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# An experimental overview of the effects of hydrogen impurities on polymer electrolyte membrane fuel cell performance

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## ABSTRACT

Hydrogen quality is critical for increasing the reliability, stability, and durability of polymer electrolyte (PEM) fuel cells. In this work, several hydrogen impurities have been studied to understand their effects on PEM fuel cell performance at various operating concentrations. Our studies have shown that the following impurities suggested by industry stakeholders do not result in substantial fuel cell degradation when they are the sole impurity in hydrogen: 5 ppm formaldehyde, 2 ppm formic acid, 19 ppm chloromethane, 30 ppm acetaldehyde, 5% ethylene, 20 ppm toluene, and 10 ppm benzene. In addition, a specific mixture of impurities called the “specification concentration level cocktail” consisting of 0.2 ppm carbon monoxide, 4 ppb hydrogen sulphide, 0.2 ppm formic acid, 2 ppm benzene, and 0.1 ppm ammonia in hydrogen, also does not show significant effects on cell performance. In comparison, when a cocktail having five times the specification concentration is introduced into the cell, significant performance loss is evident.

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## Introduction

Hydrocarbon fuels are not a renewable resource. As the economies of the world grow, the available stores of these fuels are being consumed at ever increasing rates. As a result, additional greenhouse gases are released as by-products of combustion leading to rapidly increased pollution. Therefore,

sources of clean and sustainable energy are needed to make better use of available resources. The polymer electrolyte membrane (PEM) fuel cell is a promising energy conversion device which emits almost no pollutants and has high working efficiency. The PEM fuel cell is believed to be one of the best near term solutions for the energy problem [1].

Many challenging issues must be addressed and solved before large-scale commercial applications of PEM fuel cells

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would be feasible. Generally, the performance of two key PEM fuel cell components needs to be significantly improved. The first of these is the catalyst. Different types of catalyst have been tested to improve their tolerance for impurities which “poison” active sites. In addition, the platinum catalyst loading must be decreased to reduce cost [2–8]. The other key component is the membrane. According to the United States Department of Energy (DOE), PEM fuel cells are required to have more than 5000 h life for cars, 20,000 h life for buses, and 60,000 h life for stationary applications [9]. However, the current PEM fuel cells only have approximately 2000 h life for cars [10,11], 10,000 h life for buses [11], and 22,000 h life for stationary applications [12]. To reach the required durability, membrane lifetimes must be improved by a factor of two to three. Many researchers are working on these issues and progress is being made [13–18].

Industry produced hydrogen is the most promising fuel for PEM fuel cells. Improving the fuel quality will extend the working life and facilitate the application of PEM fuel cells [19]. There is significant potential for modifying the current hydrogen manufacturing processes to make it better suited for fuel cell applications. In general, fewer impurities in hydrogen fuel are preferred because that reduces the contaminant effects on the catalyst and lowers the degradation rates of the membrane. Therefore, longer operating life is expected. However, fewer impurities also mean a higher price for hydrogen. Therefore, it is very important to understand the contaminant effects of each impurity and determine the concentration ranges which will not result in substantial PEM fuel cell degradation.

Multiple techniques have been employed for hydrogen production. Among them, steam methane reforming (SMR) is the dominant technique. In the United States, approximately 95% of hydrogen is made via SMR [20]. Besides SMR, coal/oil partial oxidation, coal destructive distillation, and electrolysis of water are also widely used to produce hydrogen. In these processes, nitrogen ( $N_2$ ), carbon monoxide (CO), carbon dioxide ( $CO_2$ ), methane ( $CH_4$ ), ammonia ( $NH_3$ ), sulfur compounds, benzene ( $C_6H_6$ ), toluene ( $C_7H_8$ ), and other organic chemicals can be produced as by-products. Some of these chemicals, such as hydrocarbons, contaminate PEM fuel cells either by direct adsorption, or by decomposing into carbon monoxide which adsorbs on the catalyst surface to reduce the electrochemically active surface area (ECSA) of the catalyst. Other impurities, such as ammonia which is widely used as tracer gas in natural gas distribution system, can replace protons in Nafion<sup>®</sup>, reducing the proton conductivity and mechanical properties of the membrane [21–24]. Some chemicals, such as  $N_2$ , only dilute the fuel and reduce performance. They will not cause performance degradation and are believed to be safe for PEM fuel cell operation.

Over past few years, many impurities that are produced during hydrogen production, transportation, and storage, have been studied [25–28] which include formaldehyde, formic acid, chloromethane, acetaldehyde, ethylene, toluene, benzene, carbon monoxide, hydrogen sulphide, and ammonia. However, these studies are normally focused on one or two impurities at a time at various operating conditions. So far, no work has been done to evaluate the effects of mixed impurities.

In this work, research has been done to study the effects of both single impurities and mixed impurities in hydrogen on PEM fuel cell performance based on an identical test protocol. The goal is to determine the maximum impurity concentrations that PEM fuel cells can tolerate without inducing significant performance degradation. The significance of performance degradation is decided by comparing the performance differences between impurity tests and the baselines (without impurities).

The purpose of this research was to provide experimental data as well as a possible qualitative mechanism analysis for prioritizing the contaminant effects of impurities in hydrogen. In particular, this research addressed the degradation of the fuel cell power system performance that can be restored by means of practical changes to operating conditions and/or fuel composition. Because of the requirements and priorities assigned by DOE and the US Drive Fuel Cell Tech Team – which includes critically important stakeholders such as the automakers, fuel producers, and major fuel cell developers, our team was constrained to a series of very specific screening tests that were representative of certain operational scenarios. The tests were structured to investigate the contaminant effects of impurities in hydrogen and prioritized to highlight the possible contaminants that may be generated during hydrogen production. This work was focused on several screening tests as well as some recovery tests. The mitigation strategies employed to alleviate hydrogen impurity contamination and the corresponding mechanisms have been widely studied by researchers and are addressed in the literature [21,29–37].

Many PEM researchers use the terms “recovery” and “mitigation” interchangeably in the literature, and for most research there is no ambiguity. However, for the DOE Hydrogen contaminant program that supported this research, these terms were given specific and distinct meanings. For this program, the term “recovery” was defined as “the degradation of the fuel cell power system performance that can be restored by practical changes of operational conditions and/or fuel composition [38], and all other mechanisms, both *in-situ* and *ex-situ*, for improving performance that was lost due to operating with contaminants in the fuel supply gas, were defined as “mitigation”. The reason for this distinction stems from the down selection process that was established to manage the very large number of potential fuel side contaminants. This process was used by a number of researchers, and then formalized by St. Pierre et al. [39].

The down selection of contaminant species was based on fixed criteria which included: i) atmospheric presence at a significant level, ii) expectation of reactivity within the fuel cell, iii) absence of recorded data, iv) largest range in chemical functionalities, v) compound toxicity, and vi) suggestions provided by industry and research institutions [39,40]. However, these criteria do not consider the effects of contaminant species on fuel cell performance. Therefore, St-Pierre et al. proposed two quantitative down selection criteria based on empirical methods [39]. The first criterion analyzes the performance degradation/recovery rates, performance loss induced by contamination, recoverable/irrecoverable performance losses and contaminant concentration. The second criterion examines the combination of the energy loss in

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