ChemComm

Accepted Manuscript



This is an *Accepted Manuscript*, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this Accepted Manuscript with the edited and formatted Advance Article as soon as it is available.

You can find more information about *Accepted Manuscripts* in the **Information for Authors**.

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard <u>Terms & Conditions</u> and the <u>Ethical guidelines</u> still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this *Accepted Manuscript* or any consequences arising from the use of any information it contains.



www.rsc.org/chemcomm



COMMUNICATION

Polymer gel electrolytes for application in aluminum deposition and rechargeable aluminum ion batteries

Received 00th January Xiao-Guang Sun,^{a*} Youxing Fang,^{ab} Xueguang Jiang,^{ab} Kazuki Yoshii,^c Tetsuya Tsuda,^c 20xx, Accepted 00th January Sheng Dai^{ab*} 20xx

DOI: 10.1039/x0xx00000x www.rsc.org/

Polymer gel electrolyte using AlCl₃ complexed acrylamide as a functional monomer and acidic ionic liquid based on mixture of 1ethyl-3-methylimidazolium chloride (EMImCl) and AlCl₃ (EMImCl-AlCl₃, 1-1.5, in molar) as a plasticizer has been successfully prepared for the first time *via* free radical polymerization. Aluminum deposition is successfully achieved using a polymer gel electrolyte containing 80 wt% ionic liquid. The polymer gel electrolytes are also good candidates for rechargeable aluminum ion batteries.

Aluminum surface coating has been proposed as a favorable alternative for cadmium and chromium coating because of the stringent environmental regulations on the highly toxic and carcinogenic cadmium and hexavalent chromium materials. Currently, there are various methods for aluminum surface coating such as hot dipping, ² thermal spraying, ^{2, 3} sputter deposition, vapor deposition, ² and electrodeposition. ^{2, 4-10} However, the most attractive method for aluminum surface coatings is electrodeposition, which can lead to thin, economical coatings that usually are adherent and do not affect the structural and mechanical properties of the substrate. Unfortunately, neither aluminum nor its alloys can be electrodeposited from aqueous solutions because hydrogen is evolved before aluminum is plated. Therefore, it is necessary to develop non-aqueous electrolytes for this purpose. On a commercial basis, aluminum can be plated using the SIGAL process, ¹¹ however, its scope is quite limited because of its pyrophoric and toxic nature. As an alternative, nonflammable room-temperature ionic liquids, which have high ionic conductivity, high thermal, chemical and electrochemical stability, ¹² are good candidates for electroplating of reactive elements such as Al that is impossible using aqueous or other organic solvents. ^{13, 14} Thus far,

This journal is © The Royal Society of Chemistry 20xx

most of the research on using ionic liquids for electrodeposition of aluminum and its alloys has been focused on chloroaluminate based ones, usually obtained by mixing anhydrous AlCl₃ with organic chloride salts such as 1-ethyl-3-methyl imidazolium chloride (EMImCl), 1-(1-butyl)pyridinium chloride (N-BPCl) etc. ^{10, 15} Unfortunately, because of the hygroscopic nature of AlCl₃ and the chloroaluminate, the electroplating has to be performed in an inert-gas protected atmosphere, ¹⁵ even with the newly developed ionic liquids based on complexation of AlCl₃ with neutral ligands and air and water stable ionic liquids. ^{8, 12, 16-23}



Fig. 1. Cyclic voltammograms of (a) EMImCI-AICl₃ (1-1.5, in molar ratio) and those of the mixture of equal volume of EMImCI-AICl₃ (1-1.5) with (b) Acetone, (c) Acetonitrile, (d) Tetrahydrofuran, (e) Toluene, and (f) dichloromethane on a Pt electrode (2 mm in diameter) under a scan rate of 100 mV/s at room temperature (AI wire was used as the counter and reference electrode).

One of the effective ways to reduce the moisture sensitivity of the chloroaluminate based ionic liquids is to soak them in a polymer matrix, i.e. formation of polymer gel electrolyte, which can act as a protection shield for moisture. The polymer gel electrolytes are usually obtained by impregnating liquid electrolytes into the

^{a.} Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA. E-mail: dais@ornl.gov; sunx@ornl.gov; Fax: +1 865 576 5235; Tel: +1 865 576 7303 .

^b Department of Chemistry, University of Tennessee, Knoxville, Tennessee 37996, United States

^c Department of Applied Chemistry, Graduate School of Engineering, Osaka University, Suita, Osaka 565-0871, Japan

[†] Footnotes relating to the title and/or authors should appear here.

Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/x0xx00000x

COMMUNICATION

preformed electrolytes or co-cast polymer and liquid electrolytes, $^{\rm 24,\ 25}$ or by copolymerization of monomers in the presence of plasticizers.²⁶ The ultimate polymer gel electrolytes with reversible aluminum deposition/stripping not only can alleviate the moisture sensitivity issue of the chloroaluminate based ionic liquids for aluminum deposition but also can benefit the development of shape-flexible rechargeable aluminum ion batteries. In the latter case, reversible aluminum deposition/stripping with high coulombic efficiency is the key to achieve long cycle stability. Currently, rechargeable aluminum ion batteries have been intensively studied because they have high theoretical specific capacity of 2980 mAh/g and volumetric capacity of 8040 mAh/cm³, and the advantage of aluminum being cheap and naturally abundant. 27-36 Although rechargeable aluminum ion batteries have challenges of finding suitable aluminum intercalating hosts, identifying new suitable electrolytes supporting reversible aluminum deposition/stripping is crucial for the eventual commercialization of this challenging energy storage device. So far, there are no reports in the open literature on polymer electrolytes exhibiting reversible aluminum deposition/stripping. Herein, we report, for the first time, on synthesis of new polymer gel electrolytes exhibiting reversible aluminum deposition/stripping, which can not only be used for aluminum deposition but also for rechargeable aluminum ion batteries.

To obtain polymer gel electrolytes, suitable solvents have to be used in which both ionic liquids and monomers are soluble, but no reaction (or interaction) between the solvent and the gel electrolyte components is preferred. Thus, we have selected common solvents with low boiling points such as acetone, acetonitrile, tetrahydrofuran (THF), toluene and dichloromethane (DCM) as a potential diluent for the acidic eutectic mixture of EMImCl-AlCl₃ (1-1.5). Cyclic voltammetry (CV) is used as a screening technique for the selection of solvents. Fig. 1 shows the CVs at a scan rate of 100 mV/s for EMImCl-AlCl₃ (1-1.5) and its mixtures with equal volume of the aforementioned solvents. Without any diluent solvents, the CV of the pure ionic liquid, EMImCl-AlCl₃ (1-1.5), shows well defined Al deposition and stripping peaks, similar to that reported in the literature. ³⁷ However, after being mixed with equal volume of organic solvents, the corresponding CVs show dramatic changes. As is well established in the literature that the reversible aluminum deposition/stripping in the acidic ionic liquid of EMImCl-AlCl₃ (1-1.5) is mainly due to the excess AlCl₃, resulting in the formation of electrochemically active species of Al_2Cl_7 .¹⁵ The electron-deficient AlCl₃ or Al₂Cl₇ tends to interact or react with molecules bearing lone pair electrons. Indeed, when the ionic liquid is mixed with such solvents like acetone, acetonitrile or THF, exothermal reaction happens. As a result, the interaction significantly changes the electrochemical activity of the solution, especially regarding aluminium deposition/stripping. For example, the addition of acetone results in one big positively shifted reduction peak at 0 V with onset potential at 0.4 V and a small oxidation peak at 0.5 V. Similarly, the addition of acetonitrile results in reduction and oxidation peaks at -0.4 and 0.9 V, respectively. With the addition of THF, only one big over-potential deposition peak at -1.5 V and small over-potential stripping peak at 1.2 V are observed. It is noted that the current density of the acetonitrile based solution is almost one order magnitude lower than those of acetone and THF based solutions, indicating the interaction in the former is much stronger than the latter ones. On the other hand, when toluene and DCM are added, reversible aluminum deposition/stripping are well maintained. Also, compared to pure ionic liquid, the addition of toluene and DCM enhances current density by 13 % and 10 %,

respectively. Considering the lower viscosity of DCM (0.44 cP) than that of toluene (0.59 cP), the lower current density in the former might be attributed to the weak interaction of the lone pair electrons of the chlorine atom in DCM with AlCl₃. However, the interaction between the ionic liquid and DCM is much weaker than those between the ionic liquid and acetone, acetonitrile and THF.



Fig. 2. ²⁷Al NMR spectra of ionic liquid EMImCl-AlCl₃ (1-1.5, in molar ratio) and its mixtures with equal volume of different organic solvents

To confirm the interactions, ²⁷AI NMR spectra were obtained for the ionic liquid and its mixtures with different organic solvents. As shown in Fig. 2. one broad peak at 103 ppm and an apparent shoulder at 97 ppm for the pure ionic liquid can be assigned to AlCl₄ and Al₂Cl₇, respectively.^{16, 38, 39} No peak change is observed when toluene is added, apparently attributed to the lack of interactions. With the addition of DCM, the spectrum becomes narrower and sharper, and at the same time the signal at 97 ppm decreases due to the weak interaction as mentioned above. However, with the addition of acetone, acetonitrile or THF, the signal at 97 ppm totally disappears, because Al_2Cl_7 (or excess $AlCl_3$) is completely consumed for the complex formation with the organic solvents, resulting in new peaks being observed. The above ²⁷Al NMR spectra are consistent with the CV data in Fig. 1, as well as previous reports of the complexes between AlCl₃ and organic solvents. 39-42

The tendency of Al_2Cl_7 to complex with organic solvents bearing lone pair electrons and the consequent loss of electrochemical activity towards aluminum deposition/stripping deems careful selection of suitable solvents and potential polymer hosts for preparation of polymer gel electrolytes. The routine polymers used to prepare polymer gel electrolytes for lithium ion batteries such as polyethylene oxide (PEO), polyacrylonitrile (PAN), polymethyl methacrylate (PMMA), and polyvinylidene fluoride (PVdF) ^{24, 25} all have functional groups that are either similar to or stronger than the aforementioned organic solvents. For example, PEO and PAN have the same functional groups as THF and acetonitrile, respectively, while PMMA and PVdF have stronger functional groups than acetone and DCM, respectively. Therefore, they are not good candidates for preparation of polymer gel electrolytes for aluminum deposition and rechargeable aluminum ion batteries. Fortunately, there was a recent report on the formation of a low melting eutectic mixture from equal molar acetamide and AlCl₃, which could be used for aluminum deposition. ¹⁶ Inspired by the report we here exploit the possibility of using a double bond containing analog of acetamide, acrylamide, as the active monomer to prepare polymer gel electrolyte. The product resulted from the complexation of acrylamide and AICl_{3.} with the latter either from the acidic EMImCl-AlCl₃ ionic liquid or from fresh addition, will be polymerized in the presence of the ionic liquid to obtain the polymer gel electrolyte.²⁰



Fig. 3. FTIR spectra of acrylamide, acrylamide-AlCl₃(1-1, in molar ratio), poly(acrylamide-AlCl₃, 1-1 in molar ratio), EMImCl-AlCl₃ (1-1.5, in molar ratio) and polymer gel electrolyte based on poly(acrylamide-AlCl₃, 1-1 in molar ratio) containing 80wt% EMImCl-AlCl₃ (1-1.5, in molar ratio).

The complexation between acrylamide and AlCl₃ can be confirmed by analysis of the FTIR spectra. As shown in Fig. 3, the N-H stretching frequencies are shifted from 3350 and 3160 in pure acrylamide to 3420 and 3340 cm⁻¹ in the complexed mixture.⁴³ The C-N stretching frequency and the NH₂ scissoring frequency are shifted from 1420 and 1350 to 1480, and 1450 cm⁻¹, respectively.⁴³ Similarly, the C=O, C=C and C-C stretching frequencies are shifted from 1670, 1650 and 1605 to 1660, 1622 and 1560 cm⁻¹, respectively. ⁴³ These frequency shifts are clearly due to the complexation of the electron-deficient AICl₃ with the lone pair electrons of the oxygen in the amide group that are delocalized through the C=C double bond and the NH₂ group. As expected, the double bond signal disappears for the spectrum of the polymerized complex of acrylamide and AlCl₃, poly(acrylamide-AlCl₃). For the spectrum of the polymer gel electrolyte containing 80 wt% ionic liquid, it clearly combines the features of both poly(acrtylamide-AlCl₃) and the acidic ionic liquid. A typical picture of self-standing membrane containing 60 wt% of ionic liquid is shown in Fig. S1.



Fig. 4. Temperature dependence of ionic conductivities of the gel polymer electrolytes based on polyacrylamide containing different amount of EMImCI-AICI₃ (1-1.5, in molar ratio)

Fig. 4 shows the ionic conductivities of the polymer gel electrolytes containing different amount of ionic liquid. Since pure poly(acrylamide-AlCl₃) was a fragile solid, its conductivity was not measured. In addition, the gel electrolytes containing less than 50 wt% of ionic liquid were not prepared because of the expected low ionic conductivities. As shown in Fig. 4, at 20 °C the ionic conductivities of the polymer electrolytes containing 50, 60, 70, and 80 wt% of ionic liquid are 5.29×10^{-5} , 2.00×10^{-4} , 8.87×10^{-4} , 1.66×10^{-3} S cm⁻¹, respectively. Usually, an ambient ionic conductivity above 10^{-3} S cm⁻¹ is good enough for practical applications.²⁴ in addition, if needed, higher ionic conductivities can be easily

achieved at high temperatures. For example, the ionic conductivity of the polymer gel membrane with 60 wt% ionic liquid is 1.02×10^{-3} S cm⁻¹ at 50 °C, more than five folds of that at 20 °C.



Fig. 5. Cyclic voltamograms of the electrolyte containing 60 wt% of EMImCl-AlCl₃ (1-1.5, in molar ratio) at 50 $^{\circ}$ C under a scan rate of 100 mV/s. The experiment was carried out inside the glove box, with Cu and Al plates being used as working and counter electrode, respectively.

The electrochemical property of the polymer gel electrolyte was evaluated using a two-electrode system with Cu plate being used as the working electrode and Al plate being used as both counter and reference electrode. Fig. 5 shows the CVs of the polymer gel electrolyte with 60 wt% of ionic liquid on a Cu working electrode under a scan rate of 100 mV/s at 50 °C. The electrolyte exhibits a well-defined AI deposition/stripping peaks during the initial CV scan. A low coulombic efficiency around 80 % is observed for these CV scans, probably due to some side reactions. It is noticed that the current densities increase with increasing scan cycles, indicating an activation process, probably due to the gradual removal of the residual surface oxide on the Al plate. In addition, it is noticed that both the cathodic peak and anodic peak shift with cycling, i.e. shifting from initial -300 mV and 130 mV to -360 mV and 170 mV after 15 cycles, respectively. This might be related to the fact that the same aluminum plate was used as both counter and reference electrode, on which the residual surface oxide layer being removed on successive cycles as evidenced by the current increase with cycling. To demonstrate the improvement of moisture sensitivity in polymer gel electrolyte, both liquid electrolyte and a polymer gel electrolyte containing 80 wt% ionic liquid were exposed to the air, after which they were taken inside the glovebox and CVs were scanned. As shown in Fig. S2, the electrochemical activity is totally lost for the liquid electrolyte after being exposed to the air for only 3 min (Fig. S2a). As a comparison, reversible aluminium deposition/stripping are still observed for the polymer gel electrolyte after being exposed to the air for 10 min (Fig. S2b). For practical applications, the polymer gel membrane can be directly casted onto suitable aluminium foil and if the plating surface has been treated appropriately, 10 min is long enough to allow the polymer gel electrolyte being taken out of a protection media and directly put onto the substrate for immediate electrodeposition.

Finally, to check the possibility of electroplating Al using this new polymer gel electrolyte, a constant voltage of -300 mV was applied at 50 °C for 4 hours, with copper plate as the working electrode and Al as both counter and reference electrode. After electrodeposition, the membrane was peeled off, and the deposit was washed with acetone. As shown in Fig. 6a that after electrodeposition the Cu substrate is covered by an off-white deposit (yellowish). Clearly, some parts of the deposit have been removed during post-deposition treatment. The SEM image in Fig. 6b reveals fine grain-like Al crystals. The relatively large crystal size

COMMUNICATION

is related to the initial low current density, as compared to those of liquid electrolytes, ^{18, 19} ¹⁹ resulting in fewer crystal seeds. To confirm the purity of the deposition, the film was further analysed with x-ray diffraction and energy–dispersive x-ray spectroscopy. As shown in Fig. 6c and d, there are only signals of the copper substrate and the electrodeposited aluminum.



Fig.6. Digital image (A), scanning electron micrograph (B), X-ray diffraction pattern (C), and energy-dispersive X-ray spectroscopy analysis (D) of the electrodeposited aluminum on copper substrate.

In summary, polymer gel electrolytes containing acidic ionic liquid, EMImCl-AlCl₃ (1-1.5, in molar ratio), have been successfully prepared for the first time. It has been shown that the selection of organic solvents is crucial in preparing polymer gel electrolytes, owing to the strong complexation of electron-deficient AlCl₃ with the lone pair electrons of the functional groups of organic solvents. The ionic conductivities of the polymer gel electrolytes increase linearly with the amount of the ionic liquid, with a high ionic conductivity of $1.66 \times 10^{-3} \text{ S cm}^{-1}$ at 20 °C being achieved for the one containing 80 wt% of ionic liquid. The cyclic voltammetry of the new polymer gel electrolytes exhibits reversible aluminum deposition/stripping at 50 °C. A successful AI deposition has been achieved at -300 mV for 4 hrs at 50°C. The significance of polymer gel electrolytes exhibiting reversible aluminum deposition/stripping is not only to achieve aluminum deposition with less moisture sensitivity but also to realize rechargeable aluminum ion battery with more flexibility in cell configuration.

This work was funded by the Strategic Environmental research and Development Program (SERDP) of the department of defence DOD (WP2316).

Notes and references

1. R. Mason, M. Neidbalson, M. Klingenberg, P. Khabra and C. Handsy, *Metalfinishing*, 2010, 12-20.

2. X. Xiang, X. Wang, G. Zhang, T. Tang and X. Lai, *Intern. J. Hydrogen Energy*, 2015, 40, 3697-3707.

3. S. Rech, A. Trentin, S. Vezzu, E. Vedelago, J.-G. Legoux and E. Irissou, J. Therm. Spray Tech., 2014, 23, 1237-1250.

4. B. D. Falola and I. I. Suni, Curr. Opin. in Solid State & Mater. Sci., 2015, 19, 77-84.

5. A. P. Abbott and K. J. McKenzie, *Phys. Chem. Chem. Phys.*, 2006, 8, 4265-4279.

6. A. P. Abbott, G. Frisch and K. S. Ryder, *Ann. Rev. Mater. Res.*, 2013, 43, 335-358.

7. L. Barchi, U. Bardi, S. Caporali, M. Fantini, A. Scrivani and A. Scrivani, *Progress in Organic Coatings*, 2010, 67, 146-151.

8. F. Endres, Zeitschrift Fur Physikalische Chemie-Intern. J. Research in Phys. Chem. & Chem. Phys. , 2004, 218, 255-283.

9. W. Simka, D. Puszczyk and G. Nawrat, *Electrochimica Acta*, 2009, 54, 5307-5319.

10. Y. G. Zhao and T. J. VanderNoot, *Electrochimica Acta*, 1997, 42, 3-13. 11. *US Pat.*, 4417954, 1983.

12. M. Armand, F. Endres, D. R. MacFarlane, H. Ohno and B. Scrosati, *Nat. Mater.*, 2009, 8, 621-629.

13. A. Corma and H. Garcı'a, Chem. Rev. , 2003, 103, 4307-4365.

P. Hapiot and C. Lagrost, *Chem. Rev.*, 2008, 108, 2238-2264.
Q. Liao, W. R. Pitner, G. Stewart, C. L. Hussey and G. R. Stafford, *J.*

Electrochem. Soc., 1997, 144, 936-943. 16. H. M. Abood, A. P. Abbott, A. D. Ballantyne and K. S. Ryder, Chem.

Commun. (Camb), 2011, 47, 3523-3525.

17. F. Coleman, G. Srinivasan and M. Swadzba-Kwasny, Angewandte Chemie-International Edition, 2013, 52, 12582-12586.

18. Y. X. Fang, K. Yoshii, X. G. Jiang, X. G. Sun, T. Tsuda, N. Mehio and S. Dai, *Electrochimica Acta*, 2015, 160, 82-88.

19. Y. X. Fang, X. G. Jiang, X. G. Sun and S. Dai, *Chem. Comm.*, 2015, 51, 13286 – 13289.

20. J. M. Hogg, F. Coleman, A. Ferrer-Ugalde, M. P. Atkins and M. Swadzba-Kwasny, *Green Chemistry*, 2015, 17, 1831-1841.

21. A. P. Abbott, R. C. Harris, Y.-T. Hsieh, K. S. Rydera and I. W. Sun, *Physical Chemistry Chemical Physics*, 2014, 16, 14675-14681.

22. F. Endres, Chemphyschem, 2002, 3, 144-154.

23. S. Z. El Abedin and F. Endres, Chemphyschem, 2006, 7, 58-61.

24. J. Y. Song, Y. Y. Wang and C. C. Wan, J. of Power Sources, 1999, 77, 183-197.

25. A. M. Stephan, Eur. Polym. J., 2006, 42, 21-42.

26. C. Liao, X. G. Sun and S. Dai, Electrochimica Acta, 2013, 87, 889-894.

27. P. R. Gifford and J. B. Palmisano, J. Electrochem. Soc., 1988,, 135, 650-654.

28. F. M. Donahue, S. E. Mancini and L. Simonsen, J. Appl. Electrochem., 1992, 22, 230-234.

29. N. S. Hudak, J. Phys. Chem. C, 2014, 118, 5203-5215.

30. M. P. Paranthaman, G. M. Brown, X. G. Sun, J. Nanda, A. Manthiram and A. Manivannan, presented in part at the 218th ECS Meeting, Las Vegas, NV, October 2010.

31. M. P. Paranthaman, H. Liu, X. G. Sun, S. Dai and G. M. Brown, in *Advances in batteries for medium and large-scale energy storage*, ed. A. Davies, Woodhead Publishing Ltd., 2015, ch. 14.

32. N. Jayaprakash, S. K. Das and L. A. Archer, *Chem. Commun.*, 2011, 47, 12610-12612.

33. W. Wang, B. Jiang, H. Sun, Z. Lin, J. Tu, J. Hou, H. Zhu and S. Jiao, *Nature Scientific Reports*, 2013, 3, 1-6.

34. J. V. Rani, V. Kanakaiah, T. Dadmal, M. S. Rao and S. Bhavanarushi, J. Electrochem. Soc., 2013, 160, A1781–A1784.

L. D. Reed and E. Menke, *J. Electrochem. Soc.*, 2013, 160, A915-A917.
M. C. Lin, M. Gong, B. Lu, Y. Wu, D. Y. Wang, M. Guan, M. Angell, C. Chen, J. Yang, B. J. Hwang and H. Dai, *Nature*, 2015, 520, 324-328.

37. J. S. Wilkes, J. A. Levisky, R. A. Wilson and C. L. Hussey, *Inorg. Chem.*, 1982, 21, 1263-1264.

38. J. S. Wilkes, J. S. Frye and G. F. Reynolds, *Inorg. Chem.*, 1983, 22, 3870-3872.

39. M. Dalibart, J. Derouault and P. Granger, *Inorg. Chem.*, 1982, 21, 2241-2246.

40. M. Dalibart, J. Derouault, P. Granger and S. Chapelle, *Inorg. Chem.*, 1982, 21, 1040-1046.

41. J. Derouault and M. T. Forel, Inorg. Chem. , 1977, 16, 3207-3213.

42. J. Derouault, P. Granger and M. T. Forel, *Inorg. Chem.*, 1977, 16, 3214-3218.

43. N. Sundaraganesan, N. Puviarasan and S. Mohan, *Talanta*, 2001, 54, 233-241.

4 | J. Name., 2012, 00, 1-3

This journal is C The Royal Society of Chemistry 20xx

Graphic abstract

Polymer gel electrolyte using AlCl₃ complexed acrylamide as a functional monomer and acidic ionic liquid based on mixture of 1-ethyl-3-methylimidazolium chloride (EMImCl) and AlCl₃ (EMImCl-AlCl₃, 1-1.5, in molar) as a plasticizer has been successfully prepared for the first time *via* free radical polymerization. The polymer gel electrolyte can be used for aluminium deposition and rechargeable aluminium ion batteries.

