

## Integrating Reactive Chlorine Species Generation with H<sub>2</sub> Evolution in a Multifunctional Photoelectrochemical System for Low Operational Carbon Emissions Saline Sewage Treatment

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**Abstract :** Organic pollutants, ammonia, and bacteria are major contaminants in sewage, which may adversely impact ecosystems without proper treatment. Conventional wastewater treatment plants (WWTPs) are operated to remove these contaminants from sewage but suffer from high carbon emissions and are powerless to remove emerging organic pollutants (EOPs). Herein, we have developed a low operational carbon emissions multifunctional photoelectrochemical (PEC) system for saline sewage treatment to simultaneously remove organic compounds, ammonia, and bacteria, coupled with H<sub>2</sub> evolution. A reduced BiVO<sub>4</sub> (r-BiVO<sub>4</sub>) with improved PEC properties due to the construction of oxygen vacancies and V<sup>4+</sup> species was developed for the multifunctional PEC system. The PEC/r-BiVO<sub>4</sub> process could treat saline sewage to meet local WWTPs' discharge standard in 40 minutes at 2.0 V vs. Ag/AgCl and completely degrade carbamazepine (one of the EOPs), coupled with significant evolution of H<sub>2</sub>. A remarkable reduction in operational carbon emissions was achieved by the PEC/r-BiVO<sub>4</sub> process compared with large-scale WWTPs, attributed to the restrained direct carbon emissions from the generation of greenhouse gases. Mechanistic investigation revealed that the PEC system could activate chloride ions in sewage to generate reactive chlorine species and facilitate •OH production, promoting contaminants removal. The PEC system exhibited operational feasibility at different pH and total suspended solids concentrations and has outstanding reusability and stability, confirming its promising practical potential. The study combined the simultaneous removal of three major contaminants from saline sewage and H<sub>2</sub> evolution in a single PEC process, demonstrating a viable approach to supplementing and extending the existing wastewater treatment technologies. The study generated profound insights into the in-situ activation of existing chloride ions in sewage for contaminants removal and offered fundamental theories for applying the PEC system in sewage remediation with low operational carbon emissions. The developed PEC system can fit well with the future needs of wastewater treatment because of the following features: (i) low operational carbon emissions, benefiting the carbon neutrality process; (ii) higher quality of the effluent due to the elimination of EOPs; (iii) chemical-free in the operation of sewage treatment; (iv) easy reuse and recycling without secondary pollution.

**Keywords :** contaminants removal, H<sub>2</sub> evolution, multifunctional PEC system, operational carbon emissions, saline sewage treatment, r-BiVO<sub>4</sub> photoanodes

**Conference Title :** ICWTM 2022 : International Conference on Wastewater Technology and Management

**Conference Location :** Sydney, Australia

**Conference Dates :** December 02-03, 2022