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# Comparison of Chloroaluminate Melts for Aluminum Graphite Dual-Ion Battery Application

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Herein, we report a comparison of aluminum graphite dual-ion cells (AGDICs) electrochemical characteristics employing the conventional 1-ethyl-3-methylimidazolium chloride:aluminum trichloride (EMIMCl:$AlCl_3$) electrolyte and two popular deep eutectic solvents (DESs), namely urea:$AlCl_3$ and acetamide:$AlCl_3$. The three electrolytes' characteristics have been evaluated in terms of Al-stripping deposition capability and cycling behavior in AGDICs. The results evidence the EMIMCl:$AlCl_3$'s Al-stripping deposition and rate capability in AGDICs superior characteristics addressed to the lower viscosity and higher conductivity with respect to the urea:$AlCl_3$ and acetamide:$AlCl_3$. On the other hand, the urea:$AlCl_3$ guarantees a much higher columbic efficiency in AGDICs, thanks to the superior electrochemical window stability.

## 1. Introduction

Batteries and, in general, electrochemical storage systems are considered the key enabling technology to green energy economy transition. Lithium-ion battery (LIB) is the most mature technology, being the selected power source for electromobility, thanks to the elevated energy density and good cycle life. However, long-term, large-scale application of LIBs appears to be problematic due to the natural scarcity and limited production capacity of key materials containing Co and Ni.[1] Resources availability concerns pushed the research to explore alternative technologies based on abundant and widely available elements. Aluminum is one of the possible choices, having the advantages of being lightweight, low cost, and abundant (the most abundant metal in the Earth's crust).[2,3] Several secondary electrochemical cells using aluminum anode and chloroaluminate melt electrolytes have been proposed,[4–8] among them the most promising in terms of cycle life and rate capability are the aluminum graphite dual-ion batteries (AGDIBs).[3,4,9–12] AGDIBs are expected to achieve 65 Wh $kg^{-1}$, 92 Wh $l^{-1}$ in terms of energy density (values evaluated on active materials only), guarantee a cycle life of more than 10000 cycles with an overall cost of 0.03 €/kWh/cycle.[3,11] To tackle the cost factor of AGDIBs, several groups proposed the use of a new class of ionic liquids called deep eutectic solvents (DESs), or ionic liquid analogs (ILAs), formed through a mixture of a strongly Lewis acid metal halide ($AlCl_3$) and a Lewis basic ligand (urea, acetamide).[13–16] The substitution of the imidazolium cation of the conventional $AlCl_3$-ImidazoliumCl chloroaluminate with a cheaper and widely available chemical such as urea can substantially impact the system's overall cost. The retail price comparison of the three compounds indicates that EMIMCl is about ten times more expensive. Clearly, the retail price does not necessarily correspond to the product cost in high volume; however, it is an indication. The application of this class of electrolytes in AGDIBs is widely reported.[17–24] However, a clear comparison of DES electrolytes against the state of the art $AlCl_3$-ImidazoliumCl chloroaluminate electrolyte is not available for AGDIB systems. For this reason, here we report a comparison of the electrochemical characteristics of AGDICs employing two of the most widely used DES electrolyte, the urea:$AlCl_3$ and the acetamide:$AlCl_3$ against the conventional 1-ethyl-3-methylimidazolium chloride: aluminum trichloride (EMIMCl:$AlCl_3$) system.

## 2. Results and Discussion

Figure 1 reports the molecular structure of the three solvents used in this study. Figure 2 reports the CV results comparison of the Al stripping deposition process on Ni substrate employing (a) EMIMCl:$AlCl_3$-1:1.5, (b) Acetamide:$AlCl_3$-1:1.5, and (c) Urea:$AlCl_3$-1:1.5 electrolytes. The highest current peak value, of almost 3 mA $cm^{-2}$, is recorded employing the EMIMCl:$AlCl_3$-1:1.5 (Figure 2a), while using Acetamide:$AlCl_3$-1:1.5 (Figure 2b), the current peak value is about 1.3 mA $cm^{-2}$. The lower Al stripping deposition current peak of 0.47 mA $cm^{-2}$ is recorded using the Urea:$AlCl_3$-1:1.5 electrolyte (Figure 2c). The difference in the peak current intensity between the three electrolyte systems can be ascribed to the electron transfer kinetics, and

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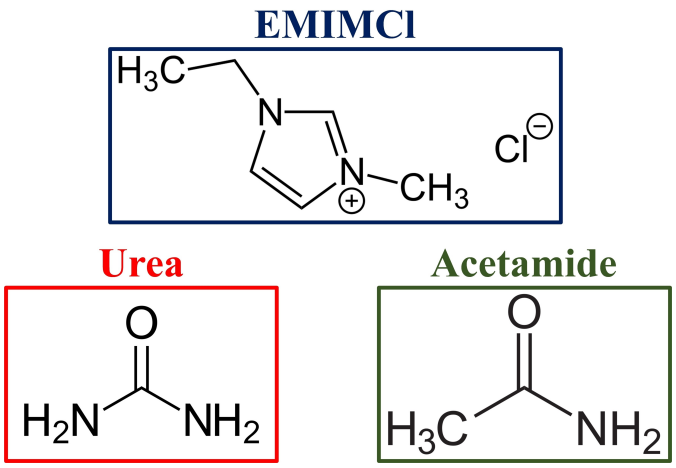



**Figure 1.** EMIMCl, Urea and Acetamide solvent molecular structure

active species concentrations, and diffusivity.[25] Indeed the higher current peak of the EMIMCl:$AlCl_3$ relates well with its higher conductivity and lower viscosity (high active species diffusivity). However, the different ionic speciation influences on the stripping deposition kinetics cannot be excluded.[13–16] The EMIMCl:$AlCl_3$ ions speciation includes $AlCl_4^-$ and $Al_2Cl_7^-$, while in the urea and acetamide compositions, we have the formation of $[AlCl_2 \cdot (ligand)_n]^+$ cations, suggested to be involved in the aluminum deposition process.[13–16] The Al deposition faradic efficiency, calculated for the second cycle, is 99.76% for EMIMCl:$AlCl_3$, 91.62% for Urea:$AlCl_3$ and 86.01% for Acetamide:$AlCl_3$. The comparison suggests the EMIMCl:$AlCl_3$-1:1.5 electrolyte as the most efficient media for the Al stripping deposition process.

Figures 3(a–b) show the voltage vs. time curves of galvanostatic cycling tests performed on symmetrical Al/Al cells using the three investigated electrolytes, carried to evaluate the long-term stability of the Al/electrolyte interphase in dynamic condition. The Al/EMIMCl:$AlCl_3$/Al cell (blue curve) has the lowest polarization, thus agreeing with the results reported in Figure 2. All the systems are characterized by stable overall overvoltage upon cycling, suggesting a stable Al/electrolyte interphase and an excellent capability to sustain long term cycling. In particular, the Urea:$AlCl_3$ system reveals a remarkable reduction of the cell overpotential during the first 5 days of measurement. This result is not in line with the CV reported in Figure 2c, where the current decreases upon cycling. The difference can be related to the different cell configuration and metal substrate used for the test. Figure 3c shows the comparison of the electrochemical stability of the three investigated electrolytes upon oxidation, evidencing the superior stability of the Urea:$AlCl_3$ system. The EMIMCl:$AlCl_3$ electrolyte also shows good stability, slightly inferior with respect to the Urea:$AlCl_3$, while the Acetamide:$AlCl_3$ shows the lowest stability, with a relevant current flow ($\geq 1\ \mu A\,cm^{-2}$) starting already at 2.2 V vs. $Al/Al^{3+}$.

The overall electrochemical characterization indicates the EMIMCl:$AlCl_3$ as the most efficient media for the Al stripping deposition process; however, the Urea:$AlCl_3$ electrolyte has better electrochemical anodic stability. The better characteristics of the EMIMCl:$AlCl_3$ electrolyte can be addressed to its higher ionic conductivity and lower viscosity in comparison with the urea and acetamide based electrolytes (Table 1).

AGDICs have been assembled to better overview the characteristics of the electrolytes. Figure 4 reports the galvanostatic cycling evaluation of the three investigated electrolytes in AGDIC, employing natural graphite (NG) electrodes with relevant loading, i.e., $\approx 5\ mg\,cm^{-2}$. Figure 4a reports the voltage signature of the first (dis-)charge cycle, revealing that the three systems are operating at a different average discharge voltage; 1.91 V for the EMIMCl:$AlCl_3$, 1.73 V for the Acetamide:$AlCl_3$ and 1.77 V for the Urea:$AlCl_3$ (see Table 2). The higher discharge voltage of the cell using the EMIMCl:$AlCl_3$ can be attributed to the electrolyte's higher conductivity and a lower viscosity.[5,17,19,22] Moreover, better Al stripping deposition properties of the EMIMCl:$AlCl_3$ can lead to improved performance. However, the different ionic speciation of DESs and the presence of $[AlCl_2(ligand)_2]^+$ species may influence the operating voltage.[17,22,26] The electrolyte composition has a relevant influence also in the AGDIC delivered capacity. The cell employing EMIMCl:$AlCl_3$ electrolyte shows the highest capacity ($\approx 110\ mAh\,g^{-1}$) with respect to the cells using Acetamide:$AlCl_3$ ($\approx 103\ mAh\,g^{-1}$) and Urea:$AlCl_3$ ($\approx 91\ mAh\,g^{-1}$) (Table 2). In this case, the different ionic speciation of the three electrolyte systems can also influence the formation of the graphite intercalation compounds (GIC), lowering the delivered capacity.[17,22,26,27] The rate capability test reported in Figure 4b confirms the superior characteristics of the EMIMCl:$AlCl_3$ in respect to the other compositions. The cell employing EMIMCl:$AlCl_3$ electrolyte is only slightly affected by the current increase, while elevated current rate ($\geq 200\ mA\,g^{-1}$) substantially reduces the other cells' delivered capacity. The urea and acetamide based electrolytes maintain good performance at $200\ mA\,g^{-1}$, while at $1000\ mA\,g^{-1}$, the capacity drops to less than 45% of the reference one for the urea, and almost to zero for the acetamide. The cell's better rate capability using the EMIMCl:$AlCl_3$ can be attributed to the electrolyte's higher conductivity and lower viscosity (Table 1).[5,17,19,22] Nonetheless, the urea composition shows the best columbic efficiency characteristic, as evidenced in Figure 4c, with values close to 100%.

**Table 1.** Viscosity, conductivity, and density of the EMIMCl:$AlCl_3$ 1:1.5, Urea:$AlCl_3$ 1:1.5, Acetamide:$AlCl_3$ 1:1.5 electrolytes. Data from Refs. [5,22,31–33].

| Electrolyte composition | Viscosity [MPa s] | | Conductivity [mS cm$^{-1}$] | | Density [g mL$^{-1}$] | | Ref. |
|---|---|---|---|---|---|---|---|
| Temperature | 20 °C | 60 °C | 20 °C | 60 °C | 20 °C | 60 °C | |
| EMIMCl $AlCl_3$ 1:1.5 | 19.42 | 8.23 | 9.19 | 20.08 | 1.3408 | 1.3052 | [31] |
| Urea $AlCl_3$ 1:1.5 | 218 | 27 | 1.6 | 4.1 | 1.564 | 1.5921 | [22,32] |
| Acetamide $AlCl_3$ 1:1.5 | 116 | 23 | 0.6 | 2.9 | 1.484 | 1.525 | [5,33] |


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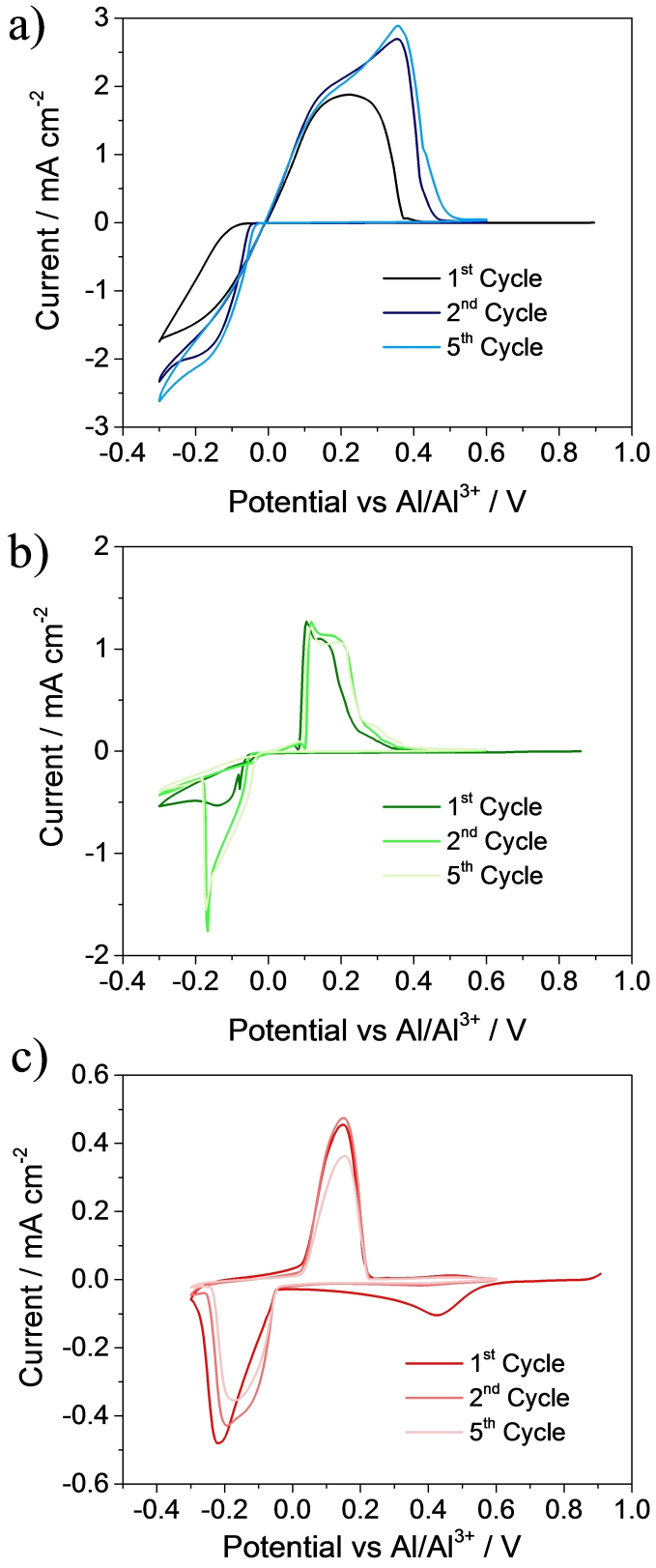



**Figure 2.** Cyclic voltammetry signatures obtained in a three-electrode configuration using Ni as working electrode and Al as counter and reference and employing a) EMIMCl:$AlCl_3$-1:1.5, b) Acetamide:$AlCl_3$-1:1.5, c) Urea:$AlCl_3$-1:1.5 electrolytes. Measurement obtained with a scan rate of 0.1 mV $s^{-1}$ at room T.

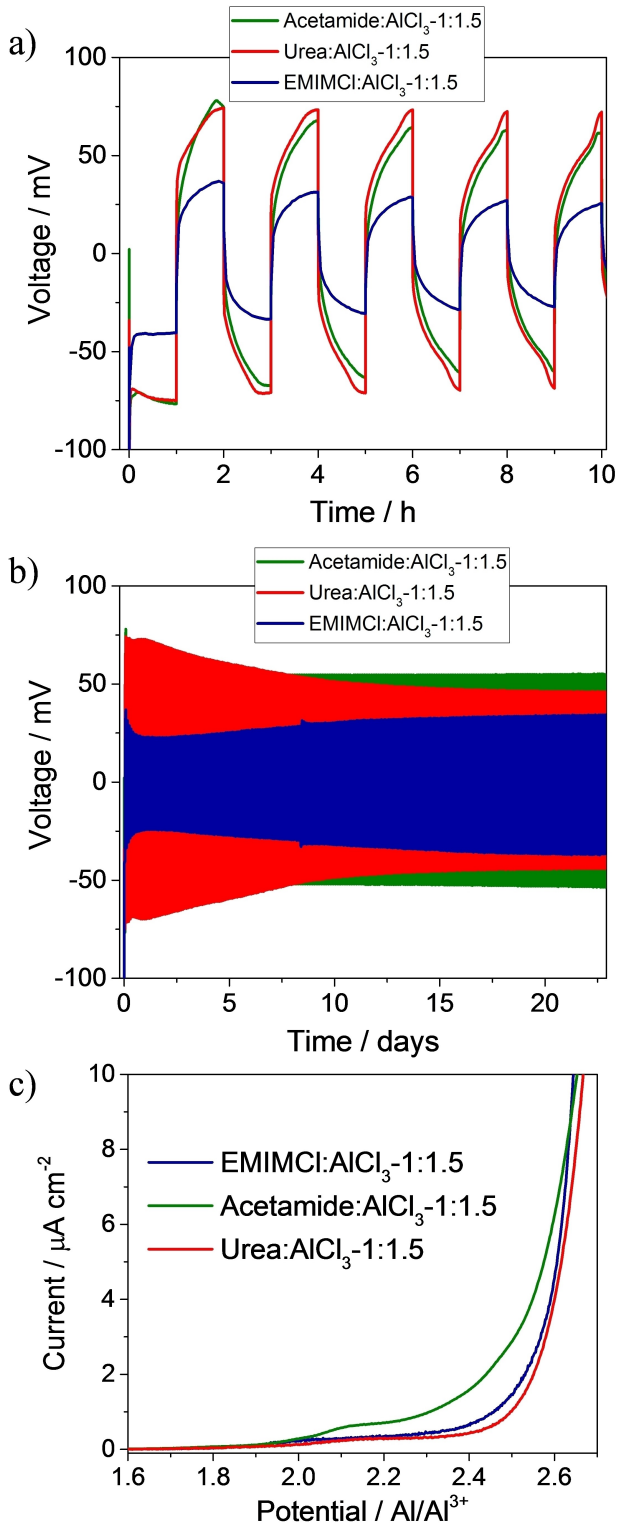



**Figure 3.** a–b) Voltage vs. time signature of the galvanostatic cycling test performed on symmetrical Al/Al cell using EMIMCl:$AlCl_3$-1:1.5 (in blue), Urea:$AlCl_3$-1:1.5 (in red) and Acetamide:$AlCl_3$-1:1.5 (in green) electrolytes. Measurement performed at 0.1 mA $cm^{-2}$ current, with a stripping deposition time of 1 h at room T. The test first 10 h are reported in (a), in (b) the full test performed for 24 days. c) Current vs. voltage signature of the linear sweep voltammetry test obtained in a three-electrode configuration using glassy carbon as working electrode and Al as counter and reference and employing EMIMCl:$AlCl_3$-1:1.5 (in blue), Urea:$AlCl_3$-1:1.5 (in red), and Acetamide:$AlCl_3$-1:1.5 (in green) electrolytes. Measurements obtained using a scan rate of 0.1 mV $s^{-1}$ at room T.


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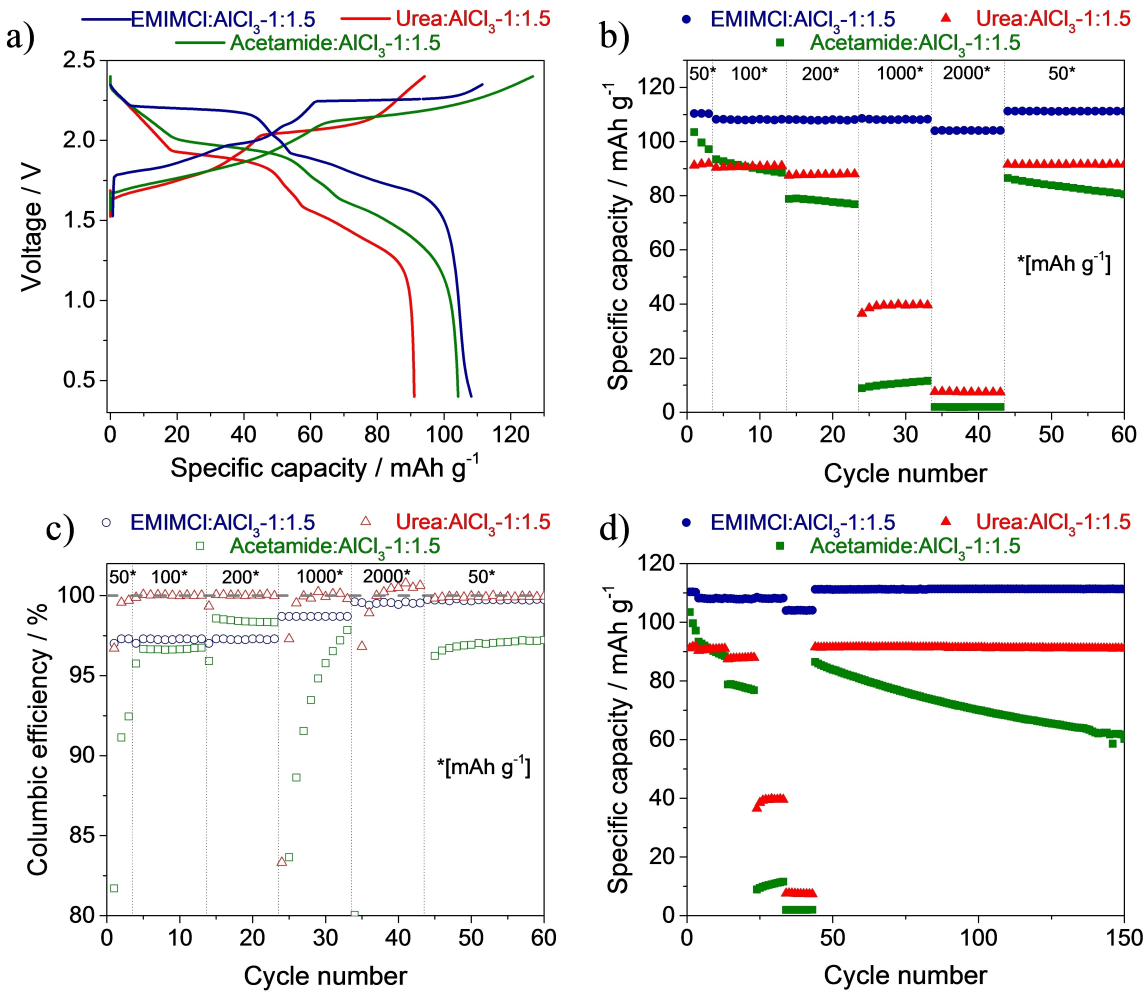



**Figure 4.** Galvanostatic cycling test results of Al/NG cells using EMIMCl:$AlCl_3$-1:1.5 (in blue), Urea:$AlCl_3$-1:1.5 (in red) and Acetamide:$AlCl_3$-1:1.5 (in green) electrolytes. a) First cycle voltage profile. b) Capacity vs. cycle number behavior of the multi-rate test. c) Coulombic efficiency vs. cycle number behavior of the multi-rate test. d) Extended cycling behavior results. Measurements obtained cycling the cells within the 0.4–2.4 V voltage window end employing specific currents of 50, 100, 200, 1000, 2000 mA $g^{-1}$ at room T.

**Table 2.** Discharge energy, average discharge voltage, cell polarization, energy efficiency, columbic efficiency, and discharge capacity of the 1st, 5th, and 100th cycle of the AGDICs using the three investigated electrolyte systems. The evaluation was obtained from the cycling test analysis reported in figure 3, for cycles performed at 50 mA $g^{-1}$ current.

| Electrolyte composition | Cycle number | Discharge energy [Wh $kg^{-1}$] | Average discharge voltage [V] | Polarization [V] | Energy efficiency [%] | Columbic efficiency [%] | Discharge capacity [mAh $g^{-1}$] |
|---|---|---|---|---|---|---|---|
| EMIMCl $AlCl_3$ | 1st | 206.60 | 1.91 | 0.16 | 90.43 | 97.02 | 110.37 |
| | 5th | 205.56 | 1.90 | 0.30 | 86.39 | 97.30 | 108.23 |
| | 100th | 211.66 | 1.90 | 0.24 | 88.63 | 99.76 | 111.39 |
| Urea $AlCl_3$ | 1st | 157.72 | 1.73 | 0.24 | 85.32 | 96.70 | 91.20 |
| | 5th | 152.94 | 1.69 | 0.27 | 86.14 | 100.06 | 90.49 |
| | 100th | 151.33 | 1.66 | 0.30 | 84.88 | 99.99 | 91.35 |
| Acetamide $AlCl_3$ | 1st | 184.83 | 1.77 | 0.26 | 71.88 | 81.70 | 103.49 |
| | 5th | 156.69 | 1.69 | 0.39 | 78.57 | 96.68 | 92.70 |
| | 100th | 103.05 | 1.47 | 0.71 | 66.11 | 97.84 | 70.08 |

The Acetamide:$AlCl_3$ system reports the lowest columbic efficiency (Table 2). The columbic efficiency behavior of the AGDICs links well with the anodic electrochemical stability of the various electrolytes (Figure 3c). The superior Urea:$AlCl_3$ stability against oxidation guarantees the best coulombic efficiency in AGDICs; result in agreement with other reports.[17,22] The Acetamide:$AlCl_3$ system shows a columbic efficiency of about 96–97 %, thus in line with this composition's lower anodic stability. The lower columbic efficiency of the Acetamide:$AlCl_3$ is reflected in the poor capacity retention in


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Al/NG cells, retaining less than the 60% after 150 cycles (Figure 4d). On the contrary, EMIMCl:$AlCl_3$ and Urea:$AlCl_3$ does not evidence a relevant capacity decay during the test's 150 cycles.

## 3. Conclusions

The overall comparison between the standard EMIMCl:$AlCl_3$ electrolyte and Urea:$AlCl_3$ and Acetamide:$AlCl_3$ DES electrolytes has been reported. The comparison evidences the superior characteristics of the EMIMCl:$AlCl_3$ electrolyte as media for the Al stripping deposition process. Additionally, the standard EMIMCl:$AlCl_3$ electrolyte reveals better characteristics in AGDICs in terms of rate capability. The better features of the EMIMCl:$AlCl_3$ over the Urea:$AlCl_3$ and Acetamide:$AlCl_3$ DES are mostly addressed to the former's lower viscosity and higher conductivity. However, AGDICs employing Urea:$AlCl_3$ system have better columbic efficiency, around 99.9% for the urea with respect to the 97% of the EMIMCl:$AlCl_3$. Indeed this is beneficial for practical cell application where the amount of electrolyte is limited, and a columbic efficiency of 97% is impractical. Additionally, the urea-based electrolyte is expected to be characterized by lower cost, considering that urea is a very common chemical widely produced and used as fertilizer. Nonetheless, the elevated viscosity and low conductivity result in a significant limitation in the AGDIB system's best characteristics, the outstanding rate capability. Improvements in the DES attributes need to be achieved to push the concept to practical application consideration.

## Experimental Section

The electrolytes prepared in a 1: 1.5-mol ratio, 1-ethyl-3-methylimidazolium chloride:aluminum trichloride (EMIMCl):$AlCl_3$, Urea:$AlCl_3$, and Acetamide:$AlCl_3$ are provided by IOLITEC. The water content of the electrolytes is lower than 100 ppm. The electrochemical measurements were performed using Teflon Swagelok® type T cells.[6,28–30] All potentials quoted in this manuscript refer to the *quasi reference* Al/$Al^{3+}$ electrode. The investigated electrolytes' capability to sustain the reversible aluminum stripping deposition process has been investigated by cyclic voltammetry (CV) of three-electrode cells using Ni foil as a working electrode and Al as counter and reference. The test has been performed at a scan rate of 0.1 $mV\,s^{-1}$ within the −0.3 V and 0.6 V vs. Al/$Al^{3+}$ voltage window. The cycling stability of the aluminum metal in the various electrolytes was evaluated by continuous stripping/deposition tests on symmetrical Al/Al cells (Al 99.99% Alfa Aesar) employing a current of 0.1 $mA\,cm^{-2}$ and a stripping/deposition time of 1 h through a Maccor 4000 battery test system. The investigated electrolytes' anodic stability has been evaluated by linear sweep voltammetry (LSV) using a three-electrode cell comprising a glassy carbon working electrode and Al-metal as counter and reference. The measurements have been performed using a scan rate of 0.1 $mV\,s^{-1}$. The evaluation of the investigated electrolytes' electrochemical behavior in Al cells has been performed assembling AGDICs comprising natural graphite (NG) positive electrodes. The natural graphite powder (NG) was provided by PLANO GmbH. NG electrodes with a loading of about 5 $mg\,cm^{-2}$ have been employed for the electrochemical characterization. The detailed electrode preparation procedure has been described in our previous paper.[11] The cycling test of the Al/NG cells has been performed using a multi-rate procedure using 50, 100, 200, 1000, 2000 $mA\,g^{-1}$ within the 0.4–2.4 voltage window. The specific capacity and current are referred to the mass of the graphite.

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## Conflict of Interest

The authors declare no conflict of interest.

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