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(71) Applicant: **NITTO DENKO CORPORATION** [JP/JP];
1-2 Shimohozumi, 1- Chome, Ibaraki-shi, Osaka 567-8680
(JP).

(72) Inventors: **STANISLAW, Rachwal**; 4877 Gardenia
Street, Oceanside, California 92057 (US). **HU, Yufen**; 3963
Via Cangrejo, San Diego, California 92130 (US). **ZHANG,
Hongxi**; 32938 Embassy Avenue, Temecula, California
92592 (US).

(74) Agent: **JOHNSON, Brent A.** et al.; Maschoff Brennan, 20
Pacifica, Suite 1130, Irvine, California 92618 (US).

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(57) Abstract: The present disclosure generally relates to ionic compositions which may be used in or as an adhesive material for selectively adhering two items together. More particularly, but not exclusively, the present disclosure relates to ionic compositions that include a cationic imidazolium compound and an anionic compound such as a sulfonyl imide compound.



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IONIC COMPOSITIONS AND RELATED USES THEREOF

Inventors: Stanislaw Rachwal, Yufen Hu, Hongxi Zhang

5 **CROSS-REFERENCE TO RELATED APPLICATIONS**

This application claims the benefit of U.S. Provisional Patent Application No. 62/466,112 filed March 2, 2017, the content of which is incorporated herein by reference in its entirety.

BACKGROUND

10 The present disclosure generally relates to ionic compositions which may be used in or as an adhesive material for selectively adhering two items together. More particularly, but not exclusively, the present disclosure relates to ionic compositions that include a cationic imidazolium compound and an anionic compound such as a sulfonyl imide compound.

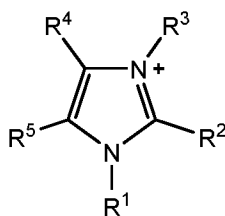
Certain compositions are known which may be used as an adhesive coating that is applied to an electrically conductive surface of a first substrate. The adhesive coating may be sandwiched between the electrically conductive surface of the first substrate and an electrically conductive surface of a second substrate in order to adhere or join the first and second substrates together. Upon the application of an electric potential, the adhesive coating is de-bonded from one or both of the first and second substrates in order to separate the first and second substrates from one another. It has been observed however that certain forms of this type of coating may have an undesired corrosive effect on the electrically conductive surfaces to which they are applied. Thus, there remains a need for further contributions in this area of technology.

The subject matter disclosed and claimed herein is not limited to embodiments that solve any disadvantages or that operate only in environments such as those described above. Rather, this background is only provided to illustrate examples of where the present disclosure may be utilized.

25 **SUMMARY**

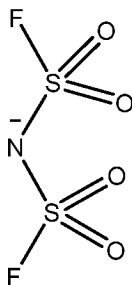
The present disclosure generally relates to ionic compositions which may be used in or as an adhesive material for selectively adhering two items together. More particularly, but not exclusively, the present disclosure relates to ionic compositions that include a cationic imidazolium compound and an anionic compound such as an anionic sulfonyl imide compound.

30 In one embodiment, a composition includes a cationic compound according to Formula (I):

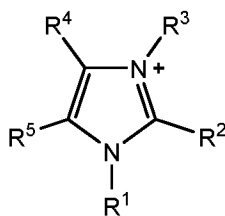


Formula (I)

wherein R¹ represents a C₁-C₈ alkyl, a C₁-C₈ hydroxyalkyl, a C₁-C₈ alkenyl, or a C₁-C₈ alkoxyalkyl, R² represents hydrogen or a C₁-C₃ alkyl, R³ represents a C₁-C₈ alkyl, a C₁-C₈ alkenyl, a C₁-C₈ alkoxyalkyl, an acetoxy C₁-C₃ alkyl, or a C₇-C₁₅ arylalkyl, R⁴ represents hydrogen or a C₁₋₄ alkyl, and R⁵ represents hydrogen or a C₁₋₄ alkyl. In one form of this embodiment, the composition further includes an anionic compound having the following structure:



In another embodiment, an apparatus includes first and second substrates which are adhered together by a composition that includes a cationic compound according to Formula (I):



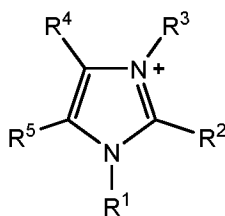
Formula (I)

10

wherein R¹ represents a C₁-C₈ alkyl, a C₁-C₈ alkenyl, or a C₁-C₈ alkoxyalkyl, R² represents hydrogen or a C₁-C₃ alkyl, R³ represents a C₁-C₈ alkyl, a C₁-C₈ hydroxyalkyl, a C₁-C₈ alkenyl, a C₁-C₈ alkoxyalkyl, an acetoxy C₁-C₃ alkyl, or a C₇-C₁₅ arylalkyl, R⁴ represents hydrogen or a C₁₋₄ alkyl, and R⁵ represents hydrogen or a C₁₋₄ alkyl.

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In another embodiment, a method involves adhering a first substrate to a second substrate with a composition that includes a cationic compound according to Formula (I):



Formula (I)

wherein R¹ represents a C₁-C₈ alkyl, a C₁-C₈ alkenyl, or a C₁-C₈ alkoxyalkyl, R² represents hydrogen or a C₁-C₃ alkyl, R³ represents a C₁-C₈ alkyl, a C₁-C₈ hydroxyalkyl, a C₁-C₈ alkenyl, a C₁-C₈ alkoxyalkyl, an acetooxy C₁-C₃ alkyl, or a C₇-C₁₅ arylalkyl, R⁴ represents hydrogen or a C₁₋₄ alkyl, and R⁵ represents hydrogen or a C₁₋₄ alkyl.

This Summary introduces a selection of concepts in a simplified form that are further described below in the Detailed Description. This Summary is not intended to identify key features or essential characteristics of the claimed subject matter, nor is it intended to be used as an aid in determining the scope of the claimed subject matter.

Additional features and advantages will be set forth in the description which follows, and in part will be obvious from the description, or may be learned by practice. The features and advantages may be realized and obtained by means of the instruments and combinations particularly pointed out in the appended claims. These and other features will become more fully apparent from the following description and appended claims.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic illustration of the use of an ionic composition described herein for adhering two substrates together.

FIG. 2 is a schematic illustration of the release or de-bonding of the two substrates of FIG. 1 upon application of an electric potential.

FIG. 3 is a schematic illustration of an apparatus used for testing adhesion properties of an ionic composition described herein.

FIG. 4 is a graphical illustration of peeling strength density vs. time of the ionic composition tested in connection with FIG. 3.

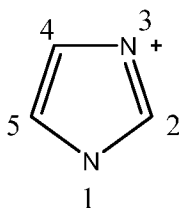
DETAILED DESCRIPTION

For purposes of promoting an understanding of the present disclosure, reference will now be made to the following embodiments and specific language will be used to describe the same. It will nevertheless be understood that no limitation of the scope of the disclosure is thereby intended, such alterations and further modifications in the described subject matter, and such further applications of the disclosed principles as described herein being contemplated as would normally occur to one skilled in the art to which the disclosure relates.

The present disclosure generally relates to ionic compositions which may be used in or as an adhesive material for selectively adhering two items together. More particularly, but not exclusively, the present disclosure relates to ionic compositions that include a cationic imidazolium compound and an anionic compound such as a sulfonyl imide compound.

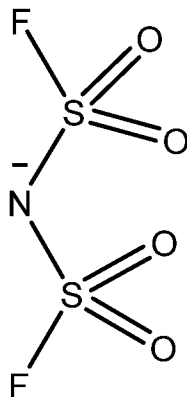
5 As used herein, when a compound or chemical structural is referred to as being “optionally substituted” it includes a feature that has no substituents (i.e. unsubstituted), or a feature that is “substituted,” meaning that the feature has one or more substituents. A substituted group is derived from the unsubstituted parent structure wherein one or more hydrogen atoms on the parent structure have been independently replaced by one or more substituent groups. A substituted group may have one or more
 10 substituent groups on the parent group structure. In one or more forms, the substituent groups may be independently selected from optionally substituted alkyl or alkenyl, alkoxy (e.g. $-\text{OCH}_3$, $-\text{OC}_2\text{H}_5$, $-\text{OC}_3\text{H}_7$, $-\text{OC}_4\text{H}_9$, etc.), alkylsulfones (e.g. $-\text{SCH}_3$, $-\text{SC}_2\text{H}_5$, $-\text{SC}_3\text{H}_7$, $-\text{SC}_4\text{H}_9$, etc.), $-\text{NR}'\text{R}''$, $-\text{OH}$, $-\text{SH}$, $-\text{CN}$, $-\text{NO}_2$, or a halogen, wherein R' and R'' are independently H or an optionally substituted alkyl.

As used herein, the term “imidazolium” refers to the ring system having the following structure:

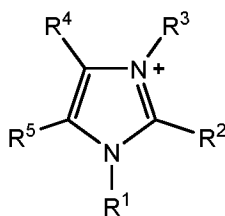


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As used herein, the terms “bis(sulfonyl)imide” and/or “sulfonyl imide” refer to a heteroatom moiety having, for example, the following structure:



20 In one embodiment, an ionic composition includes a cationic imidazolium compound according to Formula (I):



Formula (I)

wherein R¹ may represent a C₁-C₈ alkyl, a C₁-C₈ alkenyl, or a C₁-C₈ alkoxyalkyl; R² may represent hydrogen or a C₁-C₃ alkyl; R³ may represent a C₁-C₈ alkyl, a C₁-C₈ hydroxyalkyl, a C₁-C₈ alkenyl, a C₁-C₈ alkoxyalkyl, an acetoxy C₁-C₃ alkyl, or a C₇-C₁₅ arylalkyl; and R⁴ and R⁵ may independently
5 represent hydrogen or a C₁₋₄ alkyl.

In some forms, one or more of R¹, R², R³, R⁴ and R⁵ may represent a hydrophilic functional group. In a more particular form, one more of R¹, R², and R³ may represent a hydrophilic functional group. In one or more of these forms, the hydrophilic functional group may include oxygen. In some forms, the oxygen containing hydrophilic functional group may include an ether, hydroxyl, alkoxy and/or
10 ester group. In other forms, the hydrophilic functional group may include nitrogen, sulfur and/or phosphorous. In still other forms, the hydrophilic functional group may include an amino group, a sulfhydryl group or a phosphate group.

In one or more forms, one or more of R¹, R², R³, R⁴ and R⁵ may represent a hydrophobic functional group. In a more particular form, one or more of R¹, R², and R³ may represent a hydrophobic functional group. In one or more of these forms, the hydrophobic functional group may include an
15 optionally substituted alkyl group. In some forms, the optionally substituted alkyl group may include a methyl, ethyl, and/or propyl group. In other forms, the hydrophobic functional group[s] may include an optionally substituted aryl group. In some forms, the optionally substituted aryl group may include a phenyl and/or benzyl group.

In one or more forms, one or more of R¹, R², R³, R⁴ and R⁵ may be independently substituted, and the cationic compound according to Formula (I) may be asymmetrical.

As indicated above, R¹ may represent a C₁-C₈ alkyl, a C₁-C₈ alkenyl, or a C₁-C₈ alkoxyalkyl. In some more particular forms, R¹ may represent a C₁-C₃ alkyl or a C₁-C₃ alkoxyalkyl. In one or more of these forms, the C₁-C₃ alkyl may be a methyl, ethyl, n-propyl, or isopropyl group. In some forms, the C₁-
25 C₈ alkoxyalkyl may represent a methyloxymethyl group.

As indicated above, R² may represent hydrogen or a C₁-C₃ alkyl. In some more particular forms, R² may represent hydrogen. In some more particular forms, R² may represent a methyl or ethyl group.

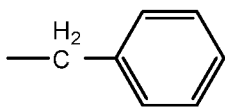
As indicated above, R³ may represent a C₁-C₈ alkyl, a C₁-C₈ hydroxyalkyl, a C₁-C₈ alkenyl, a C₁-C₈ alkoxyalkyl, an acetoxy C₁-C₃ alkyl, or a C₇-C₁₅ arylalkyl. In some more particular forms, R³ may represent a C₁-C₈ alkyl, a C₁-C₈ alkenyl, or a C₁-C₈ alkoxyalkyl. In some other forms, R³ may represent a
30 C₁-C₃ alkyl, a C₁-C₃ alkoxy, a C₁-C₃ hydroxyalkyl such as -CH₂CH₂-OH, a C₁-C₃ alkoxy, an acetoxy C₁-C₃ alkyl, or a C₅-C₇ arylalkyl such as -CH₂-phenyl. In some forms, R³ may represent a methyl or ethyl

group. In other forms, R³ may represent a C₁-C₃ alkoxy which may be, for example, a methoxy or ethoxy group. In some forms, R³ may represent C₁-C₃ hydroxyalkyl such as -CH₂CH₂-OH. In some forms R³ may represent a methyloxymethyl or ethoxymethyl group. In other forms, R³ may represent an acetoxy C₁-C₃ alkyl which may be an acetoxyethyl group. In some forms, R³ may represent a C₅-C₇ arylalkyl group which may be a benzyl group.

As indicated above, R⁴ and R⁵ may independently represent hydrogen or a C₁₋₄ alkyl. In one more particular form, at least one of R⁴ and R⁵ is a C₁₋₄ alkyl. In one particular form, R⁴ represents a C₁-C₂ alkyl and R⁵ represents hydrogen. In another form, R⁴ represents hydrogen and R⁵ represents a C₁-C₂ alkyl. In another form, R⁴ represents methyl and R⁵ represents hydrogen. In another form, R⁴ represents hydrogen and R⁵ represents methyl. In another form, one or both of R⁴ and R⁵ represent C₁-C₂ alkyls. In yet another form, one or both of R⁴ and R⁵ represent a C₁-C₄ alkyl. In still another form, R⁴ represents a C₁-C₂ alkyl or a C₁-C₄ alkyl and R⁵ represents hydrogen. In a further form, R⁵ represents a C₁-C₂ alkyl or a C₁-C₄ alkyl and R⁴ represents hydrogen. In another form, R⁴ and R⁵ both represent a C₁-C₂ alkyl which may be, for example, a methyl group and/or an ethyl group. In another form, R⁴ and R⁵ both represent CH₃.

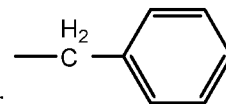
Still, other variations for the cationic compound according to Formula (I) are possible. For example, in one form, R¹ may be selected from -H, -CH₃, -CH₂-CH₃, -CH₂-(CH₃)₂, and -CH₂-O-CH₃, R² may be selected from -H, -CH₃, and -CH₂-CH₃, R³ may be selected from -H, -CH₃, -CH₂-CH₃, -

CH₂-(CH₃)₂, -CH₂-O-CH₃, -CH₂-CH₂-OH, -CH₂-CH₂-O-CH₃, -CH₂-CH₂-O-C(=O)-CH₃, or



, R⁴ may be selected from -H, -CH₃, or -CH₂-CH₃, and/or R⁵ may be selected from -H, -CH₃, and/or -CH₂-CH₃. As another example, in another form, R¹ may be selected from -CH₃, -CH₂-CH₃, -CH₂-(CH₃)₂, and -CH₂-O-CH₃, R² may be selected from -H, -CH₃, and -CH₂-CH₃, R³ may

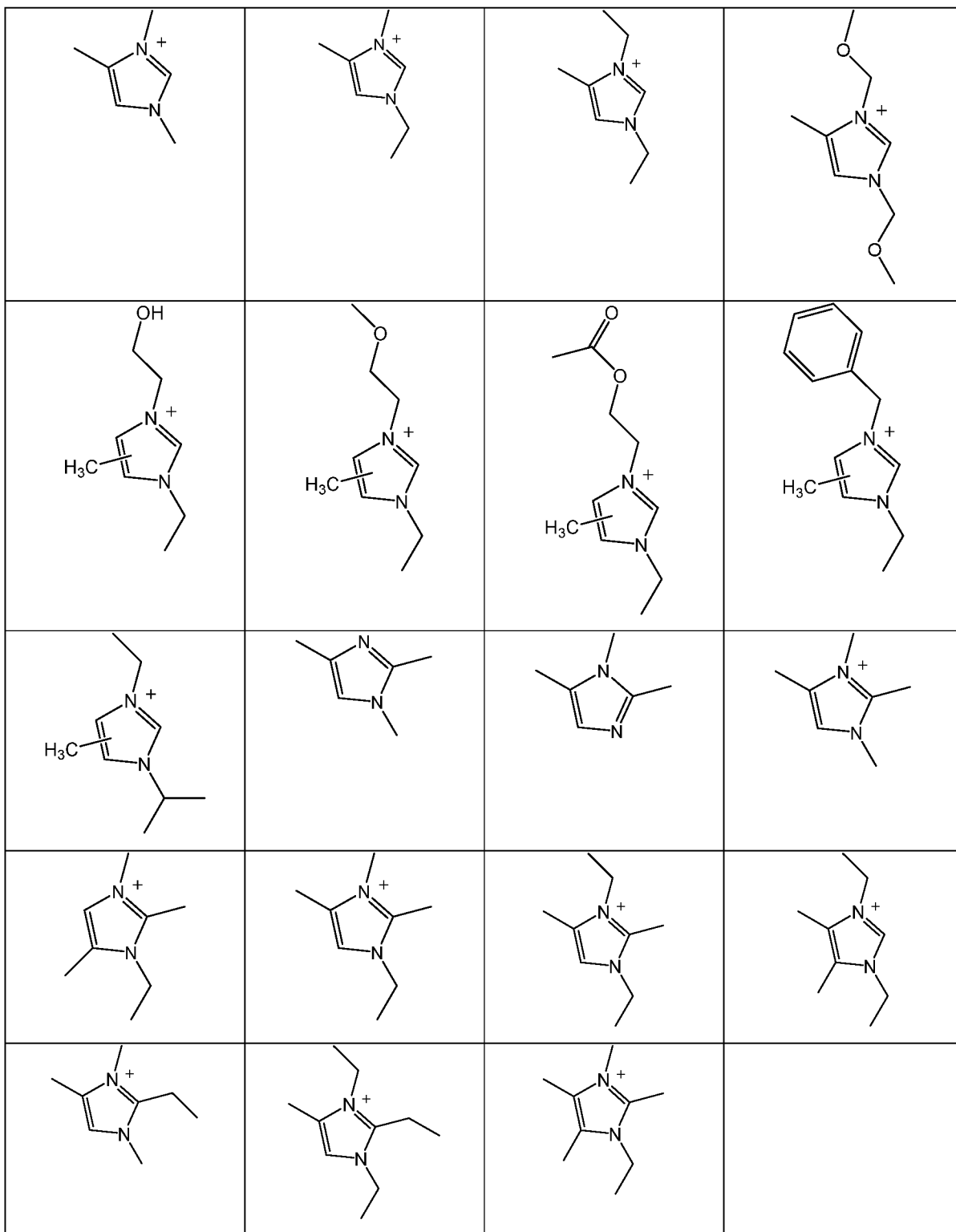
be selected from -CH₃, -CH₂-CH₃, -CH₂-O-CH₃, -CH₂-CH₂-O-CH₃, or



, R⁴ may be selected from -H or -CH₃, and/or R⁵ may be selected from -H or -CH₃. In yet a further example, in another form, R¹ may be selected from -CH₃, -CH₂-CH₃, and -CH₂-(CH₃)₂, R² may be selected from -H, -CH₃, and -CH₂-CH₃, R³ may be selected from -CH₃ and -CH₂-CH₃, R⁴ may be selected from -H or -CH₃, and/or R⁵ may be selected from -H or -CH₃.

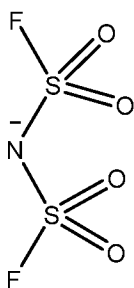
Representative compounds according to Formula (I) include, but are not limited to, those presented in Table 1:

TABLE 1



The ionic composition disclosed herein may also include an anionic compound. In one form, the anionic compound may be a bis(sulfonyl)imide anion. In a more particular form, the bis(sulfonyl)imide anion may be a bis(fluorosulfonyl)imide compound. In one form, the bis(fluorosulfonyl)imide compound may have the following structure:

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The ionic compositions described herein may be utilized as, or in, an adhesive material which may be used to bond two or more items together in a releasable fashion. Stated alternatively, the adhesive material may be used to selectively bond the items together, allowing for the adhesive material to be de-bonded from one or more of the items and facilitate separation of the items if desired. More particularly, an adhesive material according to the present disclosure may be provided on an electrically conductive surface of a first substrate, and an electrically conductive surface of a second substrate may be positioned in contact with the adhesive material in order to bond or join the first and second substrates together. In this configuration, the adhesive material is sandwiched between the first and second substrates, although other variations are contemplated. As indicated above, if desired, the adhesive material facilitates de-bonding and separation of the first and second substrates. More specifically, upon the application of an electric potential, the adhesive material will be de-bonded or released from the conductive surface of one or both of the substrates, resulting in separation of the first and second substrates from one another.

While not previously described, it should be appreciated that the compositions disclosed herein may include components in addition to the cationic and anionic compounds. For example, in one form, the compositions may also include a polymer. Non-limiting examples of polymers which could be present in the composition include those described in JP 2015-204998 and/or JP 2015-204996. In one form the polymer may have a glass transition temperature below 0 °C, although other variations are possible. In one form, the polymer may be an acrylic polymer, such as but not limited to, an acrylic polymer which contains a monomer unit derived from a C₁-C₁₄ alkyl group containing alkyl (meth)acrylate ester. In other forms however, the acrylic polymer can contain a monomer unit derived from a C₁-C₁₄ alkyl or alkoxy group. In one form, the acrylic polymer may contain an alkyl (meth)acrylate ester, and a monomer unit derived from a polar group containing monomer. In one aspect of this form, the polar group containing monomer may be a carboxyl group containing monomer. In an additional or alternative aspect of this form, the C₁-C₁₄ alkyl group containing alkyl (meth)acrylate ester is butyl (meth)acrylate.

It is contemplated that the compositions described herein could be utilized for a number of different applications, including for example those disclosed in JP 2015-204996 and/or JP20204997.

Referring now to FIGS. 1 and 2, additional details regarding the use of an ionic composition described herein for selectively bonding two substrates together in apparatus 200 will be provided. An adhesive material 203 which includes an ionic composition described herein provides a layer or coating

positioned between electrically conductive surface 206 of substrate 202 and electrically conductive surface 207 of substrate 201. In one form, one or both of substrates 201, 202 may be formed of an electrically conductive material such that one or both of electrically conductive surfaces 206, 207 is/are formed of the same material as the remainder of substrates 201, 202. However, it is possible in other forms to use one or more electrically conductive materials for electrically conductive surfaces 206, 207 which are different from the material(s) forming substrates 201, 202. Similarly, it should be appreciated that one or both of substrates 201, 202 could be formed of one or more materials which are not electrically conductive provided that surfaces 206, 207 are electrically conductive. In these forms, electrically conductive surfaces 206, 207 may be provided as a coating or layer on substrates 201, 202.

In the illustrated form, electrically conductive surfaces 206, 207 are electrically coupled to or in electrical communication with a power source 204 in a closeable electrical circuit that includes an intervening switch 205. In one form, power source 204 may be a direct current power supply that provides a DC voltage in the range of about 3V to 100V, although other variations are contemplated. When switch 205 is closed, the electrical potential is applied between electrically conductive surfaces 206, 207 in order to de-bond adhesive material 203 from one or both of electrically conductive surfaces 206, 207 and, as a result, allow substrates 201 and 202 to be physically separated from one another.

In one form, one or both of substrates 201, 202 may include an electrically conductive carbonaceous material or an electrically conductive metal. As suggested above, one or both of substrates 201, 202 may also include an electrically conductive layer which may be formed of a metallic material such as, but not limited to, aluminum. The electrically conductive layer may include a conventional material such as a metal, mixed metal, alloy, metal oxide, and/or composite metal oxide, or it may include a conductive polymer. Examples of suitable metals for the electrically conductive layer include the Group 1 metals, the metals in Groups 4, 5, 6, and the Group 8-10 transition metals. Further examples of suitable metals for the electrically conductive layer include stainless steel, Al, Ag, Mg, Ca, Cu, Mg/Ag, LiF/Al, CsF, and/or CsF/Al and/or alloys thereof. If an electrically conductive layer is present, it may have a thickness in the range of about 1 nm to about 1000 μm . In one form, the electrically conductive layer has a thickness from about 20 nm to about 200 μm , and in another form the electrically conductive layer has a thickness in the range of about 20 nm to about 200 nm.

While not previously discussed, it should be appreciated that the ionic compositions described herein may provide various properties which are desirable for certain applications. For example, in some forms, the ionic compositions disclosed herein may eliminate or reduce corrosion of the electrically conductive surfaces on which they are positioned. In one form for example, the ionic compositions disclosed herein include components which reduce the acidity of the environment immediately adjacent to the electrically conductive surfaces. In one aspect, an adhesive material may include one or more materials, in addition to the cationic and anionic compounds themselves, which may be used to reduce the corrosiveness of the ionic cations and/or anions immediately adjacent the electrically conductive surfaces. The corrosive effect of an adhesive material may be assessed pursuant to the procedures

described in ASTM G69-12 (Standard Test Method for Measurement of Corrosion Potentials of Aluminum Alloys). Additional procedures for assessing the corrosive effect of an adhesive material on the electrically conductive surfaces are described in the Examples of the subject application.

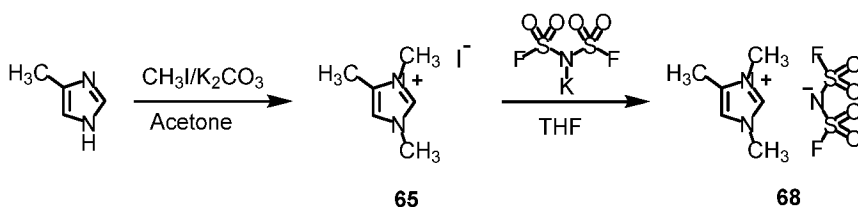
In one form, an adhesive material including an ionic composition disclosed herein may be chemically stable relative to an electrically conductive electrode or an electrically conductive material; i.e., there is a lack of (or minimal presence of) undesired reactions between a metal electrode and the adhesive material. Undesired reactions may include, for example, corrosive degradation of the metal electrode, dissolution of the metal in the selectively adherent adhesive and/or pitting of the metal electrode. An adhesive material including an ionic composition disclosed herein may be chemically stable relative to aluminum, stainless steel and/or mixtures thereof, just to provide a few examples. In one form, contact of an adhesive material including an ionic composition disclosed herein upon an electrically conductive surface may result in the absence of, or minimize, any corrosive degradation of the surface for a period of at least or greater than 15 minutes, 30 minutes, 1 hour, 3 hours, 5 hours, 7 hours, 24 hours, 50 hours, 100 hours, 125 hours, 200 hours and/or 300 hours. In some forms, direct contact of an adhesive material including an ionic composition disclosed herein upon an electrically conductive surface may minimize and/or prevent corrosive degradation of the surface for one of the time periods identified above in an environment of 85°C and 85% relative humidity. In one form, the absence of any corrosive degradation can be demonstrated by a lack of total penetration of an electrically conductive 50 nm thick sheet of aluminum foil for one of the time periods identified above and/or at the environmental conditions identified above.

In one form, an adhesive material including an ionic composition described herein may be formulated to minimize corrosion of an electrically conductive surface under conditions of prolonged high humidity and high temperature. For example, an adhesive composition may be capable of maintaining two substrates in fixed relation to each other during and after being subjected to Accelerated Aging Test Method II (preferably after exposure to 85°C and 85% relative humidity for one of the periods of time identified above). Also, while not previously discussed, it should be appreciated that the ionic compositions disclosed herein may have a molar mass that is less than or equal to about 160 grams per mole.

EXAMPLES

It should be appreciated that the following Examples are for illustration purposes and are not intended to be construed as limiting the subject matter disclosed in this document to only the embodiments disclosed in these examples.

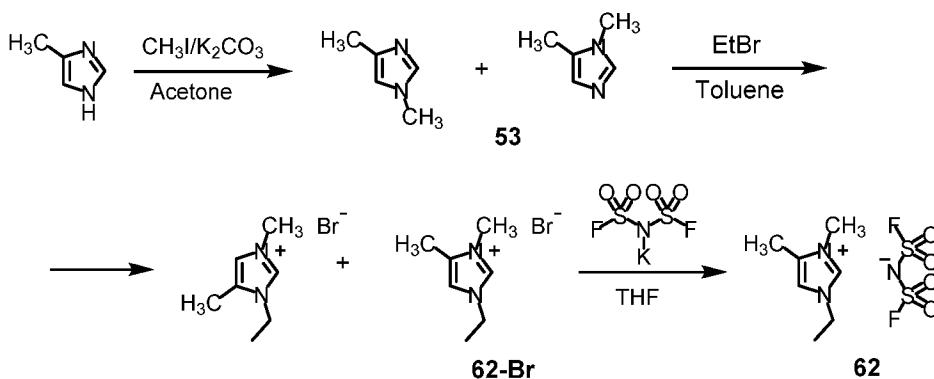
Example 1: Preparation of 1,3,4-Trimethylimidazolium bis(fluorosulfonyl)imide (68).



Iodomethane (62.2 mL, 1.0 mol) was added dropwise within 2 hours to a mixture of 4-methylimidazole (20.53 g, 250 mmol), potassium carbonate (41.40 g, 300 mmol), and acetone (300 mL) and stirred vigorously under argon. Heat from the reaction caused the temperature to rise to a boil. After the addition was complete, the mixture was stirred at 60°C for 3 hours. The solid was filtered off from the hot reaction mixture and washed with boiling acetone (2 x 300 mL). The combined filtrate and washings were concentrated under reduced pressure and triturated with THF (500 mL) to give crystalline iodide product (65) (41.16 g, 69% yield).

A mixture of iodide (65) (9.52 g, 40 mmol), KFSI (8.76 g, 40 mmol) and THF (100 mL) was stirred under argon and heated at 70°C for 4 hours. After cooling overnight, the solid was filtered off, and the solvent was removed under reduced pressure. A solution of the residue in ethyl acetate (100 mL) was washed with water (50 mL), filtered through a paper filter, and the solvent was removed under reduced pressure to give ionic liquid (68) (8.00 g, 69% yield). ¹H NMR (400 MHz, DMSO-d₆): δ 8.93 (s, 1H), 7.42 (s, 1H), 3.79 (s, 3H), 3.73 (s, 3H), 2.26 (s, 3H).

Example 2: Preparation of 3,4-Dimethyl-1-ethylimidazolium bis(fluorosulfonyl)imide (62).



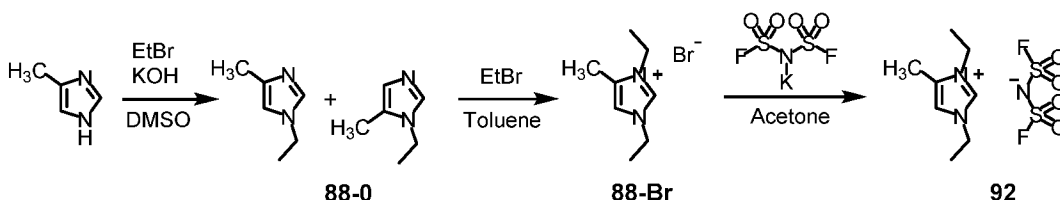
A solution of iodomethane (37.3 mL, 600 mmol) in acetone (40 mL) was added dropwise within 4 hours to a mixture of 4-methylimidazole (41.06 g, 500 mmol), potassium carbonate (103 g, 750 mmol) and acetone (500 mL). The mixture was then stirred at room temperature for 16 hours. The solid was filtered off, and the solvent was removed under reduced pressure. The residue was triturated with THF (200 mL) and set aside at room temperature overnight. The solid was removed by filtration and the solvent was evaporated under reduced pressure to give a mixture of isomers (53) (30.70 g), containing also some starting 4-methylimidazole.

A mixture of imidazoles (53) (30.00 g, 312 mmol), bromoethane (20 mL, 268 mmol) and toluene (50 mL) was placed in a pressure reactor and heated at 80°C for 16 hours. The solvent was removed

under reduced pressure. The residue was triturated with acetone (100 mL) and set aside at room temperature overnight. Obtained crystals were filtered off, washed with acetone and dried in a vacuum oven to give imidazolium bromide (62-Br) (6.43 g, 10% yield).

A mixture of imidazolium bromide (62-Br) (6.43 g, 30.7 mmol), KFSI (6.57 g, 30 mmol) and THF (100 mL) was stirred under argon and heated at 75°C for 5 hours. After cooling to 50°C, the solid was filtered off, and the solvent was removed under reduced pressure to give ionic liquid (62) (8.61 g, 94% yield). ¹H NMR (400 MHz, DMSO-d₆): δ 9.01 (s, 1H), 7.44 (s, 1H), 4.10 (q, J = 7.3 Hz, 2H), 3.79 (s, 3H), 2.29 (s, 3H), 1.38 (t, J = 7.3 Hz, 3H).

10 Example 3: Preparation of 1,3-Diethyl-4-methylimidazolium bis(fluorosulfonyl)imide (92).

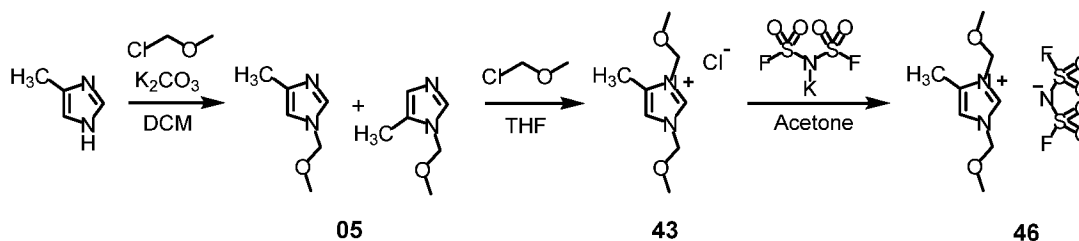


A mixture of 4-methylimidazole (39.80 g, 485 mmol), 90% KOH (31.10 g, 500 mmol) and anhydrous DMSO (150 mL) was stirred under argon for 3 hours. Bromoethane (37.2 mL, 500 mmol) was added dropwise while the reaction mixture was cooled in an ice-water bath. The mixture was then stirred at room temperature overnight. The reaction mixture was poured into ice-water (700 mL), treated with 5N NaOH (100 mL) and extracted with DCM/THF (9:1, 3 x 300 mL). The extract was washed with water (300 mL), dried over anhydrous sodium carbonate, and the solvent was removed under reduced pressure to give imidazoles (88-0) (41.97 g, 78% yield) as a mixture of two isomers in approximately equal amounts.

A mixture of imidazoles (88-0) (11.02 g, 100 mmol), bromoethane (11.2 mL, 150 mmol) and anhydrous toluene (50 mL) was sealed in a pressure reactor and heated at 80°C for 16 hours. After cooling, the bottom phase was separated and triturated with ethyl ether (100 mL) to give a crystalline material. The product was recrystallized from acetone (100 mL) to give large shiny crystals of imidazolium bromide (88-Br) (13.83 g, 63% yield).

A mixture of imidazolium bromide (88-Br) (8.57 g, 39.1 mmol), KFSI (8.57 g, 39.1 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 2 hours. The solid was filtered off, and the solvent was removed under reduced pressure. A solution of the residue in THF (100 mL) was set aside at room temperature overnight, filtered to remove a small amount of additional precipitate, and the solvent was evaporated under reduced pressure to give 1,3-diethyl-4-methylimidazolium bis(fluorosulfonyl)imide (92) (12.48 g, 100% yield). ¹H NMR (500 MHz, DMSO-d₆): δ 9.07 (s, 1H), 7.53 (s, 1H), 4.13 (q, J = 7 Hz, 2H), 4.10 (q, J = 7.0 Hz, 2H), 2.29 (s, 3H), 1.40 (m, 6H).

Example 4: Preparation of 1,3-Bis(methoxymethyl)-4-methylimidazolium bis(fluorosulfonyl)imide (46).

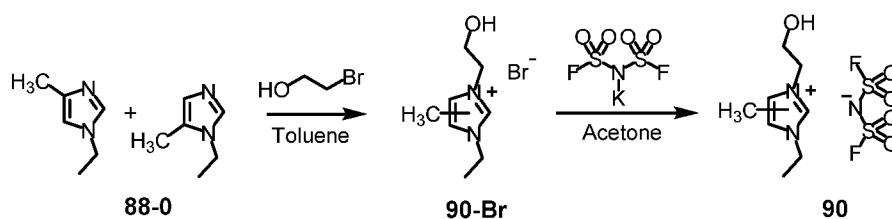


Chloromethyl methyl ether (51 mL, 634 mmol) was added dropwise within 3 hours to a vigorously stirred mixture of 4-methylimidazole (32.84 g, 400 mmol), potassium carbonate (89 g, 645 mmol) and DCM (200 mL). After addition, the mixture was stirred at room temperature overnight. The solid was filtered off and washed with acetone (300 mL). The combined filtrate and washings were concentrated under reduced pressure and the residue was chromatographed using silica gel and ethyl acetate/methanol (85:15) to give an isomeric mixture of mono-methoxy-methylated imidazoles (05) (13.48 g, 27% yield).

Chloromethyl methyl ether (11.4 mL, 150 mmol) was added dropwise to a solution of imidazoles (05) (12.00 g, 95 mmol) in anhydrous THF (50 mL) and stirred under argon at room temperature. After the addition, stirring was continued for 5 hours. The mixture was divided into two layers. The bottom layer was separated, washed with ethyl ether (200 mL) and dissolved in methanol (200 mL). The dark solution was treated with charcoal (10 g) and stirred for 20 hours. The color turned yellowish. The solution was filtered through a layer of Celite, and the solvent was evaporated under reduced pressure to give imidazolium chloride (43) (17.69 g, 90% yield).

A mixture of imidazolium chloride (43) (9.92 g, 48 mmol), KFSI (10.52 g, 48 mmol) and acetone (100 mL) was stirred under argon at 60°C for 1 hour. Charcoal (5 g) was added, and the mixture was stirred at room temperature for 2 hours. The solution was filtered through a layer of Celite, and the solvent was removed under reduced pressure to give ionic liquid (46) (15.46 g, 92% yield). ¹H NMR (400 MHz, DMSO-d₆): δ 9.45 (s, 1H), 7.69 (s, 1H), 5.55 (s, 2H, N-CH₂-O), 5.51 (s, 2H, N-CH₂-O), 3.33 (s, 3H, CH₃O), 3.32 (s, 3H, CH₃O), 2.34 (s, 3H).

Example 5: Preparation of 1-Ethyl-3-(2-hydroxyethyl)-4(5)-methylimidazolium bis(fluorosulfonyl)imide (90).

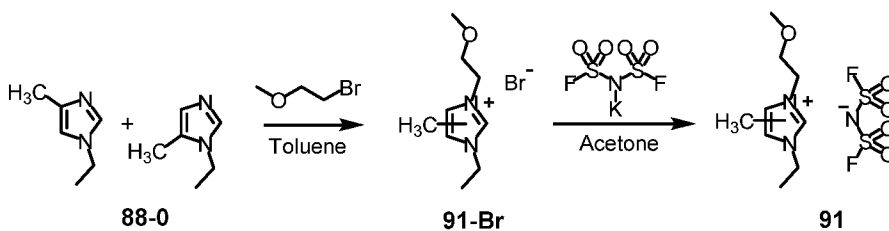


An isomeric mixture of imidazoles (88-0) (Example 3) (5.50 g, 50 mmol), 2-bromoethanol (5.3 mL, 75 mmol) and anhydrous toluene (25 mL) were sealed in a pressure reactor and heated at 100°C for

16 hours. After cooling, the bottom phase was separated and triturated with ethyl ether (50 mL) to give an oily product. The oil was dried in a vacuum oven to give a mixture of isomers (90-Br) (11.57 g, 98% yield).

A mixture of isomers (90-Br) (9.87 g, 39.6 mmol), KFSI (8.68 g, 39.6 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 2 hours. The solid was filtered off, and the solvent was removed under reduced pressure. A solution of the residue in THF (100 mL) was set aside at room temperature overnight. It was then filtered to remove a small amount of additional precipitate, and the solvent was evaporated under reduced pressure to give a mixture of 1-ethyl-3-(2-methoxyethyl)-4-methylimidazolium bis(fluorosulfonyl)imide and its 5-methylimidazolium isomer (90) (7.64 g, 97% yield). ¹H NMR of the major isomer (500 MHz, DMSO-d₆): δ 9.02 (s, 1H), 7.54 (s, 1H), 5.14 (bs, 1H, OH), 4.16 (m, 4H), 3.70 (m, 2H), 2.26 (s, 3H), 1.41 (t, J = 7.5 Hz, 3H).

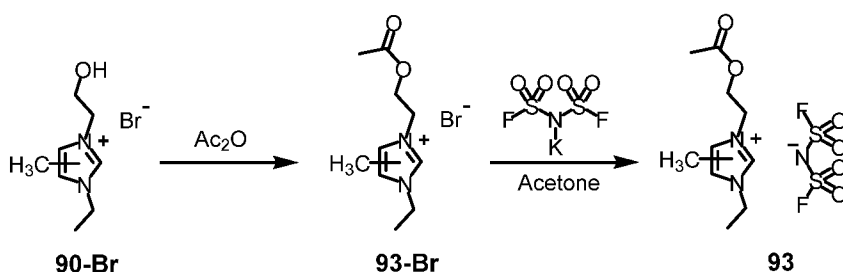
Example 6: Preparation of 1-Ethyl-3-(2-methoxyethyl)-4(5)-methylimidazolium bis(fluorosulfonyl)imide (91).



An isomeric mixture of imidazoles (88-0) (Example 3) (5.50 g, 50 mmol), 2-bromoethyl methyl ether (10.42 g, 75 mmol) and anhydrous toluene (25 mL) were sealed in a pressure reactor and heated at 80°C for 16 hours. After cooling, the bottom phase was separated and triturated with ethyl ether (50 mL) to give an oily product. The oil was dried in a vacuum oven to give a mixture of isomers (91-Br) (9.87 g, 79% yield).

A mixture of isomers (91-Br) (9.87 g, 39.6 mmol), KFSI (8.68 g, 39.6 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 2 hours. The solid was filtered off, and the solvent was removed under reduced pressure. A solution of the residue in THF (100 mL) was set aside at room temperature overnight, filtered to remove a small amount of additional precipitate, and the solvent was evaporated under reduced pressure to give a mixture of 1-ethyl-3-(2-methoxyethyl)-4-methylimidazolium bis(fluorosulfonyl)imide and its 5-methylimidazolium isomer (91) (13.57 g, 98% yield), a mixture of two isomers in an approximate molar ratio of 55:45. ¹H NMR of the major isomer (500 MHz, DMSO-d₆): δ 9.03 (s, 1H), 7.54 (s, 1H), 4.28 (t, J = 5.0 Hz, 2H), 4.16 (q, J = 7.5 Hz, 2H), 3.66 (t, J = 4.5 Hz, 2H), 3.27 (s, 3H, MeO), 2.29 (s, 3H), 1.40 (t, J = 7.5 Hz, 3H).

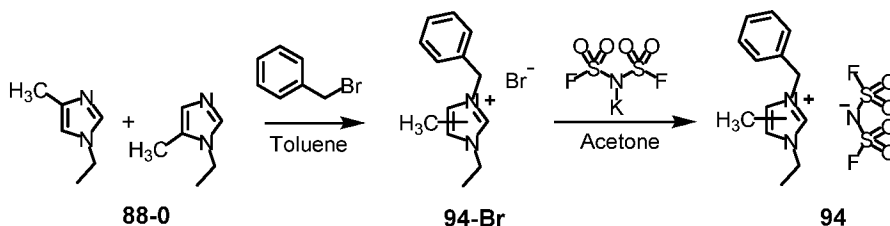
Example 7: Preparation of 1-Ethyl-3-(2-acetoxyethyl)-4(5)-methylimidazolium bis(fluorosulfonyl)imide (93).



A mixture of imidazolium bromide (90-Br) (Example 5) (6.43 g, 27.3 mmol) and acetic anhydride (20 mL) was stirred under argon and heated at 100°C for 6 hours. The mixture was diluted with m-xylene (100 mL) and concentrated under reduced pressure. Dilution with m-xylene and evaporation under reduced pressure was repeated to give product (93-Br) (7.20 g, 95% yield) as a mixture of two isomers in an approximately 1:1 ratio.

A mixture of product (93-Br) (7.00 g, 25.2 mmol), KFSI (5.54 g, 25.2 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 3 hours. After cooling, the solid was filtered off, and the solvent was removed under reduced pressure. A solution of the residue in anhydrous THF (75 mL) was set aside at room temperature overnight, then filtered and concentrated under reduced pressure to give ionic liquid (93).

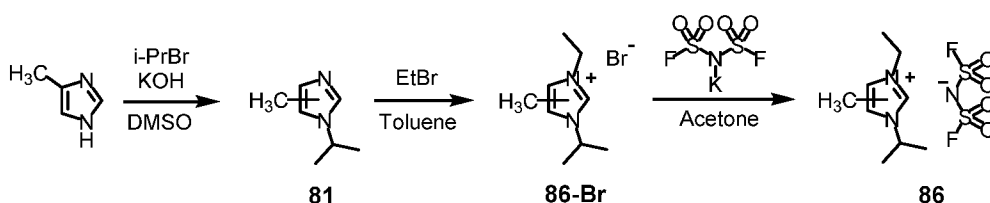
Example 8: Preparation of 1-Ethyl-3-benzyl-4(5)-methylimidazolium bis(fluorosulfonyl)imide (94).



A mixture of imidazoles (88-0) (Example 3) (8.30 g, 75 mmol), benzyl bromide (11.2 mL, 100 mmol) and toluene (100 mL) was heated at 80°C for 16 hours. After cooling, the bottom layer was separated by decantation, washed with toluene (50 mL), then washed with THF (100 mL) and dried in a vacuum oven to give bromide (94-Br) (20.85 g, 99% yield).

A mixture of bromide (94-Br) (6.70 g, 23.8 mmol), KFSI (5.22 g, 23.8 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 3 hours. After cooling, the solid was filtered off, and the solvent was evaporated under reduced pressure. The residue was dissolved in anhydrous THF, and set aside at room temperature for 4 hours. The solution was filtered, and the solvent was removed under reduced pressure to give ionic liquid (94) (8.59 g, 95% yield).

Example 9: Preparation of 3-Ethyl-1-isopropyl-4(5)-methylimidazolium bis(fluorosulfonyl)imide (86).

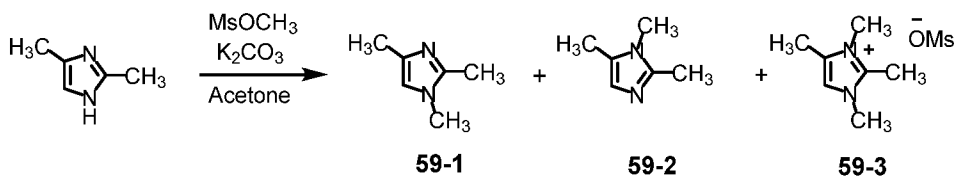


Potassium hydroxide (90%, 8.40 g, 134 mmol) was added to a solution of 4-methylimidazole in DMSO (100 mL), and the obtained mixture was stirred at room temperature for 1 hour. 2-Bromopropane (9.4 mL, 100 mmol) was then added dropwise while the mixture was cooled in an ice-water bath. After the addition, stirring at room temperature was continued overnight. The reaction mixture was poured into water (500 mL), treated with 5N NaOH (100 mL) and extracted with DCM (3 x 200 mL). The extract was washed with water (400 mL), dried over anhydrous sodium sulfate, and the solvent was removed under reduced pressure to give imidazole (81) (8.80 g, 71% yield) as a mixture of two isomers at a ratio of 2:1.

A mixture of imidazole (81) (8.50 g, 72 mmol), bromoethane (8 mL, 107 mmol) and toluene was sealed in a pressure reactor and heated at 80°C for 16 hours. After cooling, the reactor was opened and the bottom layer was separated by decantation and triturated with ethyl ether to give a crystalline product. The product was recrystallized by dissolving it in hot THF/acetone (25 + 25 mL) and pouring the solution into cold THF (150 mL). After sitting overnight at room temperature, the snow-white crystals were filtered off, washed with THF (100 mL) and dried in a vacuum oven to give imidazolium bromide (86-Br) (14.62 g, 87% yield).

A mixture of imidazolium bromide (86-Br) (5.60 g, 24 mmol), KFSI (5.26 g, 24 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 3 hours. The solid was filtered off, and the solvent was removed under reduced pressure. A solution of the residue in THF (100 mL) was set aside at room temperature overnight, then filtered and concentrated under reduced pressure to give ionic liquid (86) (8.05 g, 100% yield), a mixture of two isomers in an approximate molar ratio of 2:1. ¹H NMR of the major isomer (500 MHz, DMSO-d₆): δ 9.11 (s, 1H), 7.64 (s, 1H), 4.54 (m, 1H), 4.10 (q, J = 7.5 Hz, 2H), 2.29 (s, 3H), 1.45 (d, J = 7.0 Hz, 6H), 1.40 (t, J = 7.5 Hz, 3H).

Example 10: Methylation of 2,4-dimethylimidazole.

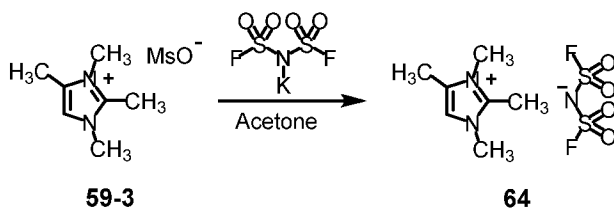


A solution of methyl mesylate (25.4 mL, 300 mmol) in acetone (50 mL) was added dropwise to a stirred mixture of 2,4-dimethylimidazole (25.00 g, 260 mmol), potassium carbonate (55.20 g, 400 mmol) and acetone (500 mL). Stirring under argon was then continued at room temperature for 16 hours. The

resulting solid was filtered off, and the solvent was removed under reduced pressure. The residue was triturated with THF (200 mL) to give crystalline product (59-3) (14.47 g, 25% yield).

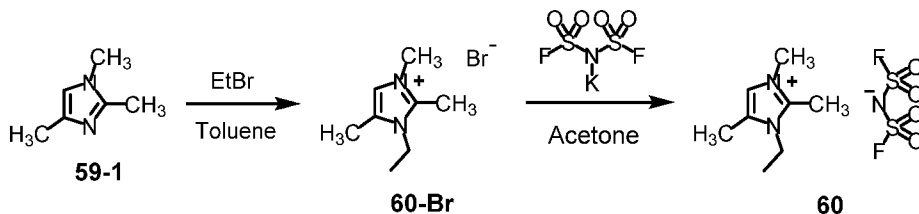
The filtrate was treated with activated charcoal, filtered through a layer of Celite, and the solvent was removed under reduced pressure. Upon storage of the residue at room temperature, some crystals
 5 were formed, and these crystals were separated, washed with THF and dried in a vacuum oven to give 1,2,4-trimethylimidazole (59-1) (4.01 g, 14% yield). Concentration of the filtrate gave a mixture of isomeric imidazoles (59-1) and (59-2) (12.19 g, 43% yield).

Example 11: Preparation of 1,2,3,4-tetramethylimidazolium bis(fluorosulfonyl)imide (64).



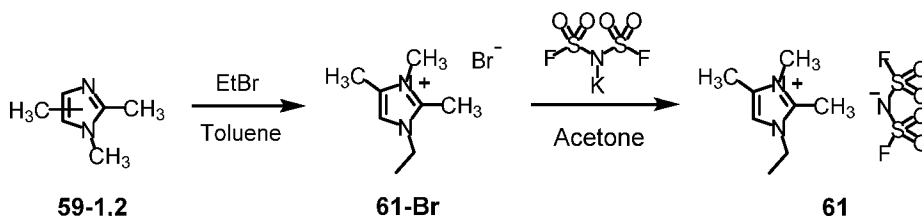
A mixture of mesylate (59-3) (Example 10) (14.47 g, 65.7 mmol), KFSI (13.59 g, 62 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 3 hours. After cooling, the mixture was diluted with THF (100 mL), the solid was filtered off, and the solvent was removed under reduced
 15 pressure to give salt (64) (17.01 g, 90% yield). ¹H NMR (400 MHz, DMSO-d₆): δ 7.33 (s, 1H), 3.71 (s, 3H), 3.62 (s, 3H), 2.55 (s, 3H), 2.24 (s, 3H).

Example 12: Preparation of 3-Ethyl-1,2,4-trimethylimidazolium bis(fluorosulfonyl)imide (60).



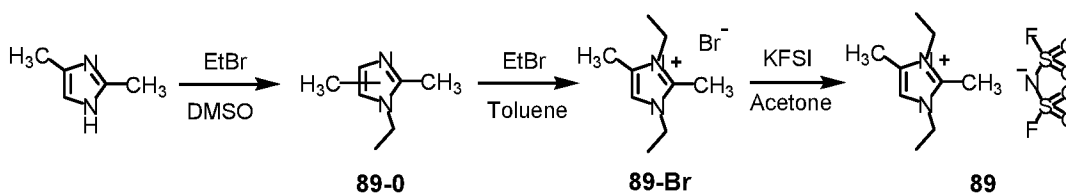
A mixture of imidazole (59-1) (Example 10) (4.00 g, 36.3 mmol), bromoethane (3.7 mL, 50
 20 mmol) and toluene (20 mL) was sealed in a pressure reactor and heated at 80°C for 3 days and nights. After cooling, the mixture was diluted with THF (20 mL). The solid was filtered off, washed with THF (20 mL) and dried in a vacuum oven to give bromide (60-Br) (7.46 g, 94% yield).

A mixture of (60-Br) (7.46 g, 34 mmol), KFSI (7.43 g, 34 mmol) and acetone (100 mL) was
 25 stirred under argon and heated at 50°C for 16 hours. After cooling to room temperature, the solid was filtered off. The filtrate was concentrated under reduced pressure to give ionic liquid (60) (11.00 g, 100% yield). ¹H NMR (500 MHz, DMSO-d₆): δ 7.33 (s, 1H), 4.10 (q, J = 7.5 Hz, 2H), 3.69 (s, 3H), 2.61 (s, 3H), 2.27 (s, 3H), 1.26 (t, J = 7.5 Hz, 3H).

Example 13: Preparation of 1-Ethyl-2,3,4-trimethylimidazolium bis(fluorosulfonyl)imide (61).

A mixture of imidazoles (59-1 and 59-2) (Example 10) (12.00 g, 109 mmol), bromoethane (11.2 mL, 150 mmol) and toluene was stirred under argon and heated at 60°C for 4 hours. After cooling, the
 5 obtained solid was filtered off, washed with THF and recrystallized from a mixture of acetonitrile and THF to give crystalline bromide (61-Br) (8.91 g, 37% yield).

A mixture of bromide (61-Br) (8.91 g, 40.6 mmol), KFSI (8.90 g, 40.6 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 3 hours. After cooling to room temperature, the solid
 10 was filtered off, and the solvent was removed under reduced pressure to give ionic liquid (61) (9.20 g, 71% yield). ¹H NMR (500 MHz, DMSO-d₆): δ 7.41 (s, 1H), 4.16 (q, J = 7.5 Hz, 2H), 3.61 (s, 3H), 2.58 (s, 3H), 2.25 (s, 3H), 1.32 (t, J = 7.0 Hz, 3H).

Example 14: Preparation of 1,3-Diethyl-2,4-dimethylimidazolium bis(fluorosulfonyl)imide (89).

15 A mixture of 2,4-dimethylimidazole (19.23 g, 200 mmol), 90% KOH (15.60 g, 250 mmol) and anhydrous DMSO (100 mL) was stirred under argon at room temperature for 3 hours. Ethyl bromide (16.4 mL, 220 mmol) was added dropwise while the mixture was cooled in an ice-water bath. After stirring at room temperature overnight, the mixture was poured into ice-water (500 mL), treated with 5N
 20 NaOH (80 mL) and extracted with DCM/THF (9:1, 2 x 300 mL). The extract was dried over anhydrous Na₂CO₃, and the solvent was removed under reduced pressure to give a mixture of two isomeric mono-ethylated products (89-0) (22.49 g, 91% yield).

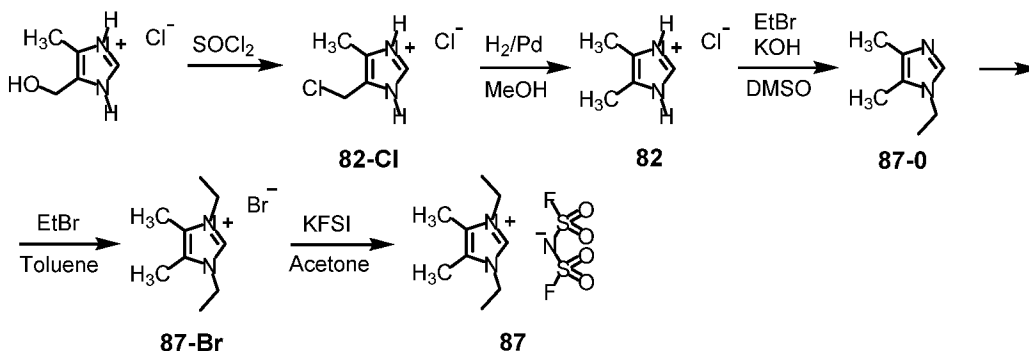
A mixture of imidazoles (89-0) (7.45 g, 60 mmol), bromoethane (7.46 mL, 100 mmol) and anhydrous toluene (50 mL) was sealed in a pressure reactor and heated at 50°C for 3 days. The obtained
 25 crystals were separated, washed with ethyl ether and dried in a vacuum oven to give imidazolium bromide (89-Br) (11.08 g, 79% yield).

A mixture of imidazolium bromide (89-Br) (5.86 g, 25.1 mmol), KFSI (5.51 g, 25.1 mmol) and acetone (75 mL) was stirred under argon and heated at 60° for 2 hours. After cooling, the solid was filtered off, and the solvent was removed under reduced pressure. A solution of the residue in anhydrous

THF (100 mL) was set aside at room temperature overnight, then it was filtered, and the solvent was removed under reduced pressure to give 1,3-diethyl-2,4-dimethylimidazolium bis(fluorosulfonyl)imide (89) (8.40 g, 100% yield). ¹H NMR (500 MHz, DMSO-d₆): δ 7.43 (s, 1H), 4.10 (m, 4H), 2.61 (s, 3H), 2.28 (s, 3H), 1.33 (t, J = 7.5 Hz, 3H), 1.27 (t, J = 7.5 Hz, 3H).

5

Example 15: Preparation of 1,3-Diethyl-4,5-dimethylimidazolium bis(fluorosulfonyl)imide (87).



In a two neck flask equipped with a reflux condenser with an attached acid trap was placed 4-hydroxymethyl-5-methylimidazolium hydrochloride (10.00 g). Thionyl chloride (40 mL) was added in one portion causing the solid to dissolve, and new precipitation started to form within a couple of minutes. The mixture was heated then at 40°C for 1 hour, diluted with ethyl ether (50 mL) and set aside at room temperature overnight. The solid was filtered off, washed with ethyl ether (2 x 50 mL) and dried in a vacuum oven to give imidazolium hydrochloride (82-Cl) (11.11 g).

15 A mixture of imidazolium hydrochloride (82-Cl) (11.00 g, 65.8 mmol), 10% Pd(C) (2 g) and methanol (200 mL) was hydrogenated at 60 psi of H₂ for 16 hours. The solid was filtered off using a Celite pad, and the solvent was removed under reduced pressure to give 4,5-dimethylimidazolium hydrochloride (82) (8.02 g, 92% yield).

20 A solution of 4,5-dimethylimidazolium hydrochloride (82) (8.00 g, 60.3 mmol) in anhydrous DMSO (100 mL) was treated with solid 90% KOH (11.20 g, 180 mmol) and bromoethane (4.9 mL, 65 mmol). The obtained mixture was stirred under argon and heated at 40°C for 16 hours. The reaction mixture was poured into ice-water (500 mL), treated with 5N NaOH and extracted with DCM/THF (9:1, 2 x 200 mL). The extract was washed with water (100 mL), dried over Na₂CO₃, and the solvent was removed under reduced pressure to give 1-ethyl-4,5-dimethylimidazole (87-0) (5.43 g, 73% yield).

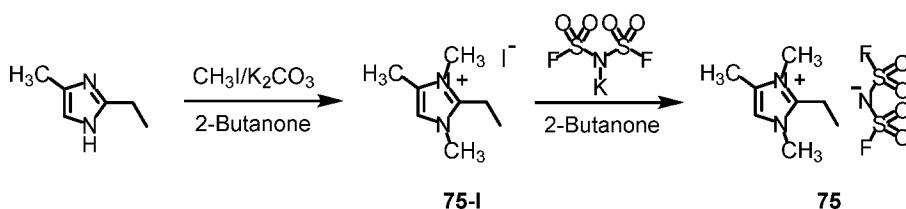
25 A mixture of 1-ethyl-4,5-dimethylimidazole (87-0) (5.40 g, 43.5 mmol), bromoethane (4.5 mL, 60 mmol) and toluene (30 mL) was placed in a pressure reactor and heated at 60°C for 20 hours. After cooling, the bottom layer was separated by decantation and recrystallized from acetone (200 mL) to give imidazolium bromide (87-Br) (6.41 g, 63% yield).

30 A mixture of imidazolium bromide (87-Br) (6.38 g, 27.4 mmol), KFSI (6.00 g, 27.4 mmol) and acetone (100 mL) was stirred under argon and heated at 60°C for 2 hours. After cooling to room

temperature, the solid was filtered off, and the solvent was removed under reduced pressure. The residue was dissolved in THF (100 mL) and set aside at room temperature overnight. A small amount of solid was found on the flask bottom. The solution was filtered, and the solvent was removed under reduced pressure to give 1,3-diethyl-4,5-dimethylimidazolium bis(fluorosulfonyl)imide (87) (9.10 g, 99% yield).

¹H NMR (500 MHz, DMSO-d₆): δ 9.01 (s, 1H), 4.09 (q, J = 7.5 Hz, 4H), 2.24 (s, 6H), 1.38 (t, J = 7.4 Hz, 6H).

Example 16: Preparation of 2-Ethyl-1,3,4-trimethylimidazolium bis(fluorosulfonyl)imide (75).

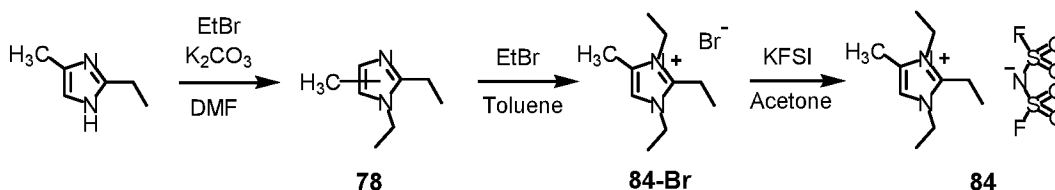


A solution of iodomethane (15.6 mL, 250 mmol) in 2-butanone (50 mL) was added dropwise to a suspension of potassium carbonate (41.40 g, 300 mmol) in a solution of 2-ethyl-4-methylimidazole (11.47 g, 100 mmol) in 2-butanone (50 mL) and stirred vigorously under argon. After the addition (3 hours), the mixture was heated under a reflux condenser at 60°C for another 3 hours. The solid was filtered off from a warm reaction mixture and washed with 2-butanone (100 mL). Combined filtrate and washings were set aside at room temperature overnight to produce crystals of iodide (75-I), which were filtered off and dried in a vacuum oven. The second crop of iodide (75-I) was obtained by concentration of the filtrate. Washing of the potassium iodide solid with boiling 2-butanone (200 mL) produced two additional crops of iodide (75-I), total 12.89 g (48% yield).

A mixture of iodide (75-I) (6.34 g, 23.8 mmol), KFSI (5.22 g, 23.8 mmol) and 2-butanone (100 mL) was stirred under argon and heated at 60°C for 4 hours. The solid was filtered off. The filtrate was diluted with ethyl acetate (400 mL), washed with 1% sodium thiosulfate (100 mL) followed by water (100 mL), dried over sodium sulfate, and the solvent was removed under reduced pressure to give ionic liquid (75). ¹H NMR (500 MHz, DMSO-d₆): δ 7.33 (s, 1H), 3.74 (s, 3H), 3.66 (s, 3H), 3.30 (q, J = 7.5 Hz, 2H), 2.24 (s, 3H), 1.16 (t, J = 7.5 Hz, 3H).

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Example 17: Preparation of 1,2,3-Triethyl-4-methylimidazolium bis(fluorosulfonyl)imide (84).



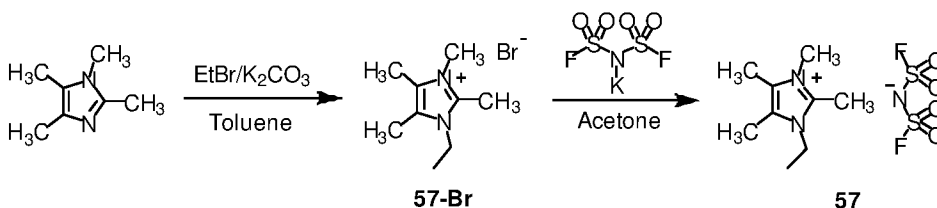
A mixture of 2-ethyl-4-methylimidazole (11.02 g, 100 mmol), potassium carbonate (13.80 g, 100 mmol), bromoethane (11.2 mL, 150 mmol) and DMF (100 mL) was stirred under argon and heated at 50°C for 40 hours. The mixture was diluted with ethyl acetate (400 mL), stirred for 15 min, and the solid

was filtered off. The solution was washed with water (2 x 100 mL), dried over anhydrous sodium sulfate, and the solvent was removed under reduced pressure to give product (78) (2.62 g, 19% yield) as a mixture of two isomers.

A solution of imidazole (78) (2.60 g, 19.8 mmol) in dry toluene (20 mL) was treated with
5 bromoethane (2 mL, 27 mmol), and the obtained mixture was heated in a pressure reactor at 80°C for 20 hours. After cooling to room temperature, the reactor was opened and the obtained crystals were filtered off, washed with toluene and dried in a vacuum oven to give imidazolium bromide (84-Br) (3.11 g, 64% yield).

A mixture of imidazolium bromide (84-Br) (3.10 g, 12.5 mmol), KFSI (2.75 g, 12.5 mmol) and
10 acetone (75 mL) was stirred under argon and heated at 60°C for 2 hours. After cooling, the solid was filtered off, and the solvent was removed under reduced pressure. The residue was dissolved in dry THF, and the obtained solution was set aside at room temperature for 2 hours. A small amount of precipitation was observed on the flask bottom. The solution was filtered, and the solvent was evaporated under
15 reduced pressure to give 1,2,3-triethyl-4-methylimidazolium bis(fluorosulfonyl)imide (84) (4.38 g, 100% yield). ¹H NMR (500 MHz, DMSO-d₆): δ 7.49 (s, 1H), 4.13 (m, 4H), 3.03 (q, J = 7.5 Hz, 2H), 2.29 (s, 3H), 1.37 (t, J = 7.5 Hz, 3H), 1.30 (t, J = 7.5 Hz, 3H), 1.19 (t, J = 7.5 Hz, 3H).

Example 18: Preparation of 1-Ethyl-2,3,4,5-tetramethylimidazolium bis(fluorosulfonyl)imide (57).



A mixture of 1,2,4,5-tetramethylimidazole (12.42 g, 100 mmol), bromoethane (11.2 mL, 150
20 mmol) and toluene (50 mL) was sealed in a pressure reactor and heated at 60°C for 2 hours, then at 80°C for 2 hours, and finally at 100°C for 16 hours. After cooling, the solid was filtered off, washed with THF and recrystallized from acetonitrile/acetone to give product (57-Br) (13.70 g, 59% yield).

A mixture of product (57-Br) (13.70 g, 59 mmol), KFSI (12.06 g, 55 mmol) and acetone (100
25 mL) was stirred under argon and heated at 60°C for 3 hours. After cooling, the solid was filtered off, and the solvent was removed under reduced pressure to give ionic liquid (57) (19.50 g, 58% yield). ¹H NMR (400 MHz, DMSO-d₆): δ 4.10 (q, J = 7.3 Hz, 2H), 3.60 (s, 3H), 2.60 (s, 3H), 2.24 (s, 3H), 2.21 (s, 3H).

Example 19: Corrosion Testing.

Using a micropipette set at 5μl, first and second aliquot samples of reference CE-1 (1-ethyl-3-
30 methyl-imidazolium, Sigma Aldrich, St. Louis, MO) and first and second aliquot samples of the respective experimental Examples identified in Table 2 were placed in spaced apart arrangement on a 50 nm aluminum film (Toray Advanced Film, Tokyo, Japan). The prepared film was placed in a

Temperature & Humidity Benchtop chamber, set at 85°C and 85% Relative Humidity (RH) (ESPEC North America, Hudsonville, MI, USA, Criterion Temperature & Humidity Benchtop Model BTL-433) and was periodically checked at selected times (initially hourly). Either a colored sheet of paper and/or a light source was placed behind the prepared film and visually examined for an indication of total perforation at the spot of each aliquot deposit. If total perforation was observed, the time was recorded and the sample was indicated as a corrosive item. The results are shown in Table 2, below.

TABLE 2

<u>Composition</u>	<u>Time to Film Perforation (hours)</u>
Reference Composition CE-1	> 120
Example 1 Composition (68)	> 125
Example 2 Composition (62)	> 200
Example 3 Composition (92)	< 3
Example 4 Composition (46)	< 1
Example 13 Composition (61)	> 200
Example 16 Composition (75)	< 3

10 Example 20: Preparation of Polymer Solution.

95 mass parts of n-butyl acrylate, 5 mass parts of acrylic acid and 125 mass parts of ethyl acetate were introduced into a stirring flask attached to a condenser that was equipped with a nitrogen gas inlet. The mixture was stirred at room temperature while introducing the nitrogen gas for about 1 hour to remove oxygen from the reaction system. 0.2 mass parts of azobisisobutyronitrile (AIBN) was added, which increased the temperature of the resulting mixture to about 63°±2°C, and mixed/stirred for about 5-6 hours for polymerization. After stopping the reaction, an acrylic polymer-containing solution resulted, having a solid content of about 30%. The apparent molecular weight of the polymer solution (P1) was determined to be about 800,000 and had a glass transition temperature of about -50°C.

20 Example 21: Preparation of Adhesive Sheet.

An adhesive sheet was prepared by mixing the polymer solution described in Example 20 with each of the compositions identified in Table 3 to obtain electrically de-bondable adhesive compositions. The prepared compositions were coated/deposited upon a surface treated PET separator (release liner) (MR38, made by Mitsubishi Plastics, Japan), forming an adhesive composite layer at a thickness of about 150 µm. The coated film was then heat dried at 130° C for about 3 minutes followed by aging at 50°C for 24 hours to obtain an adhesive layer/sheet having a thickness about 50 µm to obtain an adhesive layer/sheet. A PET release liner was aligned with the coated film and then aged at 50° C for about 24 hours to obtain an adhesive layer/sheet having a thickness of about 50 µm.

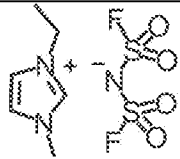
30 Example 22: Preparation of Adhesive/Glass Substrate Test Samples.

A commercially available pre-cleaned glass slide (3 x 1 inches) was used as substrate for an adhesive coating or layer. The glass slide substrate was further cleaned with acetone (Sigma Aldrich, St. Louis, MO, USA) and isopropyl alcohol (Aldrich) and then dried. A suitable amount of ethyl acetate was added to adjust the viscosity of slurry and then the slurry was casted on a pre-cleaned glass substrate with a thickness adjustable blade coater (BYK Gardner film casting knife). The cast tape was then cured at 130 °C for about 3 minutes in a preheated oven resulting in a dried coating over the substrate. A PET release liner was aligned with the coated glass and applied to the cured adhesive coated glass substrate. The release liner layered adhesive coated glass substrate was then aged/dried at about 50° C for about 24 hours and stored under ambient conditions until needed.

Just prior to the application of the nano-Al coated layer, the aforementioned release liner was removed. The aluminum described previously above (50 nm-thick aluminum coated PET film (Toray Advanced Film) was applied to the adhesive coated surface of the glass substrate.

The prepared films were placed in a Temperature & Humidity Benchtop chamber, set at 85°C and 85% Relative Humidity (ESPEC North America, Hudsonville, MI, USA, Criterion Temperature & Humidity Benchtop Model BTL-433) and were periodically checked at selected times (initially hourly). The interface between the adhesive and the aluminum foil was examined at regular predetermined time intervals, e.g., for an indication of corrosive degradation of the metal electrode, dissolution of the metal in the selectively adherent adhesive and/or pitting of the metal electrode. If corrosiveness was observed, the time was recorded and the sample was indicated as a corrosive item. The results are provided in Table 3 below.

TABLE 3

<u>Composition</u>	<u>Hours</u>
No IL	< 450
 AS-110 Chemical Formula: C ₈ H ₁₁ F ₂ N ₃ O ₄ S ₂ Molecular Weight: 291.29	< 325
Example 2 Composition (62)	< 200
Example 3 Composition (92)	< 325
Example 17 Composition (84)	> 450
Example 9 Composition (86)	> 450
Example 15 Composition (87)	> 450
Example 14 Composition (89)	< 200
Example 8 Composition (94)	< 325

Example 23: Electrical De-bonding of Adhesive Composition Testing.

Testing for electrical de-bonding or release of an adhesive composition was done in the manner as described in JP 2015-228951 and/or JP 2015-204998, and also shown in apparatus 300 of FIG. 3.

As shown in FIG. 3, adhesive material 303 (including composition (87) of Example 15) was coated upon a conductive substrate 301 which was 25 mm wide and 100 mm long. The resulting substrate 301 was laminated upon another flexible conductive layer 302 (such as aluminum foil and/or a metalized plastic film such as PET), which was 10 mm to 25 mm wide and 100 mm longer than substrate 301. The lamination was conducted by the application of rolling pressure by a 2 kg roller and roll press.

The bonding/de-bonding tester (Mark-10, Copiague, New York, USA, model ESM303 motorized tension/compression stand) was equipped with a Mark-10 force gauge (Series 7-1000) and had lower and upper clamps. The conductive substrate 301 was fixed onto the lower clamp and then electrically connected to the positive pole of a power supply 304 (Protek DC Power Supply 3006B). Flexible conductive layer 302 was fixed to the upper clamp which was connected with the negative pole of the same DC power supply. The power supply had an output range from 0 to 100 VDC. The moving/peeling speed was set at 300 mm/min. A switch 305 is present and, when closed, the electrical potential is applied between substrate 301 and layer 302.

In a dynamic test, voltage was applied a few seconds after the peeling or separation starts and the time and peeling strength readings from the force gauge were recorded by the data acquisition system (Mark-10 MESURgauge Plus). FIG. 4 shows the 180 degree peeling strength evolution with time when a 10 VDC was applied to the adhesive material that is doped with composition (87) of Example 15 at a concentration of 5 wt.%.

In a static de-bonding test, the sample was fixed on to the tester and connected to the power supply in the same way. The initial 180 degree peeling was measured at the same peeling speed. Then peeling was stopped. A DC voltage (10 VDC for example) was applied for some time (10 second for example). The peeling strength was then measured at the same peeling speed of 300 mm/min. For the same adhesive sample including composition (87) of Example 15, the initial peeling strength was 3.0 N/cm, and the residual adhesion peeling strength was ~0.5 after applying 10 VDC for 10 second.

For the processes and/or methods disclosed, the functions performed in the processes and methods may be implemented in differing order, as may be indicated by context. Furthermore, the outlined steps and operations are only provided as examples, and some of the steps and operations may be optional, combined into fewer steps and operations, or expanded into additional steps and operations.

This disclosure may sometimes illustrate different components contained within, or connected with, different other components. Such depicted architectures are merely exemplary, and many other architectures can be implemented which achieve the same or similar functionality.

The terms used in this disclosure, and in the appended claims (e.g., bodies of the appended claims) are generally intended as “open” terms (e.g., the term “including” should be interpreted as “including, but not limited to,” the term “having” should be interpreted as “having at least,” the term

“includes” should be interpreted as “includes, but is not limited to,” etc.). In addition, if a specific number of elements is introduced, this may be interpreted to mean at least the recited number, as may be indicated by context (e.g., the bare recitation of “two recitations,” without other modifiers, means at least two recitations, or two or more recitations). As used in this disclosure, any disjunctive word and/or
5 phrase presenting two or more alternative terms should be understood to contemplate the possibilities of including one of the terms, either of the terms, or both terms. For example, the phrase “A or B” will be understood to include the possibilities of “A” or “B” or “A and B.”

The terms and words used are not limited to the bibliographical meanings, but, are merely used to enable a clear and consistent understanding of the disclosure. It is to be understood that the singular
10 forms “a,” “an,” and “the” include plural referents unless the context clearly dictates otherwise. Thus, for example, reference to “a component surface” includes reference to one or more of such surfaces.

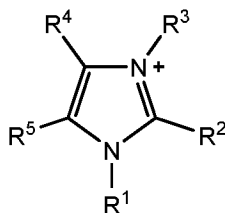
By the term “substantially” it is meant that the recited characteristic, parameter, or value need not be achieved exactly, but that deviations or variations, including for example, tolerances, measurement error, measurement accuracy limitations and other factors known to those skilled in the art, may occur in
15 amounts that do not preclude the effect the characteristic was intended to provide.

Aspects of the present disclosure may be embodied in other forms without departing from its spirit or essential characteristics. The described aspects are to be considered in all respects illustrative and not restrictive. The claimed subject matter is indicated by the appended claims rather than by the foregoing description. All changes which come within the meaning and range of equivalency of the
20 claims are to be embraced within their scope.

CLAIMS

What is claimed is:

1. A composition, comprising a cationic compound according to Formula (I):



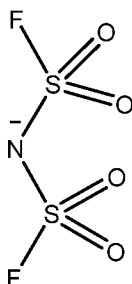
Formula (I)

5

wherein R^1 represents a C_1 - C_8 alkyl, a C_1 - C_8 alkenyl, or a C_1 - C_8 alkoxyalkyl, R^2 represents hydrogen or a C_1 - C_3 alkyl, R^3 represents a C_1 - C_8 alkyl, a C_1 - C_8 hydroxyalkyl, a C_1 - C_8 alkenyl, a C_1 - C_8 alkoxyalkyl, an acetooxy C_1 - C_3 alkyl, or a C_7 - C_{15} arylalkyl, R^4 represents hydrogen or a C_{1-4} alkyl, and R^5 represents hydrogen or a C_{1-4} alkyl.

10

2. The composition of claim 1, further comprising an anionic compound having the following structure:



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3. The composition of any preceding claim, wherein at least one of R^4 and R^5 is a C_{1-4} alkyl.

4. The composition of any preceding claim, further comprising an acrylic polymer.

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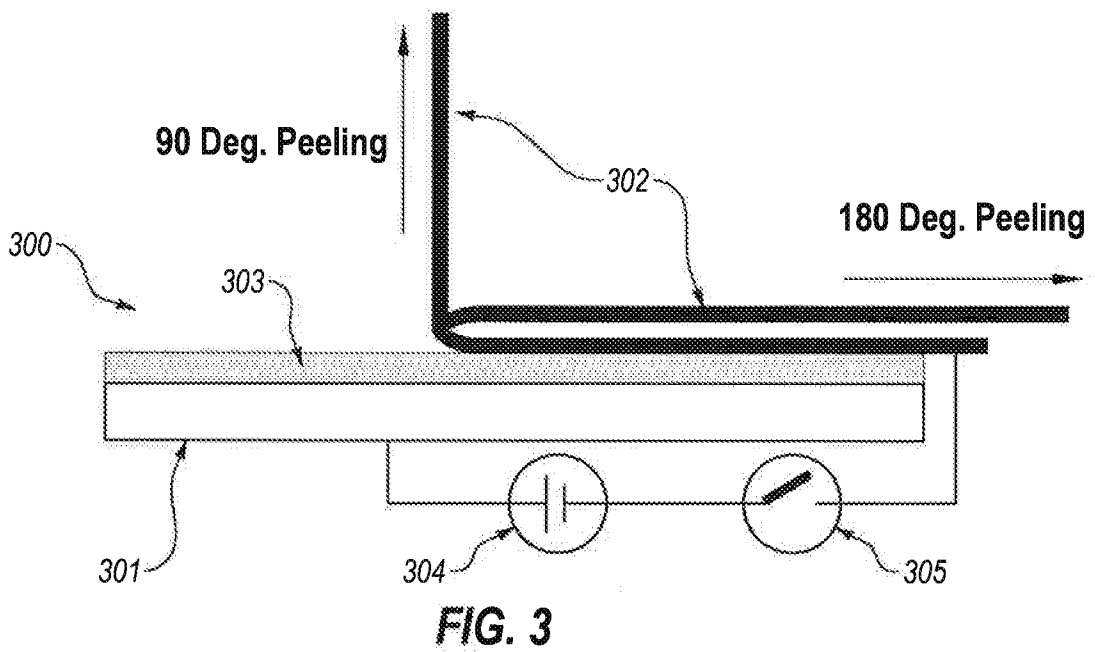
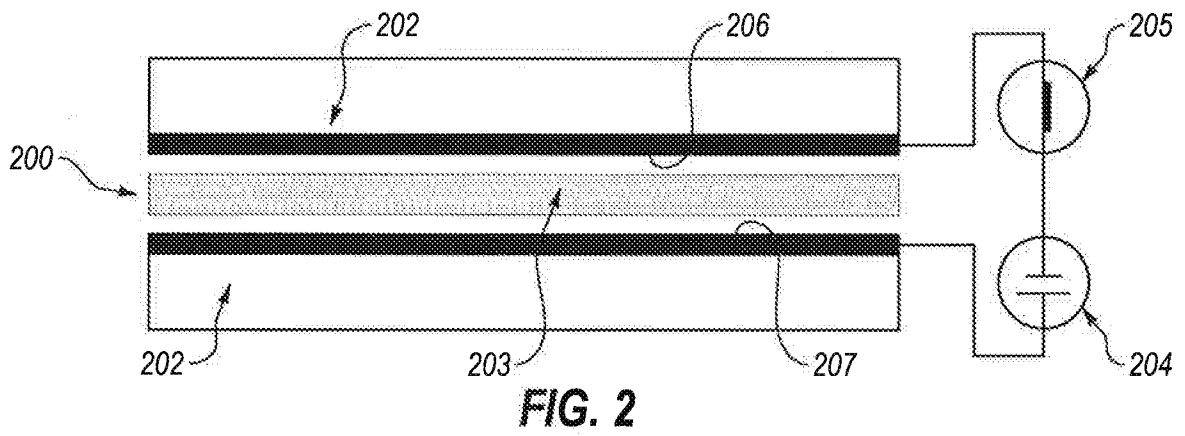
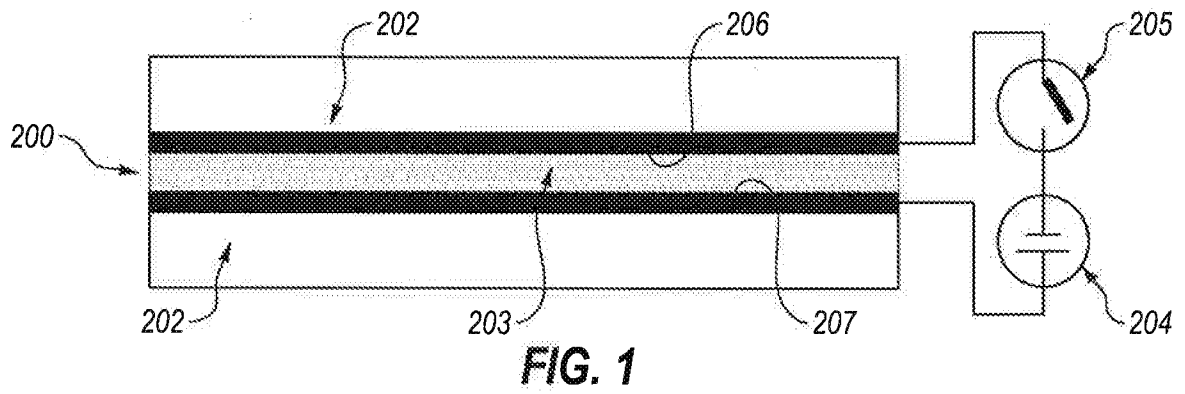
5. The composition of any preceding claim, wherein R^4 represents hydrogen and R^5 represents a C_{1-4} alkyl.

6. The composition of any one of claims 1-4, wherein R^4 represents a C_{1-4} alkyl and R^5 represents hydrogen.

25

7. The composition of claim 1 or claim 2, wherein R^1 represents $-CH_3$, $-CH_2-CH_3$, or $-CH_2-(CH_3)_2$.

8. The composition of any one of claims 1, 2 or 7, wherein R^2 represents $-H$, $-CH_3$, or $-CH_2-CH_3$.
9. The composition of any one of claims 1, 2, 7, or 8, wherein R^3 represents $-CH_3$ or $-CH_2-CH_3$.
- 5 10. The composition of any one of claims 1, 2, 7, 8, or 9, wherein R^4 represents $-CH_3$ and R^5 represents hydrogen.
11. The composition of any one of claims 1, 2, 7, 8, or 9, wherein R^4 represents hydrogen and R^5 represents $-CH_3$.
- 10 12. An apparatus, comprising a first substrate, a second substrate, and a composition according to any one of the preceding claims positioned between the first substrate and the second substrate, wherein the first substrate and the second substrate are adhered together by the composite.
- 15 13. The apparatus of claim 12, wherein the first substrate includes an electrically conductive surface, the second substrate includes an electrically conductive surface, and the composition is positioned in contact with the electrically conductive surfaces.
- 20 14. A method, comprising adhering a first substrate to a second substrate with a composition according to any one of claims 1-11.
15. The method of claim 14, further comprising applying an electrical potential between the first and second substrates and separating the first substrate from the second substrate.
- 25 16. The method of claim 14, further comprising positioning the composition on an electrically conductive surface of the first substrate.



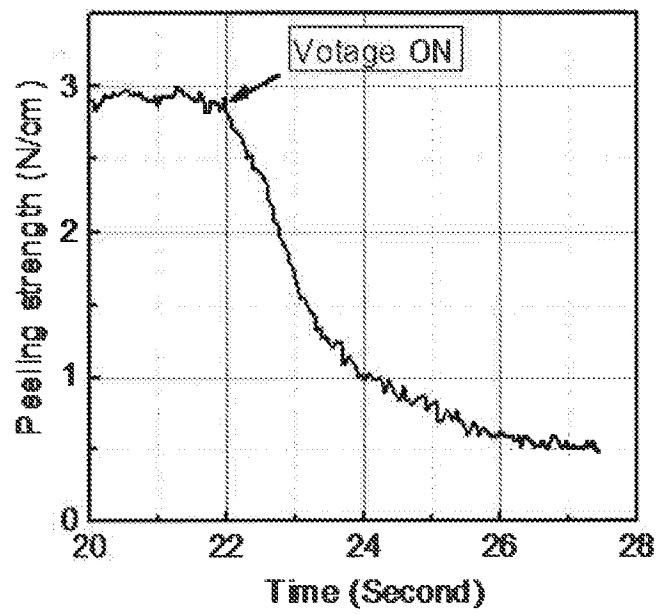


FIG. 4

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2018/020754

A. CLASSIFICATION OF SUBJECT MATTER
INV. C09J9/00 C01B21/086 C07D233/54
ADD.
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)
C09J C01B C07D

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 practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2016/135341 A1 (HENKEL AG & CO KGAA [DE]) 1 September 2016 (2016-09-01) pages 1,3-5 pages 9,12 claim 10	1,6-10, 12-16
X	----- JP 2010 037355 A (BIG TECHNOS KK; YOKOHAMA RUBBER CO LTD) 18 February 2010 (2010-02-18) paragraphs [0001], [0005] paragraphs [0036] - [0044] claims 1-7	1-16
X	----- EP 2 158 077 A1 (STORA ENSO AB [SE]) 3 March 2010 (2010-03-03) paragraphs [0001], [0009], [0038]; example 2 ----- -/--	1,7-9, 12-16

Further documents are listed in the continuation of Box C.

See patent family 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 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 24 May 2018	Date of mailing of the international search report 04/06/2018
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Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Alevizopoulou, M
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INTERNATIONAL SEARCH REPORT

International application No
PCT/US2018/020754

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>EP 2 690 149 A1 (NITTO DENKO CORP [JP]) 29 January 2014 (2014-01-29)</p> <p>paragraph [0166] -----</p>	<p>1,2,4, 7-9,12, 14</p>

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No PCT/US2018/020754

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 2016135341	A1	01-09-2016	CN 107207938 A
			EP 3262132 A1
			KR 20170122218 A
			TW 201641652 A
			US 2017355892 A1
			WO 2016135341 A1

JP 2010037355	A	18-02-2010	JP 5210078 B2
			JP 2010037355 A

EP 2158077	A1	03-03-2010	EP 2158077 A1
			WO 2008150228 A1

EP 2690149	A1	29-01-2014	CN 103571402 A
			EP 2690149 A1
			JP 6017217 B2
			JP 2014024994 A
			KR 20140014004 A
			TW 201406897 A
			US 2014030515 A1
