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Declarations under Rule 4.17:
— *as to applicant's entitlement to apply for and be granted a patent (Rule 4.17(ii)) for all designations*
— *as to the applicant's entitlement to claim the priority of the earlier application (Rule 4.17(iii)) for all designations*

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

WO 03/035609 A1

(54) Title: ZWITTERIONIC IMIDES

(57) Abstract: Zwitterionic imide compounds are provided according to the formula: R_1-SO_2-N^--SO_2-R_2^+, where R_1 and R_2^+ are any suitable groups. Typically R_1 is a highly fluorinated alkane and R_2^+ contains a quaternary ammonium group or a heteroatomic aromatic group having an cationic nitrogen, such as: pyridiniumyl, pyridaziniumyl, pyrimidiniumyl, pyraziniumyl, imidazoliumyl, pyrazoliumyl, thiazoliumyl, oxazoliumyl, or triazoliumyl. Zwitterionic liquids are provided, typically having melting points of less than 100 °C and typically having a solubility in water of less than 5% by weight.

Zwitterionic Imides

Field of the Invention

This invention relates to zwitterionic imide compounds according to the formula: R_1-SO_2-N^--SO_2-R_2^+ and zwitterionic liquids having a melting point of less than 100°C.

Background of the Invention

US 5,827,602 and US 5,965,054 disclose non-aqueous ionic liquids for use as electrolytes that are based on salts composed of separate anion and cation moieties. The cation moieties are selected from pyridinium, pyridazinium, pyrimidinium pyrazinium, imidazolium, pyrazolium, thiazolium, oxazolium, and triazolium ions.

US 6,090,895 discloses crosslinked polymers having imide crosslinking groups and methods of crosslinking polymers to form imide crosslinking groups. These crosslinked polymers may be useful as polymer electrolyte membranes (PEM's) in fuel cells.

US 6,063,522, US 4,505,997, US 5,652,072, US 5,072,040 and US 5,514,493 disclose electrolytes comprising imide functional groups.

US 5,463,005 discloses perfluorinated monomers and polymers comprising sulfonyl and carbonyl imide groups for use as solid polymer electolytes.

M.Doyle, S.K.Choi and G.Proulx, J. Electrochem. Soc., 147, 34-37, discloses the use of ethyl methyl imidazolium (EMI) triflate, a low melting ionic liquid composed of separate anion and cation moieties, together with a known sulfonated fluoropolymer (Nafion) to give a fuel cell membrane material for use at elevated temperatures.

Summary of the Invention

Briefly, the present invention provides a zwitterionic imide compound according to the formula: R_1-SO_2-N^--SO_2-R_2^+, where R_1 and R_2^+ are any suitable

groups. Typically R_1 is a highly fluorinated alkane and R_2^+ is a heteroatomic aromatic group having an cationic nitrogen, such as: pyridiniumyl, pyridaziniumyl, pyrimidiniumyl, pyraziniumyl, imidazoliumyl, pyrazoliumyl, thiazoliumyl, oxazoliumyl, or triazoliumyl. Typically the zwitterionic imide has a melting point of less than 100 °C. Typically the zwitterionic imide compound has a solubility in water of less than 5% by weight.

In another aspect, the present invention provides a zwitterionic liquid having a melting point of less than 100 °C and typically having a solubility in water of less than 5% by weight.

In another aspect, the present invention provides an electrochemical device comprising the zwitterionic liquid or the zwitterionic imide according to the present invention, such as a battery or a fuel cell.

What has not been described in the art, and is provided by the present invention, is an ionic liquid having a melting point of less than 100 °C that is a zwitterionic compound, particularly a zwitterionic imide compound.

In this application:

"highly halogenated", means containing halogen in an amount of 40 wt% or more, but typically 50 wt% or more, and more typically 60 wt% or more; and

"highly fluorinated", means containing fluorine in an amount of 40 wt% or more, but typically 50 wt% or more, and more typically 60 wt% or more; and

"substituted" means, for a chemical species, substituted by conventional substituents which do not interfere with the desired product or process, e.g., substituents can be alkyl, alkoxy, aryl, phenyl, halo (F, Cl, Br, I), cyano, nitro, etc.

It is an advantage of the present invention to provide an electrolyte for use in electrochemical devices such as batteries and fuel cells at temperatures in excess of 80 °C. The zwitterionic liquids can be more stable in use than conventional (non-zwitterionic) ionic liquids, since they do not form a neutral, volatile, water soluble species upon protonation of the anion.

Detailed Description of Preferred Embodiments

The present invention provides zwitterionic imide compounds according to the formula: R_1-SO_2-N^--SO_2-R_2^+, where R_1 and R_2^+ are any suitable groups. Typically R_1 is a highly fluorinated alkane and R_2^+ contains a heteroatomic aromatic group having an cationic nitrogen, such as: pyridiniumyl, pyridaziniumyl, pyrimidiniumyl, pyraziniumyl, imidazoliumyl, pyrazoliumyl, thiazoliumyl, oxazoliumyl, or triazoliumyl.

The present invention additionally provides a zwitterionic liquid having a melting point of less than 100 °C and typically having a solubility in water of less than 5% by weight, which may be a zwitterionic imide compound according to the present invention.

Electrochemical devices such as batteries and fuel cells employ electrolyte materials to conduct ions participating in electrochemical reactions. Common aqueous based electrolytes are not suitable for use at high temperatures in excess of 100 °C, since the water component can evaporate. Drying of the electrolyte can become a problem at temperatures of 80 °C or greater.

Polymer electrolyte membranes (PEM's) may be used in electrochemical devices, particularly in fuel cells. PEM's are composed of a polymer backbone, typically fluorinated, having pendant acidic groups such as sulfonic acid groups. Examples include Nafion® (DuPont Chemicals, Wilmington DE) and Flemion™ (Asahi Glass Co. Ltd., Tokyo, Japan). Non-fluorinated PEM's include sulfonated polysulfone and sulfonated PEEK. In a fuel cell, the PEM conducts H+ ions (protons) while forming a barrier to the passage of reactant gasses (H_2 and O_2). The PEM is electrically non-conductive. However, membranes of these polymers must contain a liquid electrolyte, typically water, to function electrolytically. Even though water is produced during the electrochemical reactions carried out in a hydrogen fuel cell, drying of the PEM can become a problem at temperatures of 80 °C or greater and the cell cannot operate with a water electrolyte at temperatures in excess of 100 °C.

Conventional (non-zwitterionic) ionic liquids can be of limited utility in extreme environments where the anion can become protonated. The resulting neutral acid may be volatile and water soluble. This is a particular concern for fuel cell use,

since a volatile, water soluble species will be washed out of the cell, rendering it inactive.

The zwitterionic liquids according to the present invention can be used as electrolytes in electrochemical devices such as batteries and fuel cells. The zwitterionic liquids may be used neat or with a solvent, diluent or additive. Additives may be used to alter melting point, viscosity, ionic conductivity, and the like. Further, the zwitterionic liquids according to the present invention can be absorbed into polymer electrolyte membranes for use in electrochemical devices, especially for use in fuel cells. The zwitterionic liquids may be substituted for water in a typical polymer electrolyte membrane by any suitable method. In one method, the zwitterionic liquid is applied to the membrane and/or the membrane is immersed in the zwitterionic liquid, with these steps being repeated as necessary.

The zwitterionic liquids according to the present invention typically have a melting point of less than 100 °C, more typically less than 80 °C, more typically less than 60 °C, and most typically less than 40 °C. The zwitterionic liquids according to the present invention typically have a solubility in water of less than 5% by weight, more typically less than 1% by weight, and most typically less than 0.1% by weight.

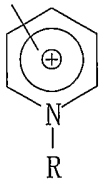
In one embodiment, the zwitterionic liquid according to the present invention is a zwitterionic imide compound according to formula (I):

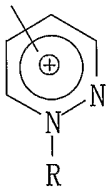
$$R_1\text{-}SO_2\text{-}N^-\text{-}SO_2\text{-}R_2^+ \qquad (I)$$

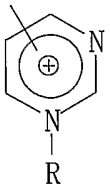
where R_1 and R_2^+ are any suitable groups.

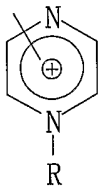
R_1 may be any group which does not interfere with the desired characteristics of the compound. R_1 may be selected from straight-chain, branched, cyclic and aromatic groups, including saturated and unsaturated groups, including heteroatomic groups, and including any of the above which are substituted. Typically R_1 is a highly halogenated alkane, more typically a highly fluorinated alkane. More typically R_1 is a perhalogenated alkane, and most typically a perfluorinated alkane. Typically R_1 contains between 1 and 20 carbon atoms, more typically between 1 and 8 carbon atoms, and most typically between 1 and 4 carbon atoms.

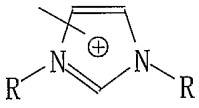
R_2^+ may be any cationic group which does not interfere with the desired characteristics of the compound. R_2^+ may be selected from straight-chain, branched, cyclic and aromatic groups, including saturated and unsaturated groups, including heteroatomic groups, and including any of the above which are substituted. Typically, R_2^+ is an aromatic group, more typically an aromatic heterocyclic group. More typically, R_2^+ is a cationic heteroatomic aromatic group including nitrogen. More typically, R_2^+ is selected from the following:

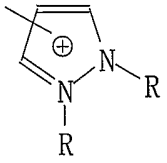
pyridiniumyl 

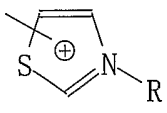
pyridaziniumyl 

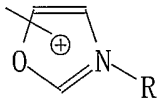
pyrimidiniumyl 

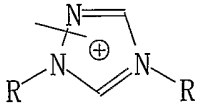
pyraziniumyl 

imidazoliumyl 

pyrazoliumyl 

thiazoliumyl 

oxazoliumyl 

triazoliumyl 

where R groups are independently selected from H, alkyl, haloalkyl, fluoroalkyl, or any group which does not interfere with the desired characteristics of the compound. R may be selected from straight-chain, branched, cyclic and aromatic groups, including saturated and unsaturated groups, including heteroatomic groups, and including any of the preceding which are substituted. Typically R contains between 0 and 20 carbon atoms, more typically between 0 and 8 carbon atoms, and most typically between 0 and 4 carbon atoms. The above-listed R_2^+ groups may be additionally substituted at any available carbon. Substituents may join with each other or with R groups to form additional aromatic or non-aromatic rings. Alternately, the open valence of the R_2^+ group, i.e., the imide attachment point, may be on the R group or other substituent, rather than on the aromatic ring as depicted above. Typically, the zwitterionic imide contains an imide bound directly to an aromatic group. The imide may be bound to a carbon or to a nitrogen of the aromatic group.

Alternately, R_2^+ may be a quaternary ammonium group. Examples of such quaternary ammonium groups include tetraalkyl ammonium cations. Where R_2^+ is a tetraalkyl ammonium group, alkyl substituents of the tetraalkyl ammonium group typically contain 1 to 12 carbons, more typically 1 to 8 carbons, and more typically 1 to 4 carbons. Alkyl substituents of the tetraalkyl ammonium group may be substituted or may link to form ring structures containing the ammonium nitrogen. Alkyl substituents of the tetraalkyl ammonium group may be substituted with polymerizable functional groups. The imide may be bound to a carbon or to the nitrogen of the ammonium group.

The zwitterionic imide according to the present invention can be made by any suitable method. Known imide syntheses may be performed using the appropriate starting materials containing the desired R_1 and R_2^+ groups, including syntheses

described in: US 6,063,522, US 4,505,997, US 5,652,072, US 5,072,040, US 5,514,493 and US 5,463,005. Useful synthetic routes may include:

(I) $R_1\text{-}SO_2\text{-}NH_2 + Cl\text{-}SO_2\text{-}R_2^+ \rightarrow R_1\text{-}SO_2\text{-}N^-\text{-}SO_2\text{-}R_2^+$

(II) $R_1\text{-}SO_2\text{-}Cl + H_2N\text{-}SO_2\text{-}R_2^+ \rightarrow R_1\text{-}SO_2\text{-}N^-\text{-}SO_2\text{-}R_2^+$

It will be recognized that reactions (I) and (II) are typically carried out in the presence of additional base such as $(Et)_3N$.

If desired, the cationic group of R_2^+ may be converted with a protecting group during synthesis and unprotected after synthesis.

This invention is useful as an electrolyte in devices such as batteries and fuel cells, particularly high-temperature devices for operation at temperatures in excess of 100 °C.

Various modifications and alterations of this invention will become apparent to those skilled in the art without departing from the scope and principles of this invention, and it should be understood that this invention is not to be unduly limited to the illustrative embodiments set forth hereinabove.

We claim:

1. A zwitterionic imide compound according to formula (I):

$$R_1\text{-}SO_2\text{-}N^-\text{-}SO_2\text{-}R_2^+ \qquad (I)$$

wherein R_1 is selected from the group consisting of straight-chain, branched, cyclic and aromatic groups, including saturated and unsaturated groups, including heteroatomic groups, and including any of the above which are substituted; and

wherein R_2^+ is any cationic group selected from the group consisting of straight-chain, branched, cyclic and aromatic groups, including saturated and unsaturated groups, including heteroatomic groups, and including any of the above which are substituted.

2. The zwitterionic imide compound according to claim 1, wherein R_2^+ is an aromatic group.

3. The zwitterionic imide compound according to claim 2, wherein R_2^+ is a heterocyclic group.

4. The zwitterionic imide compound according to claim 3, wherein R_2^+ contains a cationic nitrogen atom.

5. The zwitterionic imide compound according to claim 1, wherein R_2^+ contains a functional group selected from the group consisting of: pyridiniumyl, pyridaziniumyl, pyrimidiniumyl, pyraziniumyl, imidazoliumyl, pyrazoliumyl, thiazoliumyl, oxazoliumyl, and triazoliumyl.

6. The zwitterionic imide compound according to claim 1, wherein R_2^+ contains a quaternary ammonium cation.

7. The zwitterionic imide compound according to claim 1, wherein R_2^+ contains a tetraalkyl ammonium functional group.

8. The zwitterionic imide compound according to claim 7, wherein alkyl substituents of said tetraalkyl ammonium functional group contain 1 to 8 carbons.

9. The zwitterionic imide compound according to claim 1, wherein R_1 is a highly halogenated hydrocarbon group.

10. The zwitterionic imide compound according to claim 1, wherein R_1 is a highly fluorinated hydrocarbon group.

11. The zwitterionic imide compound according to claim 8, wherein R_1 is a highly halogenated hydrocarbon group.

12. The zwitterionic imide compound according to claim 8, wherein R_1 is a highly fluorinated hydrocarbon group.

13. The zwitterionic imide compound according to claim 1 having a melting point of less than 100°C.

14. The zwitterionic imide compound according to claim 10 having a melting point of less than 100°C.

15. The zwitterionic imide compound according to claim 12 having a melting point of less than 100°C.

16. The zwitterionic imide compound according to claim 1 having a solubility in water of less than 5% by weight.

17. The zwitterionic imide compound according to claim 12 having a solubility in water of less than 5% by weight.

18. The zwitterionic imide compound according to claim 15 having a solubility in water of less than 5% by weight.

19. A zwitterionic liquid having a melting point of less than 100°C.

20. The zwitterionic liquid according to claim 19 which is an aromatic zwitterionic liquid.

21. The zwitterionic liquid according to claim 19 having a solubility in water of less than 5% by weight.

22. The zwitterionic liquid according to claim 21 which is an aromatic zwitterionic liquid.

23. A polymer electrolyte membrane having absorbed therein the zwitterionic imide compound according to claim 1.

24. A polymer electrolyte membrane having absorbed therein the zwitterionic imide compound according to claim 8.

25. A polymer electrolyte membrane having absorbed therein the zwitterionic imide compound according to claim 12.

26. A polymer electrolyte membrane having absorbed therein the zwitterionic liquid according to claim 19.

27. An electrochemical device comprising the polymer electrolyte membrane according to claim 23.

28. An electrochemical device comprising the polymer electrolyte membrane according to claim 24.

29. An electrochemical device comprising the polymer electrolyte membrane according to claim 25.

30. An electrochemical device comprising the polymer electrolyte membrane according to claim 26.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 02/24603

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C07C311/00 C07D213/71 C07D237/18 C07D239/18 C07D241/18
C07D233/54 C07D231/18 C07D277/20 C07D263/46 C07D249/12
H01M6/00 H01M6/22

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)
IPC 7 C07D C07C 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)
BEILSTEIN Data, CHEM ABS Data, EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WOLFGANG SUNDERMEYER ET AL: "Zur Synthese perfluorierter Sulfimide, RfN=SO2, und deren Stabilisierung durch tertiäre Amine" CHEM.BER., vol. 120, 1987, pages 1191-1195, XP001117928 compounds 17 and 20 page 1192 ---	1-5
X	RUDOLF ALBRECHT ET AL: "Umsetzung mit Pyridin-N-Oxiden und Diphenylnitron" CHEM.BER., vol. 98, no. 4, 1965, pages 1205-1209, XP001119421 compounds IIIa, IIIb, Va, Vb page 1206 -page 1207 --- -/--	1-5

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"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 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	Date of mailing of the international search report
8 November 2002	15/11/2002

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, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Österle, C

INTERNATIONAL SEARCH REPORT

Ir ational Application No
PCT/US 02/24603

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
X	FRIEDRICH BOBERG ET AL: "Reaktionen von Thioxoheterocyclen mit N-Chloramiden IV. N-Phenylsulfoximidate und N-(Phenylsulfonyl)Pyridinium-Sulfonamidate " PHOSPHORUS, SULFUR, AND SILICON, vol. 57, 1991, pages 235-247, XP001118786 *compounds 3a-3e, 6a-6f* examples	1-5
X	FRIEDRICH BOBERG ET AL: "Reaktionen von Thioxoverbindungen mit Chloramiden VI. Thiochinolone, Dihydrothiazolthione und Dihydroisothiazolthione mit Natrium-N-Chlorbenzolsulfonamiden" PHOSPHORUS, SULFUR, AND SILICON, vol. 108, 1996, pages 203-220, XP001119419 *compounds E*	1-5
X	FRIEDRICH BOBERG ET AL: "Reaktionen von Thioxoverbindungen mit N-Chloramiden VII. 5,5-Dimethyl-3-Dimethylamino-2-Cyclohexen-1-Thion und 3-Dimethylamino-1-Phenyl-2-Propen-1-Thion mit Natrium-N-Chlorbenzolsulfonamiden" PHOSPHORUS, SULFUR, AND SILICON, vol. 112, 1996, pages 91-99, XP001119418 *compound 7 alpha*	1-5
X	R. APPEL ET AL: "Derivate des Imidobissulfamids" ANGEWANDTE CHEMIE, vol. 79, no. 21, 1967, pages 937-938, XP001118787 *compounds 1a-1c	1-8,13, 16
A	PATENT ABSTRACTS OF JAPAN vol. 2000, no. 25, 12 April 2001 (2001-04-12) & JP 2001 233974 A (SUMITOMO ELECTRIC IND LTD), 28 August 2001 (2001-08-28) abstract	1-18, 23-25, 27-29
A	WO 99 49529 A (MINNESOTA MINING & MFG) 30 September 1999 (1999-09-30) cited in the application *the whole document*	1-18, 23-25, 27-29
A	US 5 965 054 A (MCEWEN ALAN B ET AL) 12 October 1999 (1999-10-12) cited in the application *the whole document*	1-18, 23-25, 27-29

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INTERNATIONAL SEARCH REPORT

Ir itional Application No
PCT/US 02/24603

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 463 005 A (DESMARTEAU DARRYL D) 31 October 1995 (1995-10-31) cited in the application *the whole document*	1-18, 23-25, 27-29

INTERNATIONAL SEARCH REPORT

ernational application No.
PCT/US 02/24603

Box I Observations where certain claims were found unsearchable (Continuation of item 1 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:

2. ☒ Claims Nos.: 19-22,26,30
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:
see FURTHER INFORMATION sheet PCT/ISA/210

3. ☐ 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 II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

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

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

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.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box I.2

Claims Nos.: 19-22,26,30

Present claims 19-22, 26 and 30 relate to a compound/product/method/apparatus defined by reference to the following parameter(s):

P1: melting point of less than 100°C

The use of these parameters in the present context is considered to lead to a lack of clarity within the meaning of Article 6 PCT. It is impossible to compare the parameters the applicant has chosen to employ with what is set out in the prior art. The lack of clarity is such as to render a meaningful complete search impossible. Consequently, the search has been restricted to the subject-matter of claims 1-18, 23-25 and 27-29.

The applicant's attention is drawn to the fact that claims, or parts of claims, relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

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