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A new option in CO_2 recycle: conversion to long-chain alcohols and hydrocarbons

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Background and Justification for Acceptance

Effective recycle of CO_2, a necessary option alternative to storage, is still a challenge, notwithstanding the increasing number of studies. We proposed a novel concept based on a gas-phase photoelectrocatalytic devices [1,2], e. g. different from the conventional liquid phase PEC systems and closer to PEM fuel cells. In this device, one side is composed from a nanostructure TiO_2-based thin film where gaseous water is splitted using solar light to produce O_2, protons and electrons, while the anode side is based on novel nanostructured carbon-based electrode also operating in gas phase. On the anode side, CO_2 is converted using protons passing through a Nafion® membrane and electrons (a wire connects the two sides of the cell). The characteristics of the electrode are critical aspect and we showed that at room temperature and pressure, using Pt nanoparticles confined in conductive carbon nanopores, it is possible to form long-chain hydrocarbons [2]. We report here new results, using Fe or Pt nanoparticles on carbon nanotubes (CNT) and operations at slightly higher temperature (60°C), where we show that one order of magnitude higher productivities are possible, with a selective synthesis of alcohols (particularly isopropanol).

Results

Electrocatalytic activity tests are carried out in a continuous flow electrocatalytic reactor at room temperature and T=60°C where the working electrode operates in contact with CO_2 fed continuously, and protons, provided by an electrolyte ($KHCO_3$, 0.5 M), diffuse to catalyst through a Nafion® membrane. Experiments are conducted galvanostatically with modulations in current to improve desorption of the products from the working electrode (± 20 mA). Fe or Pt / CNT (10% wt) on gas diffusion electrodes assembled with Nafion® 112 membranes were tested in the CO_2 electrocatalytic conversion. Reaction products are mainly alcohols and (>C5) hydrocarbons. Distribution of products depends on the nature of the metal ion and reaction temperature. At room temperature mainly C5-C8 hydrocarbons are formed, whereas at higher temperatures preferentially oxygenates prevails. Fe catalysts show a higher selectivity into isopropanol, whereas Pt electrocatalysts are more selective towards C1-C2 oxygenate products. TEM characterizations of the electrocatalysts support structure-activity relationships.

References

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