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An Experimental Study on the Performance of Proton Exchange Membrane Fuel Cells with Marine Ion Contamination

Shian Li , Li Zhang, Gaokui Chen, Ruiyang Zhang, Aolong Liu, Guogang Yang and Qiuwan Shen *

Marine Engineering College, Dalian Maritime University, Dalian 116026, China
* Correspondence: shenqiuwan@dlmu.edu.cn

Abstract: Proton exchange membrane fuel cells (PEMFCs) have the advantages of high efficiency, a low operating temperature, and a pollution-free reaction. Therefore, PEMFCs have emerged as a viable clean energy solution for ships to reduce their carbon emissions. When PEMFCs operate in marine salt spray environments, foreign ions entering the cathodes of fuel cells with air can cause a decline in cell performance. In this study, the effects of the cation type (K^+, Na^+, Mg^{2+}, and Ca^{2+}) and concentration (0.25 M and 0.5 M) on cell performance in terms of the polarization curve were systematically investigated using a fuel cell test system. Cell performance degradation was observed due to the existence of cations. The influence of the four cations on cell performance followed the rule of $Ca^{2+} > Mg^{2+} > Na^+ > K^+$. Meanwhile, cell performance decreased with an increase in concentration. When the fuel cell was not contaminated, the voltage was 0.645 V at a current density of 1 A/cm^2. When the concentration was 0.5 M, the corresponding voltages were 0.594 V, 0.583 V, 0.559 V, and 0.300 V, respectively. In addition, fuel cells contaminated by $NaNO_3$ and NaCl were compared. Due to the existence of Cl^-, more severe performance degradation was observed when the fuel cells were contaminated by NaCl.

Keywords: PEMFC; marine salt spray environment; cell performance; ion contamination

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1. Introduction

In the context of promoting greenhouse gas emissions reductions in the international shipping industry, it is crucial to find solutions that reduce air pollution [1,2]. Replacing traditional fossil fuels with clean energy is one of the main approaches [3,4]. Among numerous clean energy sources, hydrogen is the most widely distributed. Due to its various production methods and diverse application scenarios, it has received widespread attention from governments and research institutes around the world [5,6]. Proton exchange membrane fuel cells (PEMFCs) use hydrogen as a fuel. The electrochemical reaction only produces water, which makes it pollution-free and carbon-free. PEMFCs can provide power in the shipping industry to improve the marine environment and reduce carbon emissions [7,8]. In addition, PEMFCs also have the characteristics of high working efficiency, high energy density, and a low working temperature [9,10]. Therefore, PEMFCs are considered one of the most promising energy conversion devices and have become an emerging fossil fuel substitute in ship power systems [11,12]. When PEMFCs operate in marine salt spray environments, foreign ions entering the cathodes of fuel cells with air can cause a decline in cell performance [13]. To effectively alleviate the negative effects of salt spray on cell performance, air filters are often used on the cathode sides of fuel cells [14]. However, air filters cannot completely eliminate the adverse effects of salt spray. In severe cases, they can directly lead to cell failure, causing the entire system to collapse [15,16].

Scholars have conducted various studies related to the effect of foreign ion contamination on cell performance. Shu et al. [17] introduced Cu^{2+} solutions with five different concentrations into a fuel cell cathode and found that as the concentration of pollutants increased the cell performance showed a decreasing trend and that the voltage decay was greater at higher current densities. Yan et al. [18] used Na^{+} and Ca^{2+} solutions with different concentrations and introduced them into a fuel cell cathode. They found that both cations affected cell performance to varying degrees. Mikkola et al. [19] found a significant decrease in fuel cell performance, with a decrease in the current density of about 30%, when a NaCl solution was injected into a fuel cell. Uddin et al. [20] introduced a Ca^{2+} solution, along with air, into a cell cathode and found that a fuel cell operating in both constant-current and constant-voltage modes exhibited performance degradation. Sulek et al. [21] immersed a proton exchange membrane in solutions containing Al^{3+}, Fe^{2+}, and Cr^{3+}. Subsequently, a membrane electrode assembly (MEA) was fabricated, and the corresponding polarization curves were obtained and compared. It was found that the degree of contamination with the three cations affected cell performance as follows: $Al^{3+} >> Fe^{2+}$ and Cr^{3+}. Qi et al. [22] introduced K^{+}, Ba^{2+}, Ca^{2+}, and Al^{3+} solutions into a cell's cathode side. It was observed that all four cations resulted in declines in cell performance. Zhu et al. [23] applied Mg^{2+} solutions with varying concentrations to a fuel cell. Their results indicated that both increasing the Mg^{2+} concentration and extending the contamination duration intensified the degradation of the cell's performance. Wen et al. [24,25] investigated an MEA in a marine salt spray environment using a salt spray test chamber. Their findings showed that the Na^{+} concentration in the membrane increased with time and the solution concentration, which likely contributed to the observed performance degradation. Li et al. [26] introduced a Cl^{-} solution to both the anode and cathode of an operating cell. Regardless of whether the Cl^{-} entered through the cathode or the anode, the cell performance exhibited a sharp decline. Li et al. [27] found that reducing the humidity of the inlet air enhanced the toxic effect of Cl^{-} and accordingly accelerated the decline in fuel cell performance. Matsouka et al. [28] mixed HCl solutions with air. After contamination, they analyzed the polarization curves and observed a downward trend in performance. Uddin et al. [29] introduced a Cl^{-} solution from the cathode and mixed it with air. A significant decrease in the cell's output voltage was observed after a certain period. Unnikrishnan et al. [30] introduced Cl_2 into a fuel cell from the cathode side and found that Cl_2 dissociated into Cl^{-}, which then adsorbed onto the surface of the platinum catalyst, negatively affecting the cell's performance. Baturina et al. [31] placed a PEMFC in a salt spray test chamber to simulate a marine salt spray environment. Their results indicated that Cl^{-} caused a decline in the cell's performance. It was reported that the longer the exposure, the greater the degree of performance degradation.

As mentioned above, cations (K^{+}, Na^{+}, Ba^{2+}, Ca^{2+}, Fe^{2+}, Mg^{2+}, Cr^{3+}, and Al^{3+}) and anions (Cl^{-}, F^{-}, and $NO_3{}^{-}$) were selected as research objects by different researchers. According to the results reported in the above studies, it was concluded that foreign ion contamination can lead to a decrease in cell performance. Different methods were adopted to analyze foreign ion contamination in the experimental schemes, and different fuel cells were used in the experimental studies by different researchers. These differences increase the difficulty in comparing the results. In a marine salt spray environment, foreign ions inevitably enter a PEMFC cathode along with air, and cell performance can be significantly affected. Therefore, it is necessary to perform studies to improve our understanding of fuel cells in marine salt spray environments.

This study focused on PEMFCs used in a marine salt spray environment and the performance of fuel cells with marine ion contamination. However, research in this area is still limited. Most studies focus on a single ion, and comprehensive comparative analyses of the effects of different ions are also needed. Therefore, it is essential to investigate the effects of various types of foreign ion contamination on cell performance. The objective of this work was to examine the effects of the cation type (K^+, Na^+, Mg^{2+}, and Ca^{2+}) and concentration (0.25 M and 0.5 M) on cell performance. Meanwhile, fuel cells contaminated by $NaNO_3$ and NaCl were also compared. Comparative studies based on the polarization curve are comprehensively demonstrated. This work can provide valuable insights into cell performance degradation due to foreign ion contamination.

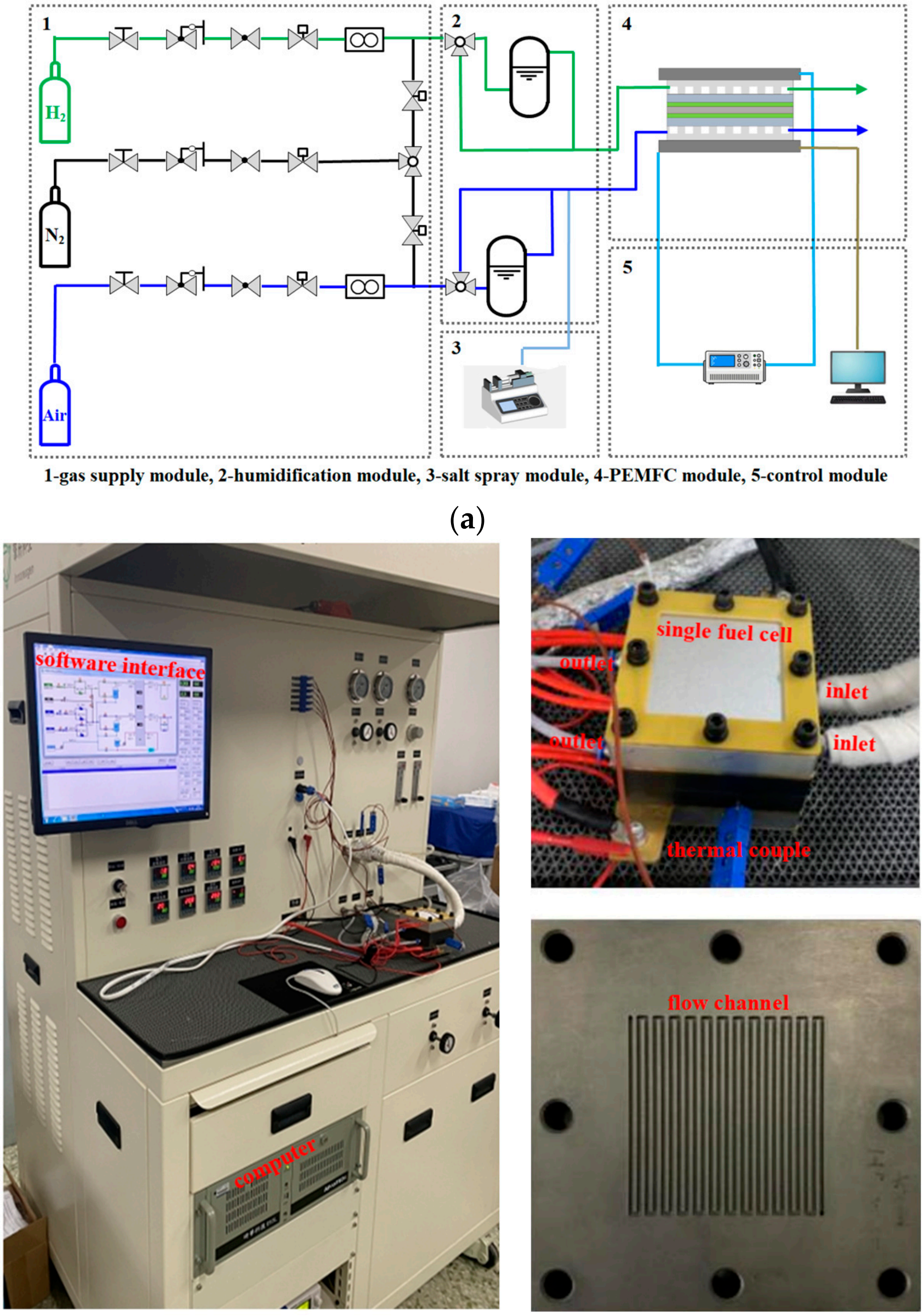
2. Experiments

In this study, experimental measurements were carried out to study the performance of PEMFCs with ion contamination. Cell performance was evaluated by the polarization curve, which can be obtained by a fuel cell test system. A diagram of the PEMFC test system is shown in Figure 1a. The fuel cell test system consisted of five modules. The gas supply module allowed for precise regulation of hydrogen and air flow into the cell. The gases could be humidified with deionized water in the humidification module prior to entering the cell. In the salt spray module, the solution was mixed with air and introduced into the cathode of the fuel cell. The solution flow rate was controlled by a micropump (SPLab01) manufactured by Baoding Shencheng Pump Industry (Baoding, China). A single fuel cell was used in the PEMFC module, and the fuel cell test system was governed by the control module. The test platform (QYC-A200) was manufactured by Dalian Innoreagen Technology Co., Ltd. (Dalian, China). An electronic load (N62401) manufactured by Hunan Next Generation Instrumental T&C Tech. Co., Ltd (Changsha, China). was used to obtain the polarization curve. The corresponding experimental setup used in this work is shown in Figure 1b. The operating parameters, including the pressure, temperature, mass flow rate, and so on, could be entered directly into the control software.

The single fuel cell and the flow field plate are shown in Figure 1c. The cross-sectional area of the channel in the graphite plate was 1 mm × 1 mm. The single fuel cell consisted of several components, including the flow field plate, a current collector plate, an MEA, an insulating gasket, an end plate, bolts, and nuts [32]. Eight bolts and nuts were used to assemble the various components. The MEA was the core component of the fuel cell, which consisted of five parts: the anode/cathode gas diffusion layer (GDL), the anode/cathode catalyst layer (CL), and the proton exchange membrane (PEM). The parameters of the MEA are summarized in Table 1. The effective active area of the MEA was 25 cm^2, and a GORE membrane with a thickness of 12 μm was used. The anode's Pt loading was 0.1 mg/cm^2, while the cathode's Pt loading was 0.4 mg/cm^2. The thickness of the anode/cathode GDL was 168 μm. The fuel cell's operating conditions are presented in Table 2. The stoichiometric ratios of the anode and cathode's reactants were 1.5 and 2.0, respectively. The operating temperature was 80 °C, and the back pressure was 1.0 bar during the experiments.

Table 1. MEA parameters.

Parameter	Value	Unit
Effective surface area	25	cm^2
Thickness of PEM	12	μm
Anode/cathode Pt loading	0.1/0.4	mg/cm^2
Thickness of GDL	168	μm



(**a**)

(**b**) (**c**)

Figure 1. (**a**) A diagram of the PEMFC test system, (**b**) a photo of the experimental setup, and (**c**) the single fuel cell and the flow field plate.

Table 2. Fuel cell's operating conditions.

Parameter	Value	Unit
Anode/cathode stoichiometric ratio	1.5/2	
Anode/cathode relative humidity	100%	
Anode/cathode humidification temperature	80	°C
Cell temperature	80	°C
Back pressure	1	bar

3. Experimental Procedures

The experimental procedures are presented in this section. The above-mentioned test platform was used to measure the polarization curves of fuel cells. The operating parameters summarized in Table 2 were first entered into the software in the computer. When the fuel cell was in a steady state, the current density was varied from 0 to 1.5 A/cm^2. The platform automatically obtained the corresponding cell voltages. This process was repeated three times to obtain an average value. A description of the test platform can also be found in our previous paper [32].

Park et al. [33] experimentally investigated the NaCl poisoning mechanism in PEMFCs. In their study, a 0.5 M NaCl solution was injected into a fuel cell for 60 min. Matsuoka et al. [28] measured the cell performance when a HNO_3 solution was introduced into fuel cells. It was reported that NO_3^- showed no effect on the performance of PEMFCs, and no changes in the MEA were observed. Therefore, solutions of KNO_3, $NaNO_3$, $Mg(NO_3)_2 \bullet 6H_2O$, and $Ca(NO_3)_2 \bullet 4H_2O$ were prepared using deionized water in this study to examine the effects of cations on cell performance. In addition, the effect of the solution concentration (0.25 M and 0.5 M) was investigated. The solutions were injected into the cathode of the fuel cell by a micropump at a rate of 0.8 mL/min for 2 h. Subsequently, polarization curves were measured to assess the effects of K^+, Na^+, Mg^{2+}, and Ca^{2+} on cell performance. Meanwhile, the corresponding polarization curves of fuel cells with NaCl contamination were also analyzed.

4. Results and Discussion

Solutions of K^+, Na^+, Mg^{2+}, and Ca^{2+} were mixed with air and introduced into the cathode of the PEMFC. The polarization and power density curves of fuel cells were analyzed after contamination to examine the effect of foreign cation contamination.

The effect of monovalent cations (Na^+ and K^+) on cell performance is presented in this section. The effect of K^+ contamination on cell performance in terms of the polarization curve and power density curve is shown in Figure 2. It was observed that cell performance could be greatly affected by increasing the K^+ concentration and current density. When the fuel cell was not contaminated, the voltage was 0.645 V at a current density of 1 A/cm^2. When the K^+ concentration was 0.25 M, the cell voltage was 0.628 V. When the K^+ concentration was 0.5 M, the cell voltage was 0.594 V. Compared to the fuel cell without contamination, the cell voltages decreased by 17 mV and 51 mV, respectively. Meanwhile, the corresponding power densities decreased by 2.636% and 7.906%, respectively. The polarization and power density curves of fuel cells with Na^+ contamination are demonstrated in Figure 3. When the Na^+ concentrations were 0.25 M and 0.5 M, the corresponding cell voltages were 0.616 V and 0.583 V, respectively. Compared to the fuel cell without contamination, the cell voltages decreased by 29 mV and 62 mV, respectively. The corresponding power densities decreased by 4.496% and 9.612%, respectively. The protons generated in the cathode CL were transported to the anode CL through the membrane. The ohmic loss of the fuel cell was mainly caused by the proton transport in the membrane. Foreign cations have a higher affinity for the sulfonic acid group than for H^+ protons. When the fuel cells were contaminated by foreign cations, the protonic conductivity was reduced. The ohmic loss accordingly increased, and cell performance degradation was also observed.

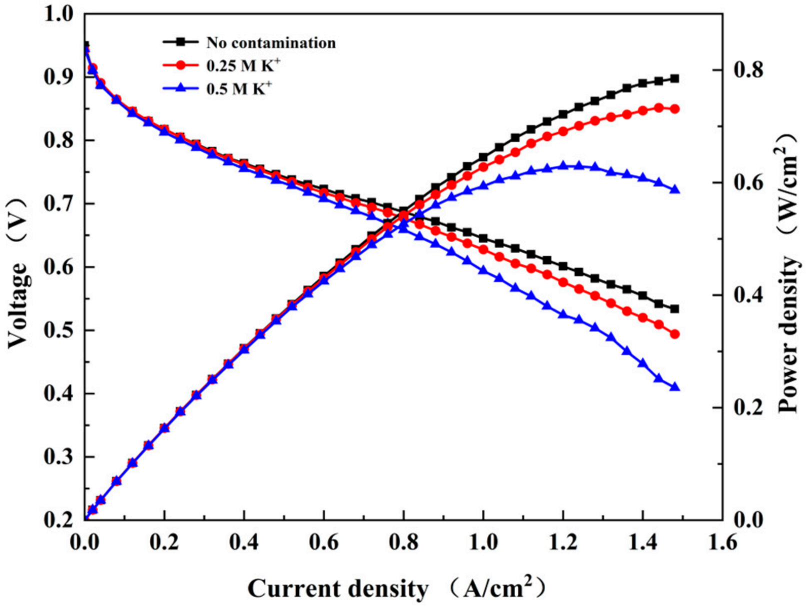


Figure 2. Effect of K^+ contamination on cell performance.

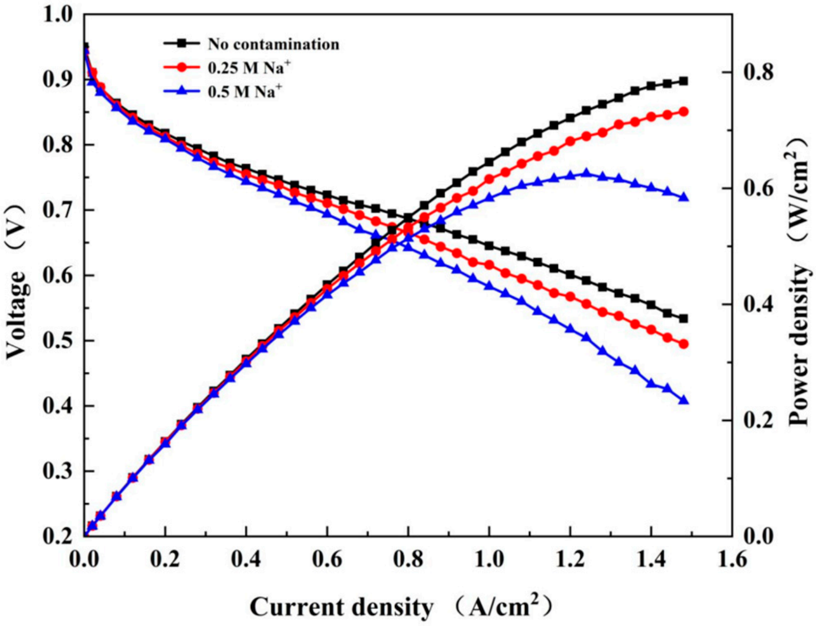


Figure 3. Effect of Na^+ contamination on cell performance.

The effect of divalent cations (Ca^{2+} and Mg^{2+}) on cell performance is presented in this section. Figure 4 presents the performance of fuel cells with Mg^{2+} contamination. When the Mg^{2+} concentrations were 0.25 M and 0.5 M, the corresponding cell voltages were 0.594 V and 0.559 V, respectively. Compared to the fuel cell without contamination, the cell voltages decreased by 51 mV and 86 mV, respectively. The corresponding power densities decreased by 8.062% and 13.333%, respectively. The performance of fuel cells with Ca^{2+} contamination is illustrated in Figure 5. When the Ca^{2+} concentrations were 0.25 M and 0.5 M, the cell voltages were 0.560 V and 0.300 V, respectively. Compared to the fuel cell without contamination, the cell voltages decreased by 85 mV and 345 mV, respectively. The corresponding power densities decreased by 13.137% and 53.488%, respectively.

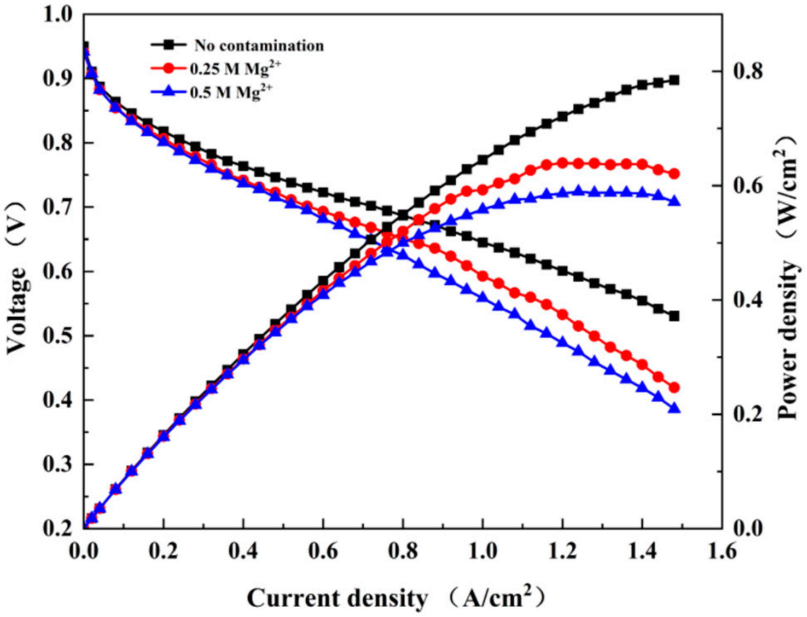


Figure 4. Effect of Mg^{2+} contamination on cell performance.

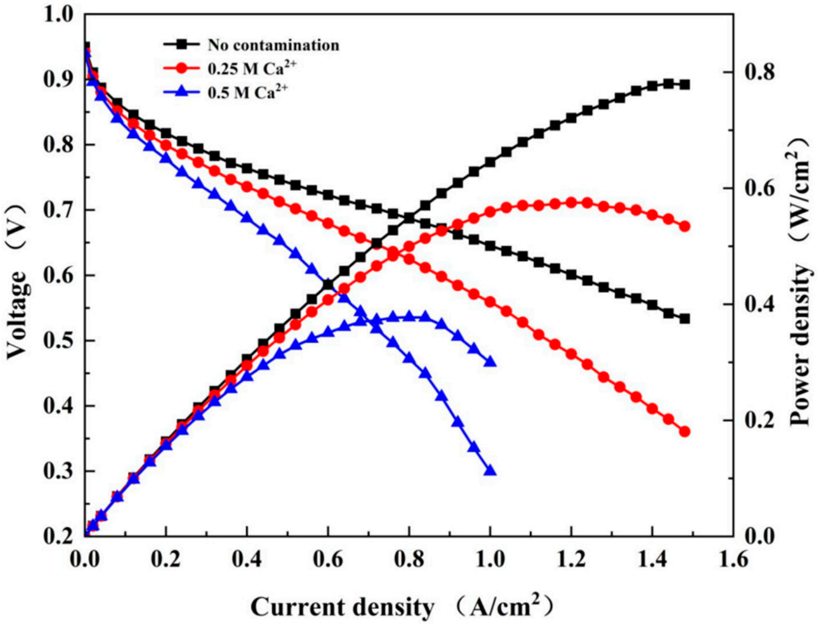


Figure 5. Effect of Ca^{2+} contamination on cell performance.

Meanwhile, the effect of the cation contamination type on cell performance was also investigated. The corresponding polarization and power density curves were summarized and compared, as illustrated in Figures 6 and 7. The cation concentrations were 0.25 M and 0.5 M, respectively. It was observed that cell performance was significantly affected by the cation type. The worst cell performance was observed when the fuel cell was contaminated by Ca^{2+}, followed by Mg^{2+}, Na^{+}, and K^{+}. It was also noticed that the effect of Ca^{2+} on cell performance became more pronounced when the cation concentration increased. The degradation of fuel cell performance caused by contamination by divalent cations (Ca^{2+} and Mg^{2+}) was more severe than that caused by contamination by monovalent cations (Na^{+} and K^{+}). The existence of cations can reduce proton conductivity, increase electro-osmotic drag, and reduce the water content [34].

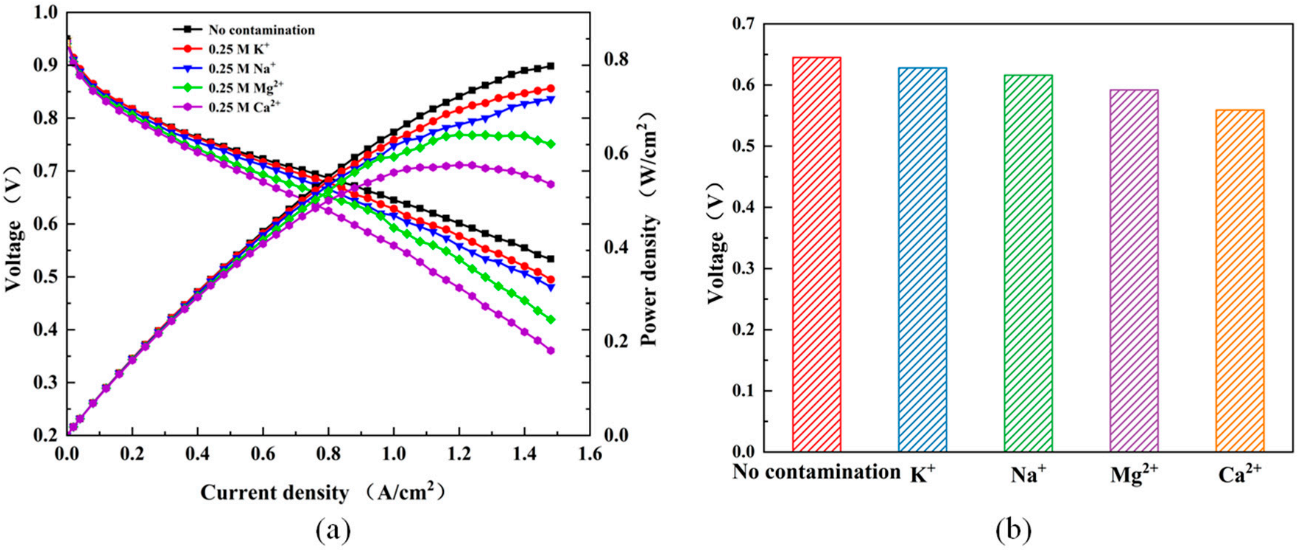


Figure 6. A comparison of fuel cell performance when the cation concentration was 0.25 M: (**a**) polarization curves and power density curves, (**b**) the cell voltage at a current density of 1.0 A/cm^2.

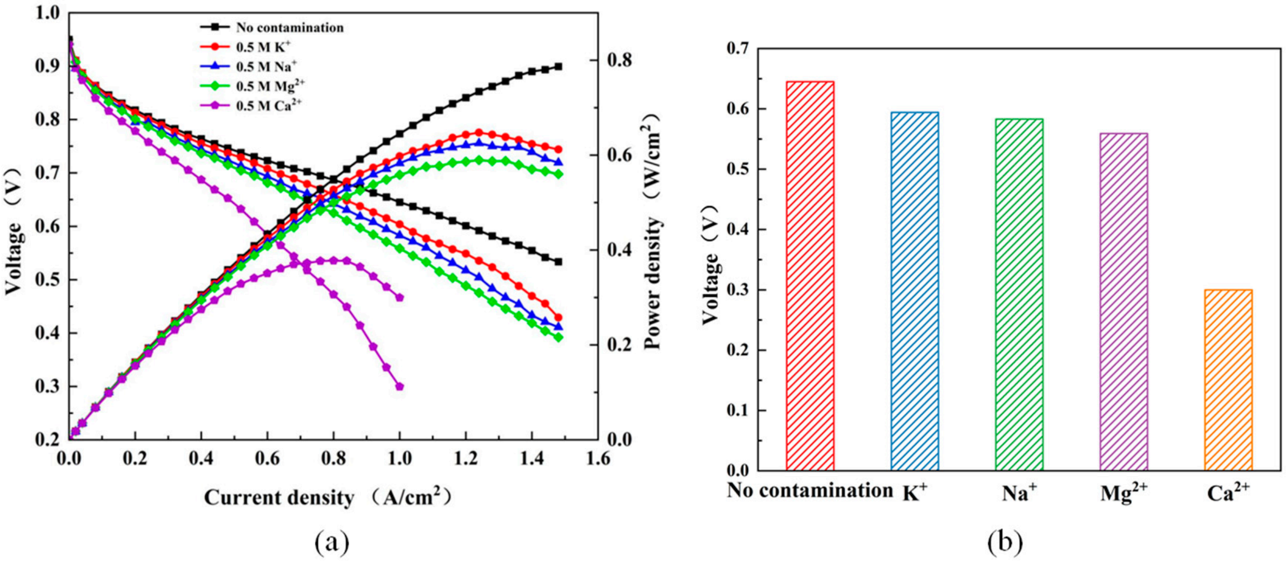


Figure 7. A comparison of fuel cell performance when the cation concentration was 0.5 M: (**a**) polarization curves and power density curves, (**b**) the cell voltage at a current density of 1.0 A/cm^2.

NaCl is the main component of seawater, so the effect of NaCl contamination on cell performance was also investigated. As shown in Figure 8, cell performance degradation was observed when the fuel cell was contaminated by NaCl. The degree of performance degradation increased with an increase in the NaCl concentration. When the NaCl concentrations were 0.25 M and 0.5 M, the corresponding cell voltages were 0.599 V and 0.477 V, respectively. Compared to the fuel cell without contamination, the cell voltages decreased by 46 mV and 168 mV, respectively. The corresponding power densities decreased by 7.132% and 26.047%, respectively.

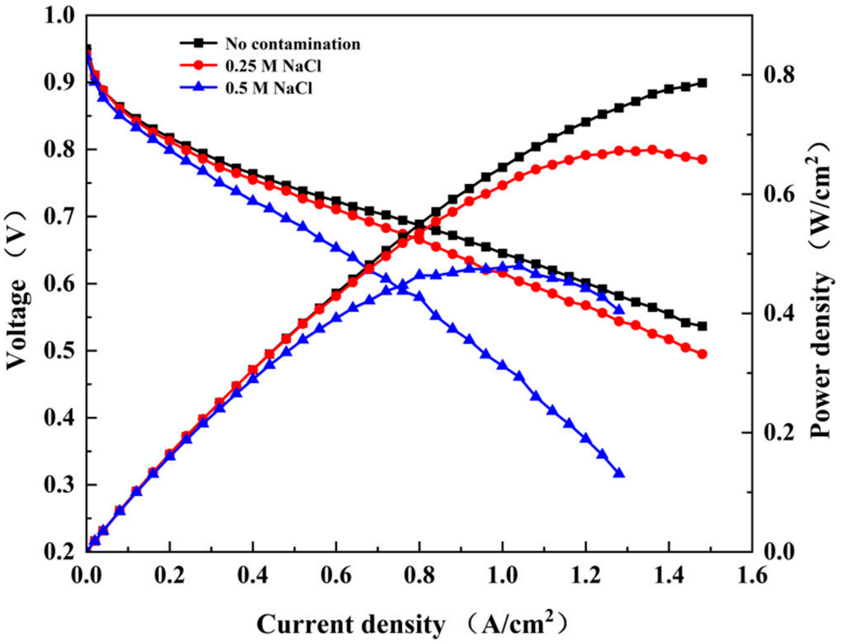
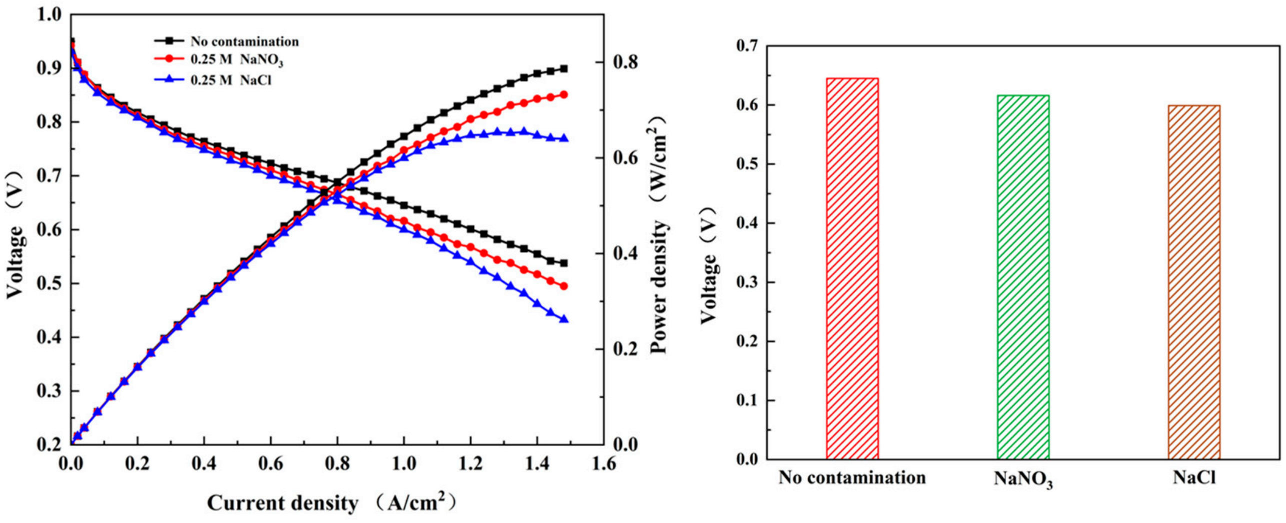


Figure 8. Effect of NaCl contamination on cell performance.

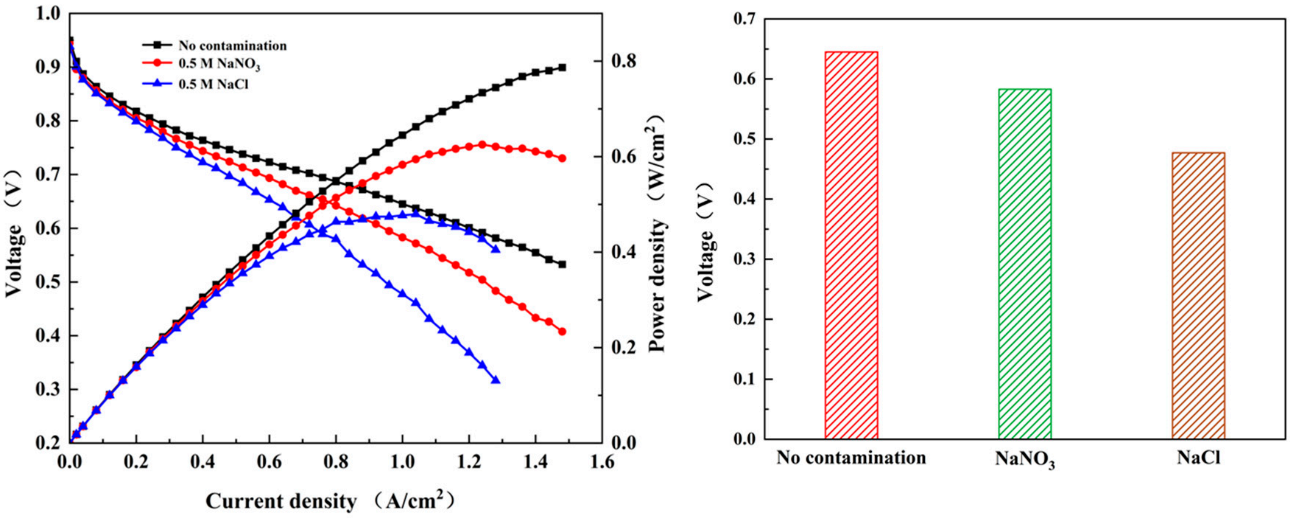
Fuel cells contaminated by 0.25 M and 0.5 M $NaNO_3$ and NaCl are compared in Figures 9 and 10. Compared to the fuel cell without contamination, the voltages of fuel cells contaminated by 0.25 M $NaNO_3$ and NaCl were reduced by 29 mV and 45 mV, respectively. When the concentration was increased from 0.25 M to 0.5 M, the corresponding voltages were reduced by 62 mV and 168 mV, respectively. It can be seen that the performance of the fuel cell contaminated by NaCl was much worse than that of the fuel cell contaminated by $NaNO_3$. Due to the existence of Cl^-, more severe performance degradation was observed. This was because Cl^- was easily adsorbed on the surface of the Pt catalyst and the active sites for the electrochemical reactions were reduced. Meanwhile, Cl^- could also induce dissolution of the Pt catalyst [33].



(a)

(b)

Figure 9. The performance of fuel cells contaminated by 0.25 M $NaNO_3$ and NaCl: (**a**) polarization curves and power density curves, (**b**) the cell voltage at a current density of 1.0 A/cm^2.



(a) (b)

Figure 10. The performance of fuel cells contaminated by 0.5 M $NaNO_3$ and NaCl: (**a**) polarization curves and power density curves, (**b**) the cell voltage at a current density of 1.0 A/cm^2.

5. Conclusions

In this study, experiments were carried out to examine the effect of ion contamination on PEMFC performance. The corresponding polarization and power density curves were presented and compared. The following conclusions were reached:

Cell performance can be significantly affected by the cation type and the concentration. The worst cell performance was observed when the fuel cell was contaminated by Ca^{2+}, followed by Mg^{2+}, Na^+, and K^+. The degradation of fuel cell performance caused by contamination by divalent cations (Ca^{2+} and Mg^{2+}) was more severe than that caused by contamination by monovalent cations (Na^+ and K^+). When the concentration increased, cell performance deteriorated more severely. The proton transport in the membrane was affected because cations have a higher affinity for the sulfonic acid group than for H^+ protons. The proton conductivity, electro-osmotic drag, and water content in the membrane were accordingly influenced. Fuel cells contaminated by $NaNO_3$ and NaCl were also compared. Due to the existence of Cl^-, more severe performance degradation was observed when the fuel cell was contaminated by NaCl. The active sites for the electrochemical reactions were reduced because Cl^- was easily adsorbed on the surface of the Pt catalyst. Meanwhile, Cl^- could also induce dissolution of the Pt catalyst. The obtained results can improve our understanding of the performance of fuel cells in marine salt spray environments.

Only a single fuel cell was investigated in this study, and a long-term test could be carried out on a fuel cell stack in the future. Experimental measurements could also be performed in a real marine environment. Meanwhile, we should pay more attention to research on fuel cell design, filter design, and recovery strategies.

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