



# Study of the aromatic hydrocarbons poisoning of platinum cathodes on proton exchange membrane fuel cell spatial performance using a segmented cell system



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## HIGHLIGHTS

- Effects of aromatic air contaminants on PEMFC were studied with a segmented cell.
- Cathode exposure to 2 ppm C<sub>6</sub>H<sub>6</sub> caused a moderate and recoverable performance loss.
- PEMFC poisoning by 2 ppm C<sub>10</sub>H<sub>8</sub> led to a drastic decrease in a cell voltage.
- Performance loss under arenes exposure is due to their strong chemisorption on Pt.
- Oxidation of C<sub>6</sub>H<sub>6</sub> to CO<sub>2</sub> can occur under PEMFC operating conditions.

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## ABSTRACT

Aromatic hydrocarbons are produced and used in many industrial processes, which makes them hazardous air pollutants. Currently, air is the most convenient oxidant for proton exchange membrane fuel cells (PEMFCs), and air quality is an important consideration because airborne contaminants can negatively affect fuel cell performance. The effects of exposing the cathode of PEMFCs to benzene and naphthalene were investigated using a segmented cell system. The introduction of 2 ppm C<sub>6</sub>H<sub>6</sub> resulted in moderate performance loss of 40–45 mV at 0.2 A cm<sup>-2</sup> and 100–110 mV at 1.0 A cm<sup>-2</sup> due to benzene adsorption on Pt and its subsequent electrooxidation to CO<sub>2</sub> under operating conditions and cell voltages of 0.5–0.8 V. In contrast, PEMFC poisoning by ~2 ppm of naphthalene led to a decrease in cell performance from 0.66 to 0.13 V at 1.0 A cm<sup>-2</sup>, which was caused by the strong adsorption of C<sub>10</sub>H<sub>8</sub> onto Pt at cell voltages of 0.2–1.0 V. Naphthalene desorption and hydrogenation only occurred at potentials below 0.2 V. The PEMFCs' performance loss due to each contaminant was recoverable, and the obtained results demonstrated that the fuel cells' exposure to benzene and naphthalene should be limited to concentrations less than 2 ppm.

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## 1. Introduction

Proton exchange membrane fuel cells (PEMFCs), as electrochemical replacements for internal combustion engines in portable, stationary and automotive power generation, have continued attracting research and development efforts due to recent successful commercialization attempts. However, technical challenges such as reliability, durability and sufficient performance under different environmental conditions still exist and are associated with fuel cell degradation. One significant factor contributing to

fuel cell performance loss and failure is contamination caused by fuel, air or system impurities [1–4]. Currently, air is the most convenient and economic oxidant for PEMFC operation; at the same time, however, air quality remains a major concern due to the presence of different air pollutants, including nitrogen- and sulfur-containing compounds (NO<sub>x</sub>, NH<sub>3</sub>, SO<sub>2</sub>), volatile organic compounds, carbon monoxide and particulates. Airborne contaminants damage fuel cell performance mainly by affecting catalysts because platinum readily chemisorbs a large variety of inorganic and organic compounds, thereby reducing the electrochemically active area (ECA) and altering sluggish oxygen reduction reaction (ORR) from a 4-electron to a 2-electron pathway [5]. Moreover, the contaminants and the products of electrochemical/chemical transformations may also affect membrane conductivity and the

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hydrophilic properties of gas diffusion layers (GDLs), resulting in performance loss due to an increase in ohmic and mass transfer overpotentials.

Extensive research efforts have been dedicated to examining PEMFC cathode contamination caused by inorganic pollutants such as SO<sub>2</sub> [3,6–15], NO<sub>x</sub> [3,7,8,11,12,14–18], NH<sub>3</sub> [7,8,11,13,19,20], H<sub>2</sub>S [7,8,11,12,21] and Cl<sup>-</sup> [13,22–26]. However, studies on the effects of organic compounds such as air contaminants are smaller in scope and limited by benzene [15,27,28], 1,3-butadiene [28], toluene [29,30] and chemical warfare agents [27]. Because organic compounds have the potential to decrease PEMFC performance, a list of 260 possible airborne contaminants provided by the Environmental Protection Agency has been evaluated. Several contaminants have been selected for further detailed studies of their effects on fuel cells [31]. The chosen contaminants belong to different classes of organic substances: alkenes (C<sub>3</sub>H<sub>6</sub>), alkynes (C<sub>2</sub>H<sub>2</sub>), arenes (C<sub>6</sub>H<sub>6</sub>, C<sub>10</sub>H<sub>8</sub>), alcohols (1-C<sub>3</sub>H<sub>7</sub>OH), nitriles (CH<sub>3</sub>CN), esters (CH<sub>2</sub>=C(CH<sub>3</sub>)COOCH<sub>3</sub>) and alkyl halides (CH<sub>3</sub>Br). The selected compounds are widely used as chemical reagents, solvents, welding fuels and pesticides, and they cause negative effects on PEMFC performance [31–35]. According to our studies, these effects are accounted for different mechanisms of interaction between membrane electrode assembly (MEA) components and the contaminants, depending on their chemical nature and properties.

Aromatic compounds (benzene and naphthalene) are considered possible air pollutants because they are produced or used in many industrial processes. Benzene is a natural component of crude oil, with a concentration ranging from 0.001 to 0.4%, and petroleum refineries are the primary source of C<sub>6</sub>H<sub>6</sub>. Other commercial sources of benzene include direct extraction from the light oil formed during coal coking and from gas-well condensates [36]. Benzene is predominantly used as a starting material for the chemical synthesis of ethylbenzene, cumene, cyclohexane and other substituted aromatic hydrocarbons. More than half of all benzene produced is processed into ethylbenzene, a precursor to styrene, which is used for manufacturing polymers and plastics. Benzene is also used as a gasoline additive (1–5% by volume) to increase octane rating and reduce knocking. Other products that incorporate benzene include solvents and pesticides. Naphthalene is another significant arene and is commercially produced from coal tar or petroleum. Worldwide, naphthalene is mainly used for the manufacturing of phthalic anhydride by vapor-phase catalytic oxidation, a precursor for polyvinyl chloride plasticizers. Naphthalene is also used in the manufacture of a wide variety of intermediates for the dye industry, synthetic resin, surfactants, moth repellent and pesticides [37,38].

Emission sources of benzene and naphthalene are similar and may be distinguished based on generation mechanism because they are emitted as products of the incomplete combustion of wood, straw, tobacco, fossil fuels (gasoline, diesel, oil, coal) and through evaporation or sublimation of C<sub>6</sub>H<sub>6</sub>- and C<sub>10</sub>H<sub>8</sub>-containing materials such as coal tar, crude oil, petroleum products and moth repellent. Emissions can be also classified as natural sources (wildfires) and as anthropogenic sources, which are a much larger and more significant sources of exposure.

Benzene is emitted during its production and from coke ovens. The other major sources of benzene in air are motor vehicle exhaust and evaporation losses during handling, distribution and storage of gasoline. The benzene concentration in ambient air generally ranges from 1 to 30 µg m<sup>-3</sup> (0.3 ppb–0.01 ppm), depending on the proximity of traffic or industrial objects [39,40]. The extensive use of naphthalene as an intermediate in the chemical industry and its presence as a major component of coal tar has led to its occurrence in industrial effluents, outdoor and indoor environments. In addition to these sources, the emission of naphthalene is similar to that

of benzene and occurs through wood and stationary fossil fuel combustion and burning. Air concentrations of naphthalene in cities typically vary from 0.5 to 1.0 µg m<sup>-3</sup> (0.1–0.2 ppb), whereas the maximum outdoor concentration can reach up to 5–25 µg m<sup>-3</sup> (1–5 ppb) [37]. Tobacco smoking has been found to be a notable indoor benzene and naphthalene emission source, producing 90–500 µg benzene/cigarette and 15–18 µg naphthalene/cigarette [41].

The adsorption of benzene and its derivatives onto Pt has been a subject of fundamental studies due to considerable interest in various electrochemical syntheses involving aromatic hydrocarbons and for the purification of effluent from organic contaminants [42–51]. However, data concerning the effects of benzene and naphthalene on PEMFC performance and oxygen reduction are rare [15,27,35]. It was shown that even 2 ppm of C<sub>6</sub>H<sub>6</sub> in air caused a fuel cell performance loss of ~150 mV at 1.0 A cm<sup>2</sup> and 80 °C, whereas exposure to 2 ppm C<sub>10</sub>H<sub>8</sub> resulted in a drastic decrease in cell voltage to 0.1 V under the same conditions. However, no explanation for the observed results was provided. Moreover, further detailed analysis and assessment are required because the evaluation of PEMFCs by single-cell testing usually provides only average values of current, voltage and impedance, whereas valuable information about local performance is beyond reach. In contrast, a segmented cell system provides spatially resolved voltage, current and impedance data and is a powerful tool for gaining a detailed understanding of fuel cell performance under various operating conditions, i.e., exposure to different fuel or air impurities [33,34,52–59]. The current work is a continuation of our previous studies regarding the effects of airborne pollutants on fuel cell performance and spatial PEMFC behavior under cathode exposure to benzene and naphthalene contaminants to assess relative transport processes along the flow field. The contaminant concentration used in this study was 2 ppm, which is higher than that observed under real conditions, to enhance and accelerate the effects of pollutants on fuel cells.

## 2. Experimental

The experiments were conducted on a GRanalytix single fuel cell test station using a segmented cell system [60]. This diagnostic tool was used in previous studies [61–64] and allows simultaneous measurements of spatial electrochemical impedance spectra (EIS), spatial linear sweep voltammetry (LSV) and cyclic voltammetry (CV) to be performed. The system has ten current channels in a high-current (standard) mode and ten channels in a low-current mode. The standard mode enables the measurement of currents up to 15 A for each channel. The low-current mode allows for the measurement of currents as low as 375 mA, which is typical for CV and LSV experiments.

The segmented cell hardware is based on a 100 cm<sup>2</sup> cell design. The hardware contains a segmented flow field, which consists of ten cell segments forming a continuous path along ten parallel serpentine channels. Each segment has an area of 7.6 cm<sup>2</sup> and its own distinct current collector and gas diffusion layer (GDL). The same channel designs are used for the segmented cathode and standard anode flow fields (the reactant streams are arranged in a co-flow configuration). The segmented cell hardware is applicable to either the anode or the cathode.

The segmented cell was operated with a commercially available 100 cm<sup>2</sup> MEA provided by Gore. The anode and cathode were made of Pt/C catalysts with a loading of 0.4 mg<sub>Pt</sub> cm<sup>-2</sup>. Sigracet 25 BC was used for the anode and cathode GDLs. The cathode used a segmented GDL and gasket configuration, whereas a single GDL was applied at the anode, and the total active area of the MEA was 76 cm<sup>2</sup>. The gasket material was made of Teflon, with a thickness of

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