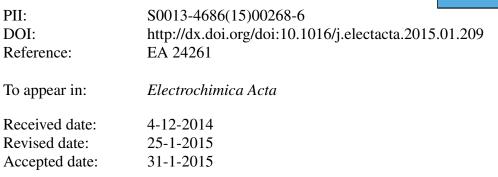
Accepted Manuscript

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Please cite this article as: P.Bumroongsakulsawat, G.H.Kelsall, Tinned graphite felt cathodes for scale-up of electrochemical reduction of aqueous CO2, Electrochimica Acta http://dx.doi.org/10.1016/j.electacta.2015.01.209

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ACCEPTED MANUSCRIPT

Tinned graphite felt cathodes for scale-up of electrochemical reduction of aqueous CO₂

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Abstract

Three-dimensional cathodes with large volumetric surface areas for CO₂ reduction in aqueous solutions were fabricated by depositing Sn on graphite felt. CO₂ was reduced electrochemically in aqueous solutions of 1 M NaClO₄ + 0.5 M NaOH saturated with CO_2 to pH ca. 7.8 and circulated though the tinned graphite felt cathodes to enhance mass transport rates. Gaseous product bubbles, presumably of CO and H₂, generated within the threedimensional cathodes decreased effective conductivities of the interstitial electrolyte solutions, increasing potential drops in the ionically conducting phase. This caused reaction rates to decay within the cathodes in the direction of current flow. Hence, the effect of increasing solution flow rate was not only to enhance transport rates, but also to decrease gas fractions within the cathode, increasing effective conductivities of the electrolyte solution and so changing the spatial distribution of potential and current density. An optimal superficial current density and charge yield of 971 A m^{-2} and 0.58, respectively, were obtained at -1.62 V (AgCl|Ag) and 99 ml min⁻¹ solution flow rate; current densities were increased by a factor of 27 compared with the behaviour of a 2D electrode. A one-dimensional mathematical model was developed that was able to predict with adequate accuracy the effects of electrode potential and electrolyte solution flow rates on cross-sectional current densities, charge yields, and potential drops within the three-dimensional cathodes in the direction of current flow.

Keywords: CO₂; electrochemical reduction; Sn; formate; bubble

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