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# Green Energy for Autonomous Devices: Surfactant-Enhanced Membraneless Hydrogen Peroxide Fuel Cells*

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***Abstract*— The amphipathic properties of surfactant macromolecules, characterized by their hydrophilic and hydrophobic terminal groups, are ubiquitously used to modulate the surface tension of interfaces and clean surfaces. Previously, the crucial role of surfactants was reported for the performance of catalytic nano/- and micromotors. In this study, we extrapolate this concept to the membraneless hydrogen peroxide ($H_2O_2$) Fuel Cells (FCs), which exhibit performance dependence on the addition of different types of surfactants: cationic - Sodium Dodecyl Sulfate (SDS), anionic - Benzalkonium Chloride (BACl), and nonionic - Tween20. Among them, charged SDS and BACl show promotion and inhibition towards fuel cell performance, respectively. Non-charged Tween20 shows a neutral effect on fuel cell performance.**

## I. INTRODUCTION

The increased urgency for adopting clean energy sources is underscored by ongoing efforts to mitigate the usage of fossil fuels, reduce the global carbon footprint, tackle the problem of global warming, bridge the widening energy gap, and curtail severe air pollution. Hydrogen peroxide ($H_2O_2$), boasting an impressive energy density of 3.0 MJ $l^{-1}$ or 2.1 MJ $kg^{-1}$ for a 60% aqueous solution, stands as a competitive alternative to hydrogen (2.8 MJ $l^{-1}$, 3.5 MJ $kg^{-1}$ at 35 MPa) [1]. The membraneless, single-compartment configuration of $H_2O_2$ Fuel Cells (FCs) presents a simpler, cost-efficient advantage, bypassing the need for an electrolytic membrane (e.g., Nafion). Recent designs featuring three-dimensional electrodes in $H_2O_2$ FCs have accomplished a maximum areal power density of up to 300 W $m^{-2}$ [2]. Despite these advancements, the development of $H_2O_2$ FCs has encountered a bottleneck – the challenge of achieving technologically significant power outputs. This bottleneck can be attributed to the practical open circle potential (OCP) of membraneless $H_2O_2$ FCs being less than the theoretical value (1.09 V), stemming from simultaneous reduction and oxidation reactions of $H_2O_2$ on the cathode, leading to a mixed potential. Potential solutions may lie in developing a more selective electrocatalytic anode and cathode for $H_2O_2$ electrooxidation and electroreduction respectively, facilitating fuel-oxidant separation using laminar co-flows, ensuring prompt delivery and removal of fuel/oxidant and reaction products, implementing redox couples, and applying high-surface area electrodes. A significant impediment in the progress of this approach is attributed to the energy-intensive requirement of fuel/oxidant pumping through high-surface-area electrodes, which is exacerbated by the elevated viscosity of laminar fluids. Addressing this issue, Mallouk and Sen introduced an innovative solution utilizing bimetallic catalytic nano- motors and pumps, harnessing the decomposition of $H_2O_2$ [3]. The mechanism involves the combination of protons and electrons with $H_2O_2$, resulting in the production of two $H_2O$ molecules at the Au surface. Concurrently, the Pt surface undergoes $H_2O_2$ oxidation, leading to proton generation and subsequent transformation into $O_2$. The Pt and Au surfaces function as the cathode and anode, respectively, with the former facilitating $H_2O_2$ reduction and proton consumption and contributing to proton generation. These protons are then shuttled to the Pt surface (cathode) for utilization. Solovev and colleagues presented an efficient, miniaturized micro- motors and pump powered by the recoil of oxygen microbubbles [4]. The design employs optimal physicochemical properties such as reaction-diffusion phenomena, surfactants, and a specifically architected catalytic micropump, thus contributing to the advancement of this scientific domain. Progressing this concept further, the Hess group demonstrated the integration of self-powered catalytic pumps into a porous, membraneless $H_2O_2$ FC [5]. However, the research indicated that bubble nucleation on the electrode, particularly within ceramic membrane pores, at elevated concentrations of $H_2O_2$ adversely affected the FC's performance. Whitesides' group [6] demonstrated the concept of a membraneless laminar flow–based fuel cell (LFFC) operating at low Reynold numbers. In their design, fuel and oxidant are separated by the interface of laminar flow, eliminating the use of a proton exchange membrane (PEM), simplifying the fuel cell architecture. However, the delivery of laminar fuels/oxidants requires high pumping pressure, leading to increased energy cost.

Hydrogen peroxide ($H_2O_2$) has recently emerged as a focus of intense scientific interest due to its unique, environmentally friendly attributes.[7-9] The decomposition of $H_2O_2$ yields

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water ($H_2O$) and oxygen ($O_2$), aligning with sustainability goals. Furthermore, its remarkable disproportionation reaction [10] enables $H_2O_2$ to act as fuel and oxidant, obviating the need for a Proton Exchange Membrane (PEM) and laminar flow conditions. In a seminal work, Yamasaki *et al.* [8] elucidated the function of their Membraneless $H_2O_2$ Fuel Cells (MHPFCs) demonstrated that the electrooxidation and electroreduction reactions selectively localized at the cathode and anode, respectively. Since then, numerous subsequent studies [11-14] have been conducted to enhance the performance of MHPFCs. The specific reactions governing the operation of these FCs can be represented as follows (E vs. SHE) [15, 16]:

Reactions in the acidic media:

$$\text{Anode: } H_2O_2 \rightarrow O_2 + 2H^+ + 2e^-;\ E_{a1} = 0.68\ \text{V} \quad (1)$$

$$\text{Cathode: } H_2O_2 + 2H^+ + 2e^- \rightarrow 2H_2O;\ E_{a2} = 1.77 \quad (2)$$

$$\text{Total: } 2H_2O_2 \rightarrow 2H_2O + O_2 \quad (3)$$

Reactions in the basic media:

$$H_2O_2 + OH^- \rightarrow HO_2^- + H_2O$$

$$\text{Anode: } HO_2^- + OH^- \rightarrow O_2 + H_2O + 2e^-;\ E_{b1} = 0.15 \quad (4)$$

$$\text{Cathode: } HO_2^- + H_2O + 2e^- \rightarrow 3OH^-;\ E_{b2} = 0.87\ \text{V} \quad (5)$$

$$\text{Total: } 2H_2O_2 \rightarrow 2H_2O + O_2$$

It is known that the theoretical open circuit potential (OCP) of MHPFC is 1.09 V (in acidic media) and 0.62 V (in basic media) [15, 16]. However, experimental results show far below OCPs due to the limited selectivity of electrodes and self-decomposition of $H_2O_2$ on electrodes [15,17]. And power output of MHPFC is not enough to satisfy practical applications.

In this study, we explored the influence of surfactants in modulating the performance of the MHPFC. The Ni foam and Au electroplated carbon fiber cloth showed excellent selectivity and catalytic performance towards the ERR and EOR, respectively. A comprehensive analysis has been performed using anionic (SDS), cationic (BACl), and nonionic (Tween20) surfactants.

## II. Materials and Methods

This section introduces the primary materials, reagents, and methods for installing the MHPFC system. In subsection A, all materials and reagents are listed in detail. Procedures for obtaining electrodes of MHPFC are included in subsection B. In subsection C, the structural characterization of electrodes is illustrated. In the last subsection, MHPFC performance characterization is shown.

### *A. Materials and Reagents*

Commercially available Nickel-foam and carbon fiber cloth, with dimensions of 25mm x 25mm, were utilized. A Gold Chloride Trihydrate ($HAuCl_4$) solution facilitated the electroplating of gold onto carbon fiber cloth, creating a high surface area electrode. The electrolyte comprised $H_2O_2$, surfactants (SDS, BACl, and Tween20), and pH regulators (NaOH, HCl). Experimental parameters involved setting $H_2O_2$ concentration at 1M and varying surfactants' concentrations at 0 mM, 0.1 mM, 1 mM, 10 mM, and 100 mM.

### *B. Electrodes Fabrication Procedure*

A facile electroplating method was adopted to fabricate a three-dimensional, high-surface area cathode. The carbon fiber cloth was pre-processed by sequentially rinsed with acetone, alcohol, and deionized water thrice. The Electrochemical Station (ECS, Shanghai Chenhua Instrument Co., Ltd., CHI600E) was used as an electroplating device with three electrodes method: carbon fiber cloth as the Working Electrode (WE), graphite rod as the Counter Electrode (CE), and Ag/AgCl (saturated 1 M KCl solution) as the Reference Electrode (RE). The electroplating parameters were established using Multi-Potential Steps: $E_{work}$: 0.3 V, 0.2 s; $E_{rest}$: 0.1 V,0.1 s. The rest period was set for the uniform diffusion of $Au^{3+}$ in diluted $HAuCl_4$ solutions. A circle consisted of one work and rest period with 1600 circles (*Fig. 1*).

### *C. Structural Characterization of Electrodes*

To characterize the performance of Au electroplated carbon fiber cloth, X-Ray Diffraction (XRD, Bruker Corporation, D8 Advance) and Scanning Electron Microscopy (SEM, Carl Zeiss AG, EVO 10) were used. XRD was taken to confirm Au on carbon fiber cloth. SEM was conducted to characterize distributions of Au nanoparticles (NP).

### *D. Performance Characterization of MHPFCs*

Performance characterizations were conducted using an electrochemical station. TMHPFC performance was evaluated under varying solution conditions established across multiple experimental groups. The first group explored the effects of proton/hydroxyl ion $H^+/OH^-$ concentration (pH=1, 3, 5, 7, 9, 11, 13) on MHPFC performance. The second group was designed to analyze the type of surfactant (cationic, anionic, and nonionic) on MHPFC performance. The final group aimed to elucidate the influence of specific surfactant concentrations on MHPFC performance.

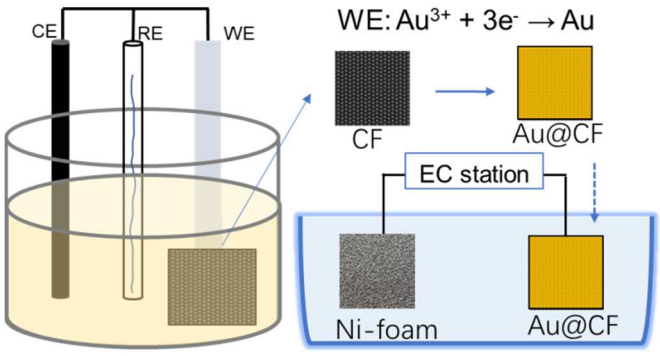


Figure 1. Electroplating method for fabricating Au@CF cathode of MHPFCs. Electroplating electrolyte consists of 5 mM $HAuCl_4$, 0.1M HCl and 0.1 M KCl.

## III. Results

*Fig. 2* shows the structural characterization results of XRD and SEM images. In *Fig. 2(a)*, XRD results indicate clear peaks at 38.19°, 44.39°, 64.58° and 77.60° corresponding to the (111), (200), (220), and (311) crystal planes of face-centered cubic (fcc) Au. SEM images shown in *Fig. 2(b-c)* have seen detectable Au nanoparticles grown and distributed on the carbon fiber cloth. All results have proved successfully electroplated onto the carbon fiber cloth.

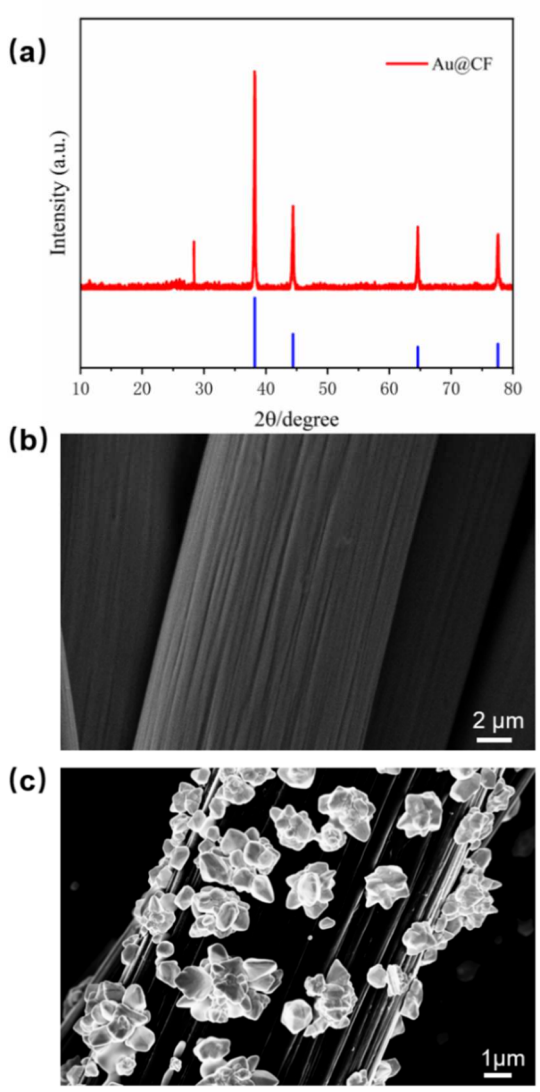


Figure 2. structural characterization of Au@CF. (a) XRD result of Au@CF, (b) SEM images of CF after pre-process, (c) SEM image of Au@CF distributed with Au nano particles.

Furthermore, the optimization of pH conditions is critical for the stability and efficacy of the H2O2 decomposition reaction. Based on reactions (1)(2) and (4)(5), it can be inferred that both $H^+/OH^-$ ions can act as catalysts to enhance the reaction rate of $H_2O_2$ electrochemical reactions. Experimental results, as shown in *Fig. 3*, validate the hypothesis that the MHPFC performance is superior in acidic (pH = 1) and basic (pH = 13) solutions compared to intermediate pH levels. The electrochemical activity at distinct pH levels, as revealed by cyclic voltammetry, underscores the distinct catalytic pathways these ions undertake. Under pH = 13 conditions, the Open Circuit Potential (OCP) and Maximum Current Density (MCD) are lower than under pH = 1 conditions. This observation is attributed to the intense self-decomposition process of $H_2O_2$, as observed in our experiments.

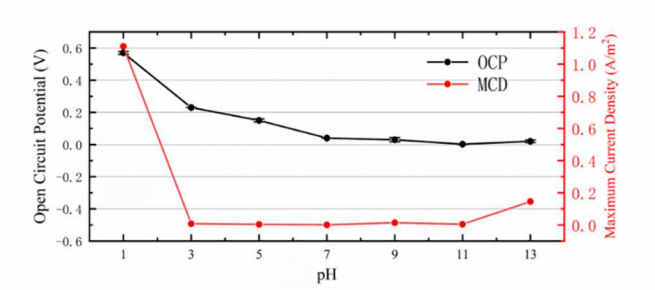


Figure 3. OCP and MCD of MHPFC under different pH in 1M $H_2O_2$ solutions

The results demonstrate a significant influence of surfactants presence in solution on MHPFCs performance seen in *Fig. 4*. The addition of SDS to the MHPFC electrolyte enhanced the performance, with the maximum power output being three times higher than in a non-SDS electrolyte, as shown in *Fig. 4(a)*. Conversely, including BACl, even in minute quantities (0.1 mM), decreased MHPFC performance, particularly in power output, with a continuous decrease observed with adding more BACl, as shown in *Fig. 4(b)*. The non-ionic surfactant Tween20, in *Fig. 4(c),* did not noticeably affect the MHPFC performance. Our preliminary hypotheses suggest that charged surfactants can accumulate at the interface between the electrolyte and electrode and modify interactions between $H_2O_2$ molecules and electrodes and facilitate the electron transfer in between.

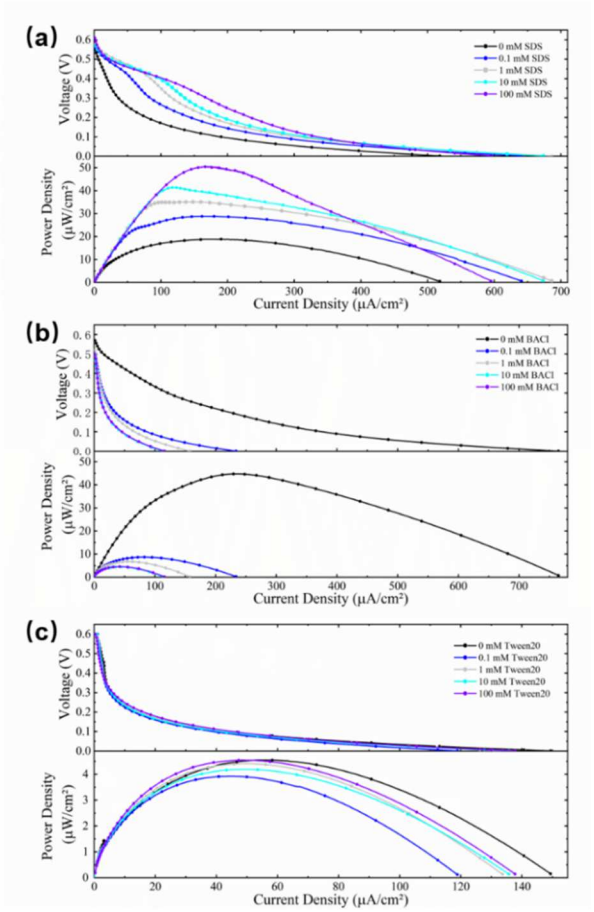


Figure 4. MHPFC performance. (a) OCP and MCD of MHPFC under different pH in 1M $H_2O_2$ solutions; polarization and power output curves of (A) SDS, (B) BACl, (C) Tween20 in 1 M $H_2O_2$, pH=1 solutions.

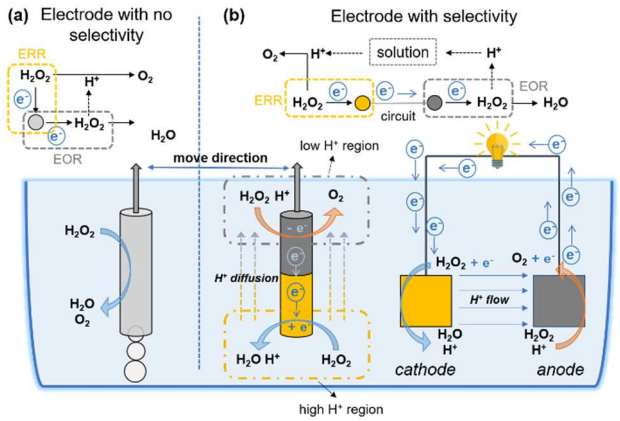


Figure 5. Micro- motors and fuel cell powered by hydrogen peroxide decomposition. (a) Schematic image of $H_2O_2$-powered tubular micromotor. (b) left: schematic of $H_2O_2$-powered motor driven by self-electrophoresis; right: schematic of $H_2O_2$-powered MHPFCs.

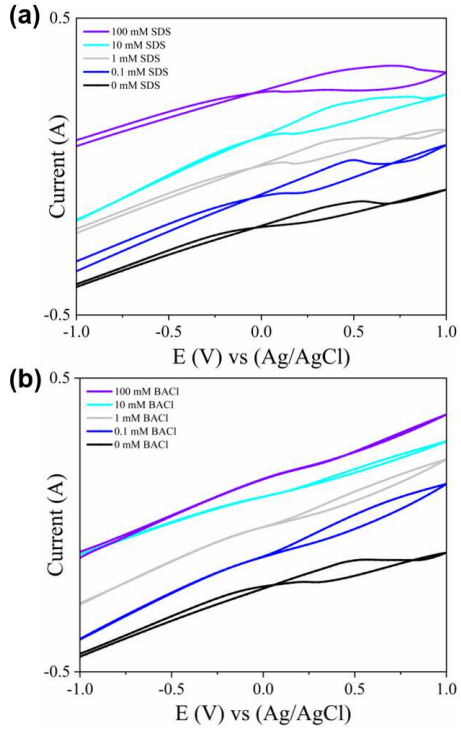


Figure 6. Cyclic voltammetry (CV) curve of Ni-foam using different concentrations of surfactants (SDS, BACl). The scan rate is 0.01 V/s.

## IV. Discussion

In 1 M aqueous solutions of $H_2O_2$ incorporating sodium dodecyl sulfate (SDS), a marginal increase in the open circuit potential (OCP) can be observed, from 0.55 V to 0.61 V (*Fig. 4*). Simultaneously, the maximum power output exhibited an increase to 50.6 μW cm$^{-2}$ in the presence of 100 mM SDS, which is nearly three-fold higher compared to the condition where SDS was not present (18.88 μW cm$^{-2}$). This outcome implies the potential for SDS to modulate the performance of MHPFCs effectively. Regarding the anionic surfactant BACl, a pronounced decrease in MHPFC performance was observed. In the absence of BACl, the maximum power output was recorded to be 44.76 μW cm$^{-2}$, whereas in the presence of only 0.1 mM BACl solutions, the power output decreased drastically to 8.69 μW cm$^{-2}$. This reduction in power output suggests a significant alteration of the interface electrochemical process, attributable to the introduction of BACl. Contrarily, the non-ionic surfactant Tween20 did not demonstrate any discernible influence on the performance of the MHPFC. The OCP and maximum power output measurements were within the range of statistical error, with an average maximum power output of 4.43 μW cm$^{-2}$ and a standard deviation of 0.42. Similarly, the OCP remained stable in the presence of Tween20, ranging between 0.52 V and 0.55 V, with an average OCP of 0.538 V and a standard deviation of 0.011. This observation suggests that Tween20 does not affect the performance of the MHPFC under the given experimental conditions.

Previously, a critical role of surfactants was reported for the operation of bubble-propelled catalytic micro/-nanomotors powered by the catalytic decomposition of $H_2O_2$.[18-20] Subsequent studies [21, 22] demonstrated the influence of surfactants on bubbles evolution (nucleation, generation, diameter, recoil frequency) with initial attempts to lower the surface tension and alter the Zeta potential (ZP) of surfaces. The $H_2O_2$ electrooxidation reaction (EOR) and electroreduction reaction (ERR) can occur at the same active point of non-selective catalyst material, shown in *Fig. 5(a)*. *Fig. 5(b)* illustrates the mechanism of using selective electrocatalysts, which separate EOR and ERR at two different materials, enabling the motion of micromotors [23] and MHPFCs [11]. One main difference between catalytic nano/-micromotors and fuel cells: anode and cathode are connected in NMs, while anode and cathode are separated in MHPFCs. Our experiments, use MHPFC commercially available Ni-foam as an anode and Au electroplated carbon fiber cloth (Au@CF) as a cathode. By controlling the amounts of surfactants added to the electrolyte, we explored how the concentration and types of surfactants influence MHPFCs performance. MHPFCs that utilized the cationic surfactant SDS demonstrated a threefold increase in maximum power output, compared to those under non-surfactant conditions, as shown in *Fig. 4(a)*. Anionic BACl resulted in a sharp decrease in power output, and nonionic Tween20 had a neutral effect on MHPFC performance, as depicted in *Fig. 4(b-c)*. Surfactants play a complex and multifaceted role in modulating the performance of MHPFCs.

Here we proposed a potential explanation of surfactants impact on fuel cells. According to research [24, 25], $Ni(OH)_2$ groups at the surface of Ni anode play a vital role in the $H_2O_2$ oxidation reaction. Besides, cyclic voltammetry (CV) results [24] indicate that the oxidation and reduction window of the $Ni(OH)_2$ group is around 0.2 V to 0.5 V (vs. Ag/AgCl). *Fig. 6* shows CV results towards the Ni electrode. It is clearly observed that with the increasing concentration of SDS, the characteristic peak of $Ni(OH)_2$ appears, as shown in *Fig. 6(a)*. In contrast, with the increasing concentration of BACl, the characteristic peak of $Ni(OH)_2$ disappears, as shown in *Fig. 6(b)*. We assume that surfactants can protect/inhibit specific groups at the electrode surface, thus changing the overall electrochemical reaction process. However, surfactants may have other significant roles in the electrochemical process. In the context of MHPFCs, the anionic SDS appears to enhance FC's performance. SDS molecules can be adsorbed on the

surface of the electrode, altering its physicochemical properties such as wettability and distribution of charge. Subsequently, it can impact the electrode-electrolyte interface, potentially facilitating the electrochemical reactions with $H_2O_2$, resulting in the power output change. Conversely, the cationic BACl surfactant negatively affects the MHPFC performance. BACl, being positively charged, can interfere with the electrostatic interactions at the electrode-electrolyte interface, possibly hindering the necessary reactions for power generation. Additionally, the surfactant can also induce an unfavorable modification of the electrode surface, potentially by adsorption, leading to a decrease in the power output. The non-ionic surfactant Tween20 does not appear to impact MHPFC performance significantly. Due to its neutral charge, Tween20 likely does not induce notable electrostatic changes at the electrode-electrolyte interface, nor does it alter the surface properties of the electrode. Thus, it is plausible that Tween20 presence does not markedly influence the electrochemical reactions involving $H_2O_2$.

Overall, our preliminary hypotheses underscore the significance of surfactants in manipulating MHPFC performance, likely through altering interfacial interactions and modulating the physicochemical properties of the electrode. Further rigorous investigations are essential to establish the exact mechanisms of surfactant action in MHPFCs unequivocally. However, it should be emphasized that the precise effects of surfactants on MHPFCs are likely to be specific to the surfactant structure, the fuel cell architecture, and the nature of the electrodes and electrolytes. Furthermore, the impact of surfactants on MHPFC performance is likely not solely due to their direct interactions at the electrode-electrolyte interface, but also their overall effect on the physicochemical properties of the system, including potential changes in the solution's viscosity, conductivity, and related parameters. While this discussion provides a feasible explanation for the observed behaviors, these are, as noted, our preliminary hypotheses. Further research is required to validate the role of surfactants and to elucidate their underlying mechanism of action, such as surface characterization of electrodes, investigation of reaction kinetics, and computational modeling to provide a more holistic understanding of the phenomena.

## V. Conclusion

As the field of nanomotors has progressively delineated itself, surfactants have been ubiquitously employed to modulate surface tension and enhance the stability of microbubbles. Intriguingly, observations suggested that the specific category of surfactant exerts a discernible influence on the kinematics of catalytic nanomotors. However, a comprehensive exploration of this phenomenon remained pending. In juxtaposition, the incorporation of surfactants in fuel cell research remains conspicuously absent. Our results accentuate the merits of a synergistic and interdisciplinary research approach. The advancements in $H_2O_2$ fuel cell (FC) technology present an innovative conceptual framework, wherein $H_2O_2$ is concurrently utilized as both fuel and oxidant within a monolithic device architecture. Our empirical investigation aimed to systematically evaluate the potential of surfactants in the electrolyte for enhancing micro- $H_2O_2$ fuel cell (MHPFC) performance. Initial findings indicated that the anionic surfactant, sodium dodecyl sulfate (SDS), could indeed augment MHPFC performance. Nevertheless, the intricate mechanistic details underlying the function of surfactants in MHPFCs necessitate further intensive investigation. The acquired knowledge could then be broadly disseminated and applied across various FC technologies, employing surfactants as performance modulating agents. Theoretically, $H_2O_2$ FCs can achieve a maximal electrochemical potential of 1.09 V, establishing a competitive edge analogous to methanol/air FCs (1.21 V) and $H_2$/air FCs (1.23 V). Moreover, $H_2O_2$ exhibits a remarkable compatibility with a diverse array of fuels, encompassing ethanol, formic acid, specific metals (e.g., magnesium), and hydrazine. Furthermore, the integration of $H_2O_2$ with alternative fuels/oxidants or the utilization of redox couples could conceivably result in power densities on the order of kW $m^{-2}$.

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