There are provided processes for preparing compounds of formula (I) AMPO₄. The processes comprise reacting a compound or formula (II) AₙHₙPO₄ₙ and a compound of formula (III) MXₙ, in the presence of an acid scavenger adapted to at least substantially solubilize the compound of formula (II) and the compound of formula (III), at a predetermined temperature, and adapted to be converted into a protic ionic liquid upon protonation. A, M, and X can be various chemical entities and n can be 1 or 2. A process for preparing electrodes is also provided.
THE TECHNICAL FIELD

[0001] The present document relates to improvements in the field of preparation of inorganic compounds. In particular, it relates to the preparation of inorganic phosphates such as phospho-olivine type compounds.

BACKGROUND OF THE INVENTION

[0002] So far, several attempts have been made so as to provide an efficient process for preparing inorganic phosphates such as phospho-olives compounds which are used in lithium batteries. These processes often required costly reagents or comprised several steps. Moreover, the processes that have been proposed so far are often very costly since they imply a high energy consumption.

[0003] It would therefore be highly desirable to be provided with a process that would overcome the above-mentioned drawbacks.

SUMMARY OF THE INVENTION

[0004] According to one aspect, there is provided a process for preparing a compound of formula (I):

\[
\text{AMPO}_4 \quad (I)
\]

wherein

A is chosen from \( \text{H}^+, \text{Li}^+, \text{Na}^+, \text{K}^+, \text{and Cs}^+; \) and

\( \text{M} \) is chosen from \( \text{Be}^{2+}, \text{Mg}^{2+}, \text{Ba}^{2+}, \text{Ca}^{2+}, \text{Sr}^{2+}, \text{Fe}^{2+}, \text{Co}^{2+}, \text{Ni}^{2+}, \text{Mn}^{2+}, \text{Zn}^{2+}, \text{Cu}^{2+}, \) and \( \text{Mo}^{2+}; \)

the process comprising reacting a compound of formula (II) with a compound of formula (III):

\[
\text{AMPO}_4 \quad (I)
\]
AH$_2$PO$_4$ (II)

MX$_n$ (III)

wherein

A is as previously defined;

M is as previously defined;

X is chosen from F$^-$, Cl$^-$, Br$^-$, I$^-$, CN$^-$, CH$_3$COO$^-$, CF$_3$SO$_3^-$, NO$_3^-$, OCN$^-$, SCN$^-$, (CN)$_2$N$^-$, CO$_3^{2-}$, ClO$_3^-$, CrO$_4^{2-}$, BrO$_3^-$, Cr$_2$O$_7^{2-}$, S$_2$O$_4^{2-}$, S$_4$O$_6^{2-}$, S$_2$O$_3^{2-}$, SO$_4^{2-}$, MnO$_4^-$, S$^{2-}$, ClO$^-$, and C$_2$O$_4^{2-}$; and

n is 1 or 2,

in the presence of an acid scavenger adapted to at least substantially solubilize the compound of formula (II) and the compound of formula (III), at a predetermined temperature, and adapted to be converted into a protic ionic liquid upon protonation.

[0005] According to another embodiment, there is provided a process for preparing an electrode comprising:

- reacting a compound of formula (II) with a compound of formula (III):

AH$_2$PO$_4$ (II)

MX$_n$ (III)

wherein

A is chosen from H$^+$, Li$^+$, Na$^+$, K$^+$, and Cs$^+$; and
M is chosen from Be\textsuperscript{2+}, Mg\textsuperscript{2+}, Ba\textsuperscript{2+}, Ca\textsuperscript{2+}, Sr\textsuperscript{2+}, Fe\textsuperscript{2+}, Co\textsuperscript{2+}, Ni\textsuperscript{2+}, Mn\textsuperscript{2+}, Zn\textsuperscript{2+}, Cu\textsuperscript{2+}, and Mo\textsuperscript{2+};

X is chosen from F\textsuperscript{-}, Cl\textsuperscript{-}, Br\textsuperscript{-}, I\textsuperscript{-}, CN\textsuperscript{-}, CH\textsubscript{3}COO\textsuperscript{-}, CF\textsubscript{3}SO\textsubscript{3}\textsuperscript{-}, NO\textsubscript{3}\textsuperscript{-}, OCN\textsuperscript{-}, SCN\textsuperscript{-}, (CN)\textsubscript{2}N\textsuperscript{-}, CO\textsubscript{3}\textsuperscript{2-}, Cl\textsubscript{O}\textsubscript{3}\textsuperscript{-}, CrO\textsubscript{4}\textsuperscript{2-}, BrO\textsubscript{3}\textsuperscript{-}, Cr\textsubscript{2}O\textsubscript{7}\textsuperscript{2-}, S\textsubscript{2}O\textsubscript{4}\textsuperscript{2-}, S\textsubscript{4}O\textsubscript{6}\textsuperscript{2-}, S\textsubscript{2}O\textsubscript{3}\textsuperscript{2-}, SO\textsubscript{4}\textsuperscript{2-}, MnO\textsubscript{4}\textsuperscript{-}, S\textsuperscript{2-}, ClO\textsuperscript{-}, and C\textsubscript{2}O\textsubscript{4}\textsuperscript{2-}; and

n is 1 or 2,

in the presence of an acid scavenger adapted to at least substantially solubilize the compound of formula (II) and the compound of formula (III), at a predetermined temperature, and adapted to be converted into a protic ionic liquid upon protonation, thereby obtaining a composition comprising a compound of formula (I) AMPO\textsubscript{4} in which A and M are as previously defined, the protonated acid scavenger, and optionally the unprotonated acid scavenger;

- depositing the composition on a substrate;
- annealing the composition deposited on the substrate so as to obtain the electrode.

[0006] For example, the acid scavenger can be liquid at room temperature. It can also be liquid at a temperature of about 0 to about 35 °C. For example, the compound of formula (II) can be reacted with the compound of formula (III) at a temperature lower than 200 °C, lower than 150 °C, lower than 100 °C, or lower than 60 °C. According to another example, they can be reacted together at room temperature, at a temperature of about 15 °C to about 60 °C, or at a temperature of about 20 °C to about 35 °C.

[0007] For example, the acid scavenger can be a compound of formula (IV):
wherein

R is chosen from hydrogen atom, C1-C12 alkyl, C3-C12 cycloalkyl, C1-C12 heterocyclyl, C2-C12 alkenyl, C2-C12 alkynyl, C6-C12 aryl, C6-C20 aralkyl, C6-C20 alkylaryl, and C1-C12 heteroaryl.

[0008] For example, R can be a polymerizable moiety. The polymerizable moiety can comprise at least one polymerizable entity chosen from a double bond, a triple bond, a carbonyl, a free radical acceptor, and a polymerizable cyclic compound. For example, the polymerizable cyclic compound can be an epoxide, an aziridine, a cyclopropene, or a thiophene. For example, the polymerizable moiety can be chosen from a cyclopropenyl, a C4-C20 alkylcyclopropenyl, a C4-C20 cyclopropenylalkyl, an epoxide, a C3-C20 alkyloxirane, a C3-C20 oxiranealkyl, C2-C20 alkenyl, and C2-C20 alkynyl. The polymerizable moiety can also be a C2-C12 alkenyl, or a C2-C12 alkynyl.

[0009] For example, the compound of formula (I) can be chosen from LiFePO4, LiBePO4, LiMgPO4, LiBaPO4, LiCaPO4, LiSrPO4, LiCoPO4, LiNiPO4, LiMnPO4, LiZnPO4, LiCuPO4, and LiMoPO4.

[0010] For example, the compound of formula (III) can be chosen from FeCl2, FeBr2, FeI2, FeF2, Fe(CN)2, Fe(SCN)2, FeSO4, Fe(CH3COO)2, and FeC2O4.
For example, a super valent metal salt chosen from Nb$^{5+}$, Al$^{3+}$, Ti$^{4+}$, Mg$^{2+}$, W$^{6+}$, and Zr$^{4+}$ can be added to the compound of formula (II), the compound of formula (III), and the acid scavenger. A carbon source can also be optionally added to the compound of formula (II), the compound of formula (III), and the acid scavenger. The carbon source can be carbon black or a polymer or rich carbon organic compound which is soluble in the acid scavenger. The carbon source can also be nanoparticles of carbon black or carbon nanotubes (multi-wall carbon nanotubes or single-wall carbon nanotubes). An organic solvent can also be further added to the compound of formula (II), the compound of formula (III), and the acid scavenger. The organic solvent can be chosen from acetonitrile, dimethylformamide, tetrahydrofuran, dimethoxymethane, and mixtures thereof.

According to another aspect, there is provided a method of scavenging protons, the method comprising contacting the protons with a compound of formula (IV):

```
R           R
\ H N R N
```

(IV)

wherein

R is chosen from hydrogen atom, C1-C12 alkyl, C3-C12 cycloalkyl, C1-C12 heterocyclic, C2-C12 alkenyl, C2-C12 alkynyl, C6-C12 aryl, C6-C20 aralkyl, C6-C20 alkaryl, and CrC12 heteroaryl.

According to another aspect, there is provided the use of a compound of formula (IV):
wherein

\[ R \text{ is chosen from hydrogen atom, } C_1-C_{12} \text{ alkyl, } C_3-C_2 \text{ cycloalkyl, } C_1-C_{12} \text{ heterocyclyl, } C_2-C_{12} \text{ alkenyl, } C_2-C_{12} \text{ alkynyl, } C_6-C_{12} \text{ aryl, } C_6-C_{20} \text{ aralkyl, } C_6-C_{20} \text{ alkylaryl, and } C_1-C_{12} \text{ heteroaryl.} \]

as an acid scavenger in the preparation of a phospho-olivine type compound.

[0014] According to another aspect, there is provided a method of using a compound of formula (IV)

\[ \text{wherein} \]

\[ R \text{ is chosen from hydrogen atom, } C_1-C_{12} \text{ alkyl, } C_3-C_{12} \text{ cycloalkyl, } C_1-C_{12} \text{ heterocyclyl, } C_2-C_{12} \text{ alkenyl, } C_2-C_{12} \text{ alkynyl, } C_6-C_{12} \text{ aryl, } C_6-C_{20} \text{ aralkyl, } C_6-C_{20} \text{ alkylaryl, and } C_1-C_{12} \text{ heteroaryl,} \]

the method comprising using the compound of formula (IV) as an acid scavenger in the preparation of a phospho-olivine type compound.

[0015] For example, the compound of formula (IV) can be contacted with protons generated during preparation of a phospho-olivine type compound. The compound of formula (IV) can further be used as a solvent.
during preparation of the phospho-olivine type compound. For example, the phospho-olivine type compound can be chosen from LiFePO₄, LiBePO₄, LiMgPO₄, LiBaPO₄, LiCaPO₄, LiSrPO₄, LiCoPO₄, LiNiPO₄, LiMnPO₄, LiZnPO₄, LiCuPO₄, and LiMoPO₄.

According to another aspect, there is provided a method for preparing LiFePO₄ comprising reacting together a lithium source, an iron source, and a phosphorus source, the improvement wherein the lithium source and the phosphorous source is LiH₂PO₄, and the iron source is FeXₙ in which X is chosen from F⁻, Cl⁻, Br⁻, I⁻, CN⁻, CH₃COO⁻, CF₃SO₃⁻, NO₃⁻, OCN⁻, SCN⁻, (CN)₂N⁻, CO₃²⁻, ClO₃⁻, CrO₄²⁻, BrO₃⁻, Cr₂O₇²⁻, S₂O₄²⁻, S₄O₆²⁻, S₂O₃²⁻, SO₄²⁻, MnO₄⁻, S²⁻, ClO⁻, and C₂O₄²⁻; and n is 1 or 2. For example, LiH₂PO₄ can be reacted with FeXₙ at a temperature lower than 200 °C, lower than 150 °C, lower than 100 °C, or lower than 60 °C. According to another example, LiH₂PO₄ can be reacted with FeXₙ at room temperature, at a temperature of about 15 °C to about 60 °C, or at a temperature of about 20 °C to about 35 °C. For example, in such a method a compound of formula (IV), as previously defined, can be used as a proton scavenger. The compound of formula (IV) can also used as a solvent.

According to another example, the compound of formula (IV) can be

![Diagram of compound](symbol)

According to another example, the compound of formula (IV) can be

![Diagram of compound](symbol)
According to another example, the compound of formula (IV) can be

\[
\begin{align*}
\text{N} & \quad \text{N} \\
\end{align*}
\]

According to another aspect, there is provided a process for preparing a compound of formula (Ia):

\[
AM^1_1M^2_{1,T}\text{PO}_4 \quad \text{(Ia)}
\]

wherein

- \( A \) is chosen from \( H^+, \text{Li}^+, \text{Na}^+, \text{K}^+, \text{and Cs}^+ \); and

- \( M^1 \) is chosen from \( \text{Be}^{2+}, \text{Mg}^{2+}, \text{Ba}^{2+}, \text{Ca}^{2+}, \text{Sr}^{2+}, \text{Fe}^{2+}, \text{Co}^{2+}, \text{Ni}^{2+}, \text{Mn}^{2+}, \text{Zn}^{2+}, \text{Cu}^{2+}, \text{and Mo}^{2+} \);

- \( M^2 \) is chosen from \( \text{Be}^{2+}, \text{Mg}^{2+}, \text{Ba}^{2+}, \text{Ca}^{2+}, \text{Sr}^{2+}, \text{Fe}^{2+}, \text{Co}^{2+}, \text{Ni}^{2+}, \text{Mn}^{2+}, \text{Zn}^{2+}, \text{Cu}^{2+}, \text{and Mo}^{2+} \); and

- \( T \) has a value of 0 to 1,

the process comprising reacting a compound of formula (II) with a compound of formula (IIia) and a compound of formula (IIib):

\[
\begin{align*}
\text{AH}_2\text{PO}_4 & \quad \text{(H)} \\
M^1_1X_1\pi_1 & \quad \text{(IIia)} \\
M^2_2X_2\pi_2 & \quad \text{(IIib)} \\
\end{align*}
\]

wherein
A is as previously defined;

\( M^1 \) and \( M^2 \) are as previously defined;

\( X^1 \) is chosen from \( P^-, Cl^-, Br^-, I^-, CN^-, CH_3COO^-, CF_3SO_3^-, NO_3^-, OCN^-, SCN^-, (CN)_2N^-, CO_3^{2-}, ClO_3^-, CrO_4^{2-}, BrO_3^-, Cr_2O_7^{2-}, S_2O_4^{2-}, S_4O_6^{2-}, S_2O_3^{2-}, SO_4^{2-}, MnO_4^-, S^2-, ClO^- \), and \( C_2O_4^{2-} \);

\( X^2 \) is chosen from \( F^-, Cl^-, Br^-, I^-, CN^-, CH_3COO^-, CF_3SO_3^-, NO_3^-, OCN^-, SCN^-, (CN)_2N^-, CO_3^{2-}, ClO_3^-, CrO_4^{2-}, BrO_3^-, Cr_2O_7^{2-}, S_2O_4^{2-}, S_4O_6^{2-}, S_2O_3^{2-}, SO_4^{2-}, MnO_4^-, S^2-, ClO^- \), and \( C_2O_4^{2-} \);

\( n^1 \) is 1 or 2; and

\( n^2 \) is 1 or 2,

in the presence of an acid scavenger adapted to at least substantially solubilize the compound of formula (II), the compound of formula (IIia), and the compound of formula (IIb), at a predetermined temperature, and adapted to be converted into a protic ionic liquid upon protonation.

[0021] Alternatively, \( T \) can have a value comprised between 0 and 1.

[0022] The person skilled in the art would clearly recognize that the process for preparing compounds of formula (Ia) is similar to the process for preparing compounds of formula (I). In fact, the particular embodiments and examples previously mentioned concerning the process for preparing compounds of formula (I), when possible, can all be applied to the process for preparing compounds of formula (Ia).

[0023] The term "alkyl" as used herein means straight and/or branched chain, saturated alkyl radicals and can include, for example, methyl, ethyl, propyl, isopropyl, n-butyl, s-butyl, isobutyl, t-butyl, 2,2-dimethylbutyl, n-pentyl, 2-methylpentyl, 3-methylpentyl, 4-methylpentyl, n-hexyl and the like.
[0024] The term "heteroaryl" has used herein refers to an aromatic
cyclic or fused polycyclic ring system having at least one heteroatom selected
from N, O, and S. For example, the heteroaryl group can be furyl, thienyl,
pyridyl, quinolinyl, isoquinolinyl, indolyl, isoindolyl, triazolyl, pyrrolyl, tetrazolyl,
imidazolyl, pyrazolyl, oxazolyl, thiazolyl, benzofuranyl, benzothiophenyl,
carbazolyl, benzoxazolyl, pyrimidiny1, benzimidazolyl, quinoxaliny1,
benzothiazolyl, naphthyridiny1, isoaxazolyl, isothiazolyl, puriny1, quinazoliny1,
and so on.

[0025] The term "heterocyclyl" includes non-aromatic rings or ring
systems that contain at least one ring having at least one hetero atom (such
as nitrogen, oxygen or sulfur). For example, this term can include all of the
fully saturated and partially unsaturated derivatives of the above mentioned
heteroaryl groups. Exemplary heterocyclic groups include pyrrolidinyl,
tetrahydrofurany1, morpholiny1, thiomorpholiny1, piperidiny1, piperaziny1,
thiazolidinyl, isothiazolidinyl, and imidazolidinyl.

[0026] The term "cycloalkyl" has used herein refers to a hydrocarbon
ring which may contain or not double bonds.

BRIEF DESCRIPTION OF DRAWINGS

[0027] In the following drawings, which represent by way of example only,
various embodiments of the invention:

Fig. 1 shows a X-ray diffraction (XRD) pattern of a LiFePO₄ sample prepared
in accordance with a process as previously defined;

Fig. 2 shows a Scanning Electron Microscope (SEM) image of the sample
analyzed in Fig. 1, wherein the scale is 500 nm; and

Fig. 3 shows an Energy Dispersive X-ray Spectroscopy (EDX) graph of the
sample analyzed in Fig. 1.

DETAILED DESCRIPTION OF VARIOUS EMBODIMENTS
Further features and advantages will become more readily apparent from the following non-limiting examples.

Examples

Example 1: Synthesis of LiFePO$_4$

Fe(CH$_3$COO)$_2$ (1.73 g, 10 mmol) was dissolved in 50 mL of anhydrous N-methylimidazole (NMI) in a 250 mL two-neck flask under argon at room temperature. LiH$_2$PO$_4$ (1.03 g, 10 mmol, 1 equiv) was dissolved in 50 mL of NMI at room temperature and was added dropwise to the solution of Fe(CH$_3$COO)$_2$ over a period of 15 minutes under argon. The resulting solution was stable against aggregation. The mixture was then heated during 2 hours at 600°C. Then, the grey particles obtained were filtered and dried at room temperature under vacuum overnight. The LiFePO$_4$ particles can eventually be annealed. The sample obtained in Example 1 was analyzed by XRD, SEM, and EDX (see Figs. 1 to 3) after being annealed at 500 0°C during 1 hour.

The lattice constants were experimentally calculated by using the distances $d_{hkil}$ between the reticular plans $(101)$, $(211)$ and $(311)$ according to an equation appropriate for an orthorhombic system.

Table 1. Experimental and theoretical lattice constants, a/b and c/b ratios, and unit cell volume of the LiFePO$_4$ of Example 1:

<table>
<thead>
<tr>
<th></th>
<th>a</th>
<th>b</th>
<th>c</th>
<th>a/b</th>
<th>c/b</th>
<th>Volume</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Experimental</strong></td>
<td>10.302</td>
<td>6.040</td>
<td>4.624</td>
<td>1.705</td>
<td>0.705</td>
<td>287.72</td>
</tr>
<tr>
<td><strong>Theoretical</strong>*</td>
<td>10.332</td>
<td>6.010</td>
<td>4.692</td>
<td>1.719</td>
<td>0.780</td>
<td>291.35</td>
</tr>
</tbody>
</table>

* : values taken from ICSD datasheet

By comparing the experimental results and the theoretical values taken from the ICSD data sheet, it can be seen that such values are very similar. The experimental crystallinity diameter is 33.0 nm.
It was calculated by using the Debye-Scherrer equation:

\[
diameter = \frac{K \lambda}{B \cos \theta}
\]

K = 0.9; \( \lambda \) is the wavelength of the radiation source \((\lambda = 1.7890 \text{ Å})\); B is the width at 50% of the height of the peak (rad) of the (101) plan.

As it can be seen from the SEM image of Fig. 2, spherical nanoparticles having a diameter of about 50 nm have been prepared.

The results shown in Fig. 3 and in Table 2 clearly demonstrate that the stoichiometry of the LiFePO\(_4\) particles obtained in Example 1. In fact, the observed ratio Fe : P was 1 : 1.1

Table 2. EDX Spectrometry of LiFePO\(_4\) (coupled to SEM via an X-Ray detector).

<table>
<thead>
<tr>
<th>Element</th>
<th>m/m %</th>
<th>Atom %</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>32.85</td>
<td>47.19</td>
</tr>
<tr>
<td>P</td>
<td>11.97</td>
<td>6.23</td>
</tr>
<tr>
<td>Cl</td>
<td>0.41</td>
<td>0.2</td>
</tr>
<tr>
<td>Fe</td>
<td>17.59</td>
<td>5.43</td>
</tr>
</tbody>
</table>

Ratio Fe : P 1 : 1.1

Example 2: Synthesis of LiMnPO\(_4\)

Mn(CH\(_3\)COO)\(_2\) (1.73 g, 10 mmol) was dissolved in 50 mL of anhydrous N-methylimidazole (NMI) in a 250 mL two-neck flask under argon at room temperature. LiH\(_2\)PO\(_4\) (1.03 g, 10 mmol, 1 equiv) was dissolved in 50 mL NMI at room temperature and was added dropwise to the solution of Mn(CH\(_3\)COO)\(_2\) over a period of 15 minutes under argon. The resulting solution was stable against aggregation. The mixture was then heated during 2 hours at a temperature of 60°C. Then, the grey particles were filtered and dried at room temperature under vacuum overnight. The LiMnPO\(_4\) particles can
eventually be annealed. The sample obtained in Example 2 was analyzed by XRD and XPS.

When the process is used for preparing LiFePO$_4$, the reaction scheme can be as follows:

\[
\text{LiH}_2\text{PO}_4 + \text{FeX}_2 \rightarrow \text{LiFePO}_4 + 2 \text{N} \equiv \text{N} \quad \text{(2 equivalents)}
\]

The reaction scheme can also be as follows:

\[
\text{LiH}_2\text{PO}_4 + \text{FeX}_2 + \text{N} \equiv \text{N} \rightarrow \text{LiFePO}_4 + 2 \text{N} \equiv \text{N} \quad \text{(more than 2 equivalents)}
\]

It was found that such processes for preparing various inorganic phosphates comprise several advantages over the prior art processes. In fact, such processes can be carried out at low temperature as opposed to processes of the prior art (for example those used for preparing LiFePO$_4$) which require high temperatures generally greater than 800 °C (such as processes involving the use of UCO$_3$ (lithium carbonate), NH$_4$PO$_4$ (ammonium phosphate) and Fe(CH$_3$COO)$_2$ (ferrous acetate). Moreover, the processes can be carried out in an organic solvent which is also an acid scavenger. For example, they permit to keep the obtained compound (for example LiFePO$_4$) under anhydrous conditions and to substantially avoid the presence of Fe(III) ions, which are contaminants detrimental for lithium batteries.

When the reaction is completed, the composition obtained which comprises the desired compound, the protonated acid scavenger, and optionally the unprotonated acid scavenger, can then readily be annealed, thereby using the scavenger as a source of carbon. Depending on the fact that an excess of acid scavenger was used or not at the beginning of the reaction, the obtained composition will be more or less viscous and easily
deposited onto a desired substrate so as to prepare an electrode having such a coating thereon.

[0040] While a description was made with particular reference to the illustrated embodiments, it will be understood that numerous modifications thereto will appear to those skilled in the art. Accordingly, the above description should be taken as specific examples and not in a limiting sense.
WHAT IS CLAIMED IS:

1. A process for preparing a compound of formula (I):

   \[
   \text{AMPO}_4 \quad (I)
   \]

   wherein

   A is chosen from H\(^+\), Li\(^+\), Na\(^+\), K\(^+\), and Cs\(^+\); and

   M is chosen from Be\(^{2+}\), Mg\(^{2+}\), Ba\(^{2+}\), Ca\(^{2+}\), Sr\(^{2+}\), Fe\(^{3+}\), Co\(^{3+}\), Ni\(^{2+}\), Mn\(^{2+}\), Zn\(^{2+}\), Cu\(^{2+}\), and Mo\(^{2+}\);

   said process comprising reacting a compound of formula (II) with a compound of formula (III):

   \[
   \text{AH}_2\text{PO}_4 \quad (II)
   \]

   \[
   \text{MX}_n \quad (III)
   \]

   wherein

   A is as previously defined;

   M is as previously defined;

   X is chosen from F\(^-\), Cl\(^-\), Br\(^-\), I\(^-\), CN\(^-\), CH\(_3\)COO\(^-\), CF\(_3\)SO\(_3\)\(^-\), NO\(_3\)\(^-\), OCN\(^-\), SCN\(^-\), (CN)\(_2\)N\(^-\), CO\(_3\)\(^2-\), ClO\(_3\)\(^-\), CrO\(_4\)\(^{2-}\), BrO\(_3\)\(^-\), Cr\(_2\)O\(_7\)\(^{2-}\), S\(_2\)O\(_4\)\(^{2-}\), S\(_4\)O\(_6\)\(^{2-}\), S\(_2\)O\(_3\)\(^{2-}\), SO\(_4\)\(^{2-}\), MnO\(_4\)\(^-\), S\(^2-\), ClO\(^-\), and C\(_2\)O\(_4\)\(^{2-}\); and

   n is 1 or 2,

   in the presence of an acid scavenger adapted to at least substantially solubilize said compound of formula (II) and said compound of formula.
(III), at a predetermined temperature, and adapted to be converted into a protic ionic liquid upon protonation.

2. The process of claim 1, wherein said acid scavenger is a compound of formula (IV):

![Diagram of (IV)]

(IV)

wherein

R is chosen from hydrogen atom, C₁⁻C₁₂ alkyl, C₃⁻C₁₂ cycloalkyl, C₁⁻C₁₂ heterocyclyl, C₂⁻C₁₂ alkenyl, C₂⁻C₁₂ alkynyl, C₆⁻C₁₂ aryl, C₆⁻C₂₀ aralkyl, C₆⁻C₂₀ Oalkylaryl, and C₁⁻C₁₂ heteroaryl.

3. The process of claim 2, wherein said acid scavenger is

![Diagram of R–N]  

![Diagram of R–N]  

4. The process of claim 1, wherein said acid scavenger is

![Diagram of N–N]  

5. The process of claim 1, wherein said acid scavenger is
6. The process of any one of claims 1 to 5, wherein said compound of formula (I) is chosen from LiFePO$_4$, LiBePO$_4$, LiMgPO$_4$, LiBaPO$_4$, LiCaPO$_4$, LiSrPO$_4$, LiCoPO$_4$, LiNiPO$_4$, LiMnPO$_4$, LiZnPO$_4$, LiCuPO$_4$, and LiMoPO$_4$.

7. The process of any one of claims 1 to 5, wherein said compound of formula (I) is LiFePO$_4$.

8. The process of claim 7, wherein said compound of formula (III) is chosen from FeCl$_2$, FeBr$_2$, FeI$_2$, FeF$_2$, Fe(CN)$_2$, Fe(SCN)$_2$, FeSO$_4$, Fe(CH$_3$COO)$_2$, and FeC$_2$O$_4$.

9. A process for preparing an electrode comprising:

- reacting a compound of formula (II) with a compound of formula (III):

$$AH_2PO_4 \quad (II)$$

$$MX_n \quad (III)$$

wherein

A is chosen from H$^+$, Li$^+$, Na$^+$, K$^+$, and Cs$^+$;

M is chosen from Be$^{2+}$, Mg$^{2+}$, Ba$^{2+}$, Ca$^{2+}$, Sr$^{2+}$, Fe$^{2+}$, Co$^{2+}$, Ni$^{2+}$, Mn$^{2+}$, Zn$^{2+}$, Cu$^{2+}$, and Mo$^{2+}$;
X is chosen from \( F^-, Cl^-, Br^-, I^-, CN^-, CH_3COO^-, CF_3SO_3^-, NO_3^-, OCN^-, SCN^-, (CN)_2N^-, CO_3^{2-}, ClO_3^-, CrO_4^{2-}, BrO_3^-, Br_2O_7^{2-}, S_2O_4^{2-}, S_4O_6^{2-}, S_2O_3^{2-}, SO_4^{2-}, MnO_4^-, S^{2-}, ClO^-, \) and \( C_2O_4^{2-} \); and

\( n \) is 1 or 2,

in the presence of an acid scavenger adapted to at least substantially solubilize said compound of formula (II) and said compound of formula (III), at a predetermined temperature, and adapted to be converted into a protic ionic liquid upon protonation, thereby obtaining a composition comprising a compound of formula (I) AMPO_4 in which \( A \) and \( M \) are as previously defined, said protonated acid scavenger, and optionally said unprotonated acid scavenger;

- depositing said composition on a substrate; and

- annealing said composition deposited on said substrate so as to obtain said electrode.

10. The process of claim 9, further comprising adding a super valent metal salt chosen from \( Nb^{5+}, Al^{3+}, Ti^{4+}, Mg^{2+}, W^{6+}, \) and \( Zr^{4+} \) to said compound of formula (II), said compound of formula (III), and said acid scavenger.

11. The process of claim 9 or 10, further comprising adding a carbon source to said compound of formula (II), said compound of formula (III), and said acid scavenger.

12. The process of claim 11, wherein said carbon source comprises carbon black, a polymer, or a carbon-rich organic compound which is soluble in said acid scavenger.
13. The process of claim 11, wherein said carbon source comprises nanoparticles of carbon black, carbon nanotubes or a mixture thereof.

14. The process of claim 11, wherein said carbon source comprises multi-wall carbon nanotubes or single-wall carbon nanotubes.

15. The process of any one claims 9 to 14, further comprising adding an organic solvent to said compound of formula (II), said compound of formula (III), and said acid scavenger.

16. The process of claim 15, wherein said organic solvent is chosen from acetonitrile, dimethylformamide, tetrahydrofuran, dimethoxymethane, and mixtures thereof.

17. The process of any one of claims 9 to 16, wherein said acid scavenger is a compound of formula (IV):

\[ \begin{array}{c}
\text{HN} \\
\text{R} \\
\text{N} \\
\end{array} \]

(IV)

wherein

\( R \) is chosen from hydrogen atom, \( \text{C}_{1-12} \text{ alkyl}, \text{C}_{3-12} \text{ cycloalkyl}, \text{C}_{1-12} \text{ heterocyl}, \text{C}_{2-12} \text{ alkenyl}, \text{C}_{2-12} \text{ alkynyl}, \text{C}_{6-12} \text{ aryl}, \text{C}_{6-20} \text{ aralkyl}, \text{C}_{6-20} \text{ alkylaryl}, \text{and C}_{6-12} \text{ heteroaryl}. \)

18. The process of claim 17, wherein said acid scavenger is

\[ \begin{array}{c}
\text{R} \\
\text{N} \\
\text{N} \\
\end{array} \]
19. The process of claim 17 or 18, wherein said acid scavenger is liquid at room temperature.

20. The process of any one of claims 17 to 19, wherein R comprises a polymerizable moiety.

21. The process of any one of claims 9 to 18, wherein said acid scavenger is

\[
\text{[Image of chemical structure]}
\]

22. The process of any one of claims 9 to 18, wherein said acid scavenger is

\[
\text{[Image of chemical structure]}
\]

23. The process of any one of claims 9 to 22, wherein said compound of formula (I) is chosen from LiFePO₄, LiBePO₄, LiMgPO₄, LiBaPO₄, LiCaPO₄, LiSrPO₄, LiCoPO₄, LiNiPO₄, LiMnPO₄, LiZnPO₄, LiCuPO₄, and LiMoPO₄.

24. The process of any one of claims 9 to 23, wherein said compound of formula (I) is LiFePO₄.

25. The process of claim 24, wherein said compound of formula (III) is chosen from FeCl₂, FeBr₂, FeI₂, FeF₂, Fe(CN)₂, Fe(SCN)₂, FeSO₄, Fe(CH₃COO)₂, and FeC₂O₄.
26. The process of any one of claims 1 to 25, wherein said compound of formula (I) is annealed after being prepared.

27. The process of any one of claims 1 to 26, wherein the compound of formula (I) is in the form of nanoparticles.

28. The process of any one of claims 1 to 27, wherein said predetermined temperature is lower than 200 °C.

29. The process of any one of claims 1 to 27, wherein said predetermined temperature is lower than 150 °C.

30. The process of any one of claims 1 to 27, wherein said predetermined temperature is lower than 100 °C.

31. The process of any one of claims 1 to 27, wherein said predetermined temperature is about 15 °C to about 60 °C.

32. The process of any one of claims 1 to 27, wherein said predetermined temperature is about 20 °C to about 35 °C.

33. Use of a compound of formula (IV):

![Compound IV](image)

wherein
R is chosen from hydrogen atom, C1-C12 alkyl, C3-C12 cycloalkyl, C1-C12 heterocyclyl, C2-C12 alkenyl, C2-C12 alkynyl, C6-C12 aryl, C6-C20 aralkyl, C6-C20 alkylaryl, and CrC2 heteroaryl, as an acid scavenger in the preparation of a phospho-olivine type compound.

34. The use of claim 33, wherein said compound of formula (IV) is

\[
\begin{array}{c}
\text{R} \\
\text{N} \\
\end{array}
\]

35. The use of claim 33 or 34, wherein said phospho-olivine type compound is chosen from LiFePO\(_4\), LiBePO\(_4\), LiMgPO\(_4\), LiBaPO\(_4\), LiCaPO\(_4\), LiSrPO\(_4\), LiCoPO\(_4\), LiNiPO\(_4\), LiMnPO\(_4\), LiZnPO\(_4\), LiCuPO\(_4\), and LiMoPO\(_4\).

36. The use of claim 35, wherein said phospho-olivine type compound is LiFePO\(_4\).

37. The use of any one of claims 33 to 36, wherein said compound of formula (IV) is

\[
\begin{array}{c}
\text{N} \\
\end{array}
\]

38. The use of any one of claims 33 to 36, wherein said compound of formula (IV) is

\[
\begin{array}{c}
\text{N} \\
\end{array}
\]
39. A method of using a compound of formula (IV)

![Formula IV](image)

wherein

R is chosen from hydrogen atom, d-C\textsubscript{-12} alkyl, C\textsubscript{3}-C\textsubscript{2} alkynyl, C\textsubscript{1}-C\textsubscript{12} cycloalkyl, C\textsubscript{1}-C\textsubscript{12} heterocyclyl, C\textsubscript{2}-C\textsubscript{12} alkenyl, C\textsubscript{6}-C\textsubscript{2} aryl, C\textsubscript{6}-C\textsubscript{2} aralkyl, C\textsubscript{6}-C\textsubscript{2}0 alkylaryl, and C\textsubscript{1}-C\textsubscript{12} heteroaryl,

said method comprising using said compound of formula (IV) as an acid scavenger in the preparation of a phospho-olivine type compound.

40. The method of claim 39, wherein said compound of formula (IV) is contacted with protons generated during the synthesis of said phospho-olivine type compound.

41. A method of scavenging protons, said method comprising contacting said protons with a compound of formula (IV):

![Formula IV](image)

wherein
R is chosen from hydrogen atom, C1-C12 alkyl, C3-C12 cycloalkyl, C1-C12 heterocyclyl, C2-C12 alkenyl, C2-C12 alkynyl, C6-C12 aryl, C6-C2O aralkyl, C6-C2O alkylaryl, and C1-C12 heteroaryl.

42. The method of claim 42, wherein said compound of formula (IV) is contacted with protons generated during preparation of a phospho-olivine type compound.

43. The method of claim 42, wherein said compound of formula (IV) is further used as a solvent during preparation of said phospho-olivine type compound.

44. The method of claim 39, 40, 42, or 43, wherein said phospho-olivine type compound is chosen from LiFePO4, LiBePO4, LiMgPO4, LiBaPO4, LiCaPO4, LiSrPO4, LiCoPO4, LiNiPO4, LiMnPO4, LiZnPO4, LiCuPO4, and LiMoPO4.

45. The method of claim 44, wherein said phospho-olivine type compound is LiFePO4.

46. In a method for preparing LiFePO4 comprising reacting together a lithium source, an iron source, and a phosphorus source, the improvement wherein said lithium source and said phosphorous source is LiH2PO4, and said iron source is FeXn in which X is chosen from F-, Cl-, Br-, I-, CN-, CH3COO-, CF3SO3-, NO3-, OCN-, SCN-, (CN)2N-, CO32-, ClO3-, CrO42-, BrO3-, Cr2O72-, S2O82-, S4O62-, S2O32-, SO42-, MnO4-, S2-, ClO-, and C2O42-; and n is 1 or 2.

47. The method of claim 46, wherein LiH2PO4 is reacted with FeXn at a temperature lower than 200 °C.

48. The method of claim 46, wherein LiH2PO4 is reacted with FeXn at a temperature lower than 150 °C.
49. The method of claim 46, wherein LiH$_2$PO$_4$ is reacted with FeX$_n$ at a temperature lower than 100 °C.

50. The method of claim 46, wherein LiH$_2$PO$_4$ is reacted with FeX$_n$ at a temperature lower than 60 °C.

51. The method of claim 46, wherein LiH$_2$PO$_4$ is reacted with FeX$_n$ at room temperature.

52. The method of claim 46, wherein LiH$_2$PO$_4$ is reacted with FeX$_n$ at a temperature of about 15 °C to about 60 °C.

53. The method of claim 46, wherein LiH$_2$PO$_4$ is reacted with FeX$_n$ at a temperature of about 20 °C to about 35 °C.

54. The method of any one of claims 46 to 53, said method being characterized in that a compound of formula (IV):

![Formula (IV)]

wherein

R is chosen from hydrogen atom, C$_1$-C$_{12}$ alkyl, C$_3$-C$_{12}$ cycloalkyl, C$_1$-C$_{12}$ heterocyclcyl, C$_2$-C$_{12}$ alkenyl, C$_2$-C$_{12}$ alkynyl, Ce-C$_{12}$ aryl, Ce-C$_{20}$ aralkyl, C$_6$-C$_2$0 alkylaryl, and C$_1$-C$_{12}$ heteroaryl,

is used as a proton scavenger.
55. The method of claim 54, wherein said compound of formula (IV) is also used as a solvent.

56. The method of any one of claims 39 to 45 and 54, wherein said compound of formula (IV) is

\[
\begin{array}{c}
R \quad N \\
\end{array}
\]

57. The method of any one of claims 39 to 45 and 54, wherein said compound of formula (IV) is

\[
\begin{array}{c}
N \\
\end{array}
\]

58. The method of any one of claims 39 to 45 and 54, wherein said compound of formula (IV) is

\[
\begin{array}{c}
\backslash \\
N \\
\end{array}
\]

59. A process for preparing a compound of formula (Ia):

\[ AM^1tM_{1-t}^2PO_4 \] (Ia)

wherein

A is chosen from H\(^+\), Li\(^+\), Na\(^+\), K\(^+\), and Cs\(^+\); and

M\(^1\) is chosen from Be\(^2+\), Mg\(^2+\), Ba\(^2+\), Ca\(^2+\), Sr\(^2+\), Fe\(^2+\), Co\(^2+\), Ni\(^2+\), Mn\(^2+\), Zn\(^2+\), Cu\(^2+\), and Mo\(^2+\);

\[
R \quad N \\
\]

\[
\begin{array}{c}
N \\
\end{array}
\]

\[
\begin{array}{c}
\backslash \\
N \\
\end{array}
\]
M² is chosen from Be²⁺, Mg²⁺, Ba²⁺, Ca²⁺, Sr²⁺, Fe²⁺, Co²⁺, Ni²⁺, Mn²⁺, Zn²⁺, Cu²⁺, and Mo²⁺; and

T has a value of 0 to 1,

said process comprising reacting a compound of formula (II) with a compound of formula (IIia) and a compound of formula (IIib):

\[ \text{AH}_2\text{PO}_4 \quad \text{(II)} \]

\[ M^1(\text{X}^1)_{n1} \quad \text{(IIia)} \]

\[ M^2(\text{X}^2)_{n2} \quad \text{(IIib)} \]

wherein

A is as previously defined;

M¹ and M² are as previously defined;

\[ \text{X}^1 \text{ is chosen from } \text{F}^-, \text{Cl}^-, \text{Br}^-, \text{I}^-, \text{CN}^-, \text{CH}_3\text{COO}^-, \text{CF}_3\text{SO}_3^-, \text{NO}_3^-, \text{OCN}^-, \text{SCN}^-, (\text{CN})_2\text{N}^-, \text{CO}_3^{2-}, \text{ClO}_3^-, \text{CrO}_4^{2-}, \text{BrO}_3^-, \text{Cr}_2\text{O}_7^{2-}, \text{S}_2\text{O}_4^{2-}, \text{S}_4\text{O}_6^{2-}, \text{S}_2\text{O}_3^{2-}, \text{SO}_4^{2-}, \text{MnO}_4^-, \text{S}^2-, \text{ClO}^-, \text{and } \text{C}_2\text{O}_4^{2-}; \]

\[ \text{X}^2 \text{ is chosen from } \text{F}^-, \text{Cl}^-, \text{Br}^-, \text{I}^-, \text{CN}^-, \text{CH}_3\text{COO}^-, \text{CF}_3\text{SO}_3^-, \text{NO}_3^-, \text{OCN}^-, \text{SCN}^-, (\text{CN})_2\text{N}^-, \text{CO}_3^{2-}, \text{ClO}_3^-, \text{CrO}_4^{2-}, \text{BrO}_3^-, \text{Cr}_2\text{O}_7^{2-}, \text{S}_2\text{O}_4^{2-}, \text{S}_4\text{O}_6^{2-}, \text{S}_2\text{O}_3^{2-}, \text{SO}_4^{2-}, \text{MnO}_4^-, \text{S}^2-, \text{ClO}^-, \text{and } \text{C}_2\text{O}_4^{2-}; \]

n¹ is 1 or 2;

n² is 1 or 2;

in the presence of an acid scavenger adapted to at least substantially solubilize said compound of formula (II), said compound of formula
(Ilia), and said compound of formula (Hlb), at a predetermined temperature, and adapted to be converted into a protic ionic liquid upon protonation.

60. The process of claim 59, wherein T has a value comprised between 0 and 1.

61. The process of claim 59 or 60, wherein said compound of formula (la) is annealed after being prepared.

62. The process of any one of claims 59 to 61, wherein the compound of formula (la) is in the form of nanoparticles.

63. The process of any one of claims 59 to 62, wherein said predetermined temperature is lower than 200 °C.

64. The process of any one of claims 59 to 62, wherein said predetermined temperature is lower than 150 °C.

65. The process of any one of claims 59 to 62, wherein said predetermined temperature is lower than 100 °C.

66. The process of any one of claims 59 to 62, wherein said predetermined temperature is about 15 °C to about 60 °C.

67. The process of any one of claims 59 to 62, wherein said predetermined temperature is about 20 °C to about 35 °C.
LiFePO$_4$
INTERNATIONAL SEARCH REPORT

A CLASSIFICATION OF SUBJECT MATTER


According to International Patent Classification (IPC) or to both national classification and IPC

B FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
COIB 25/26, COIB 25/30, COIB 25/32, COIB 25/37, HOIM 10/36, HOIM 4/04, HOIM 4/58 (all 2006 01)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic database(s) consulted during the international search (name of database(s) and, where practicable, search terms used)
Canadian Patent Database, Delphion, Google Scholar (N-methylimidazole, N-methyl imidazole, ionic liquid, lithium, iron, phosphate, olivine, ohvmo, solvent, phospho, synthesis, solvent, imidazole, scavenger)

C DOCUMENTS CONSIDERED TO BE RELEVANT

<table>
<thead>
<tr>
<th>Category*</th>
<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
<th>Relevant to claim No</th>
</tr>
</thead>
<tbody>
<tr>
<td>X</td>
<td>Pure Appl Chem, Vol 78, No 8, pp 1587-1593, 2006</td>
<td>4 1</td>
</tr>
<tr>
<td>Y</td>
<td>ANDREAS HEINTZ ET AL Whole Document</td>
<td>33-45</td>
</tr>
<tr>
<td>X</td>
<td>US7008566B2 (BARKER ET AL)</td>
<td>46 and Al'</td>
</tr>
<tr>
<td>Y</td>
<td>7 Mar 2006 (07-03-2006) Columns 1-6, Claims</td>
<td>33-45</td>
</tr>
<tr>
<td>X</td>
<td>US6794084B2 (ADAMSON ET AL)</td>
<td>46 and 47</td>
</tr>
<tr>
<td>A</td>
<td>US2005/0194567A1 (ADAMSON ET AL)</td>
<td>1-67</td>
</tr>
<tr>
<td></td>
<td>8 Sept 2005 (08-09-2005) Whole Document</td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>US6960331B2 (BARKER ET AL)</td>
<td>1-67</td>
</tr>
</tbody>
</table>

[X] See patent family annex

A document defining the general state of the art which is not considered to be of particular relevance
E earlier application or patent but published on or after the international filing date
L 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)
O document referring to an oral disclosure, use exhibition or other means
P document published prior to the international filing date but later than the priority date claimed

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 invention
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
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 documents such combination being obvious to a person skilled in the art
& document member of the same patent family

Date of the actual completion of the international search
27 June 2008 (27-06-2008)

Date of mailing of the international search report
21 July 2008 (21-07-2008)

Name and mailing address of the ISA/CA Authorized officer
Canadian Intellectual Property Office
Place du Portage I, Cl 14 – 1st Floor, Box PCT
50 Victoria Street
Gatineau, Quebec K1A 0C9
Facsimile No 001-819-953-2476
Stewart Parsons 819-997-4378
### Observations where certain claims were found unsearchable (Continuation of item 2 of the first sheet)

<table>
<thead>
<tr>
<th>Box No.</th>
<th>Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>II</td>
<td></td>
</tr>
</tbody>
</table>

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. [ ] Claim Nos because they relate to subject matter not required to be searched by this Authority, namely

2. [ ] Claim 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

3. [ ] Claim Nos because they are dependant claims and are not drafted in accordance with the second and third sentences of Rule 6-4(a)

### Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

<table>
<thead>
<tr>
<th>Box No.</th>
<th>Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>III</td>
<td></td>
</tr>
</tbody>
</table>

This International Searching Authority found multiple inventions in this international application, as follows:

(See Supplemental Box)

1. [ ] As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims

2. [X] As all searchable claims could be searched without effort justifying 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 claim 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 claim Nos

**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
The claims are directed to the following groups

**Group A**
Claims 1 and 6-8(m part) are directed to a specific process for forming compound (I) (a phospho olivine type compound) comprising the use of a generic acid scavenger
Claims 9-16 and 21-32(m part) are directed to a specific process for forming compound (I) comprising the use of a generic acid scavenger, and further depositing the compound on a substrate and annealing to form an electrode
Claim 59-67 is directed to a specific process for forming compound (Ia) (a mixed-metal phospho olivine type compound) comprising the use of a generic acid scavenger

**Group B**
Claims 2-5, and 6-8(m part) are directed to a specific process for forming compound (I) comprising the use of acid scavenger (IV)
Claims 17-20 and 21-32(m part) are directed to a specific process for forming compound (I) comprising the use of acid scavenger (IV), and further depositing the compound on a substrate and annealing to form an electrode

**Group C**
Claims 33-38 are directed to the use of a compound of formula (IV) (a mono-alkyl-substituted imidazole) as an acid scavenger in a generic synthesis of a phospho-olvino type compound
Claims 39, 40, 44(m part), 45(m part) and 56-58(m part) are directed to a method of using compound (IV) as an acid scavenger in a generic synthesis of a phospho-olvino type compound
Claims 42, 43, 44(m part) and 45(m part) are directed to a method for scavenging protons comprising using compound (IV) as an acid scavenger in a generic synthesis of a phospho-olvino type compound
Claims 54, 55 and 56-58(m part) are directed to a generic process for preparing LiFePO₄ with the use of compound (IV) as an acid scavenger

**Group D**
Claims 41 is directed to a method for scavenging protons comprising using acid scavenger (IV)

**Group E**
Claims 46-53 are directed to a generic process for preparing LiFePO₄

Groups A, B and C possess unity

The remaining groups lack unity as follows

Group D is unrelated to the remaining groups as compound (IV) is a known acid scavenger in general (see D1 for instance)

Group E is unrelated to the remaining groups as generic syntheses of LiFePO₄ are known (see D2 and D3 for instance)
<table>
<thead>
<tr>
<th>Patent Document Cited in Search Report</th>
<th>Publication Date</th>
<th>Patent Family Member(s)</th>
<th>Publication Date</th>
</tr>
</thead>
<tbody>
<tr>
<td>US 7008566B2</td>
<td>07-03-2006</td>
<td>CA 2521366A1</td>
<td>28-10-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CN 1780788A</td>
<td>31-05-2006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>JP 2006023930T</td>
<td>19-10-2006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>KR 20050120789A</td>
<td>23-12-2005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>US 2004206938A1</td>
<td>21-10-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>US 2005091363A1</td>
<td>04-05-2006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WO 2004092065A1</td>
<td>28-10-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CA 2493224A1</td>
<td>05-02-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CN 1672277A</td>
<td>21-09-2005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CN 100334757C</td>
<td>29-08-2007</td>
</tr>
<tr>
<td></td>
<td></td>
<td>JP 2005533742T</td>
<td>10-1-1-2005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>KR 20050029239A</td>
<td>24-03-2005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>US 200401 135A1</td>
<td>29-01-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WO 200401 1403A2</td>
<td>05-02-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WO 200401 1403A3</td>
<td>08-07-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CN 1871726A</td>
<td>29-1-1-2006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>DE 12004001997T5</td>
<td>26-10-2006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>US 7348100B2</td>
<td>25-03-2008</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WO 2005043647A2</td>
<td>12-05-2005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WO 2005043647A3</td>
<td>11-05-2006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>AU 1779902A</td>
<td>11-06-2002</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CA 2428201A1</td>
<td>06-06-2002</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CN 1703370A</td>
<td>30-1-1-2005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>DE 60133405D1</td>
<td>08-05-2008</td>
</tr>
<tr>
<td></td>
<td></td>
<td>EP 1343720A2</td>
<td>17-09-2003</td>
</tr>
<tr>
<td></td>
<td></td>
<td>EP 1343720B1</td>
<td>26-03-2008</td>
</tr>
<tr>
<td></td>
<td></td>
<td>EP 1574477A2</td>
<td>14-09-2005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>EP 1574477A3</td>
<td>09-1-1-2005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>JP 20045 14639T</td>
<td>20-05-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>KR 200701 12297A</td>
<td>22-1-1-2007</td>
</tr>
<tr>
<td></td>
<td></td>
<td>KR 200701 12298A</td>
<td>22-1-1-2007</td>
</tr>
<tr>
<td></td>
<td></td>
<td>KR 200701 12299A</td>
<td>22-1-1-2007</td>
</tr>
<tr>
<td></td>
<td></td>
<td>RU 20031 19145A</td>
<td>27-12-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>TW 549967B</td>
<td>01-08-2003</td>
</tr>
<tr>
<td></td>
<td></td>
<td>US 6645452B1</td>
<td>11-1-1-2003</td>
</tr>
<tr>
<td></td>
<td></td>
<td>US 2004126300A1</td>
<td>01-07-2004</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WO 0244084A2</td>
<td>06-06-2002</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WO 0244084A3</td>
<td>15-08-2002</td>
</tr>
</tbody>
</table>