

Carbon neutral electrochemical conversion of carbon dioxide mediated by [M: N+(cyclam)Cl_n] (M = Ni²⁺ and Co³⁺) on mercury free electrodes and ionic liquids as reaction media

Honores, J.

Quezada, D.

García, M.

Calfumán, K.

Muena, J. P.

Aguirre, M. J.

Arévalo, M. C.

Isaacs, M.

© 2017 The Royal Society of Chemistry. In this work, the electrochemical reduction of carbon dioxide using [Mⁿ⁺(cyclam)Cl_n] (M = Ni²⁺ and Co³⁺) as electrocatalysts has been studied in 1-butyl-3-methylimidazolium tetrafluoroborate and 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (BMImBF₄ and BMImNTf₂ respectively) ionic liquids as reaction media. Complexes were characterized electrochemically in these salts and relevant parameters, such as a heterogeneous electron transfer rate was calculated. Results indicate a faster M(ii)/M(i) redox process in BMImBF₄ despite its higher viscosity compared to BMImNTf₂. Cyclic voltammetry experiments demonstrated that [Ni(cyclam)Cl₂] is the most active macrocycle, towards the reaction under survey. For this compound, potential controlled electrolysis was carried out at -1.4 V vs. Ag/AgCl in BMImBF₄ as a solvent, yielding only CO as a reaction product, with a turnover frequency (TOF) of 0.73 h⁻¹. NMR spectra for the ionic liquids, after