

Acetamide: a low-cost alternative to alkyl imidazolium chlorides for aluminium-ion batteries

Nicolò Canever,^a Nicolas Bertrand^b and Thomas Nann^{*a}

EXPERIMENTAL SECTION

Acetamide, 1-ethyl-3-methylimidazolium chloride, aluminium chloride and dichloromethane were purchased from Sigma-Aldrich. Molybdenum foil, aluminium foil and pyrolytic graphite paper were purchased from MTI Corp. Acetamide, 1-ethyl-3-methylimidazolium chloride ([EMIm]Cl) and aluminium chloride were dried by heating at 100 °C in a vacuum oven for 48 hours. Dichloromethane was dried over activated molecular sieves (3Å) for 48 hours.

To prepare the electrolytes, the previously dried chemicals were transferred into a nitrogen-filled glove box with the O₂ and H₂O levels kept below 1 ppm. Briefly, the electrolytes were synthesized by gradually adding AlCl₃ (1.1, 1.3 or 1.5 equivalents, depending on the desired molar *r* ratio) to one equivalent of either [EMIm]Cl or acetamide under magnetic stirring at room temperature. An exothermic reaction takes place, resulting in a pale-yellow liquid for Acetamide-based electrolytes, or a brown liquid for the [EMIm]Cl-based electrolyte. Any residual AlCl₃ was reacted by heating the ionic liquids to 120 °C over a hotplate for 1 hour. Dilutions of the electrolytes with dichloromethane were performed by simply adding the desired volume fraction to the electrolytes. This step was also performed inside the glovebox.

The cyclic voltammetry and galvanostatic charge-discharge experiments were conducted in an electrochemistry cell using a three-electrode configuration. The cell was composed of a glass vessel and plastic top piece with an O-ring joint. The counter and pseudo-reference electrode consisted of two small sheets of aluminium foil, and the working electrode was either a third sheet of Al foil or pyrolytic graphite paper. The electrodes were connected via alligator clips to custom-built electrode shafts inserted into the cell and sealed with O-rings. Due to the brittleness of pyrolytic graphite paper, the sheet was sandwiched between two small Mo sheets to prevent the alligator clips from damaging it (**Figure S1**). A glassy carbon electrode was used as the working electrode to obtain the cyclic voltammograms in **4**.

The assembled electrochemical cell was connected to a Schlenk line, and was flushed with nitrogen by performing three vacuum-N₂ cycles. The electrolytes were taken out of the glovebox in a seal vial with a rubber septum cap and immediately transferred into the electrochemical cell using a syringe. The as-prepared cell was then used for the electrochemical tests.

Cyclic and linear sweep voltammetry experiments were run using a potentiostat (Metrohm Autolab PGSTAT128N) using a voltage ramp of 80 mV s⁻¹. Exchange current data from **Figure S7** was obtained by performing Tafel analysis on linear sweep voltammeteries between 1.25 and 1.75 V (vs. Al foil) with a sweep rate of 5 mV s⁻¹. Galvanostatic charge-discharge experiments were performed using a battery analyser system (NEWARE BTS CT-4008-5V10mA-164, MTI Corp.) using a rate of 83 mA g⁻¹ unless differently specified.

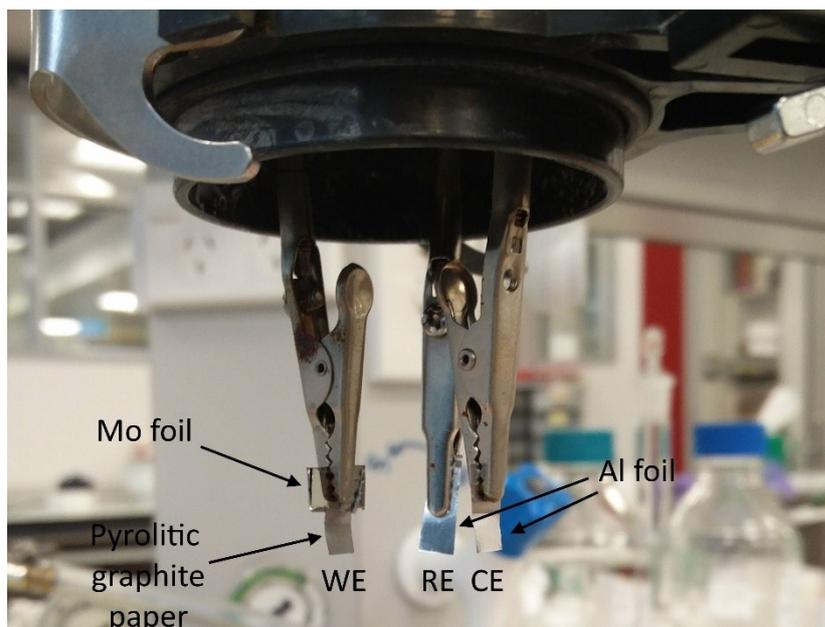


Figure S1. Digital image of a partially assembled three-electrode cell setup used to perform the electrochemical tests.

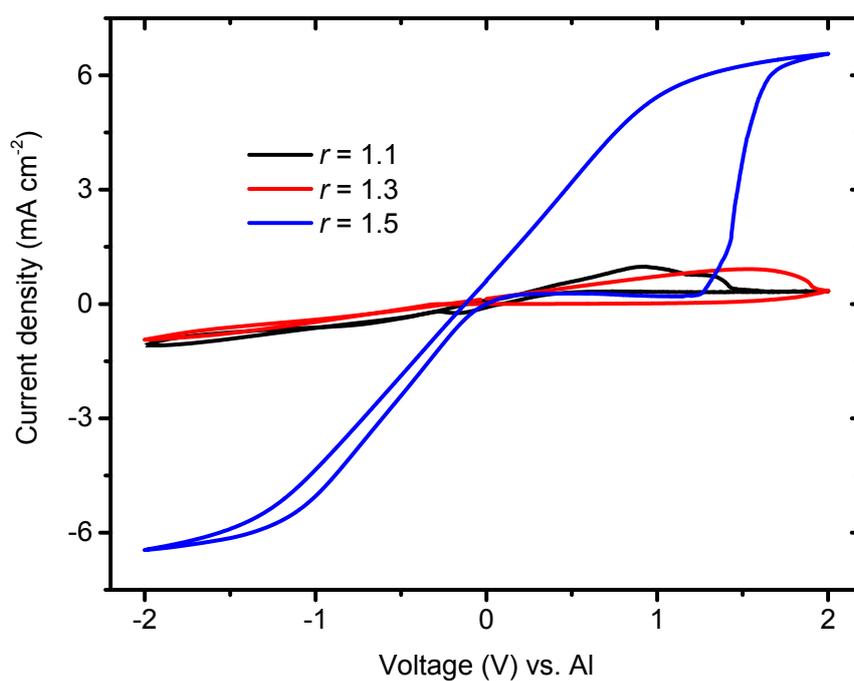


Figure S2. Electroplating cyclic voltammograms for acetamide electrolytes with different r values, using Al foil as the working electrode.

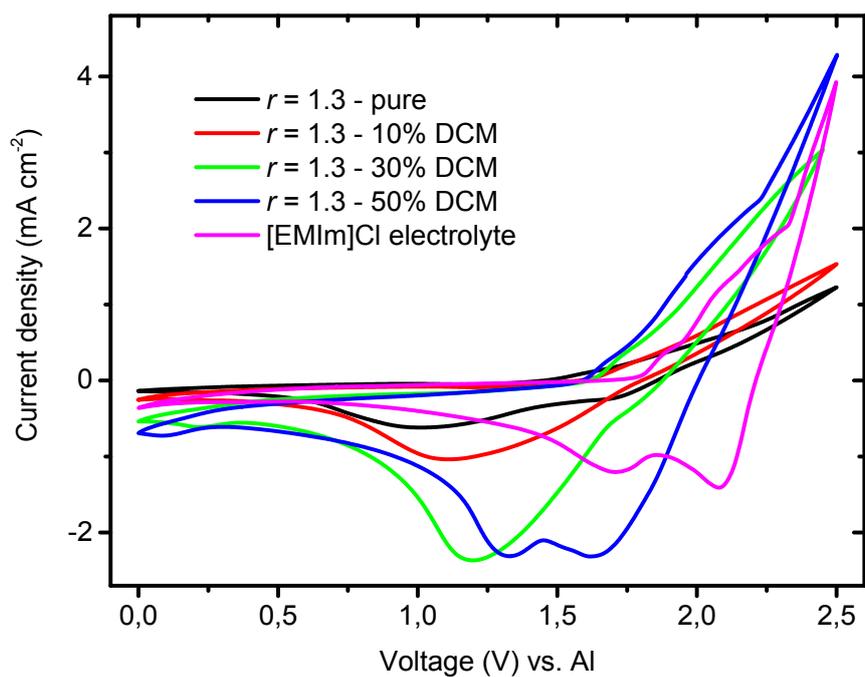


Figure S3. Cyclic voltammograms of an acetamide- AlCl_3 ionic liquid with $r = 1.3$, diluted with different volume fractions of dichloromethane (DCM), compared with the [EMIm]Cl-based electrolyte.

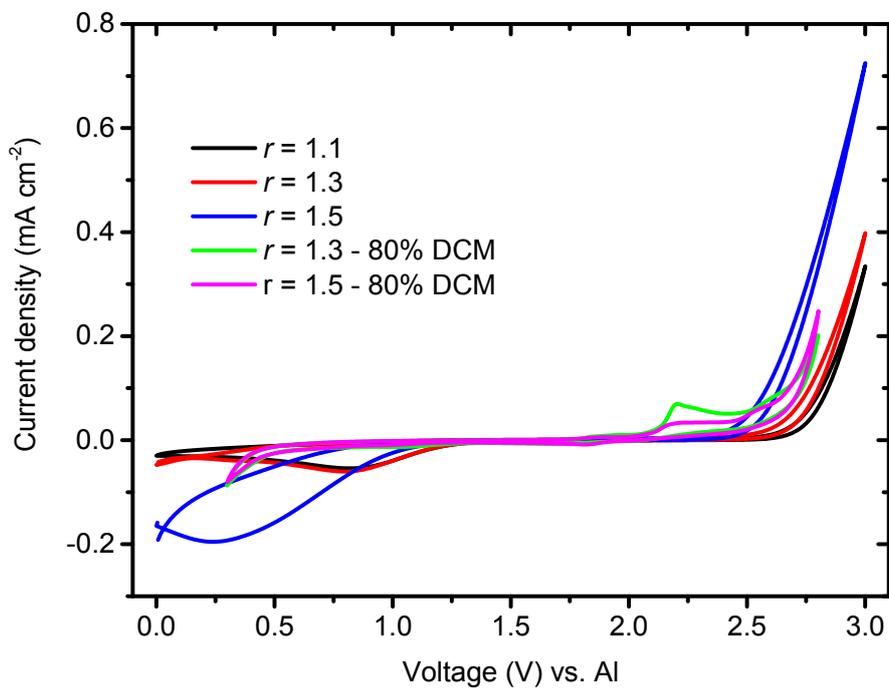


Figure S4. Cyclic voltammograms for the determination of the electrochemical voltage window of the electrolytes prepared, performed using a glassy carbon working electrode.

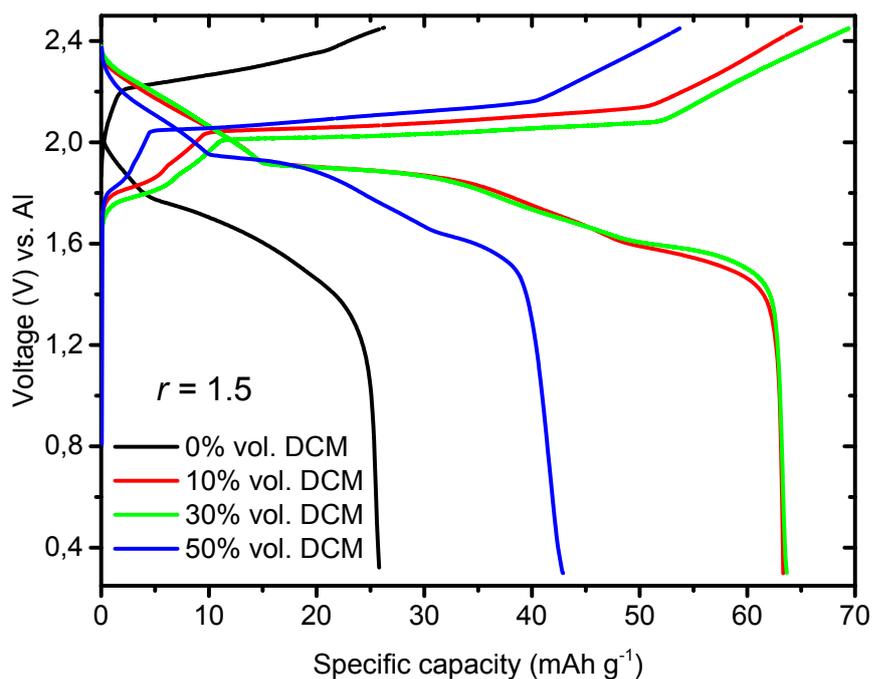


Figure S5. Typical galvanostatic charge-discharge profiles for an acetamide- AlCl_3 ionic liquid with $r = 1.5$, diluted with different volume fractions of dichloromethane (DCM).

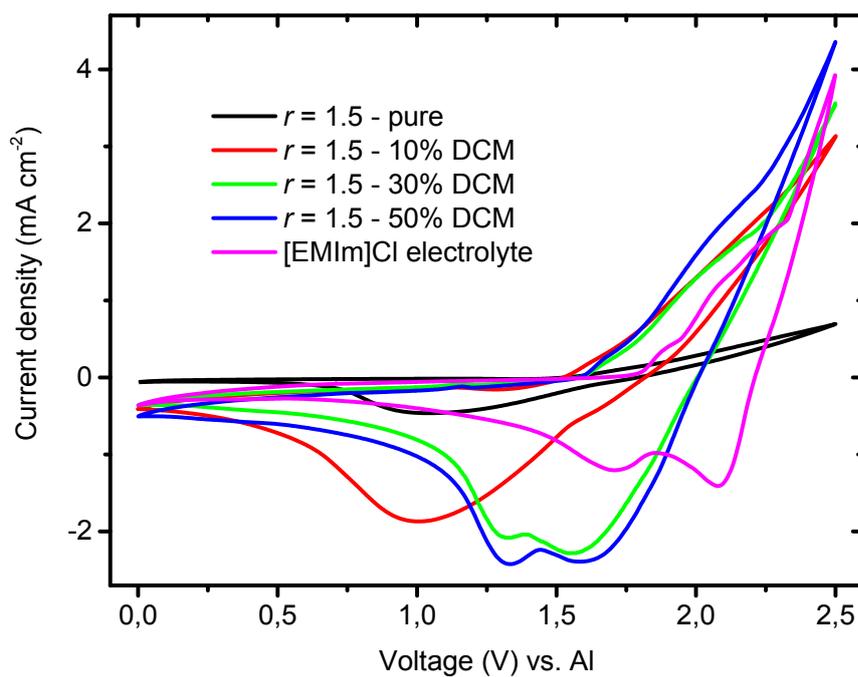


Figure S6. Cyclic voltammograms of an acetamide- AlCl_3 ionic liquid with $r = 1.5$, diluted with different volume fractions of dichloromethane (DCM), compared with the [EMIm]Cl-based electrolyte.

Figure S7. Exchange current densities of AcAm-AlCl₃ ionic liquids with different *r* values and diluted with different volume fractions of dichloromethane.

