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Decreasing the Energy Consumption of the CO_2 Electrolysis Process Using a Magnetic Field

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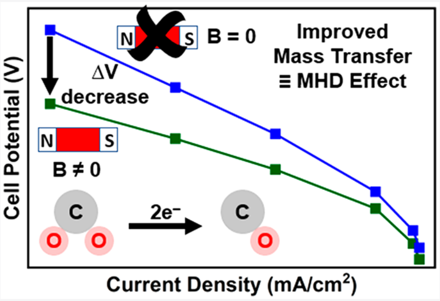
ABSTRACT: The renewable electricity-powered electrolysis of CO_2 could be a viable carbon-neutral method for producing carbon-based value-added chemicals like carbon monoxide, formic acid, ethylene, and ethanol. A typical CO_2 electrolyzer suffers, however, from the high power requirements, mainly due to the energy-intense anode reaction. In this work, we decrease the anode overpotential and thus reduce the overall cell energy consumption by using a NiFe-based bimetallic catalyst at the anode and applying a magnetic field. For a CO_2 electrolysis process producing CO in a gas diffusion electrode-based flow electrolyzer, we demonstrate that power savings in the range from 7% to 64% can be achieved at CO partial current densities exceeding −300 mA/cm^2 using a NiFe catalyst at the anode and/or by using a magnetic field at the anode. We achieve a maximum CO partial current density of −565 mA/cm^2 at a full cell energy efficiency of 45% with 2 M KOH as the electrolyte.



The renewable electricity-powered electrochemical CO_2 reduction reaction (CO_2RR) holds promise for the carbon-neutral production of various hydrocarbon fuels, chemicals, and intermediates such as carbon monoxide, formic acid, methane, methanol, ethylene, and ethanol, products that are traditionally obtained from energy-intensive carbon-positive fossil-fuel based processes.[1,2] In the 1980s and 1990s, Hori's seminal work demonstrated that transition metal catalysts such as Ag, Au, and Cu are excellent catalysts for CO_2RR.[3,4] Since then, researchers have developed better catalysts with significant improvements in electrochemical performance—current densities and Faradaic efficiencies (FEs). However, the overall CO_2 electrolysis process, comprising the CO_2RR at the cathode and an oxidation reaction at the anode, typically, the oxygen evolution reaction (OER), remains hampered by insufficient full cell energy efficiencies (EEs), high overall power requirements, as well as electrode durability and hence lacks feasibility at scale.[5]

Specifically, for CO_2RR to CO, at CO partial current densities (j_{CO}) exceeding −400 mA/cm^2, the EEs reported in literature are typically less than 40% and the cell power requirements exceed 1200 mW/cm^2.[6] The low EEs and high cell power requirements at high production rates are a direct result of the high cell potentials required. The high cell potentials are determined by the cathode and anode overpotentials, and significant improvements in cathode and/or anode overpotentials would improve the EEs and decrease the cell power requirements especially at high CO production rates or current densities. For CO_2RR to CO, efforts directed at improving the EEs and/or decreasing cell power requirements have used approaches such as changing cathode catalyst morphology,[7] using optimal electrolyte concentration and composition,[8] or using a less energy-intense anode reaction.[9]

For the CO_2 electrolysis process, the anode offers the most opportunity for improvement because the high anode overpotentials required for the OER result in the anode consuming well more than 50% of the energy supplied at operating cell potentials.[8,9] For example, when CO_2RR to CO at the cathode is coupled with OER at the anode, prior work shows that, based on thermodynamic equilibrium potentials, greater than 90% of the energy supplied is consumed by the anode reaction and less than 10% is consumed by the cathode reaction (the reaction of interest).[9] As the cell potentials are increased, these numbers change; however, the anode still consumes more energy than the cathode under the CO_2 electrolysis operating conditions.[8] Water electrolysis research

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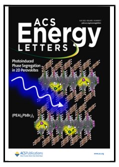

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has shown that Ir-based catalysts are better than Pt-based catalysts for OER.[10,11] Consequently, the CO_2 electrolysis field also favors Ir-based anode catalysts.[12,13] Ir is a precious metal, however, and thus not desirable in light of achieving economically feasibility for CO_2 electrolysis at scale. Many researchers have researched OER catalysts based on more abundant nonprecious metals.[10,11] For example, Ni- and Fe-based catalysts require lower overpotentials and hence perform better than Ir-based catalysts in alkaline media.[11,14] Some of us recently demonstrated an additive-assisted electrodeposited NiFe-based bimetallic catalyst that exhibits remarkable performance for OER.[15] The additive, 3,5-diamino-1,2,4-triazole (DAT), is an inhibitor that allows for a uniform electrodeposition, achieving a 1:1 molar ratio of Ni:Fe, and the creation of a fractal-like morphology of the NiFeDAT catalyst layer. To date, this NiFeDAT catalyst has not been evaluated at high current densities in a gas diffusion electrode (GDE)-based flow cell.

Use of a GDE-based flow electrolyzer allows for a gaseous CO_2 feed and thus circumvents diffusion limitations due to the low solubility of CO_2 in aqueous media.[16] Mass transfer in these systems can still be enhanced further by using reactor engineering or process intensification approaches such as optimizing fluid flow characteristics and/or optimizing the electrolyte composition.[5,8,17−19] An underexplored process intensification technique in electrocatalysis is the use of magnetic fields and is as such the focus of the work here. The role of magnetic fields in electrocatalytic systems has been studied but is not fully understood.[20,21] Some studies suggest that a magnetic field enhances the electrochemical performance of a system by influencing the local electromagnetic field near the electrode surface, thereby altering the spin states of the reaction intermediates.[20,22,23] Other studies suggest that enhancements in the electrochemical performance in the presence of a magnetic field are caused by improved mass transfer due to the magnetohydrodynamic (MHD) effect.[20,24] Studies on the use of magnetic fields in the CO_2 electrolysis process are relatively rare.[22,25]

In this work, we seek to reduce the energy consumption of the CO_2 electrolysis process by enhancing the anode performance by using a NiFeDAT catalyst for the OER while also applying a magnetic field. Specifically, in a GDE-based CO_2 flow electrolyzer, we combined CO production at the cathode (Ag catalyst) with O_2 evolution at the anode (NiFeDAT catalyst) under alkaline conditions and studied the effects of the applied magnetic field on current densities, electrochemical kinetics and mass transfer, EEs, and overall cell power requirements.

The electrochemical performance of the entire cell for CO production via CO_2 electrolysis was evaluated using either IrO_2 or NiFeDAT catalyst at the anode in the presence and absence of a magnetic field. Figure 1a shows the cell potentials required as a function of the applied total current density (j_{total}) (which is the same as the anode current density) with IrO_2 and NiFeDAT catalysts at the anode in the absence of a magnetic field. The cell potentials required with NiFeDAT are lower than those required with IrO_2. For example, for an applied j_{total} of −300 mA/cm^2, the cell potential required decreased from −2.89 to −2.58 V, corresponding to a 310 mV decrease in cell potential when changing from IrO_2 to NiFeDAT.

Figure 1b quantifies the production of CO, in terms of j_{CO}, as a function of the cell potential with IrO_2 and NiFeDAT catalysts at the anode in the presence and absence of a magnetic field. The j_{CO} obtained with NiFeDAT is significantly higher than that obtained with IrO_2, irrespective of the presence or absence of the magnetic field. This also means that the cell potentials required to achieve the same j_{CO} would be smaller with NiFeDAT than with IrO_2. When a magnetic field was applied at the anode, the j_{CO} obtained at a certain potential increased (or the cell potential required for achieving a certain j_{CO} decreased) with both IrO_2 and NiFeDAT. For example, in the absence of a magnetic field at the anode, at −3.00 V cell potential, the j_{CO} values obtained were −345 and −515 mA/cm^2 respectively with IrO_2 and NiFeDAT. On the other hand, in the presence of a magnetic field at the anode, at −3.00 V cell potential, j_{CO} values of −444 and −565 mA/cm^2 were obtained, respectively, with IrO_2 and NiFeDAT.

Figures 1c and S1 show the anode and cathode polarization plots, respectively, with IrO_2 and NiFeDAT anode catalysts in the absence of a magnetic field. When the anode catalyst is changed from IrO_2 to NiFeDAT, the anode polarization plots show that the anode potentials decrease while the cathode potentials remain identical. This tells us that the increase in CO_2 electrolysis performance when using NiFeDAT instead of IrO_2 at the anode in the absence of a magnetic field is solely due to the reduction in anode overpotentials.

Figure 1d shows the Tafel slopes in the low overpotential region for OER at the anode with IrO_2 and NiFeDAT catalysts in the absence of a magnetic field. The decrease in Tafel slope from 78 mV/decade for IrO_2 to 47 mV/decade for NiFeDAT suggests that NiFeDAT enhances the electrochemical kinetics for OER when compared to IrO_2 and thus increases the electrochemical performance. The reduction in anode overpotentials and the decrease in Tafel slopes demonstrates that NiFeDAT is a better performing catalyst for OER than IrO_2.

Figure 1e shows the CO FE trends as a function of j_{CO} with IrO_2 and NiFeDAT catalysts at the anode in the presence and absence of a magnetic field. The CO FEs were typically greater than 95% and were not influenced by the choice of the anode catalyst or the presence or absence of a magnetic field at the anode. Because the CO FEs were similar to both anode catalysts in the presence or absence of a magnetic field, the increase in j_{total} also meant a proportional increase in j_{CO}, and thus, the trends for j_{total} will also hold for j_{CO}.

Figure 1f shows the flow electrolyzer setup used in this work. The setup was slightly modified from our prior work.[8] For applying a magnetic field at the anode, we used a Nd magnet current collector (Figure 1f). For experiments without the magnetic field at the anode, a stainless-steel current collector was used. At the cathode, no magnetic field was applied. The detailed description of the setup is provided in the SI (Supporting Information).

To determine the origin of the enhancements observed in the electrochemical performance in the presence of a magnetic field at the anode, we analyzed the polarization plots and performed a Tafel slope analysis. Figure 2a and b shows the anode polarization plots with NiFeDAT and IrO_2 catalysts at the anode in the presence and absence of a magnetic field. Corresponding cathode polarization plots are presented in Figures S2a and b, respectively. These plots indicate that when a magnetic field is applied at the anode, the anode overpotentials decrease but the cathode overpotentials remain almost constant, leading to decreased cell potentials or increased j_{total} (and consequently, j_{CO}). The enhancement in electrochemical performance is more significant in the high overpotential region (cell potentials more negative than −2.00

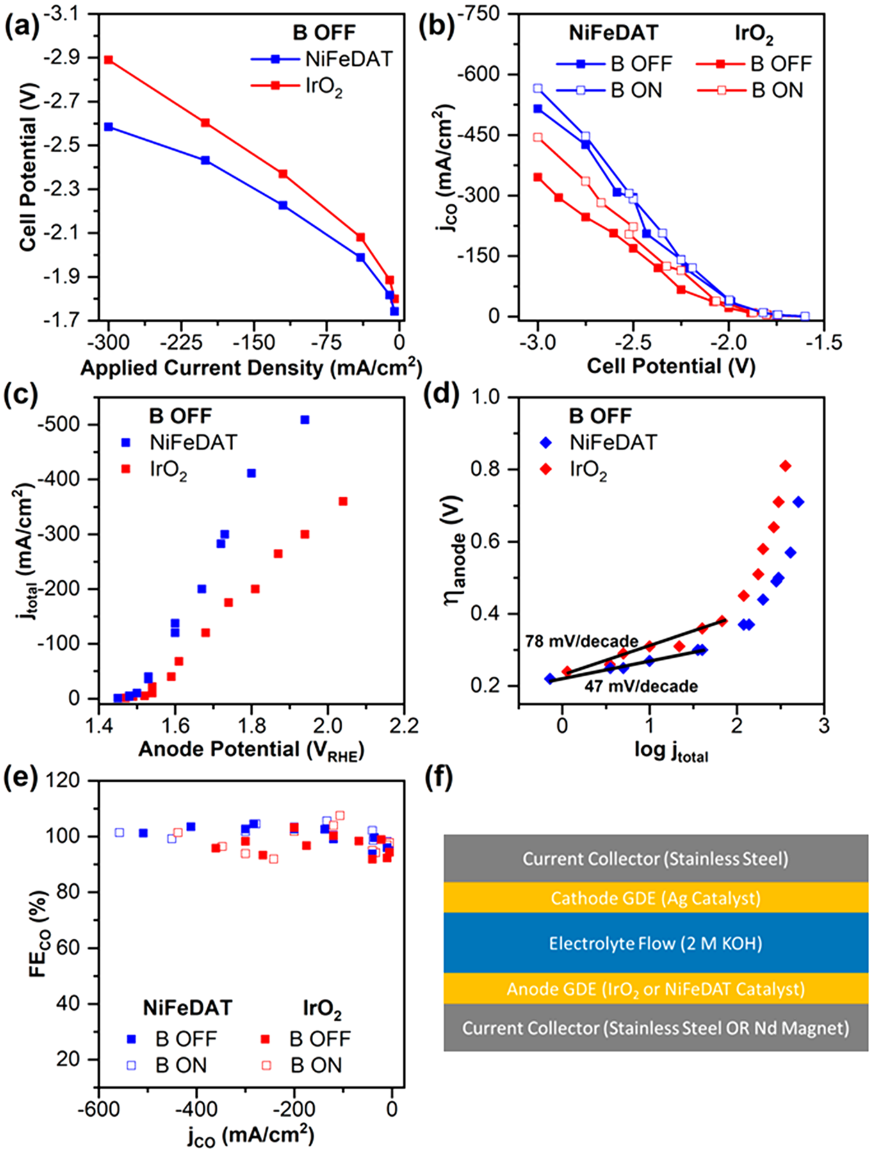

Figure 1. CO_2 Electrolysis performance. (a) $j-V$ curve demonstrating the cell potentials required for achieving the applied current densities with IrO_2 and NiFeDAT anode catalysts in the absence of a magnetic field; (b) $j-V$ curve demonstrating the j_{CO} obtained as a function of cell potentials in the presence and absence of a magnetic field with IrO_2 and NiFeDAT anode catalysts; (c) $j-V$ curve demonstrating the j_{total} obtained as a function of anode potentials measured vs RHE (V_{RHE}) with IrO_2 and NiFeDAT anode catalysts in the absence of a magnetic field; (d) Tafel plots showing the Tafel slopes in the low overpotential region with IrO_2 and NiFeDAT anode catalysts in the absence of a magnetic field; (e) CO Faradaic efficiency trends obtained as a function of j_{CO} with IrO_2 and NiFeDAT anode catalysts in the presence and absence of a magnetic field; (f) cell configuration of the flow electrolyzer used for electroanalysis experiments with or without magnetic fields. In all cases, the cathode was a Ag GDE with a catalyst loading of 1 mg/cm^2. The catalyst loadings of the IrO_2 GDE and the NiFeDAT GDE were also 1 mg/cm^2. The electrolyte, 2 M KOH, was flown at 1 mL/min. The flow rate of CO_2 was 17 sccm. The curves show that higher electrochemical performance is obtained when NiFeDAT is used as the anode catalyst instead of IrO_2 and/or when a magnetic field is applied at the anode. Moreover, the CO FEs are unaffected by the choice of anode catalyst and by the absence or presence of a magnetic field at the anode.

V) characterized by mass transfer limitations. No significant enhancement in electrolysis performance is evident in the low overpotential region (cell potentials more positive than −2.00 V) representative of kinetic limitations. The low and high overpotential regions were chosen on the basis of prior work and the change in Tafel slopes observed here.[8]

Figure 2c and d shows the Tafel slopes in the low and high overpotential regions with NiFeDAT and IrO_2 catalysts at the anode in the absence and presence of a magnetic field. In the low overpotential region, the Tafel slopes did not change significantly in the absence and presence of a magnetic field with both NiFeDAT and IrO_2. However, in the high overpotential region, the Tafel slopes decreased from 740 to 489 mV/decade with IrO_2 and from 487 to 437 mV/decade with NiFeDAT in the presence of a magnetic field, indicating improvement in mass transfer. As also pointed out in prior work,[24] a homogeneous magnetic field applied parallel to the electrode surface has no effect on the charge-transfer coefficient and thus, the enhancement in electrochemical performance cannot be due to enhancements in electrochemical kinetics.

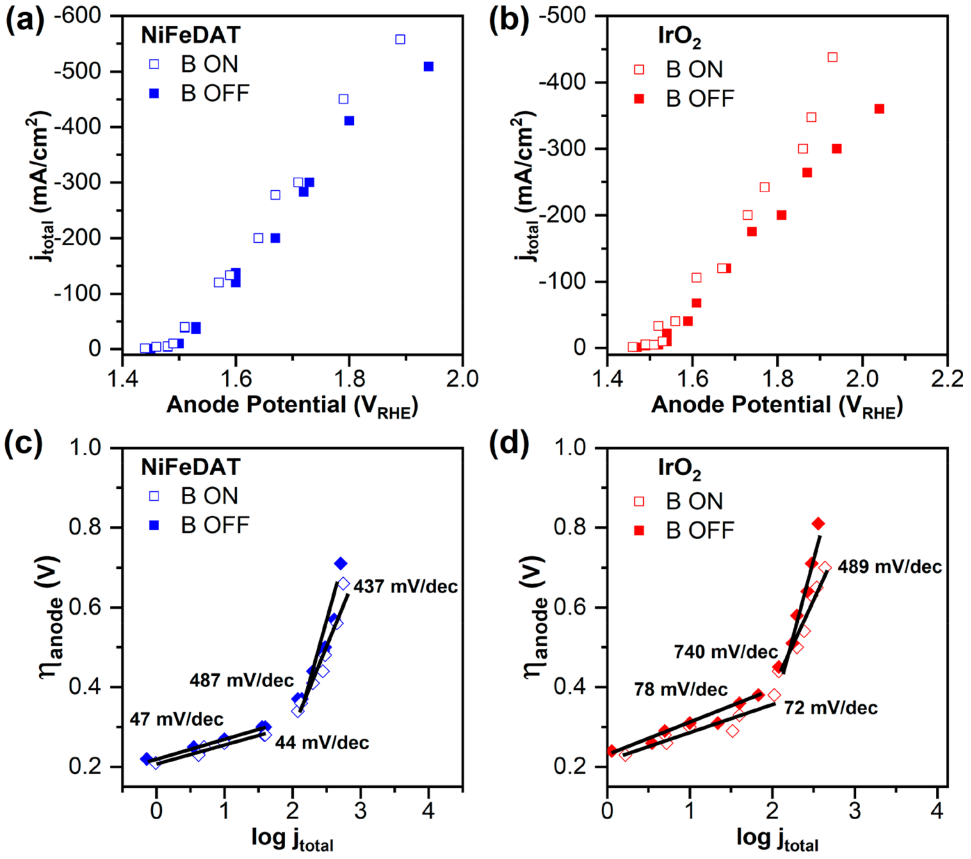

Figure 2. Polarization plots and Tafel slopes. (a) j−V curve demonstrating the j_{total} obtained as a function of anode potentials measured vs RHE (V_{RHE}) with NiFeDAT anode catalyst in the presence and absence of a magnetic field; (b) j−V curve demonstrating the j_{total} obtained as a function of anode potentials measured vs RHE (V_{RHE}) with IrO_2 anode catalyst in the presence and absence of a magnetic field; (c) Tafel plots showing the Tafel slopes in the low and high overpotential regions with NiFeDAT anode catalyst in the presence and absence of a magnetic field; (d) Tafel plots showing the Tafel slopes in the low and high overpotential regions with IrO_2 anode catalyst in the presence and absence of a magnetic field. In all cases, the cathode was a Ag GDE with a catalyst loading of 1 mg/cm². The catalyst loadings of the IrO_2 GDE and the NiFeDAT GDEs were also 1 mg/cm². The electrolyte, 2 M KOH, was flown at 1 mL/min. The flow rate of CO_2 was 17 sccm. The curves show that higher electrochemical performance obtained in the presence of a magnetic field at the anode irrespective of the anode catalyst used is due to the reduction in anode overpotentials especially in the high overpotential region. Based on Tafel slope analysis, this reduction in overpotentials can probably be attributed to enhanced mass transfer.

Combining the insights from the polarization plots (Figure 2a, b) and Tafel slopes (Figure 2c, d), we conclude that the presence of a magnetic field enhances the mass transfer of ions in the electrolyte (K^+ and OH^-) but does not influence the electrochemical kinetics. We attribute the observed enhanced mass transfer to the MHD effect. We also studied the MHD effect in flow electrolyzers by qualitatively analyzing the diffusion and hydrodynamic boundary layers and the mass transfer coefficients (Figure S5). We hypothesize that the thicknesses of the diffusion (δ_D) and hydrodynamic (δ_H) boundary layers become smaller in the presence of a magnetic field than those in the absence of a magnetic field. In the MHD effect, the Lorentz force generated by the application of a magnetic field is an external force and is responsible for improving the diffusion and momentum fluxes near the electrode.[26,27] The improvement in diffusion flux results in a higher local concentration of electroactive species in the electrolyte near the electrode and improvement in momentum flux leads to higher fluid velocities near the electrode. These higher local concentrations of electroactive species and fluid velocities mean smaller boundary layers in the presence of a magnetic field than in the absence of a magnetic field. Thus, the enhancement in electrochemical performance due to the MHD effect can be attributed to improved mass transfer of the electroactive species in the electrolyte. Calculation S1 in the SI provides order-of-magnitude estimates for these changes in the diffusion and hydrodynamic boundary layer thicknesses as well as changes in the mass transfer coefficients. Prior work has modeled the MHD effect in stationary cells by coupling the Navier−Stokes equations with the continuity equation and accounting for the Lorentz force generated due to the magnetic field.[26−32] However, the mass transfer coefficient and the velocity profile need to be corrected in these models for a flow electrolyzer system before these models can quantitatively explain the MHD effect in flow electrolyzers.[33−35] For a more detailed understanding of the MHD effect, we advise the reader to read the following literature: refs 20, 21, 26, 27, and 32.

From a systems perspective, the enhancement in electrochemical performance translates to decreased cell power requirements and higher EEs at high j_{CO}. Figure 3a shows the CO EEs as a function of j_{CO} with IrO_2 and NiFeDAT catalysts at the anode in the presence and absence of a magnetic field. The CO EEs are higher in the presence of a magnetic field because lower cell potentials were required for the same j_{CO}. In general, CO EEs obtained with NiFeDAT are

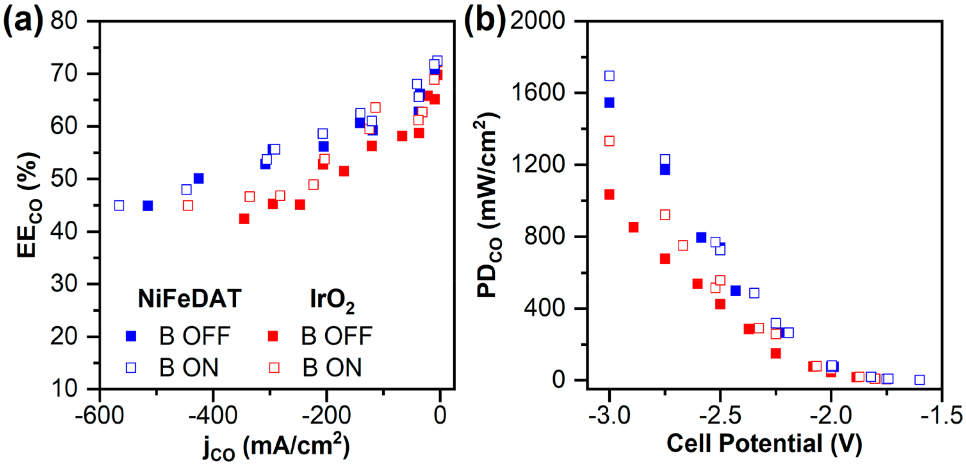

Figure 3. Energy efficiency and power density trends. (a) Full cell energy efficiencies for CO production obtained as a function of j_{CO} with IrO_2 and NiFeDAT anode catalysts in the presence and absence of a magnetic field; (b) power densities for CO production as a function of cell potentials with IrO_2 and NiFeDAT anode catalysts in the presence and absence of a magnetic field. In all cases, the cathode catalyst was a Ag GDE with a catalyst loading of 1 mg/cm^2. The catalyst loadings of the IrO_2 GDE and the NiFeDAT GDEs were also 1 mg/cm^2. The electrolyte, 2 M KOH, was flown at 1 mL/min. The flow rate of CO_2 was 17 sccm. The figure shows that higher full cell energy efficiencies can be obtained with NiFeDAT catalyst at the anode in the presence of a magnetic field while realizing significant energy savings at high j_{CO}.

higher than the CO EEs obtained with IrO_2. For example, in the absence of a magnetic field at the anode, we achieved an EE of 45% at a j_{CO} of −515 mA/cm^2 with NiFeDAT compared to an EE of 42% at a j_{CO} of −345 mA/cm^2 with IrO_2. In the presence of a magnetic field at the anode, we achieved an EE of 45% at a j_{CO} of −565 mA/cm^2 with NiFeDAT compared to an EE of 45% at a j_{CO} of −444 mA/cm^2 with IrO_2. This suggests that similar EEs can be achieved with both catalysts albeit with lower j_{CO} values with IrO_2 anode catalyst. Similarly, at an applied current density of −200 mA/cm^2, we achieved EEs of 53% and 56% with IrO_2 and NiFeDAT anode catalysts, respectively, in the absence of a magnetic field. In the presence of a magnetic field, at an applied current density of −200 mA/cm^2, we achieved EEs of 54% and 59% with IrO_2 and NiFeDAT anode catalysts, respectively. Figure 3b shows the cell power densities as a function of the cell potential with IrO_2 and NiFeDAT catalysts at the anode in the presence and absence of a magnetic field. Cell power requirement calculations show that by switching the anode catalyst from IrO_2 to NiFeDAT and using a magnetic field at the anode, power savings of up to 64% for the overall CO_2 electrolysis process can be achieved while achieving a j_{CO} of −565 mA/cm^2 at an EE of 45% with 2 M KOH as the electrolyte. In general, for j_{CO} exceeding −300 mA/cm^2, power savings in the range from 7% to 64% were observed. Detailed calculations of the power savings are provided in the SI (Calculation S3). The relative benefits of using NiFeDAT as the anode catalyst instead of IrO_2 and/or using a magnetic field at the anode are summarized in Tables S3 and S4 in the form of overall power savings.

In summary, we explored the use of magnetic fields to enhance the electrochemical performance, and thus the energetics, of the CO_2 electrolysis process. Specifically, we showed that the application of a magnetic field at the anode reduces the overpotential at the anode irrespective of the anode catalyst used. Experimental data (*j*−*V* curves, polarization plots, and Tafel slopes) indicates that the beneficial effects observed in the presence of a magnetic field are due to the enhancement of mass transfer of electroactive species caused by the MHD effect. We demonstrate that the use of the electrodeposited bimetallic NiFeDAT catalyst at the anode in the presence of a magnetic field can result in 7% to 64% savings in power consumption compared to the commonly used IrO_2 catalyst at the anode in the absence of a magnetic field at j_{CO} exceeding −300 mA/cm^2. We also achieved a j_{CO} of −565 mA/cm^2 at 45% EE in a 2 M KOH electrolyte flowing at 1 mL/min. Future work should not only focus on addressing the problem of carbonate formation during the CO_2 electrolysis process by understanding and manipulating the effects of the local environment near the electrode but also study the influence of magnetic field on the long-term performance stability of the electrodes, especially focusing on carbonate formation and catalyst etching.[36,37] Further studies would also be needed to unravel the mechanistic aspects of the observed enhancements due to the application of a magnetic field. Magnetic fields can influence the local electromagnetic fields near a wide variety of other electrode surfaces. Thus, future work should explore the use of the concept demonstrated in this study of enhancement of electrochemical performance using magnetic fields for various other electrocatalysts and/or electrochemical reactions, e.g., ethylene production via CO_2RR on Cu-based catalyst (cathode) or, more ambitiously, ammonia production via nitrogen reduction.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsenergylett.1c01029.

> Safety note; materials and methods; electrochemical performance; SQUID magnetometry; MHD effect calculations; energy calculations (PDF)

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Notes

The authors declare no competing financial interest.

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