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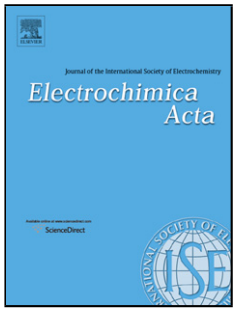

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ACCEPTED MANUSCRIPT

Improvement of the selectivity of the electrochemical conversion of CO_2 to hydrocarbons using cupreous electrodes with in-situ oxidation by oxygen

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Abstract

It is still a challenge to find catalysts for the selective electrochemical reduction of CO_2 to value added products. We present a new method for the in-situ modification of a copper catalyst by adding oxygen to the reactant gas. With an appropriate process procedure consisting of a sequence of electrolysis and purge steps, a Cu_2O layer was formed during the purge steps. When it was reduced again during the next electrolysis step, Cu^+ species remained stable on the surface and let to a higher faradaic efficiency for C_2H_4 formation and even a lower onset potential for CO_2 reduction in general. This effect was observed for a wide range of O_2 content from 10 % to 60 % in the reactant gas. In comparison to electrolysis with pure CO_2, formation of methane was largely suppressed.

Keywords: *electrochemical reduction, carbon dioxide, copper oxide, ethylene, hydrocarbons, selectivity*

1. Introduction

The insufficiently controlled emission of CO_2 into the atmosphere and the associated change of the global climate are increasingly recognized as a serious problem. In this context, the direct electrochemical conversion of CO_2 to hydrocarbons pioneered by work of Hori et al. in the 1980s found renewed interest in the past few years. Intermittently available excess electrical energy

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