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Constructing global models from past publications to improve design and operating conditions for direct alcohol fuel cells

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ABSTRACT

This work aims to analyze past publications on direct alcohol fuel cells (DAFC) in the literature using two data mining tools (artificial neural networks and decision trees) and to develop global models to predict the conditions leading to high performance of DAFC. The database constructed for this purpose contains 4682 data points over 271 polarization (IV) curves obtained from 36 publications in the literature. Decision tree classification models were used to develop heuristics to select the suitable fuel cell design and operational conditions to improve the maximum power density while artificial neural network models (ANN) were developed to test the predictability of IV curves at the conditions where experimental results were not available. The same ANN models were also used to determine the relative importance of design and operational variables to provide some insight to determine the variable to be manipulated. All these analyses were quite successful deducing some useful heuristics and models for the future studies from the continuously growing experience accumulated in the literature.

1. Introduction

Direct alcohol fuel cells (DAFC) have been extensively studied in recent years for their potential use in electronics and transportation due to their advantages like liquid fuel use and high energy densities. In the last decades, significant improvements have been achieved to make DAFCs more efficient in order to replace hydrogen fuel cells and other conventional sources; these improvements were reported in numerous studies in the literature aiming to enhance electro-oxidation of alcohols by novel catalyst preparation techniques, to reduce alcohol crossover by synthesis of new hydrocarbon and composite fluorinated membranes and to replace noble metal based catalysts with non-noble metals for cost reduction (Kamarudin et al., 2013). In addition, in order to achieve the desired performance and high durability in fuel cells, numerous studies were also performed on water and heat management, oxidant transport and on the utilization of different type of alcohols (Ho et al., 2014; Li and Faghri, 2013).

The fundamental problems regarding electro-catalytic reactions in DAFCs are: decomposition of alcohols to surface poisoning species like CO leading to the degradation of catalytic activity, increase in the complexity of oxidation mechanism with the increasing chain length of aliphatic alcohol fuel, and lack of information about electro-catalyst composition–activity relationship on numerous low-Pt (platinum) loaded or non-Pt based anode and cathode electro-catalysts (Zhao et al., 2011). In order to test more noble and non-noble metal combinations and various catalyst compositions, the combinatorial approaches have become very

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attractive for fast catalyst down selection in the last ten years; many binary like PtSn (platinum–tin), ternary like PtRuW (platinum–ruthenium–tungsten) or quaternary like $Ni_{31}Zr_{13}Pt_{33}Ru_{23}$ (nickel–zirconium–platinum–ruthenium) catalyst formulations have been tested in order to replace conventional model electro-catalysts like Pt black or PtRu, and promising results were achieved (Lamy et al., 2004; Liu et al., 2006; Zhou, 2004). The alcohol cross-over is also another important issue for DAFC, and it is closely related to the membrane structure, morphology and thickness as well as operational variables such as temperature, pressure, alcohol concentration and water content. All these complex relationships create challenges for modifying the existing membranes or developing new alternatives (Ahmed and Dincer, 2011).

In addition to experimental studies, numerous modeling works were also carried out by various investigators. For instance, the semi-empirical models were developed and used to investigate the effect of operating parameters and to predict the behavior of polarization curves (Andreadis and Tsiakaras, 2006; Argyropoulos, 2003; Tu et al., 2006); these models were found to be applicable for a wide range of experimental conditions, and able to determine the major cause of fuel cell voltage drop or conditions for minimum alcohol crossover (Tapan and Ozturk, 2009). The semi-empirical models were also developed and used to study the effects of fuel cell materials (for instance polymer electrolyte membrane) on the fuel cell performance (Springer et al., 1991). In addition to these steady state models, the dynamic models were developed to assess the stability of fuel cell performance (Chiu et al., 2011) and to optimize fuel and exergetic efficiency (Wu and Lin, 2010).

All these experimental and modeling efforts show that there is a wide experimental domain to be explored for developing high performance fuel cells and understanding the related electro-catalytic and physical issues. From morphology of membrane and composition of electrode materials to operating conditions, there are many fuel cell design and operational variables to be optimized. However, these variables are in large numbers, have wide ranges and strongly interact with each other; therefore, it is impossible to test all the possible alternatives in a single work and come up with an acceptable solution. On the other hand, the number of research papers also increases continuously creating a massive accumulation of knowledge that can be utilized to understand the complex interactions among the vast number of variables. Indeed, a simple review of the literature shows that there are numerous publications on the novel catalytic materials to boost electro-catalytic activity of anode and cathode electrodes, on the membranes or fuels to suppress cross over and enhance the performance of the fuel cell and on the models to predict physical behavior of fuel cell at different operating conditions. It is quite possible that the information we are expected to gain from a new experiment may be already hidden within the network of published data and it may be found if we use suitable tools. Thanks to the fast developments in computational resources in recent years, these tools are now available under the data mining discipline.

Data mining is a collection of algorithms and methods to extract valuable knowledge hidden in a large and complex database; by using several tasks such as clustering, classification and estimation, and tools to perform these tasks; it helps researchers to see the correlations, trends and patterns that are not easy to see with naked eyes (Alpaydın, 2004; Larose, 2005; Tan et al., 2005). For many years, various data mining tools have been successfully utilized in various fields from biology (Mamitsuka et al., 2013) and astronomy (Way et al., 2012) to customer relations and economy (Ahlemeyer-Stubbe and Coleman, 2014).

We also used some data mining tools on the relatively large databases that we created from the results reported in the past publications for some catalytic reactions, and obtained considerable success. For example, we used decision tree analysis, which is a classification method, to develop heuristics for CO oxidation (Günay and Yildirim, 2013) and water gas shift reactions (Odabaşı et al., 2014), and artificial neural networks (one of the most common estimation methods in data mining) to predict the outcome of untested conditions and understand the effects of catalyst preparation and operational variables on the catalytic performance (Günay and Yildirim, 2011, 2013; Odabaşı et al., 2014); in some cases, we also applied clustering methods to divide the dataset into smaller subsets to improve the modeling efficiency (Günay and Yildirim, 2013). There are also similar works performed by different groups in recent years for oxidative coupling of methane (Kondratenko et al., 2015; Zavyalova et al., 2011) and biodiesel production (Baroi and Dalai, 2014). To the best of our knowledge, no such study (using data in published literature to develop models and heuristics) was performed on the fuel cell literature, although successful applications of data mining tools (especially artificial neural networks) were reported in the electrochemical field by various investigators to the smaller datasets created in a single work. For example, ANN modeling was employed to model the electrochemical process in the porous cathode of solid oxide electrolysis cells (Grondin et al., 2013), to model the dynamic mechanical behavior of polymer electrolyte membrane fuel cells (Paclisan and Charon, 2013) and to predict voltages and local temperatures of solid oxide fuel cells (Razbani and Assadi, 2014). All these works made us believe that data mining on the entire DAFC literature can also be performed to complement the experimental works.

In this work, a database was constructed using the experimental data set from the works published last 12 years on direct alcohol fuel cells. Then, the database was analyzed using decision trees and artificial neural networks to construct models to determine the conditions leading to high fuel cell performance, to predict the performance of fuel cell under certain set of fuel cell and operational variables, and to determine the relative importance of these variables on the fuel cell performance.

2. Computational work

2.1. *Constructing the database*

The literature on the direct alcohol fuel cell from 2002 to 2014 was reviewed and 36 publications, which report polarization curves (IV data) at various fuel cell design and operating conditions, were selected. Then, a database consisting 4682 data points over 271 IV curves was constructed from the experimental data in these publications. List of these publications, together with the distribution of data among the papers (as the number of data points, number of IV curve, alcohol type and membrane type) is presented in Supporting information file.

We tried to model both *entire IV curve (voltages at various values of current)* and *the maximum power density* as performance (dependent) variable, since they may provide different kind