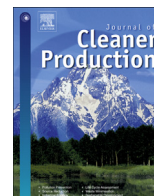




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## Global warming footprint of the electrochemical reduction of carbon dioxide to formate

A. Dominguez-Ramos<sup>a,\*</sup>, B. Singh<sup>b</sup>, X. Zhang<sup>b,c</sup>, E.G. Hertwich<sup>b</sup>, A. Irabien<sup>a</sup><sup>a</sup> Departamento de Ingeniería Química y Química Inorgánica, ETSIIyT, Universidad de Cantabria, Avda. Los Castros s/n, Santander 39005, Spain<sup>b</sup> Industrial Ecology Programme, Department of Energy and Process Engineering, Norwegian University of Science and Technology (NTNU), Trondheim 7491, Norway<sup>c</sup> State Key Lab of Multi-phase Complex Systems, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China

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## ABSTRACT

Carbon dioxide Capture and Storage (CCS) is an important technological option for climate change mitigation. Utilization (U) of this captured CO<sub>2</sub> as raw material for Electrochemical Reduction (ER) has been suggested as a valorization option to produce organics such as formate-based products. Previous work has focused on the influence of operating conditions or the selected cathodic material on the faradaic efficiency and distribution of products. The environmental sustainability of formate production through the ER of CO<sub>2</sub> has been assumed rather than investigated. In this study, we perform a life cycle assessment, focusing on resources and greenhouse gas (GHG) emissions. Even though the processes reported in the literature result in a wide range of GHG emissions for the ER of CO<sub>2</sub> to formate-based products from 32 to 519 kg CO<sub>2</sub>-eq.·(kg HCOO<sup>-</sup>)<sup>-1</sup>, this is higher than the current commercial production processes (3.1 kg CO<sub>2</sub>-eq.·(kg HCOO<sup>-</sup>)<sup>-1</sup>). The consumption of chemicals by the electrolysis and of steam in the purification of the final formate products are found to be the dominant causes of environmental burdens of the integrated process. A future scenario under very optimistic conditions suggests 0.33 kg CO<sub>2</sub>-eq.·(kg HCOO<sup>-</sup>)<sup>-1</sup> thus presenting a potential pathway to an environmentally sustainable CO<sub>2</sub> utilization option.

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## 1. Introduction

The idea of utilizing CO<sub>2</sub> captured from power plants and other point sources to produce chemicals is gaining increasing attention (Quadrelli et al., 2011). Potential products include methanol, dimethyl ether, olefins, carbonates and formic acid (Chaplin and Wragg, 2003; Gattrell et al., 2006; Gattrell et al., 2007; Gonçalves et al., 2010; Graves et al., 2011; Kuhl et al., 2012; Lee and Tak, 2001; Schizodimou and Kyriacou, 2012; Subramanian et al., 2007; Whipple and Kenis, 2010). It has even been suggested that the reduction of CO<sub>2</sub> could be used as a method of storing surplus peak electricity (Olah, 2004, 2005; Olah et al., 2009).

However, since the capture of CO<sub>2</sub> and its electrochemical reduction into organic products needs additional chemicals and energy, the net profile of the utilization of captured CO<sub>2</sub> can be negative. According to Irabien et al. (2009) the two main elements that can be used to determine the environmental sustainability of

products or processes are the *Natural Resources Sustainability* (NRS) and the *Environmental Burdens Sustainability* (EBS). On the other hand, the life cycle approach can be well adopted to perform the environmental sustainability assessment (ESA) of CO<sub>2</sub> utilization options. Life cycle assessment (LCA) is a well-established tool to check or assess for any problem-shifting in environmental impacts (Finnveden et al., 2009), providing a holistic view of the environmental sustainability of the selected scope. For sustainability concerns, it is expected that the effect from the obtained products over the whole life cycle should be lower than the actual products to be substituted. In this paper, we investigate the environmental sustainability of producing formate or formic acid (depending on pH) by electrochemical reduction of CO<sub>2</sub> and compare this production route to the conventional synthesis pathways.

The formate/formic acid product could function as a carbon based energy vector for the storage of excess of electricity from renewable sources. Direct Formic Acid Fuel Cells (DFAFCs) are envisaged for low power appliances (Cai et al., 2012; Kundu et al., 2007). While the energy density of formic acid with 5.9 MJ·(kg HCOOH)<sup>-1</sup> is lower than that of methanol 21.9 MJ·(kg CH<sub>3</sub>OH)<sup>-1</sup> (Demirci, 2007), formic acid has a relatively

\* Corresponding author. Tel.: +34 942 206749.

E-mail addresses: [domingueza@unican.es](mailto:domingueza@unican.es), [emperadorantonio@hotmail.com](mailto:emperadorantonio@hotmail.com) (A. Dominguez-Ramos).

higher electrocatalytic oxidation rate and lower membrane cross-over, which are barriers for Direct Methanol Fuel Cells (Rees and Compton, 2011; Rice et al., 2002). Also, the transport and storage of formic acid are simpler than methanol (Cai et al., 2012; Rice et al., 2002). Potential markets for formic acid are the leather industry, agriculture (BASF, 2012; Reutemann and Kieczka, 2011) and potential substitution of strong acids at steel pickling (Mantra Venture Group, 2010).

The aim of this work is to present an environmental sustainability assessment (ESA) of the production of formate-based products (formate/formic) via electrochemical reduction (ER) of CO<sub>2</sub> captured at a coal combustion plant (as CO<sub>2</sub> source) equipped with a carbon dioxide capture unit, leading to a Carbon Capture, Storage and Utilization system (CCS&U). The quantification of the life cycle GHG emissions could help in the detection and understanding of the critical steps, thus identifying the potential possibilities for improvement of the ER process. The present work is divided into four sections including this introduction. The second section describes the system and methodology used in the assessment. A shared structure for the CCS&U system is divided into three levels: 1st reaction; 2nd reaction and purification; and 3rd reaction and purification within the post-combustion power plant. The individual inventories and hypothesis for each level are described. The third section is devoted to the results and discussion. The NRS at the reaction level is firstly presented as an introduction to the EBS analysis focused on the greenhouse gas emission of the reaction and purification level, which is the core of this work and therefore described in detail. A long-term future improvement in the purification step together with a complete reduction in the consumption of chemicals was included in order to evaluate the theoretical potential of the technology. A proposal to improve the environmental sustainability of formate production via ER route is made. Main conclusions are presented in the last section.

## 2. Methodology

This study assumes a hypothetical facility which includes the electrochemical reduction of CO<sub>2</sub> to formate-based products connected to a coal-fired power plant equipped with a carbon capture unit (we assume post-combustion based on amine as solvent). A detailed flowchart of the process structure for the carbon dioxide capture, transport, storage and utilization (CCS&U) is presented in Fig. 1, which shows four main sections: combustion; capture; transport and storage; and electrochemical reduction (including electrochemical reactor and purification sub-sections which are the main focus of this work).

The captured CO<sub>2</sub> is split into two fractions: (i) the fraction for use as raw material for the ER process and (ii) the fraction for compression, transport and storage. The fraction of the captured CO<sub>2</sub> sent to the ER process is defined as the *Derivation Ratio* (DR). The stream is assumed to be pure CO<sub>2</sub> with enough pressure for the ER process. The larger CO<sub>2</sub> fraction is supplied to compression, transport and storage, and only relatively low values of DR are found to be technically plausible for conversion to formate. In the ER process, the CO<sub>2</sub> is reduced to formate-based products in the cathodic compartment in the presence of chemicals acting as supporting electrolytes. A parallel cathodic reaction produces hydrogen. At the anodic compartment the main involved reaction is the production of oxygen. The electricity produced by the generator is partially split into: (i) final electricity distributed to the grid; (ii) energy for the capture process (including capture, desorption and compression); and (iii) energy for the ER process. Alternatively, the electricity needed in the ER process can also be supplied by PV or wind power. Additional steam is needed for the distillation of the aqueous formate produced in the ER process. The energy for transport and storage is withdrawn from external sources (corresponding grid mix).

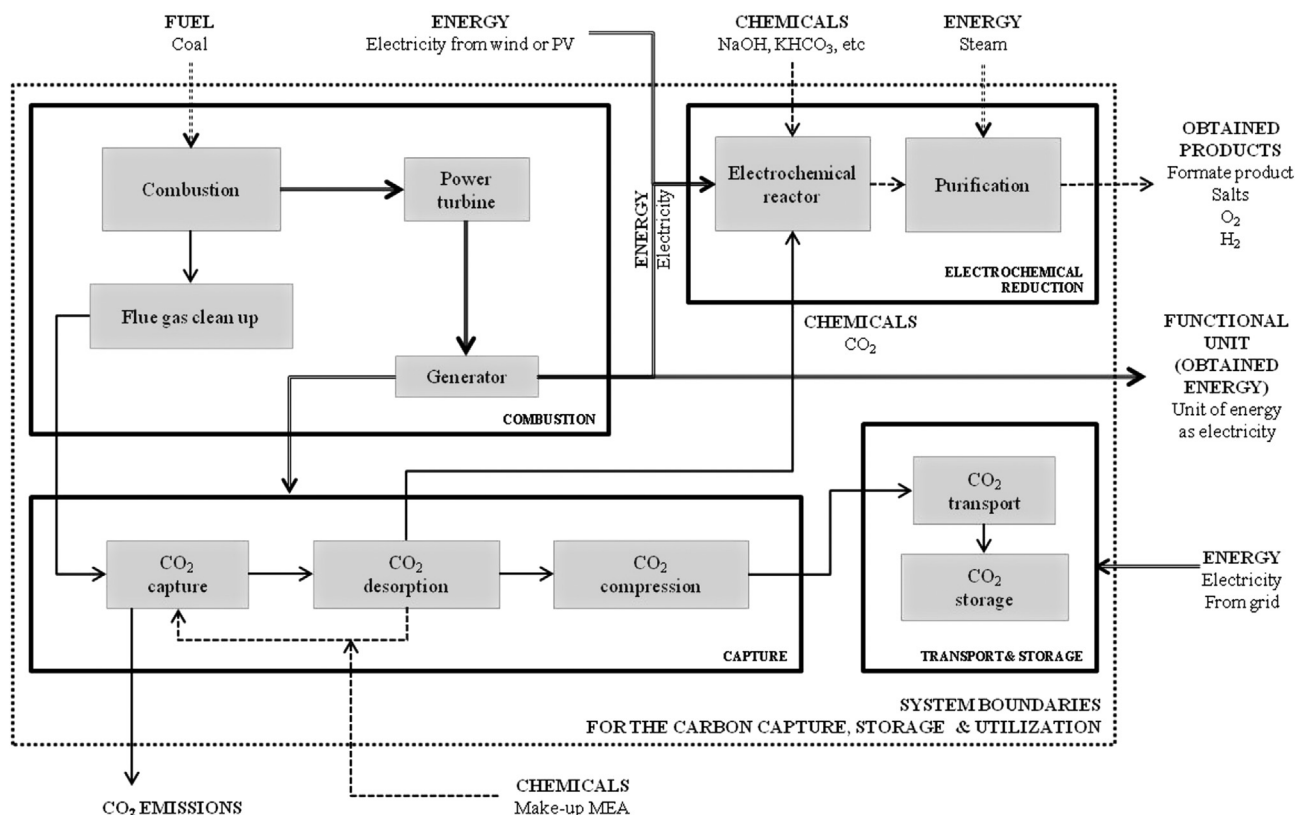


Fig. 1. Process flowchart of the suggested process structure for the combustion plant and the CCS&U unit (including system boundaries).

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