Aprotic phosphonium-based ionic liquid as electrolyte for highly CO2 electroreduction to oxalate

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Abstract

In this study, a new electrolyte system consisting of tetrabutylphosphonium 4-(methoxycarbonyl) phenol ([P4444][4-MF-PhO]) ionic liquid and acetonitrile (AcN) was developed as CO2 electroreduction electrolyte to produce oxalate, and the mechanism was studied. The results showed that using the new ionic liquid-based electrolyte, the reduction system exhibits 93.8% Faradaic efficiency and 12.6 mA cm-2 partial current density of oxalate at -2.6 V (vs. Ag/Ag+). The formation rate of oxalate is 234.4 μmol cm-2 h-1, which is better than that reported in the literature. The mechanism study using density functional theory (DFT) calculation revealed for the first time that [P4444][4-MF-PhO] IL can effectively activate CO2 molecules through ester and phenoxy double active sites, stabilize the reaction intermediate. The potential barriers of the key intermediates *CO2- and *C2O42- formation by induced electric-field was reduced in the phosphonium-based ionic environment, which greatly facilitates the activation and conversion of CO2 molecules to oxalate.

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