Mechanistic Link to Electrode Magnetism in Magneto-Electrocatalytic Oxygen Evolution

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Significance and Relevance

Controlling electrocatalytic water-splitting with magnetic fields (MFs) could offer a simple approach to unlocking higher performance in industrial electrolysis. This work presents novel results for ferromagnetic Ni and paramagnetic Pt as working electrodes in magneto-electrocatalytic oxygen evolution. By investigating the catalytic performance at various MF strengths and orientations, a qualitative assessment is made of the governing magneto-electric mechanisms in these systems. A model of a thorough experimental approach for proper analysis of magnetic effects is demonstrated.

Preferred and 2nd choice for the topic:	Sustainable and clean energy production and transport / Advanced process with electrocatalysis and plasma utilization
Preferred presentation:	Oral preferred or Short Oral

Introduction and Motivations

Magneto-electrocatalytic water-splitting has been widely researched but is not yet well understood due to the complicated nature of magneto-electric (ME) mechanisms. These mechanisms tend to originate from Lorentz force, Kelvin force, and electrode spin-polarization; they present as magnetohydrodynamics (mass transport), magnetoresistance (charge transport), and promoted $^3O_2$ formation.[1] The reported ME effects on performance vary wildly, with some systems demonstrating enhancements of up to 650% and some succumbing to negative effects.[2,3] The large disparity between reports can be attributed to interference of ME mechanisms, where the dominant mechanism heavily depends on factors such as: electrode composition, relative MF orientation, and MF strength.

Materials and Methods

Electrochemical measurements were performed using a 3-electrode system with a Hg/HgO reference electrode and Pt sheet (1x1 cm^2) counter electrode in 1M KOH electrolyte. For the working electrode (1x1 cm^2), Ni foam, Ni sheet, and Pt sheet were compared. Using a CHI640E Potentiostat, chronoamperometry measurements were performed and magnetic effects were calculated from changes in current density. MFs up to 800 mT were imposed using an electromagnet.

Results and Discussion

Ni is a strong ferromagnet and Pt is a weak paramagnet; this comparison facilitates understanding of how spin polarization and local magnetic flux density link to the domination of certain ME mechanisms. Initial studies (Fig. 1A) show a positive correlation between effect magnitude and MF strength, particularly for Ni foam and Pt sheet. The MF enhances Ni performance and worsens Pt performance. It could be assumed from this result that, for Ni, the strong magnetic flux density and greater spin-alignment favors ME mechanisms such as: Kelvin force attracting reactive radicals in the electrolyte, magnetohydrodynamics shortening the diffusion layer, and promoted formation of paramagnetic $^3O_2$. The Pt, on the other hand, should have a weak affinity to these enhancement mechanisms and so may have succumbed to increased charge-carrier resistance within the electrode, known as positive magnetoresistance.

To investigate further, the magnetic effects were studied for each system in four orientations (Fig. 1B & 1C): 0° (original), 90°, 180°, 270°. Considering the Ni electrodes first, the strong dependence on orientation which leads to a negative effect at 180° and 270° suggests that Kelvin force and favored $^3O_2$ formation are not dominant mechanisms. The orientation-dependent Lorentz force, therefore, is the likely origin of dominant ME mechanisms – magnetohydrodynamics and intra-electrode charge-carrier movement – in the Ni systems. The persistent negative effect exhibited by the Pt supports the hypothesis of positive magnetoresistance dominating this system.

The optimal conditions demonstrated in this work (ferromagnet, 90°) should be considered in future studies pushing similar systems towards industrialization.

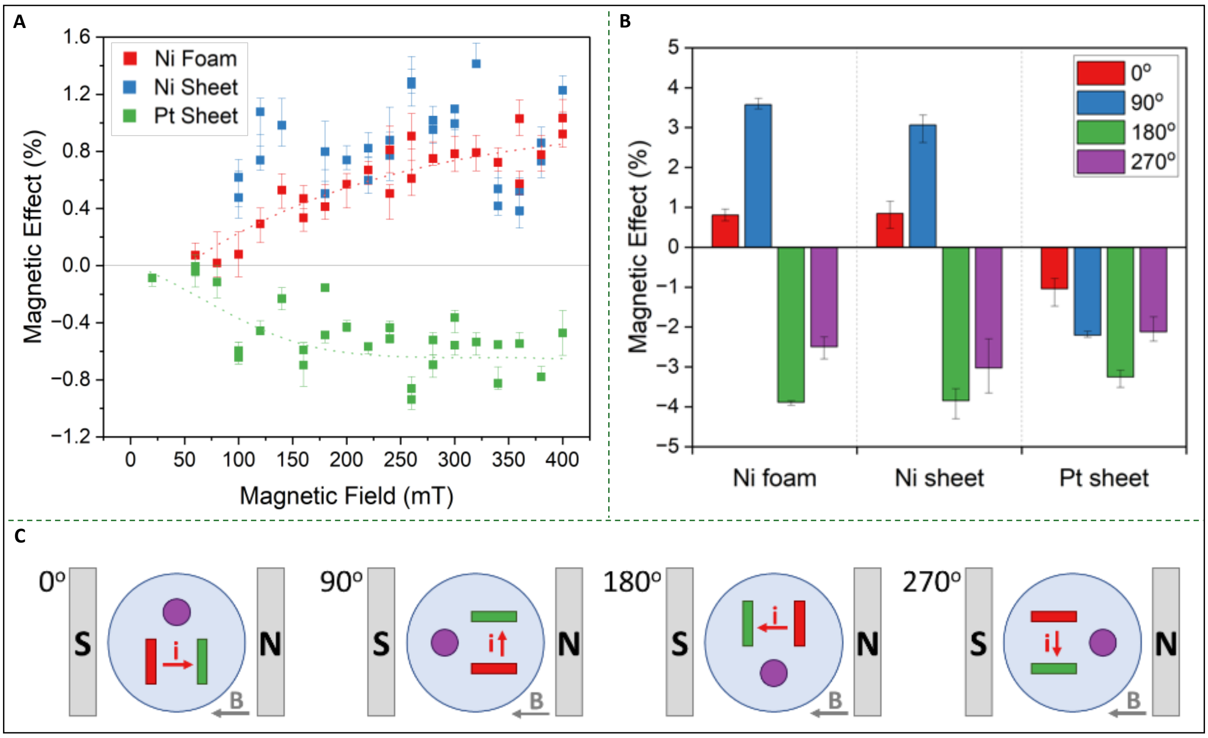

Figure 1. [A] Plots of magnetic effect vs MF strength in 0°-orientation for each working electrode. Magnetic field was ramped in 20 mT intervals from 0 to 400 mT. [B] Graph showing magnetic effect at 800 mT for four orientations for each working electrode. [C] Schematic showing a top-down view of the electrocatalytic setup in the four orientations relative to the MF, B. The red, green, and purple shapes represent the working, counter, and reference electrodes, respectively.

References

1. S Luo, K. Elouarzaki, Z. Xu, *Angew. Chem.* **2022**, *61*, e202203564
2. J. Saha, R. Ball and C. Subramaniam, *ACS Sustain. Chem. Eng.*, **2021**, *9, 7792–7802*
3. M. Lin, L. Hourng, C. Kuo, *Int. J. Hydrogen Energy*. **2012**, *37,* 2