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[Continued on next page]

(54) Title: LI-ION BATTERY WITH BLENDED ELECTRODE

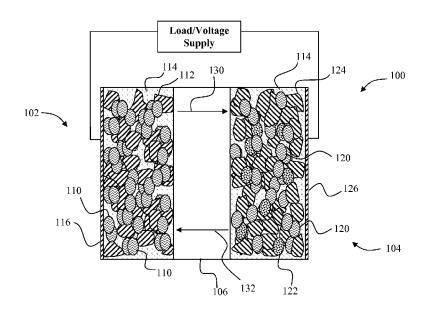


FIG. 1

(57) Abstract: An electrochemical cell (100) with a blended cathode (104) in one embodiment includes a negative electrode (102) including a form of lithium, a positive electrode (104) spaced apart from the negative electrode (102), a separator (106) positioned between the negative electrode (102) and the positive electrode (104), a first active material (120) in the positive electrode (104) including a form of lithium, and a second active (122) material in the positive electrode (104) including a form of sulfur.



TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, Published: EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

- with international search report (Art. 21(3))
- before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments (Rule 48.2(h))

Li-ION BATTERY WITH BLENDED ELECTRODE

Cross-reference is made to U.S. Utility Patent Application Serial No. 12/437,576 entitled "Li-ion Battery with Selective Moderating Material" by John F. Christensen et 5 al., which was filed on May 8, 2009; U.S. Utility Patent Application Serial No. [Attorney Docket No. 1576-0297] entitled "Li-ion Battery with Variable Volume Reservoir" by John F. Christensen et al., which was filed on May 8, 2009; U.S. Utility Patent Application Serial No. [Attorney Docket No. 1576-0299] entitled "Li-ion Battery with Over-charge/Over-discharge Failsafe" by John F. Christensen et al., which was filed on 10 May 8, 2009; U.S. Utility Patent Application Serial No. [Attorney Docket No. 1576-0301] entitled "System and Method for Pressure Determination in a Li-ion Battery" by John F. Christensen et al., which was filed on May 8, 2009; U.S. Utility Patent Application Serial No. [Attorney Docket No. 1576-0302] entitled "Li-ion Battery with Load Leveler" by John F. Christensen et al., which was filed on May 8, 2009; U.S. Utility 15 Patent Application Serial No. [Attorney Docket No. 1576-0303] entitled "Li-ion Battery with Anode Coating" by John F. Christensen et al., which was filed on May 8, 2009; U.S. Utility Patent Application Serial No. [Attorney Docket No. 1576-0304] entitled "Li-ion Battery with Anode Expansion Area" by Boris Kozinsky et al., which was filed on May 8, 2009; U.S. Utility Patent Application Serial No. [Attorney Docket No. 1576-0305] entitled "Li-ion Battery with Porous Silicon Anode" by Boris Kozinsky et al., which was 20 filed on May 8, 2009; U.S. Utility Patent Application Serial No. [Attorney Docket No. 1576-0306] entitled "Li-ion Battery with Rigid Anode Framework" by Boris Kozinsky et al., which was filed on May 8, 2009; U.S. Utility Patent Application Serial No. [Attorney

Docket No. 1576-0308] entitled "System and Method for Charging and Discharging a Liion Battery" by Nalin Chaturvedi et al., which was filed on May 8, 2009; and U.S. Utility Patent Application Serial No. [Attorney Docket No. 1576-0310] entitled "System and Method for Charging and Discharging a Li-ion Battery Pack" by Nalin Chaturvedi et al., which was filed on May 8, 2009, the entirety of each of which is incorporated herein by reference. The principles of the present invention may be combined with features disclosed in those patent applications.

Field of the Invention

10 **[0001]** This invention relates to batteries and more particularly to lithium-ion batteries.

Background

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[0002] Batteries are a useful source of stored energy that can be incorporated into a number of systems. Rechargeable lithium-ion batteries are attractive energy storage systems for portable electronics and electric and hybrid-electric vehicles because of their high specific energy compared to other electrochemical energy storage devices. In particular, batteries with a form of lithium metal incorporated into the negative electrode afford exceptionally high specific energy (in Wh/kg) and energy density (in Wh/L) compared to batteries with conventional carbonaceous negative electrodes.

[0003] When high-specific-capacity negative electrodes such as lithium are used in a battery, the maximum benefit of the capacity increase over conventional systems is realized when a high-capacity positive electrode active material is also used.

Conventional lithium-intercalating oxides (e.g., LiCoO₂, LiNi_{0.8}Co_{0.15}Al_{0.05}O₂, Li_{1.1}Ni_{0.3}Co_{0.3}Mn_{0.3}O₂) are typically limited to a theoretical capacity of ~280 mAh/g (based on the mass of the lithiated oxide) and a practical capacity of 180 to 250 mAh/g. In comparison, the specific capacity of lithium metal is about 3863 mAh/g. The highest 5 theoretical capacity achievable for a lithium-ion positive electrode is 1168 mAh/g (based on the mass of the lithiated material), which is shared by Li₂S and Li₂O₂. Other highcapacity materials including BiF₃ (303 mAh/g, lithiated) and FeF₃ (712 mAh/g, lithiated) are identified in Amatucci, G.G. and N. Pereira, Fluoride based electrode materials for advanced energy storage devices. Journal of Fluorine Chemistry, 2007. 128(4): p. 243-10 262. All of the foregoing materials, however, react with lithium at a lower voltage compared to conventional oxide positive electrodes, hence limiting the theoretical specific energy. The theoretical specific energies of the foregoing materials, however, are very high (> 800 Wh/kg, compared to a maximum of ~500 Wh/kg for a cell with lithium negative and conventional oxide positive electrodes).

- 15 **[0004]** Lithium/sulfur (Li/S) batteries are particularly attractive because of the balance between high specific energy (i.e., >350 Wh/kg has been demonstrated), rate capability, and cycle life (> 50 cycles). Only lithium/air batteries have a higher theoretical specific energy. Lithium/air batteries, however, have very limited rechargeability and are still considered primary batteries.
- 20 [0005] Li/S batteries also have limitations. By way of example, the United States Advanced Battery Consortium has established a goal of > 1000 cycles for batteries used in powering an electric vehicle. Li/S batteries, however, exhibit relatively high capacity fade, thereby limiting the useful lifespan of Li/S batteries.

[0006] One mechanism which may contribute to capacity fade of Li/S batteries is the manner in which the sulfur reacts with lithium. In general, sulfur reacts with lithium ions during battery discharge to form polysulfides (Li_xS), which may be soluble in the electrolyte. These polysulfides react further with lithium (i.e., the value of x increases from 1/4 to 1/3 to 1/2 to 1) until Li₂S₂ is formed, which reacts rapidly to form Li₂S. In Li/S batteries described in the literature, both Li₂S₂ and Li₂S are generally insoluble in the electrolyte. Hence, in a system in which intermediate polysulfides are soluble, each complete cycle consists of soluble-solid phase changes, which may impact the integrity of the composite electrode structure.

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[0007] Specifically, Li₂S may deposit preferentially near the separator when the current through the depth of the positive electrode is non-uniform. Non-uniformity is particularly problematic at high discharge rates. Any such preferential deposition can block pores of the electrode, putting stress on the electronically conducting matrix and/or isolating an area from the composite electrode. All of these processes may lead to capacity fade or impedance rise in the battery.

[0008] Moreover, soluble polysulfides are mobile in the electrolyte and, depending on the type of separator that is used, may diffuse to the negative electrode where the soluble polysulfides may becoming more lithiated through reactions with the lithium electrode. The lithiated polysulfide may then diffuse back through the separator to the positive electrode where some of the lithium is passed to less lithiated polysulfides. This overall shuttle process of lithium from the negative electrode to the positive electrode by polysulfides is a mechanism of self discharge which reduces the cycling efficiency of the battery and which may lead to permanent capacity loss.

[0009] Some attempts to mitigate capacity fade of Li/S batteries rely upon immobilization of the sulfur in the positive electrode via a polymer encapsulation or the use of a high-molecular weight solvent system in which polysulfides do not dissolve. In these batteries, the phase change and self-discharge characteristics inherent in the above-described Li/S system are eliminated. These systems have a higher demonstrated cycle life at the expense of high rate capability and capacity utilization.

[0010] What is needed therefore is a battery that exhibits the high energy density of a Li/S cathode while providing high rate capability and capacity utilization. A further need exists for a battery that exhibits reduced capacity fade due to internal discharge.

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Summary

[0011] In accordance with one embodiment, an electrochemical cell with a blended cathode includes a negative electrode including a form of lithium, a positive electrode spaced apart from the negative electrode, a separator positioned between the negative electrode and the positive electrode, a first active material in the positive electrode including a form of lithium, and a second active material in the positive electrode including a form of sulfur.

[0012] In accordance with another embodiment, an electrochemical cell includes a negative electrode, a positive electrode spaced apart from the negative electrode, a separator positioned between the negative electrode and the positive electrode, a first active material in the positive electrode including a form of sulfur, and a second active material in the positive electrode that does not include a form of sulfur.

Brief Description of the Drawings

[0013] FIG. 1 depicts a schematic of a battery system including an electrochemical cell with one electrode including a form of lithium and another electrode including an active material with a form of lithium and another active material with a form of sulfur;

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[0014] FIG. 2 depicts a schematic of a battery system including an electrochemical cell with one electrode including a form of lithium and a fully dense electrode including an active material with a form of lithium along with active material with a form of sulfur.

10 <u>Description</u>

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[0015] For the purposes of promoting an understanding of the principles of the invention, reference will now be made to the embodiments illustrated in the drawings and described in the following written specification. It is understood that no limitation to the scope of the invention is thereby intended. It is further understood that the present invention includes any alterations and modifications to the illustrated embodiments and includes further applications of the principles of the invention as would normally occur to one skilled in the art to which this invention pertains.

[0016] FIG. 1 depicts a lithium-ion cell 100, which includes a negative electrode 102, a positive electrode 104, and a separator region 106 between the negative electrode 102 and the positive electrode 104. The negative electrode 102 includes active materials 110 into which lithium can be inserted, inert materials 112, electrolyte 114 and a current collector 116.

[0017] The negative electrode 102 may be provided in various alternative forms. The negative electrode 102 may incorporate dense Li metal or a conventional porous composite electrode (e.g., graphite particles mixed with binder). Incorporation of Li metal is desired since the Li metal affords a higher specific energy than graphite.

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- [0018] The positive electrode 104 includes active materials 120 and 122 into which lithium can be inserted, inert materials 124, the electrolyte 114 and a current collector 126. The separator region 106 includes an electrolyte with a lithium cation and serves as a physical and electrical barrier between the negative electrode 102 and the positive electrode 104 so that the electrodes are not electronically connected within the cell 100. The active material 120 includes a form of sulfur and may be entirely sulfur. The active material 122 does not incorporate sulfur and preferably incorporates a form of lithium such as lithium titanium oxide, or $\text{Li}_{4+x}\text{Ti}_5\text{O}_{12}$. The active materials 120 and 122 are preferably selected to be materials which react with lithium in similar voltage windows. While the active materials 120 and 122 are depicted in a similar fashion in FIG. 1, the sulfur chemistry may react in a substantially different manner. For example, sulfur tends to dissolve into the electrolyte as it becomes lithiated. The sulfur then becomes more
- [0019] Under moderate power demands, the lithium-ion cell 100 operates in a manner similar to the lithium-ion battery cell disclosed in U.S. Patent Application Serial No.

 11/477,404, filed on June 28, 2006, the contents of which are herein incorporated in their entirety by reference. In general, electrons are generated at the negative electrode 102 during discharging and an equal amount of electrons are consumed at the positive electrode 104 as lithium and electrons move in the direction of the arrow 130 of FIG. 1.

lithiated and then re-precipitates on the carbon surface.

[0020] In the ideal discharging of the cell 100, the electrons are generated at the negative electrode 102 because there is extraction via oxidation of lithium ions from the active material 110 of the negative electrode 102, and the electrons are consumed at the positive electrode 104 because there is reduction of lithium ions predominantly into the active material 122 of the positive electrode 104. Oxidation of lithium ions occurs predominantly in the active material 122 because the active material 122 is a material that reacts with and/or incorporates lithium into its structure more rapidly than the first active material 120. During discharging, the reactions are reversed, with lithium and electrons moving in the direction of the arrow 132.

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- 10 [0021] In various applications, however, the cell 100 may be subjected to sudden increases in power demand. When used in vehicles, for example, acceleration of the vehicle places an increased demand on the cell 100. As the demand exceeds the rate of Li reduction supported by the active material 120, the reduction of lithium ions into the active material 122 of the positive electrode 104 increases.
- 15 [0022] Accordingly, for moderate power demand, power is provided primarily through reactions including the active material 122 which does not implicate the same mechanisms of capacity fade implicated by the active material 120. Cycling of the active material 120 is thus reduced, thereby minimizing the deleterious effects of rapid lithiation of the sulfur. The balance of active material 120 and active material 122 may thus be adjusted to provide the surge capability desired for a particular application while maximizing the life of the cell 100.
 - [0023] Another active material may also be incorporated into a sulfur cathode to simplify state-of-charge determination. Li_xS₈ has a characteristic plateau over a

particular SOC range (x > 4), and it is difficult to ascertain the SOC of the material from the voltage. By combining Li_xS₈ with a material that has a sloping potential in the same potential window, SOC determination for the cell based upon the cell voltage is simplified. Lithium titanate, however, has a flat potential and would not provide simplified SOC determination.

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[0024] While one form of a negative electrode 102 and a positive electrode 104 is depicted in FIG. 1, a blended electrode 104 may be provided in variety of alternative embodiments. Such embodiments may include a dense Li negative electrode or a porous composite negative electrode. The blended positive electrode 104 may likewise be a dense electrode or a porous electrode

[0025] The manner in which a particular negative electrode 102/ positive electrode 104 is manufactured can vary based upon the particular form selected for the negative electrode 102 and positive electrode 104. Lithium titanate, for example, is a ceramic material. Accordingly, the lithium titanate can be integrated as a compact (i.e., nonporous) layer. As a compact layer, however, winding of the cell layers for a wound cell type battery may present challenges. Alternatively, the positive electrode 104 may consist of a porous composite (just as in a conventional electrode) that consists of small lithium titanate and sulfur particles and binder (e.g., polyvinyl difluoride). The pores in this embodiment may be filled with a Li-cation containing electrolyte (just as in any porous electrode).

[0026] A porous lithium titanate layer, however, typically requires a substrate in order to be handled. For example, conventional porous electrodes are deposited as slurries on a current collector, the solvent of the slurry is removed (leaving pores in place

of the solvent), and the composite electrode is "calendared" (compressed with a roll press) to achieve the desired thickness and porosity and to improve particle-to-particle contact. Usually the electrodes are double sided (i.e., with porous electrodes on either side of the current collector).

- 5 [0027] A porous-composite negative electrode/ porous-composite lithium titanate/LiS electrode may be obtained by forming the electrode in a conventional manner followed by a further deposition of slurry including the lithium titanate and LiS. Subsequently, the solvent in the lithium titanate/LiS slurry may be evaporated and the formed layers calendared.
- 10 [0028] FIG. 2 depicts an alternative embodiment of a lithium-ion cell 200 which operates in substantially the same manner as the lithium-ion cell 100. The lithium-ion cell 200, however, is a solid state cell which includes a negative electrode 202, a positive electrode 204, and an electrolyte layer 206 between the negative electrode 202 and the positive electrode 204. The negative electrode 202 includes an active layer 210 including inert materials and active materials into which lithium can be inserted, and a current collector 216.
 - [0029] The positive electrode 204 includes an active layer 220 including inert materials and active materials into which lithium can be inserted, and a current collector 222. The electrolyte layer 206 supplies lithium cation and serves as an electrical barrier between the negative electrode 202 and the positive electrode 204 so that the electrodes are not electronically connected within the cell 200. The active material used in the active layer 220 includes a form of sulfur and may be entirely sulfur. Another active

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material within the active layer 220 does not incorporate sulfur and preferably incorporates a form of lithium such as lithium titanium oxide, or $Li_{4+x}Ti_5O_{12}$.

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[0030] In a further embodiment, one of the anode and the cathode may be solid while the other of the anode and the cathode is porous. By way of example, the lithium-ion cell 200 may be modified to replace the solid positive electrode 204 with the porous positive electrode 104 of FIG. 1.

[0031] While the invention has been illustrated and described in detail in the drawings and foregoing description, the same should be considered as illustrative and not restrictive in character. It is understood that only the preferred embodiments have been presented and that all changes, modifications and further applications that come within the spirit of the invention are desired to be protected.

Claims

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Claim 1. An electrochemical cell, comprising:

- a negative electrode including a form of lithium;
- 5 a positive electrode spaced apart from the negative electrode;
 - a separator positioned between the negative electrode and the positive electrode;
 - a first active material in the positive electrode including a form of lithium; and
 - a second active material in the positive electrode including a form of sulfur.
- 10 Claim 2. The electrochemical cell of claim 1, wherein the second active material includes $\text{Li}_x S_8$, and $0 \le x \le 16$.
 - Claim 3. The electrochemical cell of claim 2, wherein the first active material includes a lithium oxide.
 - Claim 4. The electrochemical cell of claim 2, wherein the first active material includes $Li_{4+x}Ti_5O_{12}$.
- Claim 5. The electrochemical cell of claim 1, wherein the first active material includes a lithium oxide.
 - Claim 6. The electrochemical cell of claim 1, wherein the first active material includes $Li_{4+x}Ti_5O_{12}$.

Claim 7. An electrochemical cell, comprising:

a negative electrode;

sulfur.

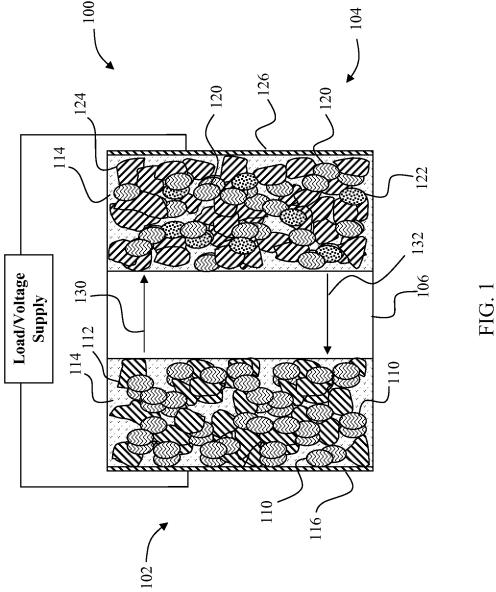
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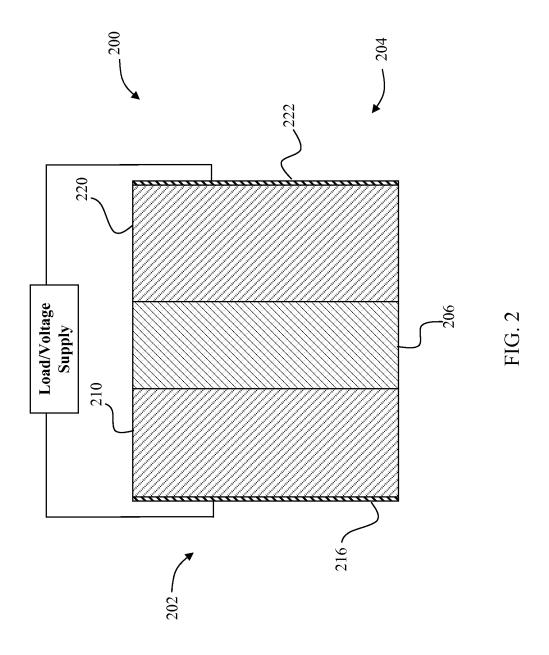
- a positive electrode spaced apart from the negative electrode;
- a separator positioned between the negative electrode and the positive electrode;
 - a first active material in the positive electrode including a form of sulfur; and
 - a second active material in the positive electrode that does not include a form of
- 10 Claim 8. The electrochemical cell of claim 7, wherein the first active material includes $\text{Li}_x S_8$, and $0 \le x \le 16$.
 - Claim 9. The electrochemical cell of claim 8, wherein the second active material includes a form of lithium.
 - Claim 10. The electrochemical cell of claim 9, wherein the second active material includes $\text{Li}_{4+x}\text{Ti}_5\text{O}_{12}$.
- Claim 11. The electrochemical cell of claim 9, wherein the second active material includes lithium oxide.
 - Claim 12. The electrochemical cell of claim 7, wherein the negative electrode includes a form of lithium.

Claim 13. The electrochemical cell of claim 7, wherein the second active material includes a form of lithium.

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- Claim 14. The electrochemical cell of claim 13, wherein the second active material includes $Li_{4+x}Ti_5O_{12}$.
- Claim 15. The electrochemical cell of claim 13, wherein the second active material includes lithium oxide.





INTERNATIONAL SEARCH REPORT

International application No PCT/US2010/034051

A. CLASSIFICATION OF SUBJECT MATTER INV. H01M4/58 H01M1 H01M10/0525 H01M4/485 H01M4/131 H01M4/136 ADD. H01M4/36According to International Patent Classification (IPC) or to both national classification and IPC B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) H01M Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal C. DOCUMENTS CONSIDERED TO BE RELEVANT Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. χ US 2007/292764 A1 (SOMA MASANORI [JP] ET 1,5,6 AL) 20 December 2007 (2007-12-20) paragraph [0008] paragraph [0013] paragraph [0015] paragraph [0038] - paragraph [0040] paragraph [0060] - paragraph [0061] WO 97/01191 A2 (POLYSTOR CORP [US]) 9 January 1997 (1997-01-09) page 13, line 19 - line 24 X 1,5,6 claims 15, 18 -/--Further documents are listed in the continuation of Box C. See patent family annex. Special categories of cited documents: "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone filing date document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such docu-"O" document referring to an oral disclosure, use, exhibition or other means ments, such combination being obvious to a person skilled in the art. document published prior to the international filing date but later than the priority date claimed "&" document member of the same patent family Date of the actual completion of the international search Date of mailing of the international search report 2 September 2010 10/09/2010 Name and mailing address of the ISA/ Authorized officer European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016 Fernandez Morales, N

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2010/034051

		PCT/US2010/034051							
C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT									
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.							
Х	EP 1 143 547 A1 (MATSUSHITA ELECTRIC IND CO LTD [JP]) 10 October 2001 (2001-10-10) paragraph [0009] - paragraph [0010] paragraph [0023] paragraph [0040] examples 1-4	1,2							
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International application No. PCT/US2010/034051

INTERNATIONAL SEARCH REPORT

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)								
This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:								
1. Claims Nos.: because they relate to subject matter not required to be searched by this Authority, namely:								
Claims Nos.: because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:								
Claims Nos.: because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).								
Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)								
This International Searching Authority found multiple inventions in this international application, as follows:								
see additional sheet								
As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.								
As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of additional fees.								
3. As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:								
4. No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.: 1-6								
Remark on Protest The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee. The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.								
No protest accompanied the payment of additional search fees.								

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-6

An electrochemical cell, comprising: a negative electrode including a form of lithium; a positive electrode spaced apart from the negative electrode; a separator positioned between the negative electrode and the positive electrode; a first active material in the positive electrode including a form of lithium; and a second active material in the positive electrode including a form of sulfur.

2. claims: 7-15

An electrochemical cell, comprising: a negative electrode; a positive electrode spaced apart from the negative electrode; a separator positioned between the negative electrode and the positive electrode; a first active material in the positive electrode including a form of sulfur; and a second active material in the positive electrode that does not include a form of sulfur.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
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