

Green synthesis of hydrogen peroxide in a coaxial microwave plasma reactor using water and argon

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Abstract: We demonstrate the advantages of nanosecond microwave pulsations for H₂O₂ synthesis in a water-argon plasma at atmospheric pressure. The analysis of reactive species via spectroscopy gave hints into reaction pathways. Additionally, plasma zone quenching enhanced H₂O₂ recovery.

1. Introduction

Green production of hydrogen peroxide (H₂O₂) has been explored during the last two decades involving technical approaches like direct synthesis, electrolysis and plasmas. [1] Plasmas route represent an interesting platform for the electrification of processes owing to their capacity of fast on-off times, fast kinetics and lower capital investments. Microwave plasmas, in particular, have shown promise to efficiently activate chemicals. [2] However, due to their relatively high temperatures, their application for hydrogen peroxide synthesis is limited, as hydrogen peroxide is a thermally sensitive product. During the last years, Karlsruhe Institute of Technology (KIT) has been applying nanosecond pulsation of microwaves in order to control the time of activation and chemical reactions. [3] In this work, we present the concept and progress in the production of hydrogen peroxide using this approach. By tuning the energy supply, plasma cooling and microwave pulsation, we have managed to reach concentrations beyond 50mM..

2. Methods

In this work, we directly use water (dropwise) in combination with argon plasma to produce H₂O₂ by means of a microwave plasma reactor. The reactor consists of a coaxial torch based on microwaves modulated with nanosecond pulsations. The concentration of the liquid H₂O₂ product was measured by UV-Vis Spectroscopy and the active chemical species were analyzed by Optical Emission Spectroscopy (OES).

3. Results and Discussion

A first scanning of the microwave parameters (power and pulsation time) revealed that nanosecond scale of microwave pulsations had a significant influence enhancing the H₂O₂ formation in the millimolar range in comparison with continuous microwave plasma, which only yielded micromolar concentrations. The variation of the time of pulsations had a slightly higher influence over the formation of H₂O₂ than the input power variation, but the power had a greater influence over the H₂O₂ concentration in comparison to the ducty clycle variation. Furthermore, lowering the water flow rate had the greatest influence allowing higher concentrations of H₂O₂ beyond 50 mM. Lastly, the comparison among different cooling temperatures revealed that lower temperatures could favour a faster stabilization of the product with a slightly improvement in the final concentration. This could be due

to avoiding ion-electron interactions further related to the decomposition of H₂O₂. A qualitative analysis by OES of the plasma-water environment showed the presence of hydrogen phases alpha and beta, atomic oxygen and hydroxyl radical spectra regions, providing hints of possible reaction mechanisms. Moreover, increasing the water residence time led to a higher H₂O₂ yield possibly connected to a higher plasma-water contact area. A comparison of our results with literature reports is shown in Figure 1.

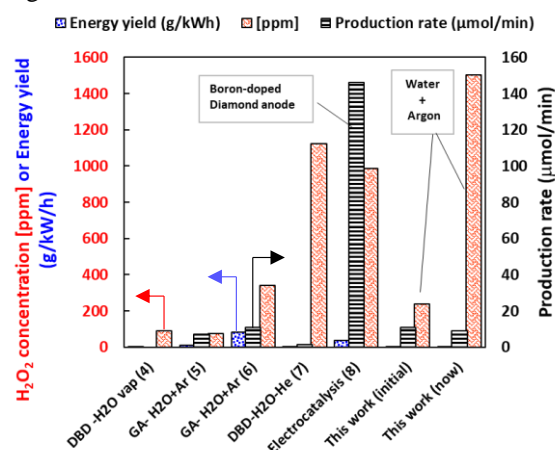


Fig. 1. Comparison of H₂O₂ synthesis via water and plasma: Literature vs. This Work.

4. Conclusion

The potential of the nanosecond pulsed microwave plasma technology was shown for H₂O₂ synthesis directly from Ar and H₂O. Lower water flow rates led to higher H₂O₂ concentrations, and the quenching was related to a different reaction pathway, as interpreted by OES.

References

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