)$, NH , O, S, $NH-L'-NH$, $O-L'-O$, $S-L'-S$

L' = CH_2 , $(CH_2)_2$, $(CH_2)_3$, $(CH_2)_4$, $(CH_2)_6$, 

FIG. 4

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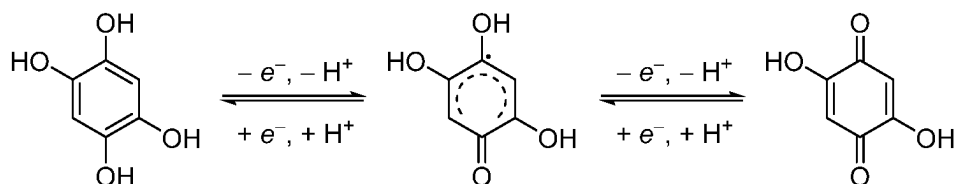


FIG. 5

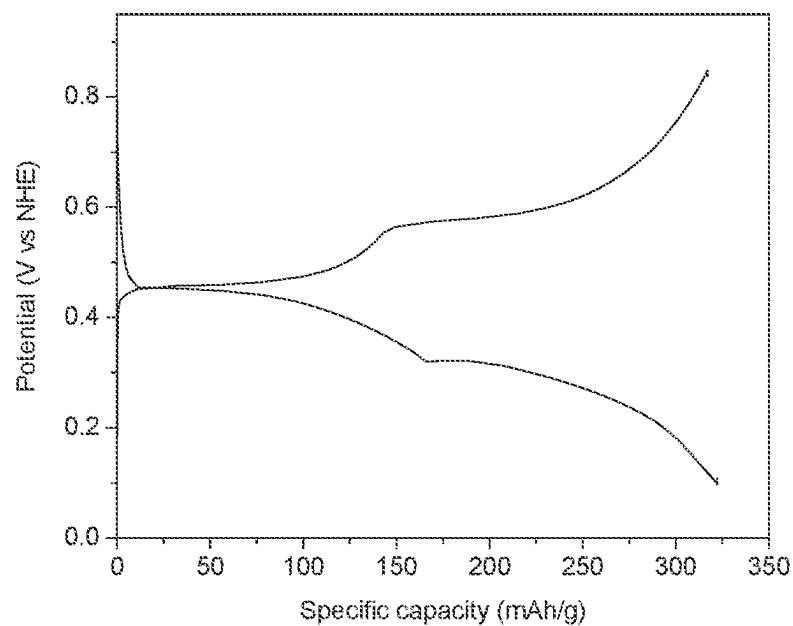


FIG. 6

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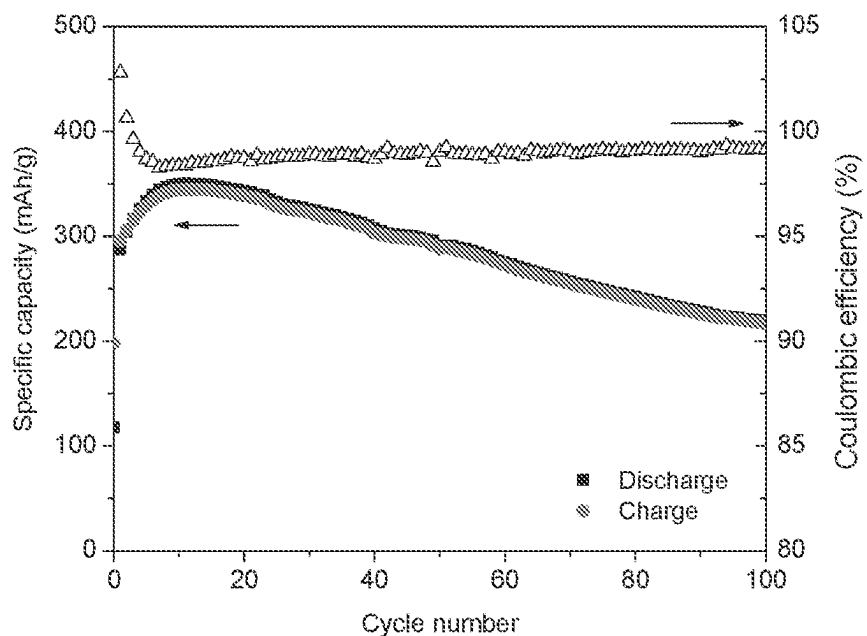


FIG. 7

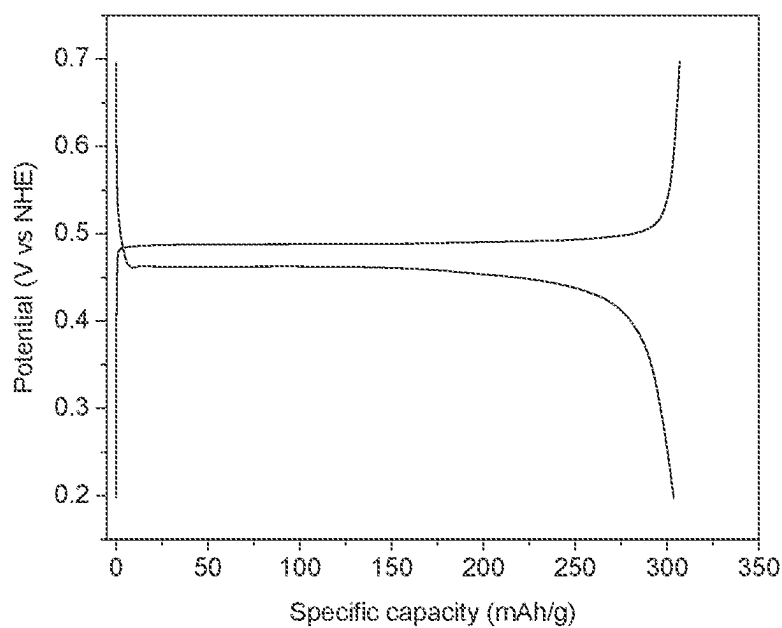


FIG. 8

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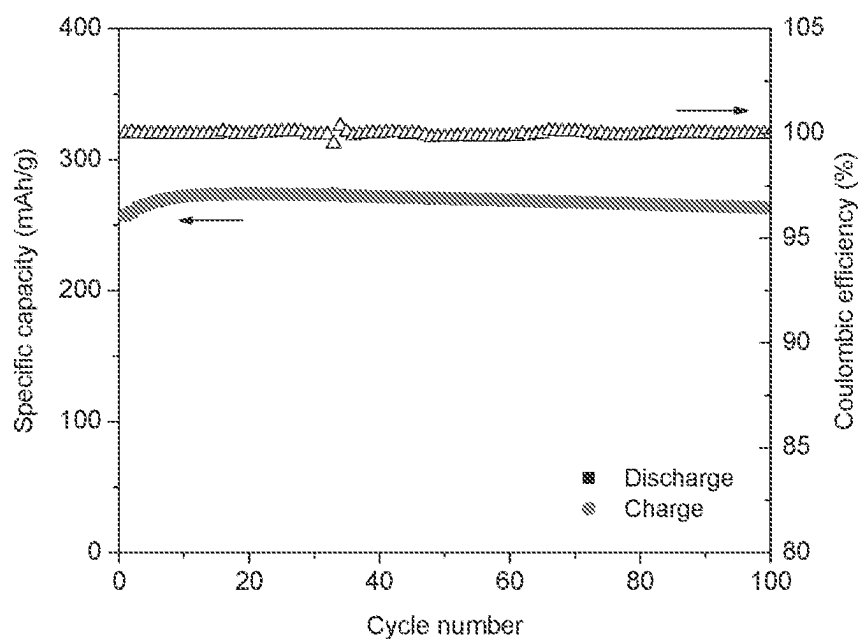


FIG. 9

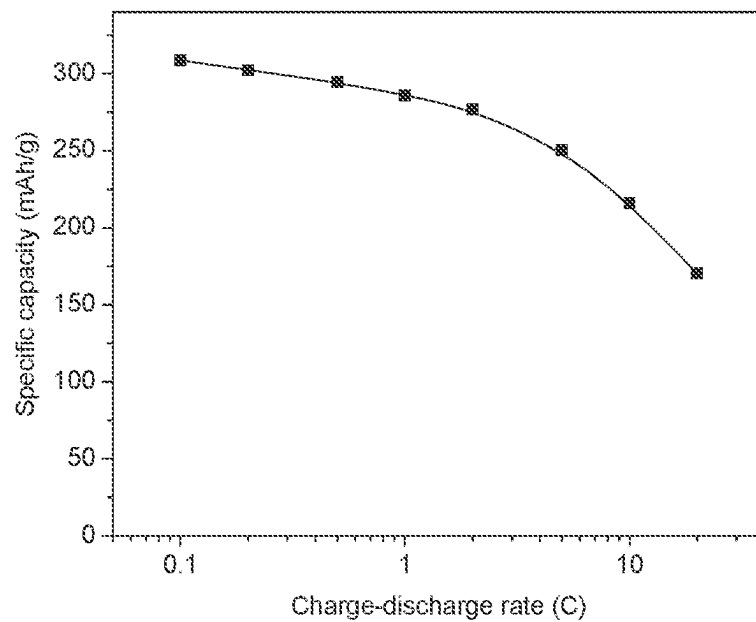


FIG. 10

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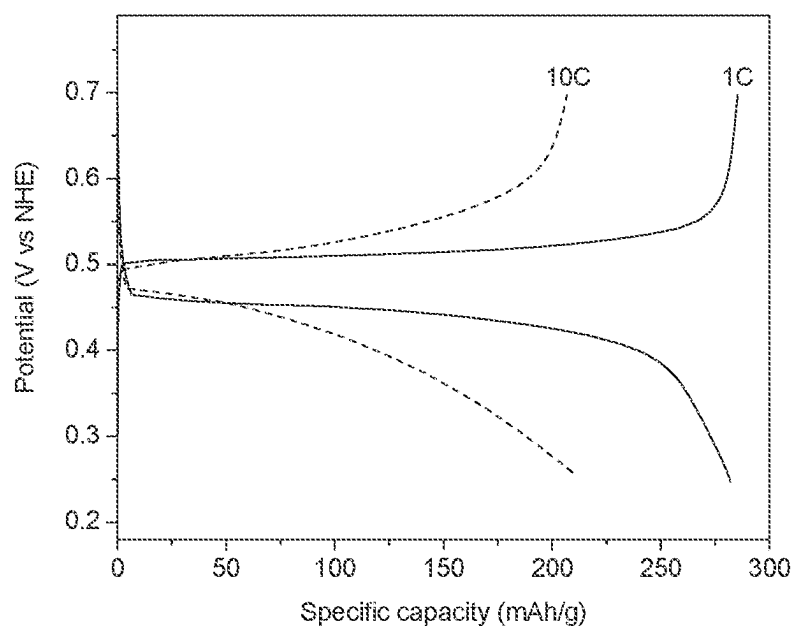


FIG. 11

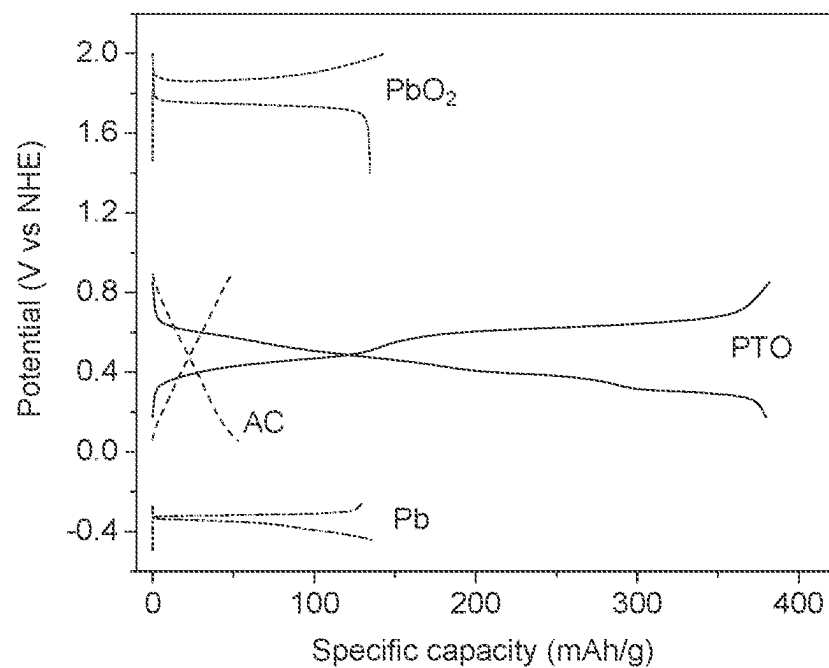


FIG. 12

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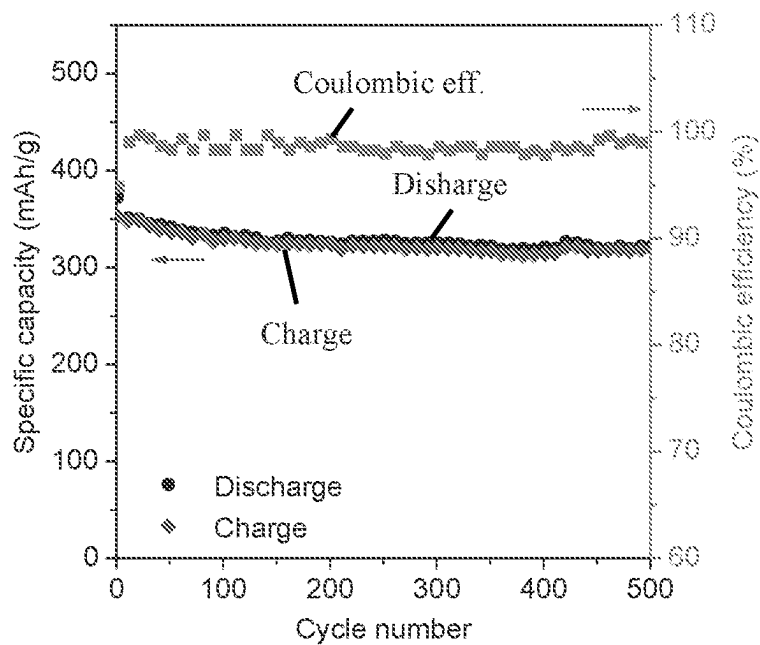


FIG. 13

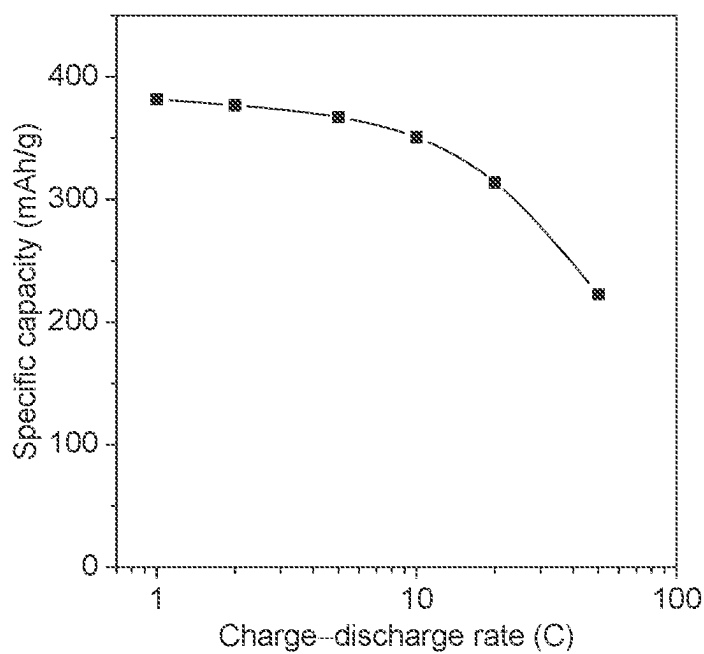


FIG. 14

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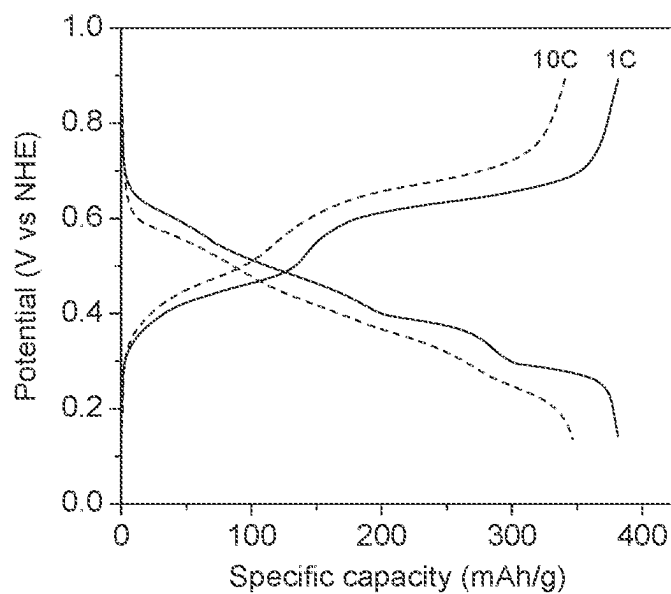


FIG. 15

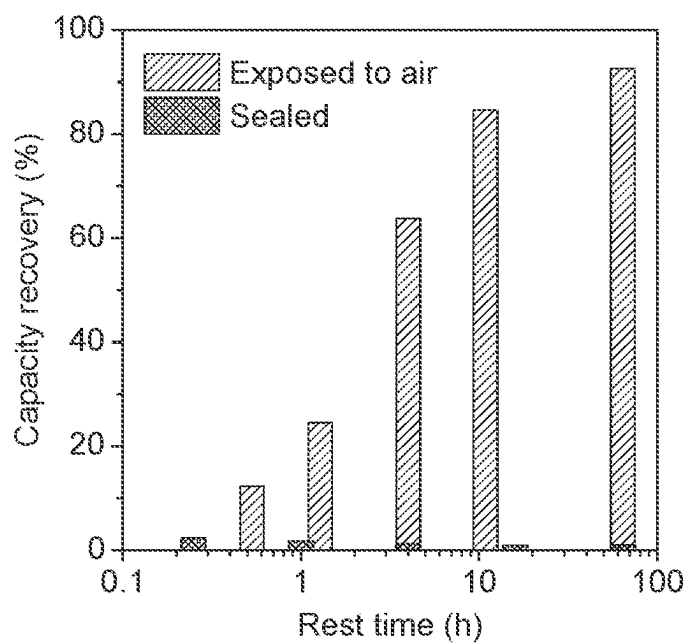


FIG. 16

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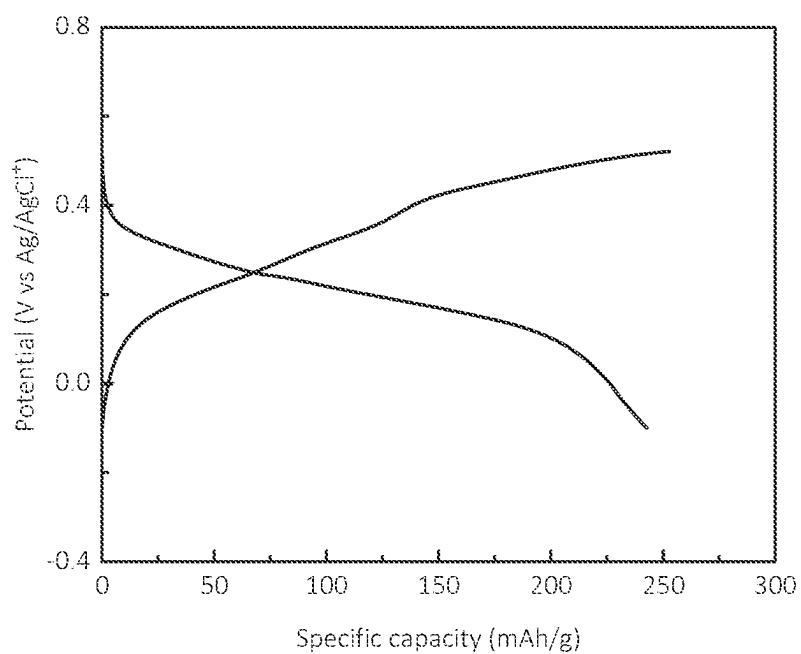


FIG. 17

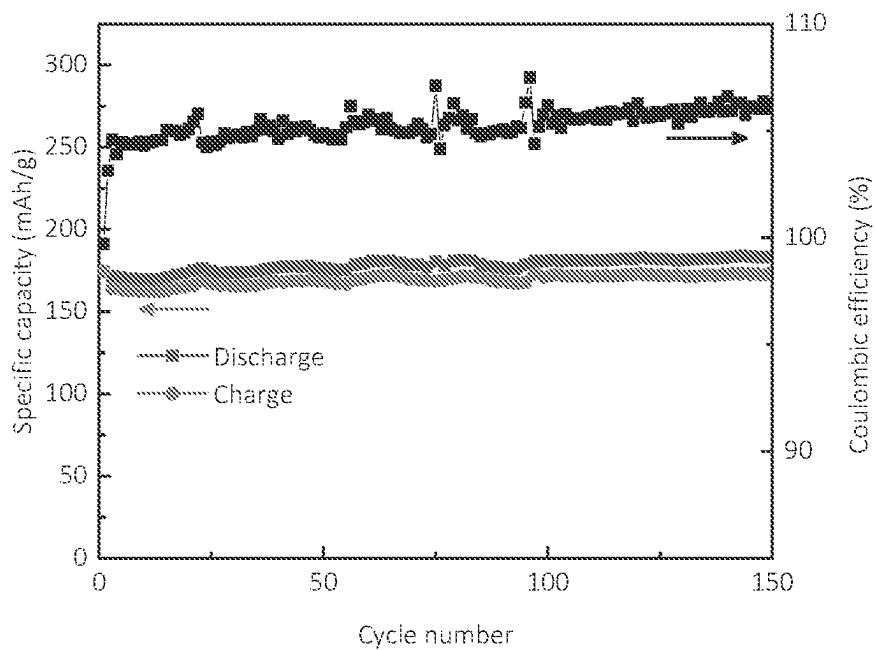


FIG. 18

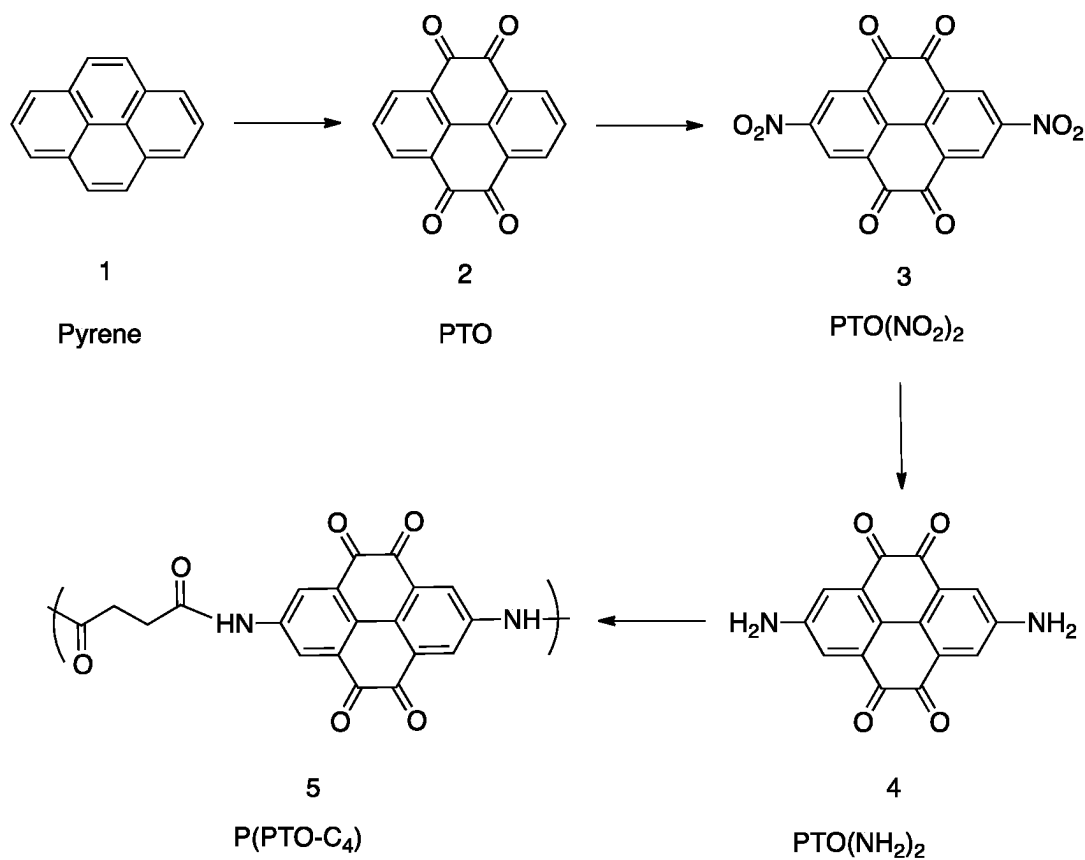


FIG. 19

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**LEAD-ACID BATTERIES WITH FAST
CHARGE ACCEPTANCE****RELATED APPLICATIONS**

This application claims the benefit of U.S. Provisional Patent Application No. 62/165,377 filed on May 22, 2015, which is incorporated herein by reference.

FIELD OF THE INVENTION

This invention relates to lead acid batteries (LABs). More particularly, to LABs with improved electrodes and charge acceptance.

BACKGROUND OF INVENTION

The lead acid battery (LAB) industry represents more than \$30B in annual sales globally. A common arrangement for LABs is to utilize lead oxide (PbO_2) as a cathode, lead as an anode, and 2-8 M sulfuric acid as the electrolyte. Some features of LABs are: high discharge power, excellent high and low temperature operation, ultra-safe, and excellent existing recycling programs that are highly economical. However, LABs may utilize heavy metals that are or may become banned in the future due to toxicity and/or environmental concerns. For example, Europe threatens to extend the ban on heavy metals to lead acid batteries, including those used for vehicles. Further, in some applications, higher charge acceptance than current LABs provide may be desirable. For example, micro hybrid and stop/start technologies require higher charge acceptance that current lead acid battery technology cannot provide.

The charge acceptance problem in LABs is related to the negative electrode. As an example, the mechanism of LABs may include $PbSO_4$ forming large crystals on the negative electrode. This leads to a sulfated negative electrode or a negative electrode that is at a lower state-of-charge than the positive. This failure mechanism is common in start-stop vehicles. Under the high-rate partial state-of-charge conditions that are required by mild/full hybrid electric vehicles, the lead electrode is known to suffer premature capacity loss due to aggressive sulfation at the electrode surface. The use of additives with high surface area alleviates this problem but does not eliminate it. This leads to a sulfated negative electrode or a negative electrode that is at a lower state-of-charge than the positive. This failure mechanism is common in start-stop vehicles.

Lead-carbon (PbC) batteries and UltraBattery® have been developed by several companies to address this challenge, which substitute activated carbon for the lead anode. Charge/discharge cycle testing of these batteries at 1C further confirmed that a standard positive electrode in an LAB could be charged and discharged at high rate. However, the negative electric double-layer capacitor-type electrode in the PbC battery had very low capacity and a capacitive voltage profile (i.e. sloping). Further, these batteries required a complicated negative electrode design that resulted in high resistance preventing adequate current for cold starting of vehicles.

SUMMARY OF INVENTION

A lead-organic (PbOrg) battery is discussed herein, which refers to a lead-acid battery that utilizes at least one organic material as an electrode. In one embodiment, a PbOrg battery may provide ten or more times higher charge accep-

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tance than other lead-acid batteries. The PbOrg battery may provide positive and negative electrodes in an electrolyte. The PbOrg battery may provide an electrode formed with one or more organic materials, such as, but not limited to, organic compounds comprising cyclic substructures with hydroxyl and/or carbonyl groups. The cyclic substructures may be a carbocycle, heterocycle, or aromatic. In some embodiments, the organic compound may be a molecule. In some embodiments, the organic compound may be a coordination polymer. In some embodiments, the organic compound may be a polymer. In some embodiments, the PbOrg battery may decrease the use of lead in comparison to prior lead acid battery designs by 50% or more.

The foregoing has outlined rather broadly various features of the present disclosure in order that the detailed description that follows may be better understood. Additional features and advantages of the disclosure will be described hereinafter.

BRIEF DESCRIPTION OF THE DRAWINGS

For a more complete understanding of the present disclosure, and the advantages thereof, reference is now made to the following descriptions to be taken in conjunction with the accompanying drawings describing specific embodiments of the disclosure, wherein:

FIG. 1 illustrates an arrangement for a PbOrg battery;

FIG. 2 shows the structure of the reduced form of organic materials that can be used as the electrode materials for the disclosed PbOrg battery;

FIG. 3 shows the structure of the oxidized form of organic materials that can be used as the electrode materials for the disclosed PbOrg battery;

FIG. 4 shows the structure of organic polymers that can be used as the electrode materials for lead-polymer batteries;

FIG. 5 shows the reductive protonation mechanism in the reversible electrochemical reduction of an organic molecule;

FIG. 6 shows the voltage profile of an organic molecule, 2,5-dihydroxy-1,4-benzoquinone, in an acid electrolyte;

FIG. 7 shows the cycling performance of 2,5-dihydroxy-1,4-benzoquinone in an acid electrolyte;

FIG. 8 shows the voltage profile of an organic molecule, 2,5-dimethoxy-1,4-benzoquinone, in an acid electrolyte;

FIG. 9 shows the cycling performance of 2,5-dimethoxy-1,4-benzoquinone in an acid electrolyte;

FIG. 10 shows the rate performance of 2,5-dimethoxy-1,4-benzoquinone in an acid electrolyte;

FIG. 11 shows the voltage profile of 2,5-dimethoxy-1,4-benzoquinone at high current densities;

FIG. 12 compares the voltage profile of PbO_2 , Pb, activated carbon, and an organic molecule, pyrene-4,5,9,10-tetraone, in a sulfuric acid electrolyte;

FIG. 13 shows the cycling performance of pyrene-4,5,9,10-tetraone in an acid electrolyte;

FIG. 14 shows the rate performance of pyrene-4,5,9,10-tetraone in an acid electrolyte;

FIG. 15 shows the voltage profile of pyrene-4,5,9,10-tetraone at high current densities;

FIG. 16 shows the capacity recovery of pyrene-4,5,9,10-tetraone when exposed to air;

FIG. 17 shows the voltage profile of a polymer anode, poly[pyrene-4,5,9,10-tetraone], in an acid electrolyte;

FIG. 18 shows the cycling performance of poly[pyrene-4,5,9,10-tetraone] in an acid electrolyte; and

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FIG. 19 shows a synthesis route for poly[pyrene-4,5,9,10-tetraene].

DETAILED DESCRIPTION

Refer now to the drawings wherein depicted elements are not necessarily shown to scale and wherein like or similar elements are designated by the same reference numeral through the several views.

Referring to the drawings in general, it will be understood that the illustrations are for the purpose of describing particular implementations of the disclosure and are not intended to be limiting thereto. While most of the terms used herein will be recognizable to those of ordinary skill in the art, it should be understood that when not explicitly defined, terms should be interpreted as adopting a meaning presently accepted by those of ordinary skill in the art.

It is to be understood that both the foregoing general description and the following detailed description are exemplary and explanatory only, and are not restrictive of the invention, as claimed. In this application, the use of the singular includes the plural, the word “a” or “an” means “at least one”, and the use of “or” means “and/or”, unless specifically stated otherwise. Furthermore, the use of the term “including”, as well as other forms, such as “includes” and “included”, is not limiting. Also, terms such as “element” or “component” encompass both elements or components comprising one unit and elements or components that comprise more than one unit unless specifically stated otherwise.

Lead-organic (PbOrg) batteries with high charge acceptance and improved electrodes are discussed further herein. FIG. 1 illustrates an arrangement for a PbOrg battery 100. The PbOrg battery 100 may provide positive 110 and negative 120 electrodes in an electrolyte 130. The positive electrode may be formed from any suitable material, such as, but not limited to, lead, lead sulfate, and lead oxide, the valence of lead in these compounds may range from 0 to +4. The electrolyte may be any suitable electrolyte material, such as, but not limited to, any suitable acid or aqueous sulfuric acid with a concentration of 0.01-10.0 M. In some embodiments, the electrolyte may have a pH value equal to or lower than 2 ($\text{pH} \leq 2$). In some embodiments, the electrolyte material may be a solid-state electrolyte. As a result, the entirety of the battery would be comprised of materials that are solid-state, and as such, the battery may be referred to as a solid-state battery.

FIGS. 2-4 illustrate representative molecular compounds or structures of suitable electrode materials for a PbOrg battery. In some embodiments, the negative electrode of the PbOrg battery may have a lead content of 0-10 wt %. The PbOrg battery may provide negative electrodes that include at least one organic compound or organic material. As a nonlimiting example the organic compound may be selected from the compounds shown in FIG. 2. Generally, these organic materials comprise C—O—H groups in their reduced/charged state (shown in FIG. 2).

Nonlimiting embodiments of a first set of organic materials that are suitable for a negative electrode are discussed herein. In some embodiments, the organic material comprises at least one cyclic substructure with at least one hydroxyl group. In some embodiments, the organic material may be a bicyclic or multi-cyclic compound with at least one hydroxyl group. In some embodiments, the cyclic substructure may be a five/six-membered ring. The cyclic substructure may be a carbocycle with carbon atoms forming the ring or heterocyclic with both carbon and non-carbon atoms

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forming the ring. In some embodiments, the organic material may comprise aromatic rings with at least one hydroxyl group. Further, the cyclic substructure may comprise one or more R groups where $R_1, 2, \dots, 8 = M_x, CH_3, CH_2CH_3, CH_2CH_2CH_3, CH(CH_3)_2, C(CH_3)_3, CHCH_2, CCH, OM_x, OCH_3, OCH_2CH_3, OCH_2CH_2CH_3, OCH(CH_3)_2, OC(CH_3)_3, NH_2, NHCH_3, N(CH_3)_2, NHCH_2CH_2CH_3, NHCH(CH_3)_2, NHC(CH_3)_3, CN, Cl, Br, I, SO_3M_x$, where $M = H, Li, Na, K, Mg, Ca, Al$, or a mixture of these elements, $x = 0.33 \sim 1$. In some embodiments, the cyclic substructure may be fused with one or more aromatic ring systems. In some embodiments, the aromatic ring system may be a single six-/five-membered aromatic ring. Further, the aromatic ring may comprise atoms chosen from C, N, O, or S. In some embodiments, the aromatic ring system may be fused aromatic rings chosen from naphthalene and perylene. In some embodiments, the aromatic ring system may be biaryls comprising two six-/five-membered aromatic rings. Further, the aromatic rings in the biaryl may comprise atoms chosen from C, N, O, or S.

The organic materials, such as those shown in FIG. 2, may have some of their C—O—H groups reversibly transformed into C=O groups in their oxidized/discharged state. FIG. 5 shows a nonlimiting example of the electroactivity of organic compounds arises from this reversible electrochemical redox reaction between hydroxyl and carbonyl groups and the ability of the negatively charged oxygen atoms to coordinate with proton. While the two-step (de)protonation mechanism of the reversible electrochemical reduction of an organic carbonyl compound (e.g. 2,5-dihydroxy-1,4-benzoquinone) is shown, it shall be understood that similar reactions may occur for any organic compounds comprising a cyclic substructure with at least one hydroxyl/carbonyl group in a battery environment. As nonlimiting examples, for the corresponding oxidized forms of compounds shown in FIG. 2, the result of this reaction leads to a second set of organic materials that are shown in FIG. 3 and discussed further below.

Nonlimiting embodiments of the second set of organic materials that are suitable for a negative electrode are described as follows. In some embodiments, the organic material comprises at least one cyclic substructure with at least one hydroxyl group. In some embodiments, the organic molecular material may be a bicyclic or multi-cyclic compound with at least one hydroxyl group. In some embodiments, the cyclic substructure may be a five/six-membered ring. The cyclic substructure may be a carbocycle with carbon atoms forming the ring or heterocyclic with both carbon and non-carbon atoms forming the ring. In some embodiments, the organic molecular material may comprise aromatic rings with at least one hydroxyl group. Further, the cyclic substructure may comprise one or more R groups where $R_1, 2, \dots, 8 = M_x, CH_3, CH_2CH_3, CH_2CH_2CH_3, CH(CH_3)_2, C(CH_3)_3, CHCH_2, CCH, OM_x, OCH_3, OCH_2CH_3, OCH_2CH_2CH_3, OCH(CH_3)_2, OC(CH_3)_3, NH_2, NHCH_3, N(CH_3)_2, NHCH_2CH_2CH_3, NHCH(CH_3)_2, NHC(CH_3)_3, CN, Cl, Br, I, SO_3M_x$, where $M = H, Li, Na, K, Mg, Ca, Al$, or a mixture of these elements, $x = 0.33 \sim 1$. In some embodiments, the cyclic substructure may be fused with one or more aromatic ring systems. In some embodiments, the aromatic ring system may be a single six-/five-membered aromatic ring. Further, the aromatic ring may be a heterocycle with heteroatoms chosen from N, O, or S. In some embodiments, the aromatic ring system may be fused aromatic rings chosen from naphthalene and perylene. In some embodiments, the aromatic ring system may be biaryls

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comprising two six-/five-membered aromatic rings. Further, the aromatic ring in the biaryl may comprise atoms chosen from C, N, O, or S.

In some embodiments, the organic material(s) that are suitable for a negative electrode may comprise at least one material selected from the first set or second set discussed above. In some embodiments, the organic materials may comprise a combination of at least one of the organic molecular materials from the first set discussed above and at least one of the organic materials from the second set discussed above. Referring to the exemplary reaction shown in FIG. 5, it shall be understood that the organic material(s) that are suitable for a negative electrode may have both a hydroxyl and carbonyl present in some embodiments, such as in the intermediate state shown. In some embodiments, the organic material may comprise a first cyclic substructure with a hydroxyl group and a second cyclic substructure with a carbonyl group. In some embodiments, the organic material may be a cyclic substructure that comprises both a hydroxyl group and a carbonyl group.

In some embodiments, the organic material(s) for the negative electrode may further include one or more metal ions that can be included in the substitution groups. The resulted compounds are sometimes known as "coordination polymers." For the embodiments of electrode materials discussed above, these coordination polymers have an organometallic polymer structure. The coordination polymer may comprise two or more of the organic molecular material options discussed above (e.g. FIGS. 2-3) linked by metal cation centers, or more specifically metal cations and molecular anions of organic molecules. As a nonlimiting example, cyclic substructures with at least one hydroxyl (C—O—H) group and/or carbonyl (C=O) group, such as the first and second sets of organic materials discussed above, may be connected by metal cations. Further, the metal cations may be chosen from Li⁺, Na⁺, K⁺, Mg²⁺, Ca²⁺, Al³⁺, or a combination thereof. Additionally, some embodiments may have the cyclic substructures connected by metal cations via the R group(s) and/or fused aromatic ring systems. These variations do not change the molecular nature of the active organic molecules.

FIG. 4 illustrates exemplary molecular structures of suitable poly-/oligomer electrode materials in their oxidized/discharged state for a PbOrg battery, particularly lead-polymer battery. Generally, the structural formulas comprise (I) structures where electroactive cores are attached to a poly-/oligomer backbone, or (II) structures where electroactive cores are part of the poly-/oligomer backbone. In some embodiments, the electroactive core may be selected from the organic materials discussed above, such as the nonlimiting examples in FIGS. 2-3. The electroactive core may comprise cyclic substructures with at least one hydroxyl and/or carbonyl group. The electroactive core may be a carbocycle with carbon atoms forming the ring or heterocycle with both carbon and non-carbon atoms forming the ring. The electroactive core may comprise cyclic substructures that are functionalized with R groups and/or fused aromatic ring systems. Nonlimiting examples of the electroactive core are shown in FIG. 4. Note that only the oxidized forms of the active cores are shown in FIG. 4, but it shall be understood that any number of the carbonyl groups in these cores may be reversibly transformed to hydroxyl groups when desired or during battery operation. In some embodiments, the organic material may be a poly-/oligomer, where repeating unit(s) comprise at least one cyclic substructure with at least one hydroxyl group or carbonyl group. Further, the cyclic substructures may be

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functionalized with at least one R group and/or fused aromatic ring system. With respect to the structural formula (I) the chain comprises any polymer or oligomer backbone. In some embodiments, the polymer backbone is chosen from polyethylene, polypropylene, polystyrene, and polyaryls. Nonlimiting examples of the aryl structures in the polyaryls include benzene, thiophene, pyrrole, pyridine, naphthalene, anthracene, perylene, or a combination thereof. The electroactive cores may be linked to the poly-/oligomer backbone directly or via a linker, such as O atoms, ester bonds, amide bonds, or aryl groups. In structural formula (II), electroactive cores may be either directly connected by covalent bonds or linked by linkers. Linkers may comprise a moiety chosen from the group consisting of C=O, (C=O)-L'-(C=O), NH, NH-L'-NH, O, O-L'-O, S, S-L'-S, CH₂, CH₂-L'-CH₂, (CH₂)₂, (CH₂)₂-L'-(CH₂)₂, 5-6 membered (hetero)aryl, or biaryls with two identical or different 5-6 membered (hetero)aryl groups, where L' may be chosen from CH₂, (CH₂)₂, (CH₂)₃, 5-6 membered (hetero)aryl group, or biaryls with two identical or different 5-6 membered (hetero)aryl groups. Without limitation, "n" indicates the average number of repeating units in the formula. In some embodiments, the average number of repeat units for a poly-/oligomer may be at least 2.

In some embodiments, the organic materials disclosed herein for the negative electrode are characterized by (1) electrochemically reversible conversion between the C—O—H-containing form and C=O-containing form or vice versa, and/or (2) an average reduction potential of 0.6 V or less versus normal hydrogen electrode (NHE) in 4.4 M H₂SO₄.

These PbOrg batteries may provide high charge acceptance, such as over-ten-time higher charge acceptance than prior lead acid batteries, and as much as 50% less lead and sulfuric acid than current LABs. Current LABs only allow a C/8 charge rate at room temperature and C/24 at 0° C. Some vehicle manufacturers are adopting start/stop technology to meet new US, European, and/or Chinese requirements for fuel efficiency. In order to do so, LABs will need to provide power for the electrical system when the engine is shut down and then recover charge quickly to allow for repetitive charge/discharge events. Similarly, most renewable energy or energy efficiency projects also have to have a better balance of charge and discharge in order to store energy efficiently when it is available, such as brake energy recovery in vehicles, smoothing wind and solar output, and/or grid stabilization in regulatory/ancillary service. In some embodiments, the charge rate of PbOrg batteries may be 1C or greater. As a nonlimiting example, the PbOrg batteries may be charged from any state-of-charge to 80% charged within 1 hour or shorter. The high charge acceptance would immediately address issues for lead acid battery use in new start/stop and micro hybrid vehicles and would make these batteries suitable for a wide range of new applications such as hybrid vehicles and buses, light rail, and/or grid storage applications. To the best of our knowledge, organic materials have not yet been examined in lead-acid batteries. In some embodiments, the specific capacity of these PbOrg batteries may be 30 mAh/g or greater. Because the negative electrode in the improved LABs batteries does not contain lead and not consume sulfate ions, the batteries reduce the use of lead and sulfuric acid by 50% of weight. These PbOrg batteries will rejuvenate the lead-acid battery industry by enabling high charge acceptance originated from polymeric active materials.

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Experimental examples are discussed below to illustrate the ability of organic materials to be potentially sustainable, low-cost, and high-energy electrode materials.

The following examples are included to demonstrate particular aspects of the present disclosure. It should be appreciated by those of ordinary skill in the art that the methods described in the examples that follow merely represent illustrative embodiments of the disclosure. Those of ordinary skill in the art should, in light of the present disclosure, appreciate that many changes can be made in the specific embodiments described and still obtain a like or similar result without departing from the spirit and scope of the present disclosure.

Organic compounds are shown to be reliable and low-cost negative electrode materials that can be used to replace lead in a LAB. This (de)protonation reaction is an intrinsically fast and highly reversible process. Organic compounds that are potentially sustainable, low-cost, and high-capacity (200-500 mAh/g) are very attractive electrode materials. FIGS. 6&7 show the electrochemical performance of a cell based on 2,5-dihydroxy-1,4-benzoquinone tested in a 4.4 M sulfuric acid electrolyte. A specific capacity of up to 350 mAh/g is obtained, which is 91% of the theoretical capacity based on a two-electron transfer reaction, confirming the proposed mechanism. FIG. 8-11 show the electrochemical performance of 2,5-dimethoxy-1,4-benzoquinone. The average discharge voltage is 100 mV lower than that of 2,5-dihydroxy-1,4-benzoquinone, which will be translated into 100 mV increase of battery voltage. The capacity retention is 96% after 100 cycles. When charged at a constant rate of 1C from a completely discharged state, 93% of the maximum capacity is obtained. Even at a very high charging rate of 10C, 67% of the maximum capacity can be obtained with minimal loss (10 mV) in voltage. FIG. 12 compares the voltage profile of PbO₂, Pb, activated carbon, and pyrene-4,5,9,10-tetraone in a sulfuric acid electrolyte. The specific capacity of the pyrene-4,5,9,10-tetraone is 382 mAh/g, which is more than five times of that of activated carbon and two times higher than that of Pb. The voltage profile for PTO is also less sloping than that for activated carbon.

FIG. 13 shows the capacity retention of pyrene-4,5,9,10-tetraone in an acid electrolyte is 90% after 500 cycles. FIG. 14 shows 92% of the capacity of pyrene-4,5,9,10-tetraone is recovered when galvanostatically charged at 10C. FIG. 15 shows the voltage decrease for pyrene-4,5,9,10-tetraone when increasing the charging rate from 1C to 10C is 20 mV. A fully charged pyrene-4,5,9,10-tetraone electrode quickly recover capacity when exposed to air (FIG. 16). This capability indicates the organic compound can be reversibly oxidized by oxygen, hence enabling oxygen cycle within a typical lead-acid battery.

In some embodiments, a method for fabricating polymer materials that are suitable for use as an electrode in a PbOrg battery includes processing a solution that includes pyrene to form poly[pyrene-4,5,9,10-tetraone (PTO)]. The PTO may then be processed to form PTO(NO₂)₂, which is then processed to form PTO (NH₂)₂. The PTO (NH₂)₂ may be processed to form P(PTO-C₄) that is suitable to be utilized as an electrode in a PbOrg battery.

Organic polymers are shown to be reliable and low-cost negative electrode materials that can be used to replace lead in a LAB. FIGS. 17&18 shows electrochemical performance of poly[pyrene-4,5,9,10-tetraone]. A specific capacity of 250 mAh/g is found, which is four times of that of activated carbon and double of that of Pb. After 150 cycles, no sign of capacity decay can be observed. As a nonlimiting example, in some embodiments, the organic polymer for an electrode

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may be P(PTO-C₄). A nonlimiting embodiment of the synthesis route of P(PTO-C₄) that can be used in the anode is shown in FIG. 19.

As micro hybrid and stop/start technologies require higher charge acceptance, lead acid battery technology may soon be replaced. The technology discussed herein would be a direct replacement for the negative lead electrode currently used in existing lead acid battery manufacturing. This technology will solve both the charge acceptance problem of current LABs and also the low energy and sloping voltage profile of PbC batteries. Finally, this technology may be easily integrated into existing LAB manufacturing processes, which will dramatically reduce time-to-market and cost to scale-up this technology.

Embodiments described herein are included to demonstrate particular aspects of the present disclosure. It should be appreciated by those of skill in the art that the embodiments described herein merely represent exemplary embodiments of the disclosure. Those of ordinary skill in the art should, in light of the present disclosure, appreciate that many changes can be made in the specific embodiments described and still obtain a like or similar result without departing from the spirit and scope of the present disclosure. From the foregoing description, one of ordinary skill in the art can easily ascertain the essential characteristics of this disclosure, and without departing from the spirit and scope thereof, can make various changes and modifications to adapt the disclosure to various usages and conditions. The embodiments described hereinabove are meant to be illustrative only and should not be taken as limiting of the scope of the disclosure.

What is claimed is:

1. A lead acid battery comprising:

a first electrode, wherein the first electrode is a lead compound;

an electrolyte, wherein the electrolyte has a pH value equal to or lower than 2; and

a second electrode comprising at least one organic material, wherein the organic material is a molecular material and comprises at least one cyclic substructure with at least one hydroxyl (C—O—H) group and/or carbonyl (C=O) group, the second electrode has a lead content of 0-10 wt %, the second electrode has a reduction potential of 0.6 V or less versus a normal hydrogen electrode (NHE) in 4.4 M H₂SO₄, and wherein further the organic material undergoes a reversible electrochemical redox reaction between hydroxyl and carbonyl groups during charging or discharging.

2. The battery of claim 1, wherein the second electrode is electrochemically reversible between a C—O—H-containing form and a C=O-containing form or vice versa.

3. The battery of claim 1, wherein the C atoms in the at least one hydroxyl (C—O—H) group and/or carbonyl (C=O) group are part of a cyclic substructure.

4. The battery of claim 1, wherein the cyclic substructure is a carbocycle with carbon atoms forming a ring.

5. The battery of claim 1, wherein the cyclic substructure is a heterocycle with both carbon and non-carbon atoms forming a ring.

6. The battery of claim 1, wherein the cyclic substructure is an aromatic ring.

7. The battery of claim 1, wherein the cyclic substructure further comprises one or more R groups where R₁, R₂, . . . , R₈=M_x, CH₃, CH₂CH₃, CH₂CH₂CH₃, CH(CH₃)₂, C(CH₃)₃, CHCH₂, CCH, OM_x, OCH₃, OCH₂CH₃, OCH₂CH₂CH₃, OCH(CH₃)₂, OC(CH₃)₃, NH₂, NHCH₃, N(CH₃)₂, NHCH₂CH₂CH₃, NHCH(CH₃)₂, NHC(CH₃)₃,

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8. The battery of claim 1, wherein the cyclic substructure is fused with one or more aromatic ring systems.

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1.

1.

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-continued



where $\textcircled{\text{Ar}}$ =

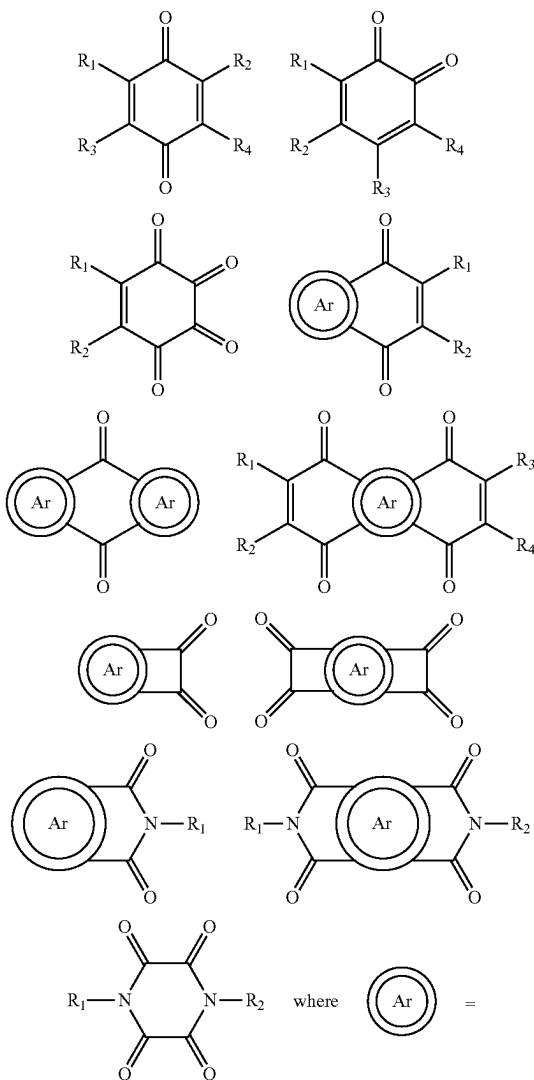
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$A_1 = NR_1$, O, S, and

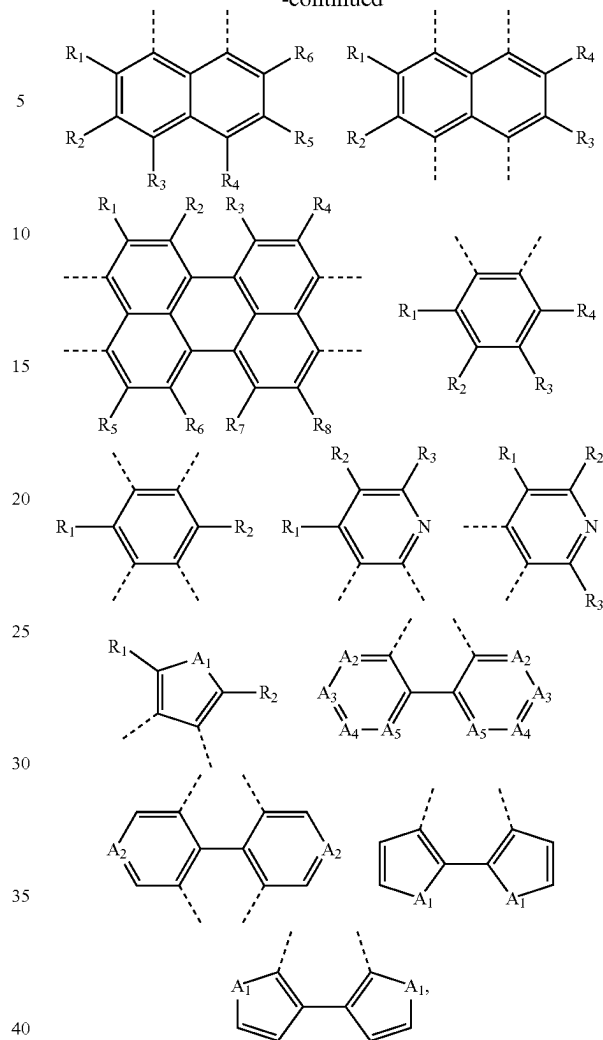
$A_{2, 3, 4, 5} = C, N$.

21. The battery of claim 1, wherein the organic material is selected from:



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-continued



and

$R_{1, 2, 3, 4} = M_x, CH_3, CH_2CH_3, CH_2CH_2CH_3, CH(CH_3)_2, C(CH_3)_3, CHCH_2, CCH, OM_x, OCH_3, OCH_2CH_3, OCH_2CH_2CH_3, OCH(CH_3)_2, OC(CH_3)_3, NH_2, NHCH_3, N(CH_3)_2, NHCH_2CH_2CH_3, NHCH(CH_3)_2, NHC(CH_3)_3, CN, Cl, Br, I, SO_3M_x$.

* * * * *