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Screening of Binder Materials for Ag-Based Gas Diffusion Electrodes for CO_2 Conversion to CO

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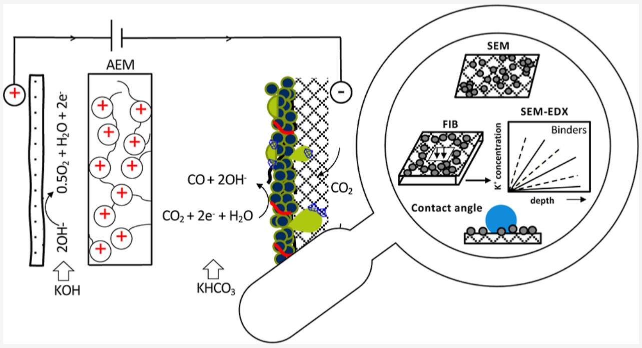
ABSTRACT: Electrochemical CO_2 reduction (CO_2R) to chemicals and fuels has made tremendous progress since the introduction of gas diffusion electrodes (GDEs) to overcome mass-transfer limitations and enable industrial-scale current densities. The advancement in the field, however, has come with new challenges that are related to the stability and degradation of the GDE due to flooding issues, which currently hinder the scale-up. Here, we investigated the effect of six different binding materials (Nafion, polytetrafluoroethylene, Fumion, Pention, poly(vinyl alcohol), and polypyrrole) on the stability and performance of Ag-based GDEs for CO_2R to CO in alkaline media. All binders show a decrease in the Faraday efficiency (FE) of CO and increase in hydrogen evolution reaction over time. The most hydrophilic GDE based on polypyrrole can uphold a higher FE of CO for longer times, which is contrary to a common belief that low wettability is required for long-term stability. By using a range of tools (SEM−EDX, SEM−FIB, X-ray diffraction, and contact angle measurements) for the postelectrolysis characterization of the GDEs, we show that the performance loss is related to flooding, bi(carbonate) precipitation, and catalyst agglomeration. These results contribute to a better understanding of the stability issues in GDE-based CO_2 electrolyzers.



■ INTRODUCTION

Electrochemical reduction of CO_2 (CO_2R) to value-added chemicals is an interesting power-to-X (P2X) concept that can help decarbonize the chemical industry. CO_2R has made a tremendous progress since the introduction of gas diffusion electrodes (GDEs) to overcome the mass-transfer limitations that impeded the liquid-based CO_2R process.[1,2] Nowadays, several CO_2R products can be obtained at high current densities (CDs), exceeding ampere per square centimeter scale, and low cell voltages.[3−10] However, the use of GDEs has also brought challenges, which are related to the stability and degradation of the electrodes, especially at high CD operation.[11] The exact degradation mechanism has yet to be elucidated, but factors that are known to play an important role include (1) the alkaline environment, which is typically used in CO_2R to suppress the competing hydrogen evolution reaction (HER) and promote C−C coupling and cause (bi)carbonate precipitation, (2) the change of hydrophobicity and wettability of the electrodes upon application of a current, which may cause electrolyte penetration into the GDE, ultimately flooding the catalyst layer (CL) and gas diffusion layers (GDLs) and blocking CO_2 supply to the catalyst, triggering the HER, and (3) other phenomena such as erosion and dissolution of GDE materials.[12] Furthermore, the degradation mechanism depends on the cell configuration [e.g., membrane electrode assembly (MEA) or flow electrolyzer] and the mode of GDE operation (i.e., flow-by or flow-through), which is controlled by the differential pressure across the GDE.[13−16] The flooding of the GDE and electrolyte precipitation and dissolution in an anion-exchange membrane (AEM)-based CO_2 electrolyzer seem to follow an oscillatory phenomenon, where CO_2R switches to HER and vice versa, resulting in salt precipitation in the former and salt dissolution in the latter.[17,18] To circumvent the flooding problem, two approaches have been proposed in the literature.[19] The first approach or the "first line of defense"

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focuses on the three-phase interface, where the GDE is in contact with gas and liquid. By controlling the wettability and hydrophobicity of the CL and GDL, the penetration of the electrolyte into the GDE can be prevented. Examples of the first approach have been reported by Reyes et al.,[20] Senocrate et al.,[21] Wu et al.,[22] and O'Brien et al.[23] The second approach or the "second line of defense" focuses more on curing the problem. The structure and composition of the GDE are modified to allow for perspiration and drainage of the excess electrolyte through the different layers. The second approach has been mostly studied by the Broekmann group.[24−27] They developed an energy-dispersive X-ray spectroscopy/inductively coupled plasma mass spectrometry (EDX/ICP−MS) approach to quantitatively monitor the electrolyte penetration into the GDE in three dimensions. Recently, Wrobel et al.[28] used a mass balance approach to monitor and quantify ions in the anolyte, catholyte, and GDE permeate during CO_2R to CO in a bipolar membrane based flow reactor. Future research must show which of the approaches are the best and whether "prevention is better than cure" for the long-term stability of GDEs. For more information on the degradation mechanisms, we refer the reader to the recent review papers on the subject.[29−31]

Recent studies show that GDE manufacturing and the ink formulation for the GDE has a significant impact on the properties and performance of the GDE.[32,33] Typically, the catalyst particles and the binding material (ionomer) are mixed in a solvent to form the ink, which is then spray-coated onto the GDL to obtain the GDE. In particular, the type and loading of the ionomer seems to play a crucial role in the flooding-induced instability of the GDE. The primary function of the ionomer is to bind the CL to the GDL and provide conductivity, but it also impacts the local environment and factors that influence CO_2R such as wettability, gas transport, electrode morphology, and pH. Nwabara et al.[34] explored the effect of different binders [Nafion, polyvinylidene fluoride, Fumion, polytetrafluoroethylene (PTFE), and Sustainion] on the stability of Ag-based GDEs for CO_2R to CO in an alkaline flow cell. These authors found that PTFE-based GDEs elude carbonate formation, while GDEs coated with a durable overlayer enhanced the stability. Chen et al.[35] studied the effect of catholyte and CL binders on the CO_2R selectivity in a BPM-based flow reactor. The composition of the catholyte and the type of binder materials were shown to have a significant effect on the selectivity of CO_2R to CO, whereas the selectivity toward formate was not influenced by these parameters. Idros et al.[36] investigated the effect of dispersion solvents (acetone and methanol) on the stability of Cu-based GDEs for CO_2R to multicarbon products. They showed that the use of acetone as a solvent in the catalyst ink formulation resulted in more uniform and flooding-resistant GDEs than when methanol was used. Wang et al.[37] studied the effect of Nafion and several anion-exchange ionomers on the performance of CO_2R to CO in an AEM-based MEA cell. The best performance was shown for the GDEs with Sustainion, which mitigated electrode flooding due to a balanced conductivity and hydrophobicity. Liu et al.[38] investigated the role of Fumion and Nafion binders on the stability of Ag-based GDEs for CO_2R to CO in a zero-gap MEA cell. These authors showed that Fumion-based GDEs are superior to Nafion-based GDEs, while the former is less hydrophobic, disproving the common opinion that nonwettability is crucial to prevent flooding. Similarly, Zhang et al.[39] concluded that only increasing the hydrophobicity of the CL is not sufficient to prevent flooding of GDEs in CO_2 electrolyzers. Recently, Zeng et al.[40] studied eight different ionomers for CO_2R to multicarbon products with Cu-based GDEs in a zero-gap MEA electrolyzer. They studied the impact of ionomer hydrophobicity on the selectivity and partial current density of C_{2+} products, but the GDE stability was not discussed. From these previous studies, no general conclusions can be drawn on the suitability of one binder type over the other for the GDE stability, which seems to be highly dependent on the reactor configuration and conditions.

In this work, we investigated the effect of various functional and nonfunctional binding materials or ionomers (Nafion, PTFE, Fumion, Pention, poly(vinyl alcohol) (PVA), and polypyrrole) on the stability and performance of Ag-based GDEs operated in a flow-by electrolyzer for CO_2 conversion to CO in alkaline conditions. We selected ionomers with anion exchange, cation exchange, and redox functionalities and ionomers without any functionality to study the effect of functional groups. The experiments were performed in the potentiostatic mode, and the Faraday efficiencies (FEs) of the products (CO and H_2) were measured as a function of time (up to 4 h). To understand the decreased CO production and increased HER in time, we performed detailed postelectrolysis characterization of the GDEs using SEM−EDX, X-ray diffraction (XRD), and contact angle measurements. We used a novel method to quantify electrolyte penetration into the GDE by employing a focused ion beam (FIB) to prepare a cross-sectional sample, which allowed for tracking the concentration of the electrolyte ions (potassium) as a function of depth using SEM−EDX. As discussed earlier, the Broekmann group also used potassium as a tracer for electrolyte seepage, but their method is based on the EDX/ICP−MS approach. These characterization results show that the loss of GDE performance is a combined effect of the flooding phenomena, induced by wetting of the GDE, (bi)carbonate precipitation, and agglomeration of the catalyst particles. Moreover, the stability does not necessarily correlate with the hydrophobicity of the CL because the GDE with the highest wettability (polypyrrole) showed the best performance.

■ EXPERIMENTAL SECTION

Preparation of the GDE. Silver (Ag) nanoparticles (<100 nm particle size, Sigma-Aldrich) and 10 wt % binding material were weighed and placed in a glass vial. Different binding materials/dispersions, Nafion (Sigma-Aldrich), Teflon (FEPD 121, Fuel Cell Store), Fumion (FAA-3-SOLUT-10, Fuel Cell Store), Pention (S72—10 wt %, Fuel Cell Store), poly(vinyl alcohol) (PVA), and polypyrrole doped with proprietary acids (5 wt % dispersion in H_2O, Sigma-Aldrich), were used in the GDE preparation. Isopropanol and Milli-Q water were added to the nanoparticles and mixed by sonicating the solution for 1 h at 20% amplitude and 0.5 cycle using an ultrasonicator (UP100H, Hielscher Ultrasonics). The ink solution was airbrushed on the carbon paper (Sigracet 28BC, Fuel Cell Store) while placed horizontally and attached to a heating plate. The loading of Ag nanoparticles on the GDE was ~1 mg/cm^2. After airbrushing, the GDE was allowed to cool, followed by rinsing with water and drying at room temperature.

Characterization of the GDE. The morphology and elemental composition of the GDEs were characterized by Field Emission Scanning Electron Microscopy (FE-SEM, JEOL, JSM-6500F). To quantify potassium penetration into

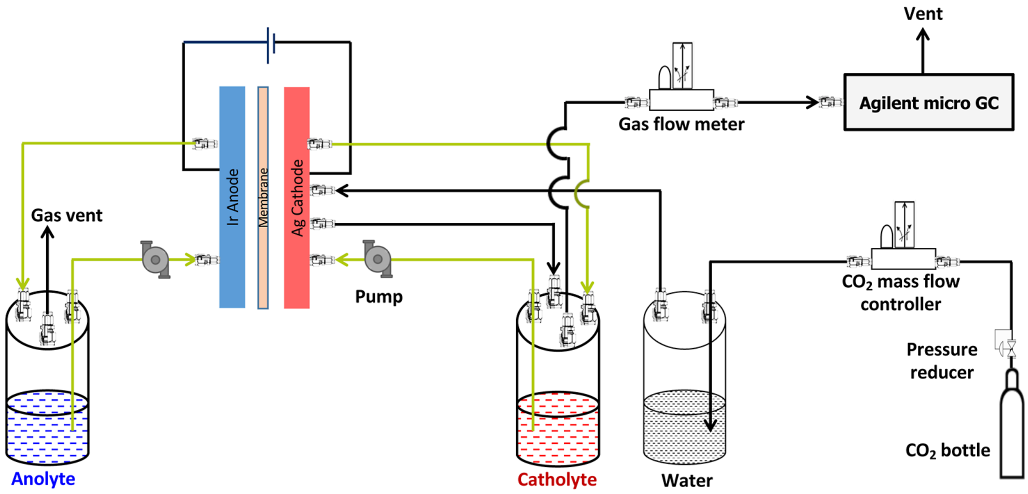

Figure 1. Electrochemical setup to convert CO_2 to CO using an Ag-based GDE. The electrolyzer is operated in the flow-by mode at 4 V. As the catholyte and anolyte, 0.5 M $KHCO_3$ and 0.5 M KOH are used, respectively. The Ag-based cathode and Ir-MMO anode are separated with an AEM.

the GDEs, a cross section was created with a Thermo Scientific Helios UXe DualBeam G4 apparatus, which was equipped with an EDX detector and a FIB. For depth profiling, these GDEs were loaded into the FE-SEM and line scans were measured vertically on the cross section. The GDEs were also characterized by powder XRD (Bruker D8) to identify the chemical compounds. The hydrophobicity and wettability of the GDEs were quantified by measuring the contact angle of a water droplet with an optical tensiometer (Theta Lite, TL100-TL101) by using the sessile drop method. The electrolyte after the reaction was analyzed by inductively coupled plasma optical emission spectroscopy (ICP-OES, Spectro-Arcos) to measure the concentration of (catalyst) metals.

Electrolyzer and Materials. A schematic diagram of the electrochemical setup used in this study is shown in Figure 1. The CO_2 electrolyzer (5 cm^2) was purchased from Dioxide Materials and modified according to our needs. As the anode and cathode, a titanium plate coated with iridium-mixed metal oxides (Ir-MMO, Magneto Special Anodes) and a Ag-based GDE were used, respectively. The anode and cathode compartments were separated by an AEM (Fumasep FAB-PK 130, Fumatech).

After assembling the electrolyzer, 0.5 M potassium bicarbonate ($KHCO_3$, Sigma-Aldrich) catholyte (100 mL) and 0.5 M potassium hydroxide (KOH, Sigma-Aldrich) anolyte (100 mL) were circulated continuously with a flow rate of 10 mL/min using peristaltic pumps (Kamoer F01A-STP). At the cathode, a CO_2 flow rate of 100 mL/min was maintained using a mass flow controller (EL-FLOW, Bronkhorst), and the output flow was monitored using a mass flow meter (CORI-FLOW, Bronkhorst). CO_2 was saturated with water through bubbling before being fed to the electrolyzer. All experiments were performed at room temperature and pressure conditions. The electrolyzer was used at a constant voltage (4 V) in a flow-by configuration, while the output current was measured using a potentiostat (PGSTAT204, Autolab). The reaction products and unreacted gases coming out of the catholyte chamber were analyzed using an Agilent 990 Micro Gas Chromatography (GC) with Molecular sieve 5A and PoraPLOT U columns connected to thermal conductivity detectors. For the GC, calibration curves were generated by plotting the concentrations of the components against the areas obtained with calibration gas mixtures of varying concentrations; see Figure S1 of the Supporting Information. By comparing the retention time and the area of the peaks, the type of component and its concentration were determined.

The FEs of the products were determined through the following equation

$$\mathrm{FE}_i = \frac{Q \times n \times F \times x_i}{I_t} \tag{1}$$

where Q (mol/s) is the molar flow at the outlet of the catholyte compartment, n_i the number of electrons required for product i, F the Faraday constant (96,485 C/mol), x_i the molar fraction of the product measured by GC, and I_t the total current (C/s) passed during the sampling time.

■ RESULTS AND DISCUSSION

Electrolysis of CO_2 to CO. The results of the CO_2 electrolysis experiments are shown in Figure 2a−f. The reproducibility of the data were checked by repeating these experiments, see Figure S2 of the Supporting Information. For all binding materials, the Faraday efficiency (FE) of CO drops after a short electrolysis time (~1 h), while the HER increases steadily over time. We note that the sum of the FEs for CO and H_2 is close to but not exactly 100%, which means that we have some uncharacterized gaseous and liquid products. Ag-based electrocatalysts are selective toward CO, but they can also produce small amounts of other products. Formate was detected in trace amounts in the catholyte compartment (Table S2 of the Supporting Information), but this cannot explain the missing electrons. It is worthwhile to mention that any produced formate will likely crossover to the anode compartment due to the use of an AEM, but this cannot be scrutinized here since the anolyte was not analyzed. Initially, the FE of CO is >80% but drops below 40% for all binders after 4 h of electrolysis time. For PVA-based GDEs, the FE of CO drops even below 30%. For all binders, a drop in CD from ~50 to ~30 mA/cm^2 is observed over time. However, the CDs on GDEs with Fumion, PVA, and polypyrrole binders show more erratic behavior, which we attribute to excessive bubble formation in the electrolyte. It is worthwhile to discuss the

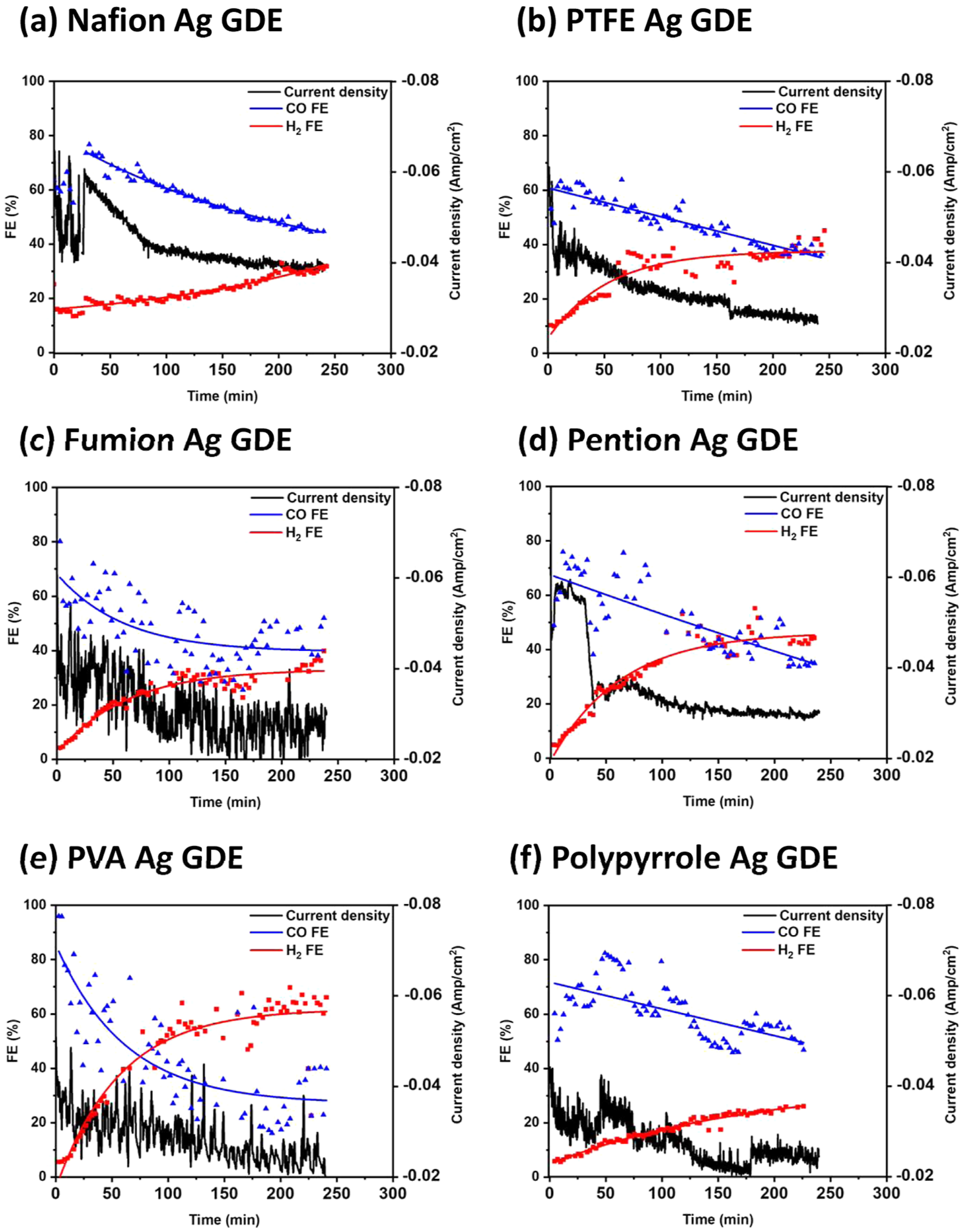

Figure 2. FE and current density as a function of time for CO_2R to CO on an Ag-based GDE with different binders. (a) Nafion, (b) PTFE, (c) Fumion, (d) Pention, (e) PVA, and (f) polypyrrole. Line fits to FE data are used to guide the eyes.

functionality of the binders in conjunction with electrolysis performance. Nafion, Fumion, Pention, and polypyrrole are functional ionomers as they bear ionic groups that can exchange ions, while PTFE and PVA are nonfunctional ionomers. Nafion has negatively charged groups and will exchange cations, whereas Fumion, Pention, and polypyrrole have positively charged groups with an anion-exchange functionality. The CDs of the Nafion-based GDE are significantly higher than those of the GDEs based on other binders. This suggests that binders with a cation-exchange functionality exhibit a higher CD than nonfunctional binders or anion-exchange binders. The lower CDs of PTFE and PVA-based GDEs can be explained due to their lower ionic conductivities. No obvious trends can be observed for the FE of CO with respect to the functionalities of the binders. The worst and best performances in terms of FE are seen for PVA and polypyrrole binders, respectively. The loss of performance, the increase of HER, and instability over time have been observed in several previous studies.[34,37,38] The exact mechanism for this instability has yet to be elucidated, but GDE degradation and flooding seem to play a crucial role herein. Nevertheless, it is clear from our experiments and previous studies[41,42] that flooding sets in and impacts the performance within a short (i.e., a couple of hours) electrolysis time. In the following, we aim to provide a deeper understanding of this unstable behavior by pre- and post-electrolysis analysis of the GDEs using multiple characterization tools.

Characterization of the GDE. The wettability and hydrophobicity of the GDE can provide useful information about the flooding phenomena. Therefore, we measured the static contact angle of a water droplet in equilibrium with the

GDE surface. The results of the contact angles measurement for the pristine and spent GDEs with different binders are presented in Figure 3a−l and Table 1. All pristine GDEs, except the one based on polypyrrole, show hydrophobic characteristics with contact angles >135°. The hydrophobicity of the pristine GDEs shows the following order for the binders: PTFE ≈ Nafion > Pention > Fumion ≈ PVA > polypyrrole. Remarkably, the pristine polypyrrole-based GDE with a contact angle of 90° exhibits a reduced hydrophobicity, even when compared to the bare GDL without a catalyst (a contact angle of >148°, see Figure S3 of the Supporting Information). More interestingly, the contact angle of all spent GDEs, regardless of the binder type, decreases substantially after 4 h of electrolysis time. This means that the GDEs lose their initial hydrophobicity and become more hydrophilic as current is applied, which is consistent with previous findings.[41] A comparison of the results in Figures 2 and 3 reveals that the CO_2R performance is not necessarily correlated with the hydrophobicity of the GDE. The most hydrophilic polypyrrole-based GDE shows the best performance, as it can uphold a high FE of CO for longer times.

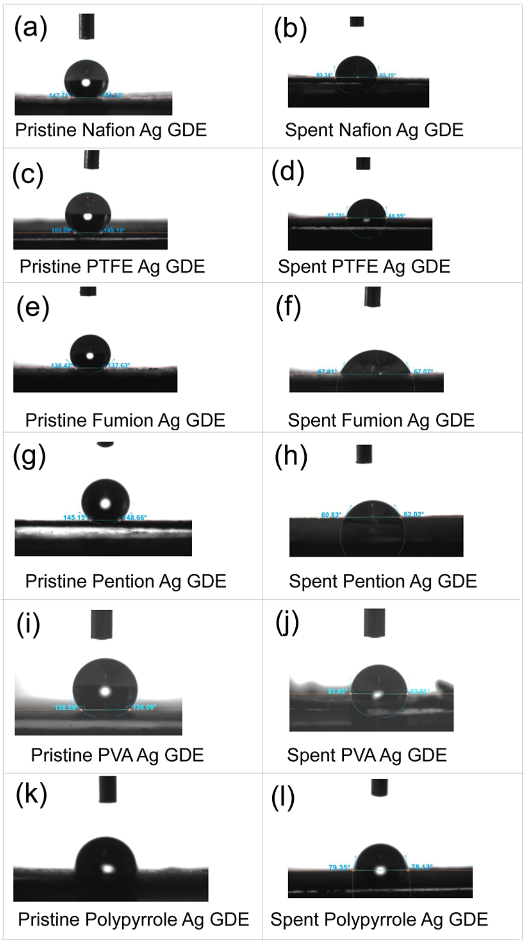

Figure 3. Contact angle measurements of pristine (left) and spent (right) GDEs of different binders. (a,b) Nafion-based GDE, (c,d) PTFE-based GDE, (e,f) Fumion-based GDE, (g,h) Pention-based GDE, (i,j) PVA-based GDE, and (k,l) polypyrrole-based GDE.

Table 1. Contact Angles of a Water Droplet on Pristine and Spent Ag-Based GDEs with Different Binders

binder	Pristine GDE/(°)	spent GDE/(°)
Nafion	149 ± 2	93 ± 5
PTFE	150 ± 2	89 ± 4
Fumion	138 ± 6	67 ± 3
Pention	147 ± 3	62 ± 5
PVA	136 ± 5	93 ± 2
polypyrrole	90 ± 7	78 ± 8

Subsequently, we used FE-SEM to characterize the pristine and spent GDEs. In Figure 4a−l, the SEM results for different binding materials are presented. The catalyst is homogeneously distributed on the pristine GDEs, while significant agglomeration is seen on the spent GDEs, in particular, for the less hydrophobic GDEs based on Fumion, Pention, PVA, and polypyrrole. We used ICP-OES analysis to quantify catalyst dissolution, but no Ag particles were found in the catholyte (see Table S1 of the Supporting Information). Hence, the nonuniform distribution of the catalyst particles observed in the SEM images of the spent GDEs is mainly due to agglomeration. The performance loss of CO_2R to CO may partly be attributed to catalyst agglomeration, as it reduces the active surface area and exposes the catholyte to the GDL triggering HER and GDE flooding.

The results discussed thus far do not provide direct evidence for GDE flooding and related performance degradation. To quantify flooding, we performed a cross-sectional FE-SEM-EDX analysis of the spent GDEs (after 4 h of electrolysis). A cross-section of the GDE was prepared with FIB, and FE-SEM-EDX was used to perform a line scan from the CL to the GDL, see Figure S4 of the Supporting Information. The potassium ions were used as a tracer for the penetration of the catholyte into the GDE, a prognostication of flooding. The results of this depth profiling for GDEs with different binders are shown in Figure 5. For all binders, the concentration of potassium increases as a function of GDE depth, which means that the catholyte accumulates inside the pores of the GDE. The highest concentration of potassium is observed for the Fumion-based GDE, which also suffered from potassium bicarbonate precipitation on the GDE surface (zero depth). XRD measurements of the white precipitates, also observed in the SEM image of the spent Fumion GDE, confirmed the presence of potassium bicarbonate on and in the GDE (Figures S5 and S6 of the Supporting Information). The concentration of potassium for the GDEs with different binders shows the following order: Fumion > Pention > PTFE ≈ Nafion ≈ PVA > polypyrrole. Note that we also studied the flooding of the catalyst-free GDE (essentially the Sigracet 28BC GDL only). Obviously, the catalyst-free GDE produced only hydrogen while completely flooding the GDL (black bullets and line in Figure 5). SEM characterization results of the pristine and spent catalyst-free GDEs can be found in Figure S7 of the Supporting Information. For the binders, the penetration of catholyte roughly correlates with the hydrophobicity of the spent GDEs, but with polypyrrole as an exception. Based on the hydrophobicity data of the pristine and spent GDEs, one would expect a high catholyte leakage for polypyrrole, but in contrast, it shows the lowest catholyte penetration. Nevertheless, the better electrolysis performance of the polypyrrole GDE in terms of FE can be explained by the lower flooding tendency. As flooding is limited for polypyrrole, the electrolysis performance decay over time for this type of GDE should come from other factors, such as catalyst agglomeration and binder degradation. We hypothesize that flooding of polypyrrole-based GDEs is inhibited, despite the low hydro-

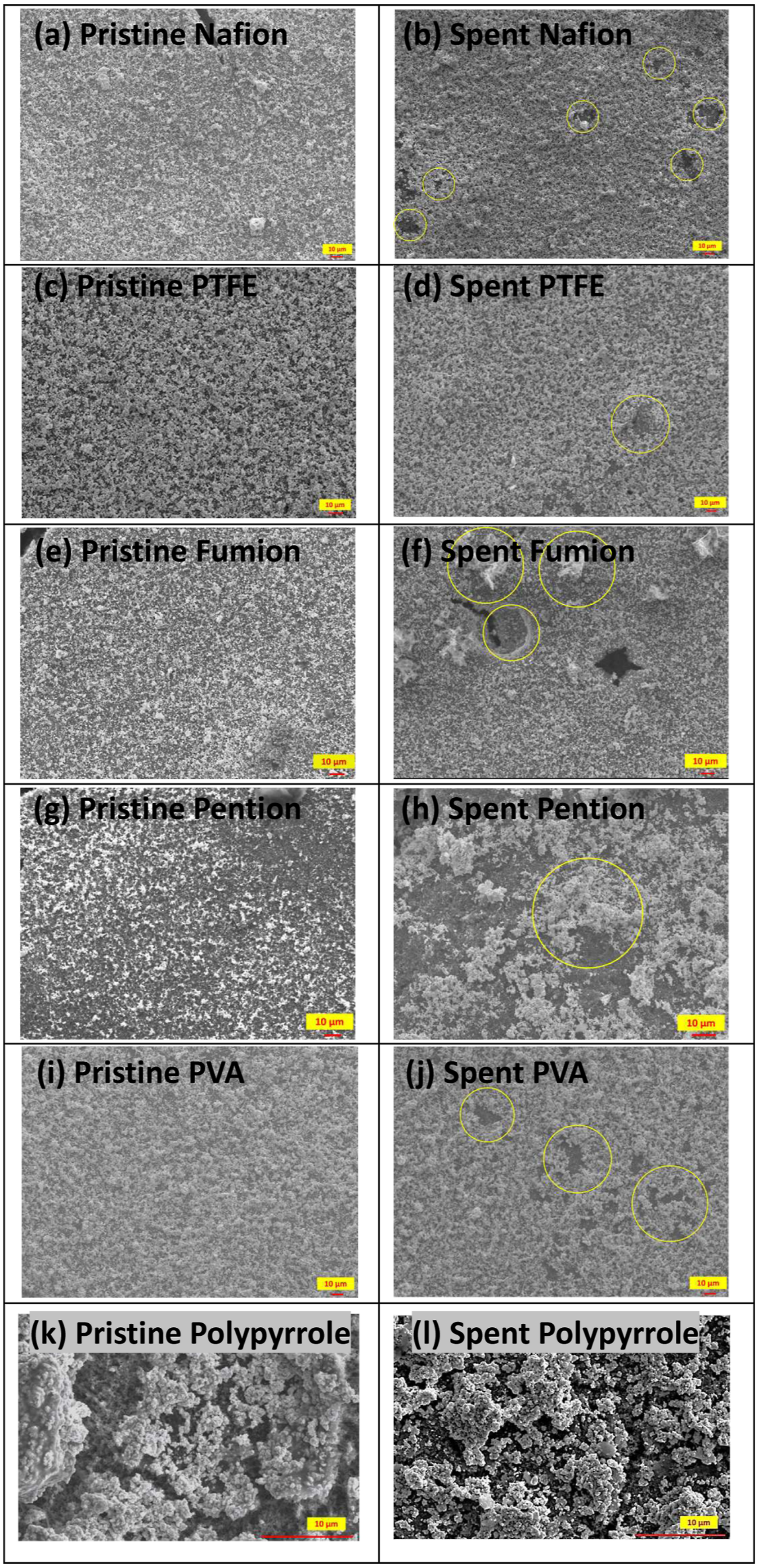

Figure 4. SEM images of pristine (left) and spent (right) GDEs of different binders. (a,b) Nafion-based GDE, (c,d) PTFE-based GDE, (e,f) Fumion-based GDE, (g,h) Pention-based GDE, (i,j) PVA-based GDE, and (k,l) polypyrrole-based GDE.

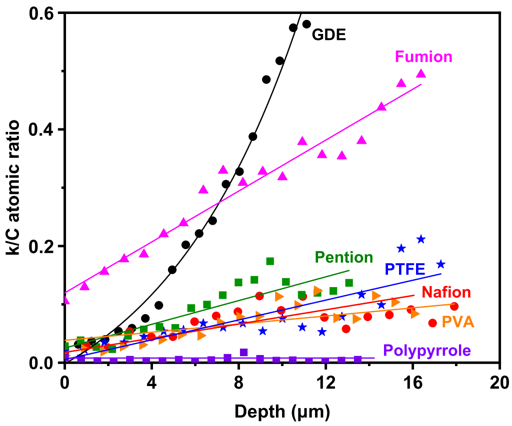

Figure 5. Depth profile of potassium (K) ions in the GDE with varying binding materials measured with SEM−EDX. The pure GDE or GDL (black bullets and line) contained no catalyst and binders. Lines are used to guide the eyes.

phobicity, due to the remarkable properties of polypyrrole coatings (i.e., high ion-exchange capacity, corrosion resistance, electrochromic effects, and redox activity).[43] Polypyrrole-based coatings are known to change wettability and morphology upon application of a redox potential.[44] For polypyrrole doped with dodecylbenzenesulfonate, the coating switch from hydrophilic to hydrophobic when an oxidation or reduction potential is applied, respectively.[45] Hence, the measured hydrophobicity for pristine and spent polypyrrole GDEs under ex situ (oxidizing) conditions does not correspond with the hydrophobicity of the GDEs under in situ (reducing) conditions. In reality, the hydrophobicity of the polypyrrole-based GDEs under CO_2 electrolysis (in situ) conditions might have been higher than the hydrophobicity derived from the contact angle measurements under ex situ conditions. The good stability performance of polypyrrole-based GDEs can be explained on the basis of this hydrophobicity switch phenomena. This plausible explanation is merely a hypothesis, which cannot be verified in the current study, as our setup did not allow for contact angle measurements under operando conditions. It is also surprising to see that Fumion has a better electrolysis performance than PVA, while the former seems to be more susceptible to flooding (i.e., it shows a higher catholyte penetration).

Our analysis shows that the loss of GDE performance is likely a combined effect of flooding, (bi)carbonate precipitation, and catalyst agglomeration. Furthermore, we show that it is not straightforward to correlate the electrolysis performance with the functionalities of the binders. The performance of functional or nonfunctional binders may be affected by many factors (e.g., reactor configuration, type of catholyte and membrane, and operational conditions), which complicate to draw general conclusions on the suitability of one binder type over the other. However, our results confirm that a high initial hydrophobicity of GDEs is not strictly needed for long-term stability, which is in agreement with recent studies of GDE degradation. Future studies should focus more on structure−property and structure−function relationships of binders to correlate functionality with CO_2 electrolysis performance. In addition, more emphasis should be paid to the catalyst ink formulation/components and its impact on GDE stability.

■ CONCLUSIONS

The use of GDEs has enabled high-rate CO_2 reduction to value-added products. Unfortunately, the gain in production rates has come with new challenges related to the long-term stability of the GDEs due to flooding-induced degradation. Here, we investigated the effect of various ionomers (also called binders) on the stability and performance of Ag-based GDEs for the electrochemical reduction of CO_2 to CO. The primary role of the ionomer is to bind the CL to the GDL and provide conductivity, but it can also impact the microenvironment and the properties of the CL such as wettability and water management, gas transport, and electrode morphology. All six investigated binding materials showed a performance loss over time (i.e., the FE of CO decreased while the HER increased over time). To investigate the root cause of this performance degradation, we performed a detailed post-electrolysis analysis of the GDEs using several characterization tools such as contact angle measurements, SEM−EDX, SEM−FIB, and XRD. The contact angles of a water droplet on pristine and spent GDEs show that the hydrophobicity of the CL changes over time as current is applied. The initial hydrophobic GDEs become hydrophilic after 4 h electrolysis time regardless of the used binders. SEM analysis of the pre- and postelectrolysis GDEs show significant catalyst agglomeration. FIB and SEM−EDX were used to perform a cross-sectional analysis of the spent GDEs to track catholyte penetration into the GDL. The concentration of potassium, which was used as a tracer for the catholyte, increased as a function of the GDE depth for all binders, except for polypyrrole, which suggests electrolyte accumulation and flooding of the GDL. For Fumion-based GDEs, XRD analysis showed that a significant amount of bicarbonate precipitates were present on the surface. No generalized conclusions can be presented on the electrolysis performance with respect to the functionality of the binders. The results show that a high hydrophobicity is not strictly needed for long-term stability as polypyrrole, the least hydrophobic but flood-resistant GDE, showed the best performance.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.iecr.4c03279.

> ICP-OES analysis of the catholyte, GC calibration curves, HPLC analysis of the catholyte, repeated CO_2R experiments, contact angle measurements of pristine and spent GDL, FIB-SEM-EDX analysis of spent Fumion GDE, XRD analysis of spent Fumion GDE, and SEM analysis of pristine and spent GDL (PDF)

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Notes

The authors declare no competing financial interest.

Biographies

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