

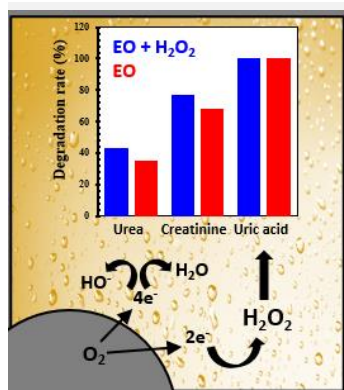
## Exploring the potential of *In Situ* electrogeneration of H<sub>2</sub>O<sub>2</sub> for advanced electro-sanitation practices

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This study investigated the impact of *in situ* hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) generation using gas diffusion electrodes (GDEs) synthesized from Printex-L6 carbon (CP-L6). As a strategy for electro-sanitation, the potential of H<sub>2</sub>O<sub>2</sub> was harnessed for the treatment of synthetic urine, specifically targeting the degradation of urea, creatinine, and uric acid. Various current densities were examined for H<sub>2</sub>O<sub>2</sub> electrogeneration in Na<sub>2</sub>SO<sub>4</sub>. In a specific condition, the amount of H<sub>2</sub>O<sub>2</sub> generated was compared with its generation in synthetic urine. Results indicated that the amount of H<sub>2</sub>O<sub>2</sub> is influenced by the compositional complexity of urine, potentially due to its consumption in degrading organic content. Among the components, the order of degradation was: uric acid > creatinine > urea. Partial mineralization exceeding 30.0% was primarily due to urea degradation. These findings highlight the crucial role of H<sub>2</sub>O<sub>2</sub> in wastewater treatment, suggesting that its application can increase treatment efficiency.

### Introduction

Human urine is a major water pollutant with a complex chemical composition, primarily consisting of urea, creatinine, and uric acid [1,2]. This complexity highlights the need for innovative technologies and catalytic materials capable of generating chemical oxidants to manage such challenging matrices. *In situ* generation of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) presents a promising solution, particularly for decentralized treatment systems, as it reduces issues related to storage, transport, and handling of the reagent. H<sub>2</sub>O<sub>2</sub> is a versatile oxidant that can be produced through the oxygen reduction reaction (ORR) via the 2-electron pathway [3]. Carbon-based materials, such as Printex-L6, are effective for facilitating selective reactions due to their large surface area and functional groups [4]. This study aimed to assess the effectiveness of H<sub>2</sub>O<sub>2</sub> electrogenerated using Printex-L6-based gas diffusion electrodes for the electrochemical treatment of synthetic urine, focusing on the degradation of urea, creatinine, and uric acid.

### Material and Methods

The synthetic urine matrix consisted of 0.0550 mol L<sup>-1</sup> urea, 0.0015 mol L<sup>-1</sup> creatinine, 0.0003 mol L<sup>-1</sup> uric acid, 0.1200 mol L<sup>-1</sup> NaCl, 0.0200 mol L<sup>-1</sup> Na<sub>2</sub>SO<sub>4</sub>, 0.0200 mol L<sup>-1</sup> Na<sub>2</sub>HPO<sub>4</sub>, and 0.0600 mol L<sup>-1</sup> KCl, with a TOC of 757.58 mg L<sup>-1</sup>, pH 5.10, and conductivity of 18.29 mS cm<sup>-1</sup>. The electrolytic system consisted of an undivided cylindrical glass cell containing an Ag/AgCl reference electrode, a dimensionally stable anode, and the GDE as the working electrode. The GDE was positioned at the base of the cell, where O<sub>2</sub> was injected directly at a flow rate of 0.05 L min<sup>-1</sup>. This electrode was developed following the standard employed by the research group [4,5]. The working volume was 250 mL. H<sub>2</sub>O<sub>2</sub> electrogeneration was analyzed at

different current densities (25-100 mA cm<sup>-2</sup>) using a Metrohm Autolab PGSTAT-302 N potentiostat, with Na<sub>2</sub>SO<sub>4</sub> as the electrolyte under the same ionic strength and pH conditions as the urine matrix, and quantified by UV-Vis absorption spectrophotometry using ammonium molybdate. All experiments were conducted in duplicate for 120 minutes under mechanical stirring and a controlled temperature of 20°C using a thermostatic bath. Energy consumption and current efficiency associated with H<sub>2</sub>O<sub>2</sub> quantification were the criteria for selecting the current density to compare H<sub>2</sub>O<sub>2</sub> generation in the urine matrix and investigate the parallel degradation of urea, creatinine, and uric acid. Urea concentration was monitored by a colorimetric method, where solutions containing urea develop a green-yellow coloration upon contact with a reagent composed of p-dimethylaminobenzaldehyde. High-performance liquid chromatography (HPLC-UV) was used to monitor the degradation of uric acid and creatinine with a Shimadzu 20A HPLC. Nitrogen species, active chlorine species, and the evolution of carboxylic acids were monitored by ion chromatography.

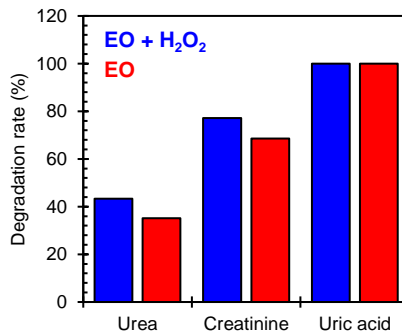
### Results and Discussion

The results obtained from H<sub>2</sub>O<sub>2</sub> electrogeneration in Na<sub>2</sub>SO<sub>4</sub> solutions, evaluated under conditions of similar pH and ionic strength as synthetic urine and at different current densities (25, 50, 75, and 100 mA cm<sup>-2</sup>), indicated a clear trend of increasing H<sub>2</sub>O<sub>2</sub> production with higher current densities. This observation is consistent with the literature and may be related to the increased electron flow available for the oxygen reduction reaction at the cathode. Correspondingly, energy consumption also increased with higher current density. However, current efficiency showed a decreasing trend with increasing current density, likely due to additional parasitic reactions such as oxygen evolution at the

anode competing with  $\text{H}_2\text{O}_2$  generation. Considering the results and viability factors, a current density of  $50 \text{ mA cm}^{-2}$  was selected for subsequent studies, where  $\text{H}_2\text{O}_2$  generation reached  $425.6 \text{ mg L}^{-1}$  in 120 min.

Under the same conditions,  $\text{H}_2\text{O}_2$  electrogeneration was analyzed in synthetic urine. The results revealed that the accumulated amount of  $\text{H}_2\text{O}_2$  in 120 minutes was  $396.0 \text{ mg L}^{-1}$ . This difference between  $\text{Na}_2\text{SO}_4$  and synthetic urine suggests that the  $\text{H}_2\text{O}_2$  generated in urine is being consumed in the degradation of organic compounds present, as degradation rates of 43.4%, 77.2%, and 100% were achieved for urea, creatinine, and uric acid, respectively (Figure 1). While these findings are crucial for assessing the efficiency of the electrogeneration process and its application in wastewater treatment, synthetic urine is a complex matrix. The presence of chloride in the solution and the use of a DSA anode lead to the formation of active chlorine species, which, like  $\text{H}_2\text{O}_2$ , can degrade organic compounds. To evaluate the potential of active chlorine species, the system was used without  $\text{H}_2\text{O}_2$  electrogeneration by injecting  $\text{N}_2$  instead of  $\text{O}_2$  into the GDE. In comparison, without  $\text{H}_2\text{O}_2$  generation, uric acid degradation remained at 100%, but the degradation rates of the other compounds were lower, with reductions of 35.0% for urea and 68.0% for creatinine. In absolute terms, this represents a significant reduction, demonstrating that in situ  $\text{H}_2\text{O}_2$  generation positively impacts the degradation of organic compounds in synthetic urine. These results indicate that the presence of  $\text{H}_2\text{O}_2$  contributes to an overall improvement in the treatment system's efficiency.  $\text{H}_2\text{O}_2$  generation appears to promote more effective degradation of

urea and creatinine, increasing the mineralization rate and reducing TOC more than the system without  $\text{H}_2\text{O}_2$ . The efficacy of  $\text{H}_2\text{O}_2$  in degrading organic compounds can be attributed to its potent oxidizing capacity, which can attack a wide range of organic molecules, facilitating their conversion into less complex and more easily removable products.



**Figure 1.** Difference in the rate of degradation of organic components of synthetic urine by electrochemical oxidation with and without hydrogen peroxide generated using a GDE based on CP-L6 in 120 min of electrolysis.

## Conclusions

This study demonstrates the effectiveness of in situ hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) electrogeneration using gas diffusion electrodes made from CP-L6 carbon for the electrochemical treatment of synthetic urine. An analysis of different current densities revealed a consistent increase in  $\text{H}_2\text{O}_2$  production with higher current densities, with the  $50 \text{ mA cm}^{-2}$  condition generating  $425.6 \text{ mg L}^{-1}$  of  $\text{H}_2\text{O}_2$  in 120 min. Comparing  $\text{Na}_2\text{SO}_4$  to synthetic urine showed that the complexity of urine leads to the consumption of part of the generated  $\text{H}_2\text{O}_2$  in the degradation of organic compounds, resulting in degradation rates of 43.4% for urea, 77.2% for creatinine, and 100% for uric acid. Without  $\text{H}_2\text{O}_2$  generation, degradation efficiency was lower, especially for urea and creatinine, resulting in reduced mineralization. The key findings of this study emphasize the crucial role of  $\text{H}_2\text{O}_2$  in the degradation of complex organic compounds in synthetic urine. The promising ability of  $\text{H}_2\text{O}_2$  to act as a powerful oxidizing agent suggests that its application could enhance the efficiency of water and wastewater treatment systems, particularly in electro-sanitation processes.

## Acknowledgments

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