

Supporting information: Accelerated screening of gas diffusion electrodes for carbon dioxide reduction

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Additional electrochemistry and product analysis data

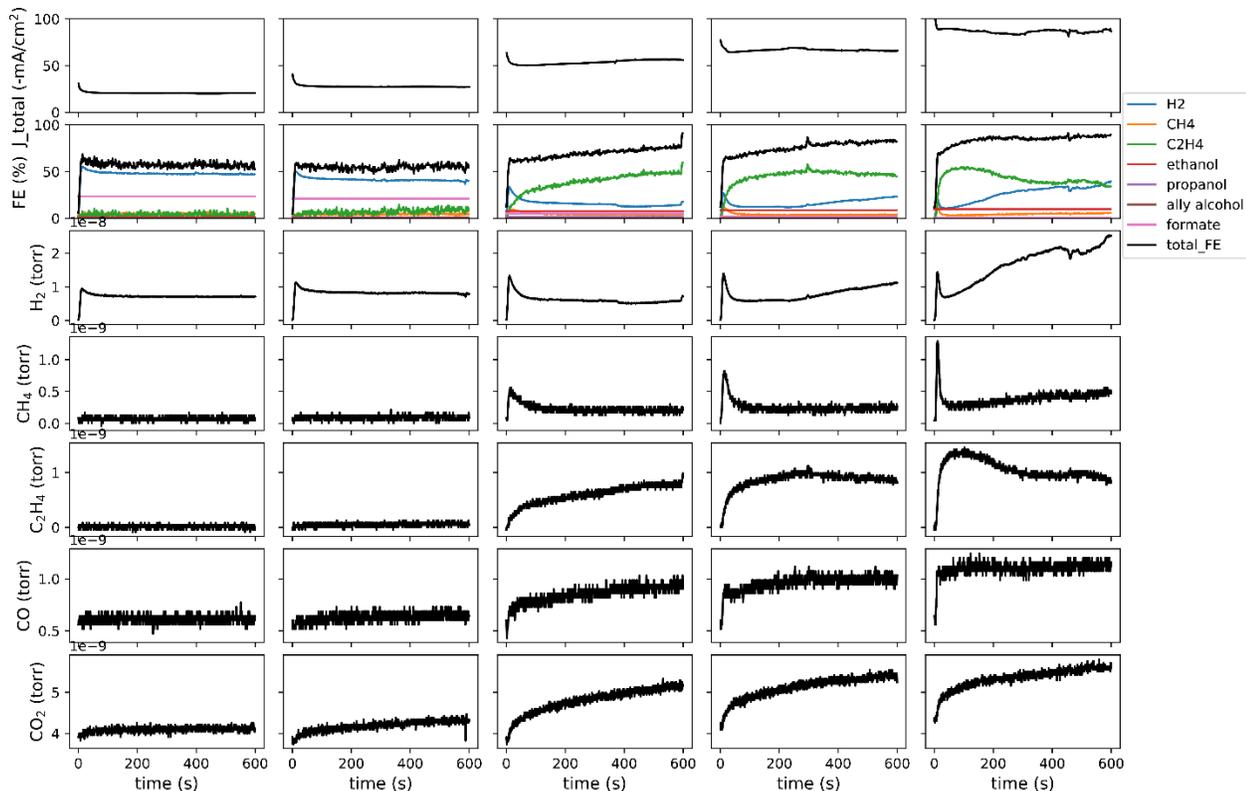


Figure S1. Chronoamperometry time series data. Each column is an experiment with fresh electrolyte performed using the same GDE sample. The potentials (left to right) are -1.5, -1.6, -1.8, -1.9, -2.0 V vs Ag/AgCl (or -0.83, -0.93, -1.13, -1.23, -1.33 V vs RHE, no resistance compensation). The electrochemical current and gaseous product data in Fig. 4 result from the time-average of these data. The data for liquid products in the Faradaic efficiency (FE) panel are horizontal lines corresponding to values assessed from the post-electrochemistry HPLC and GC measurement of aggregated product in the liquid electrolyte. CO production, which is prevalent at the lower overpotential and less so at higher overpotential, likely accounts for most of the difference between the inferred total FE and 100%.

Detailed description of AutoGDE system

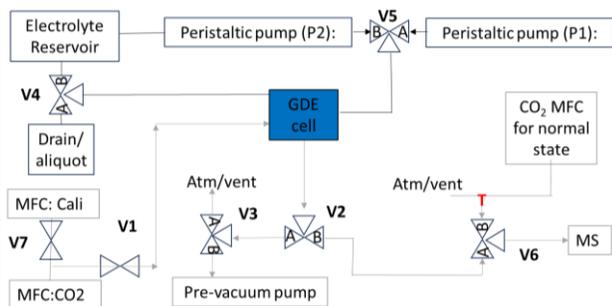


Figure S2. Pump and valve diagram showing the ancillary liquid and gas handling for the GDE cell

AutoGDE modules:

1. The Gas Handling Fixture (GHF) is intended to serve as a static platform to which all other instrument modules are aligned, and to which they apply pressure to achieve suitable seals for full GDE cell operation. It is a PEEK cylinder, with a hole bored through the center which allows the adjustable-force GDL Interface Piston (GIP) to freely actuate around its top plane surface. The PEEK cylinder also has two radial inlet ports which extend from the center bore to the circumference, and contain flat bottom fittings for creating compression seals to external gas sources.

The GHF is mounted securely to structural framing such that it allows for the GIP and WEP to have unrestricted access to both lower and upper planes, respectively. In this way, the GHF remains stationary with respect to the other modules, and allows for its assembly to the other modules to be repeatedly engaged and disengaged via electronically controlled linear actuators. In the Engaged state, the GHF creates a sealed gaseous environment between the radial gasket seal on the GIP and the GDL of the WEF. The bottom plane of the GHF is affixed with a thin compression plate, which facilitates the gasket seal around the circumference of the GIP via axial compression. In this configuration, the mobile range of the GIP is restricted and must be carefully calibrated to the working plane of the swappable Working Electrode Fixture (WEF).

2. The GIP Assembly is securely affixed to a load cell which allows for automated adjustment of the force applied between the top surface of the probe and the GDL. The pair (probe and load cell) is situated on a linear slide (Del-Tron, S3-1.5) and linear actuator with an adjustable force spring coupling them. The adjustable spring pressure allows for the transmitted pressure range to be modulated without the need for feedback from a closed loop linear actuator. This enables repeatable compression of the fully engaged cell during operation and between different working electrode chambers.

The gasket seal around the probe, which restricts axial motion when compressed to a full seal, contributes to the load cell reading, requiring application of a higher load on the GIP to achieve the intended probe-GDE applied pressure. For the demonstration reported herein, the load cell serves to identify engagement of the full AutoGDE assembly, e.g. by failing to reach the pressure indicative of an appropriately sealed cell, the instrument can automatically identify a failure of the compression mechanism and notify the user that instrument inspection is required before continuing the experimental campaign (albeit without computationally identifying the root cause). With this check of cell sealing in-place, we have routinely operated the AutoGDE without observing leaks.

3. The Working Electrode Fixture (WEF) contains the target GDL/catalyst and is assembled prior to placement within the AutoGDE instrument. The main body is constructed of PEEK, with ports to allow for liquid electrolyte handling as well as a conductive insert to enable electrical contact to the catalyst surface on the GDL.

The liquid handling ports are o-ring compression fittings which seal a PEEK tube to the main WEF body. The pumping (and reference electrode) port tube terminates approximately 12 mm from the center of the WEF. The electrolyte manifold port tube extends into the main chamber and is positioned such that the aperture is approximately 0.5 mm from the surface of the membrane when the AutoGDE is fully engaged. The tip is cut at an angle to prevent blockage by the membrane when negative pumping pressure is applied to the WEF.

The reported instrument was fabricated with a copper contact ring permanently fixed to the WEF body. When the AutoGDE is engaged, the GIP compresses the GDL against this contact ring. The position of the copper

contact ring, specifically its coplanar relation to the inner chamber diameter boundary, is paramount to proper operation of the AutoGDE. When engaged, the GDL/catalyst is in simultaneous contact with the electrical pad and the PEEK inner diameter of the WEF. The hydrophobic nature of PTFE GDL's and PEEK restrict liquid electrolyte contact to the inner diameter of the WEF and the GDL/Catalyst surface, which prevents liquid electrolyte from touching the copper contact ring and thereby excludes the contact ring from participating in electrochemistry. Careful fabrication of the contact ring, and co-assembly with the WEF, is necessary to achieve this relation in practice. Electrical access is achieved by mechanically contacting the copper ring with a brass rod, which brings the electrical connection beyond the perimeter of the WEF to enable connection to the potentiostat. Reliable conduction is assisted by using silver paste on the tip of the brass rod during installation. The brass rod is sealed to the WEF via o-ring compression fittings, and connected to the potentiostat Working Electrode during operation.

When the AutoGDE is engaged, the WEF creates a static face seal to the GHF on bottom, and to an ion exchange membrane on top. When the AutoGDE is disengaged, the WEF is able to be removed from the instrument without affecting any of the other modules. This allows quick exchange of a used GDL/Catalyst layer for a new GDL/catalyst layer with minimal human intervention (or robotic automation) by simply exchanging one WEF assembly for another WEF assembly containing a new GDL/catalyst layer.

4. The Counter Electrode Fixture (CEF) is the top most module of the AutoGDE. Similar to the GDL Interface Probe, the CEF is mounted to an adjustable spring loaded linear slide and actuator. Likewise, this configuration also allows for repeatable engagement to, and retraction from, the stationary WEF and GHF without the need for closed loop functionality from the linear actuator. The CEF consists of four main components: a porous frit, a conduit for liquid ingress to the porous frit, a sealing plate to define the electrolyte flow path, and a main body which aligns these components.

The porous frit takes the place of a planar counter electrode in a traditional electrochemical cell. It offers a high electrochemical surface area during operation, while also providing a mechanically stiff backstop to prevent bowing of the membrane during liquid filling and draining of both the CEF and WEF. The pores allow the electrolyte proximal access to the membrane, while any gaseous by-products are removed by the electrolyte recirculation using peristaltic pumps.

The base frit can be purchased commercially in disc form. The disc is then modified by adjusting the outdoor diameter as needed to create a friction fit within the housing of the CEF. Furthermore, an annulus is machined into the face of one side to mate to the electrolyte inlet conduit via mechanical pressure-fitting during fabrication. The most direct method to fabricate this assembly is by traditional subtractive manufacturing (machining), and can be equally accomplished with either milling or lathing techniques. It is paramount during fabrication that the center of the annulus is untouched by machining operations, because the machining process tends to weld the pores of the frit together creating a solid interface, and the inner part of the annulus must remain porous to enable electrolyte flow.

The conduit allows for liquid ingress to the sealed CEF via percolation through pores of the frit during operation. It also provides electrical contact to the porous frit, and is fabricated in concert with the porous frit annulus. It is machined such that the outer diameter of the mating head has press fit interference with the outer diameter of the machined frit annulus. This provides a mechanically robust connection, and allows for both liquid ingress and electrical contact to the porous frit from outside of the sealed CEF during operation.

The conduit acts as a liquid feedthrough with an o-ring compression fitting to seal it to the CEF. It extends approximately 25 mm from the top of the CEF. Flex tubing is then placed on the end of the conduit such that roughly 15 mm of the conduit remains exposed and leads to the electrolyte reservoir. The counter electrode of the potentiostat clips to the exposed portion of the conduit, thus creating electrical contact to the porous frit during operation.

Four flat-bottom ports on top of the CEF allow for compression fittings to seal o-rings around PEEK tubes. These PEEK tubes are similarly fitted with flexible peristaltic pump tubing, each leading to peristaltic pumping, which for the present work involves 4 single-channel pumps. The outflow of each pump routes to the same counter electrode electrolyte reservoir as the conduit inlet tube, which completes the electrolyte recirculation loop.

The assembly of the CEF is completed with the attachment of the flow-field sealing plate. This is necessary because spatial constraints restrict the position of the PEEK exhaust tubes with respect to the porous frit, and the sealing plate creates an environment which allows the peristaltic exhaust pumps to pull liquid through the conduit and appropriately direct it to the membrane interface.

The CEF assembly is mounted to a stiff ring-mount cage, which transmits the motion of the linear actuator and allows access to the liquid conduit and PEEK exhaust tubes for easy disassembly and diagnostics.

5. The AutoGDE instrument is intended to operate indefinitely given sufficient supplies of anolyte and catholyte. Continuous and unmonitored operation therefore requires the ability to modify the gas and liquid flow states via electronic means. Automated gas and liquid flow management is accomplished with the use of electronic pumps and valves. The pumping and valving diagram describes the liquid and gas handling components, where the states of solenoid valves and peristaltic pumps are controlled via computer digital outputs. All pumps, valves, and mass flow controllers (MFC's) are controlled by custom software developed in-house as described in the associated code repositories.

Standard Operating Procedures

Preparation

Before operating the AutoGDE, the GDL/catalyst must be inserted into the WEF. The profile and placement of the GDL is critical. Friction between the circumference of the GDL and the inner wall of the WEF housing pocket is utilized to retain the GDL within the WEF during preparation and transfer to the AutoGDE, however too large a profile can result in the GDL having a tendency to fold on itself, rendering planar contact with the PEEK sealing surface and contact ring impossible to achieve in practice. On the other hand, too small a profile and the GDL will fall out of its place making assembly of the AutoGDE difficult to achieve. Reduced GDL area also allows the possibility of incomplete coverage of the PEEK sealing surface and possible electrolyte contact with the metal contact ring during operation. The goal, therefore, is to create a GDL profile that is “just right”, ie large enough to ensure a friction fit in the WEF housing pocket, leading to full coverage of and contact with both PEEK sealing surface and contact ring, but small enough to avoid folding or otherwise creating non-planar contact with these surfaces when engaged. The authors have empirically determined that circular profiles are inadequate to satisfy all these conditions; small diameter GDLs are not held in place via friction even though they allow for flat contact with and full coverage of the sealing surface and contact ring plane, while diameters larger than the GDL housing pocket necessitate folding during insertion. Instead, the authors have found success

with octagonal profiles whose vertices are circumscribed by a circle equal to 105% of the GDL housing diameter. In the present work, the WEF housing pocket is 0.325" and the GDL profile is defined by a regular octagon circumscribed by a diameter of 0.341".

Although manual profiling (with shears or razor blades, as examples) is adequate to produce these profiles in limited numbers, improved scale up operations are required to reduce the manual labor necessary to produce large numbers of GDL's. The authors have found that machined punches are useful (i.e. leather processing tools machined to the desired profile), however they tend to wear down over time which degrades the cut quality and reproducibility of the GDL profile. They are also difficult to modify if a different sized WEF aperture is required. Therefore, the authors have demonstrated the AutoGDE with GDL profiles processed via laser cutting. Laser profiling results in precise, reproducible, easily modifiable, and mass producible GDL profiles. This technique is restricted to GDL's which are strongly absorbing in the desired laser bandwidth, which comports with the CO2 laser and PTFE GDL's reported herein.

The resting "Normal State" of the instrument is defined by the Mass Spec being isolated from the AutoGDE in a "bypass" configuration, in which the Mass Spec is exposed to a stream of pure CO2 to prevent N2/O2 (Air) spikes from overwhelming the detector. This is accomplished by diverting Mass Spec flow from the AutoGDE system to a dedicated "Normal State MFC" via V6B. This "Normal State MFC" is set manually before and turned off after a full AutoGDE campaign. Its existence is critical to proper operation so it appears in the instrument schematic, however it does not appear in the experiment code.

Operation

#. **Step Name**

Action Order

Explanation/Description

1. **Liquid Fill to GDL/Catalyst**

V2A ON→V3B ON→V4B ON→Wait→2A OFF→ 3B OFF→ 4B OFF

Electrolyte must be introduced to the WEF in such a way that liquid contact to the GDL/Catalyst surface is guaranteed. This is accomplished by pulling high flow rate (relative to MS during operation) vacuum on the GIP, which transmits into the WEF due to the porosity of the GDL. Liquid will then get pulled into the WEF via V4B until it comes into contact with the GDL/Catalyst surface. The hydrophobicity of the GDL then prevents liquid from crossing over to the GIP/GHF while guaranteeing contact between the Catalyst and Electrolyte.

2. **WEF Fill**

4A ON→5B ON→ P2ON →Wait→ P2OFF→ 5B OFF →4B OFF

Electrolyte is then pushed into the cell to ensure contact with the membrane via P2. The reference electrode is housed in this same inlet line between V5 common and the WEF to establish a 3-electrode measurement configuration..

3. **GHF Purge**

1 ON →2A ON →3A ON→ CO2 MFC ON (Purge Rate) →wait→ 2B ON→ 6B OFF 6AON →wait

CO₂ quickly flows through the GHF to purge residual gas remaining in the system. This Purge rate is faster than the CO₂ flow during measurement to decrease the time needed to flush the full volume of the cell.

4. Set CO₂ Operational Flow Rate

CO₂ MFC (Operational Rate)

The CO₂ flow rate is lowered to be commensurate with the MS inlet flow rate during operation/measurement.

5. MS Background (Normal State)

CO₂ MFC OFF → 1 OFF → 2A OFF → 3A OFF → 2B OFF → 6A OFF → 6B ON → 7 OFF

MS Signal check with direct flow of CO₂ process gas (not from AutoGDE). Helps with diagnosing issues from either the MS or the AutoGDE and gives a baseline to monitor signal changes.

6. WEF Drain

4B OFF → 5B OFF → 4A ON → 5B ON → P1 ON → Wait → P1 OFF → 5A OFF → 4A OFF

Initialization drain so that the first sample in the following loop sees the exact same filling/draining procedure as all subsequent fill/drain cycles within the loop.

7. FOR LOOP BEGIN (User Defined Range)

This loop performs diagnostic CV profiles to evaluate catalyst performance.

8. Liquid Fill

4A ON → 5B ON → P2 ON → Wait → P2 OFF → 5B OFF → 4B OFF

Fill with new liquid.

9. GHF Purge

1 ON → 2A ON → 3A ON → CO₂ MFC ON (Purge Rate) → wait → 2B ON → 6B OFF 6A ON → wait

Purge residual gas from previous CA.

10. Set CO₂ Flow Rate

CO₂ MFC ON (Operational Rate)

Set CO₂ operational Flow Rate.

11. Perform CV Fast

PStat Potential Sweep Fast

Perform a CV at 50mV/s or other electrochemistry experiment.

12. Perform CV Slow

PStat Potential Sweep Slow

Perform a CV at 20mV/s or other electrochemistry experiment.

13. Isolate WEF (Normal State)

CO₂ MFC OFF → 1 OFF → 2A OFF → 3A OFF → 2B OFF → 6A OFF → 6B ON → 7 OFF

Check MS Signal with direct flow of CO2 process gas .

14. Drain Liquid

4B OFF → 5B OFF → 4A ON → 5B ON → P1 ON → Wait → P1 OFF → 5A OFF → 4A OFF

Drain liquid to waste by pushing air via P1 through V5A and V4A.

15. FOR LOOP END → Back to 7

Repeat the loop a user defined number of times, then move on

16. .FOR LOOP BEGIN (loop for each CA)

Begin the defined CA profiles. Each CA is performed with a new liquid fill.

17. WEF Fill

4A ON → 5B ON → P2 ON → Wait → P2 OFF → 5B OFF → 4B OFF

Fill WEF with new, fresh, electrolyte in preparation for the next CA profile.

18. GHF Purge

1 ON → 2A ON → 3A ON → CO2 MFC ON (Purge Rate) → wait → 2B ON → 6B OFF 6A ON → wait

Purge residual gas from the system. Gas flows simultaneously from the GIP to atmosphere via V2A to V3A, and to the MS via V2B and V6A. During purge, a majority of the gas flows to atmosphere (Purge Rate >> MS flow rate).

19. GHF Flow Rate Set

CO2 MFC ON (Operational Rate)

CO2 flow is set only slightly higher than MS inlet flow, causing a majority of the CO2 flow to go to the MS, and a minority to atmosphere.

20. Perform CA

PStat Potential Set

Perform chronoamperometry or other electrochemistry experiment.

21. Isolate Cell (Normal State)

CO2 MFC OFF → 1 OFF → 2A OFF → 3A OFF → 2B OFF → 6A OFF → 6B ON → 7 OFF

Isolate AutoGDE from MS to allow for the WEF drain/fill cycle, and to check MS signal with a known CO2 source.

22. Liquid Drain

4B OFF → 5B OFF → 4A ON → 5B ON → P1 ON → Wait → P1 OFF → 5A OFF → 4A OFF

Use air to push the electrolyte into an archive vial via P1, V5A, and V4B..

23. FOR LOOP END → Back to 16

Repeat Above loop for each CA potential, then move on.

24. Electrolyte Fill

4A ON→5B ON→ P2ON →Wait→ P2OFF→ 5B OFF →4B OFF

Last WEF fill to perform calibration gas measurement in an environment commensurate with the operational measurement.

25. GHF Purge

1 ON →2A ON →3A ON→ CO2 MFC ON (Purge Rate) →wait→ 2B ON→ 6B OFF 6AON →wait
Purge residual gas with CO2.

26. Calibration Gas Introduction

V7 ON →Cali MFC ON (Calibration Rate)→ Wait → Cali MFC OFF→ V7 OFF

Introduce known calibration gas to the system via V7.

27. Isolate WEF (Normal State)

CO2 MFC OFF→ 1 OFF →2A OFF →3A OFF →2B OFF→ 6A OFF→ 6B ON

Isolate the MS from the AutoGDE.

28. Liquid Drain

4B OFF→ 5B OFF →4A ON→ 5B ON→ P1 ON →Wait→ P1 OFF→ 5A OFF →4A OFF

Drain remaining liquid.

29. All Off

All MFC OFF→ All Valves OFF

Turn off the system to its rest state.

30. END

Bill of Materials

Table S1. Major system components and estimate cost

Category	Item	Detail	Estimate cost (USD)	Quantity	Total
Module fabrication and hardware	WEF material and component Machining		1200	1	\$1,200
	CEF material and component Machinig		300	1	\$300
	GHF material and component Machining		150	1	\$150
	GIP Material and Component Machining		100	1	\$100

	CEF Motion Control Auxiliary Components and Machining		600	1	\$600
	GIP Motion Control Auxiliary Components and Machining		700	1	\$700
	Linear Actuators	Firgelli FA-AL-200-12-12	154.95	2	\$310
	T-Slot Railings (To mount each module and electrolyte reservoirs)	McMaster-Carr	200	1	\$200
	T-Slot Brackets and Fasteners	McMaster-Carr	50	1	\$50
	Load Cells	Omega LC8150-250-100	843.29	2	\$1,687
Gas management	Alicat MFC		1250	3	\$3,750
	Fluid valves (3 way Selection Valves, BioChem)	080T312-62-5	435.18	5	\$2,176
	Fluid Valves (Isolation Valves, NResearch)	HP225T011	91.62	2	\$183
	Flat bottom fittings with Ferrules	Cole Parmer	5.5	30	\$165
	Alicat Power Cables		55	4	\$220
	Peristaltic Pump		1018	6	\$6,108
Liquid management	Tubing/tubing for organic solvent	25ft/pack	88.5	2	\$177
	PEEK Tees and chemically compatible fluidic connectors	Tees, fittings	46.4	2	\$93
	Chemically Compatible O-rings		5	2	\$10
Computer and electronics	Computer		750	1	\$750
	5V Power Supply	Omron S8VK-G03005	76.61	1	\$77
	12V Power Supply	Omron S8VK-G06012	89.06	1	\$89
	Wago Distribution Panels	2946-2001-1401-ND	2.13	10	\$21
	Various Electrical Wiring components		50	1	\$50
	Relays	Crydom DRA	57.24	15	\$859
	Powerstrip		20	2	\$40
	NI DAQ	cDAQ-9174	1501	1	\$1,501
TOTAL					\$21,565

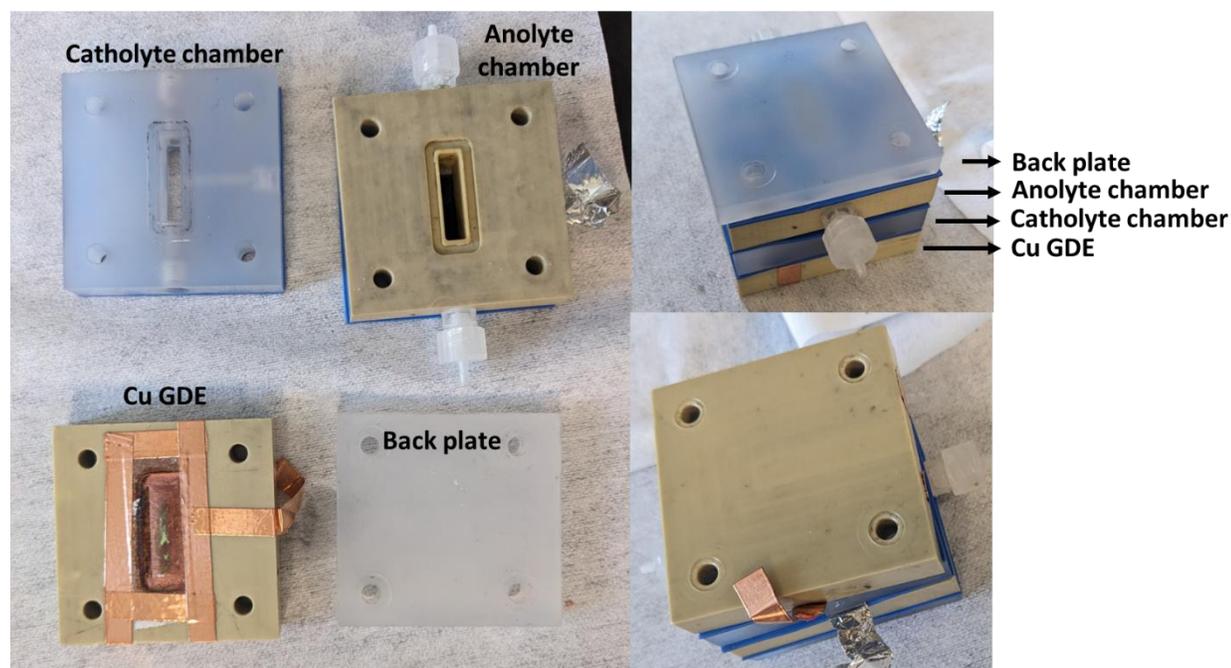
Table S2. CO₂RR performance of Cu GDE evaluated by AutoGDE and conventional GDE cells¹ in 1M KHCO₃

Potential (V vs RHE)	J (- mA/cm ²)	FE_H ₂ (%)	FE_CH ₄ (%)	FE_C ₂ H ₄ (%)	FE_alcohol (%)	FE_C _{≥2} (%)	FE_formate (%)	Reference
-0.83	20.7	47.5	4.8	3.6	0.7	4.3	23.5	This work
-0.93	27.3 ± 0.5	34.8 ± 8.6	4.0 ± 0.3	8.1 ± 1.7	2.2 ± 0.5	10.2 ± 2.2	19.3 ± 2.4	This work
-1.13	55.0 ± 2	13.7 ± 2.6	4.4 ± 0.2	39.2 ± 0.5	9.1 ± 0.9	48.3 ± 0.4	5.4 ± 0.4	This work
-1.23	73.7 ± 10	11.8 ± 5.9	3.8 ± 0.4	49.7 ± 6.5	10.1 ± 0.4	59.8 ± 6.9	2.6 ± 0.8	This work
-1.33	86.7	25.7	4.6	42.3	10.3	52.6	0.5	This work
-0.9	45 ± 1	22 ± 4	17 ± 1	19 ± 4	13 ± 1.4	32 ± 4	17.4 ± 0.4	(1)
-1.05	65 ± 1	27 ± 1	4 ± 5	17 ± 1	15.1 ± 0.9	33 ± 2	23 ± 7	(1)
-1.15	84 ± 5	15 ± 3	4.4 ± 8	22 ± 7	18 ± 5	40 ± 8	19 ± 4	(1)

Comparison of operations between conventional and AutoGDE:

Conventional GDE:

Conventional GDE design/setup may be different from one to another group, however, it generally looks like below¹:



The assembly steps are commonly described below:

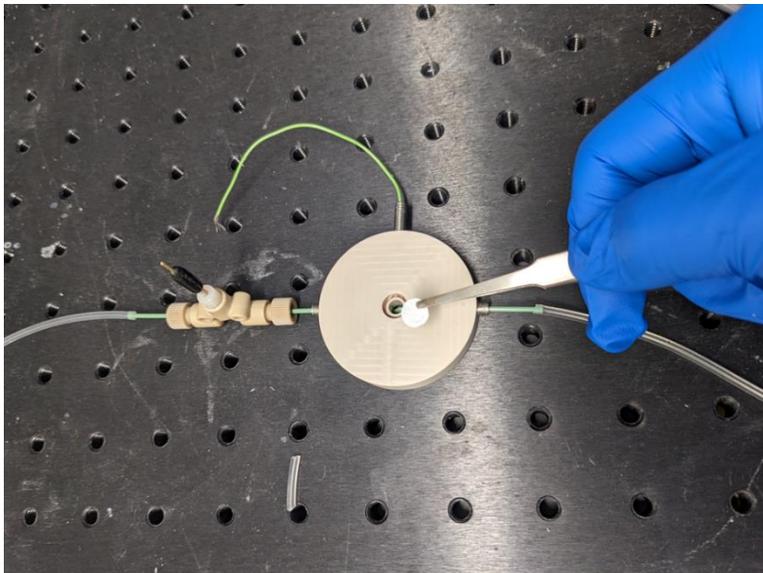
1. Cut GDE and tape it onto the WE fixture cell

2. Put (assemble) GDE cell pieces together including membranes and use screw to compress/tighten the cell. The compression force may be different from one to another run, which is different from autoGDE where the compression force by robot is consistent
3. Start pumping catholyte and anolyte and do recirculation
4. Start to flow reactant gas (i.e. CO₂) and wait for system to reach equilibrium
5. Start electrolysis experiments
6. Undo step 2 and then step 1
7. Start over from step 1 or step 4 (depending on if replacing GDEs) for the next experiment

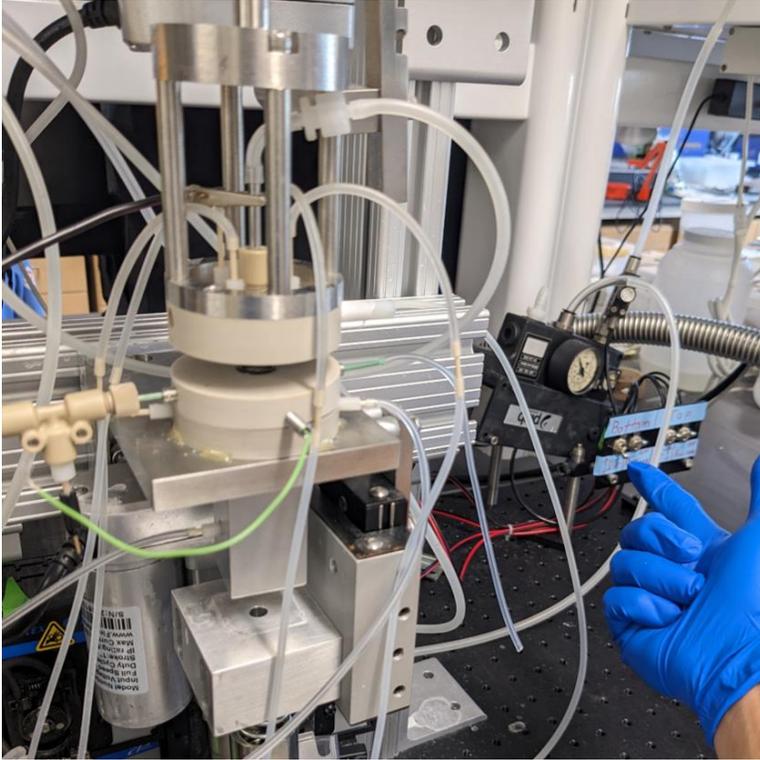
AutoGDE:

The major difference between auto and conventional GDE cells is step 1, 2 and 6 (assemble and disassemble), where conventional GDE would take up to 20 mins to complete these steps (although highly depending on operators). Since the conventional GDE cells are put together by compression with screws by human operation, a leak test is required after assembly. Furthermore, it is very difficult to have these 2 steps automated. For autoGDE, however, it takes only less than 30 (s) (before robot arm is implemented to make it full automation) to assemble/disassemble the cell. The steps are as followed:

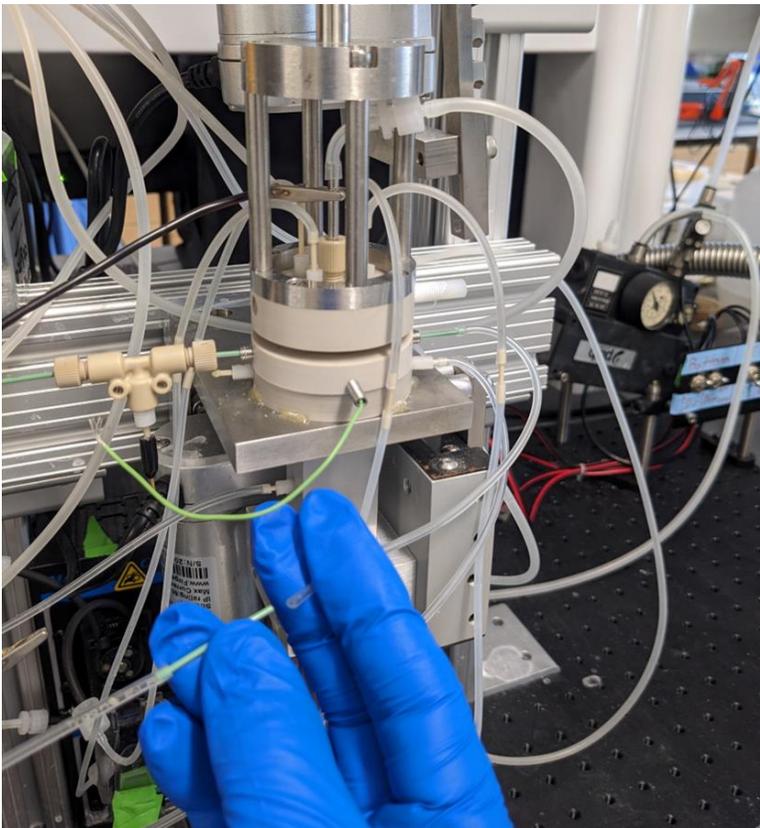
1. Place GDE into the GDE WE fixture



2. Compressed the cell by flipping two power buttons



3. Connect both ends of the tubing of the WE fixture



4. Start to flow reactant gas (i.e. CO_2) and wait for system to be equilibrium

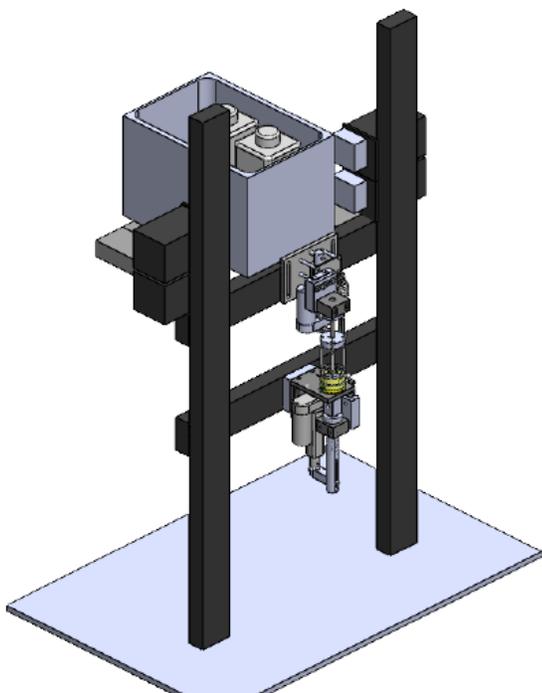
5. Start electrolysis experiments
6. Undo step 2 and then step 1
7. Start over from step 1 or step 4 (depending on if replacing GDEs) for the next experiment

The AutoGDE goes through similar steps as conventional cell for step 3 to 4, and the time for step 3 to 4 are highly depending on the downstream analytical chemistry instrument used. Step 5 electrolysis experiment commonly takes 5 to 15 mins depending on electrochemical techniques used for evaluating performance of GDEs. Comparing the time needed between conventional and autoGDE, it could be up to 4x faster (i.e. 5min electrolysis +assemble/dissemble time) per experiment for autoGDE with continuous GDE screenings.

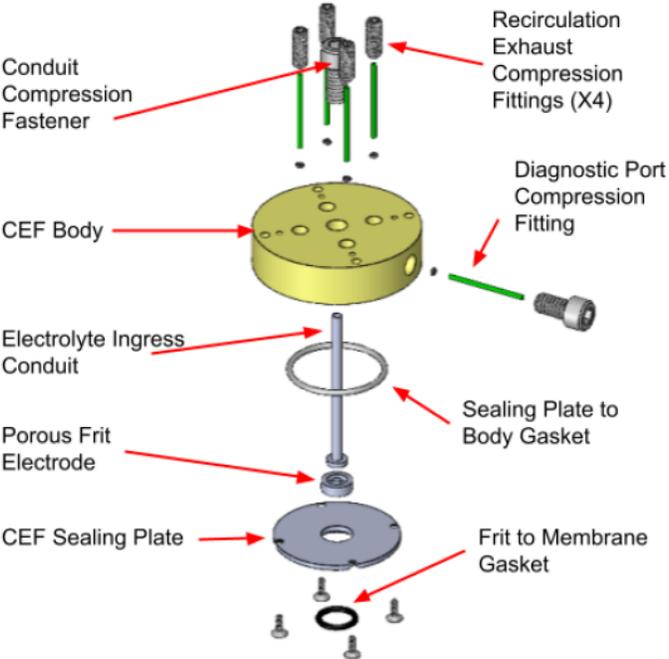
Abridged description of AutoGDE assembly:

Please see the repository (<https://data.caltech.edu/records/f40n8-cv274>) for the full assembly instructions in “AutoGDE Assembly.pdf” and additional files with machine drawings and port callouts. Key visuals for illustrating the construction of AutoGDE are provided here for convenience.

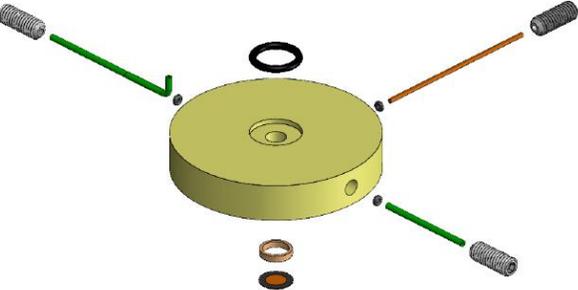
AutoGDE system including its mechanical infrastructure. The electrochemical cell from Fig. 1 of the main paper is the cylindrical stack of components (yellow):



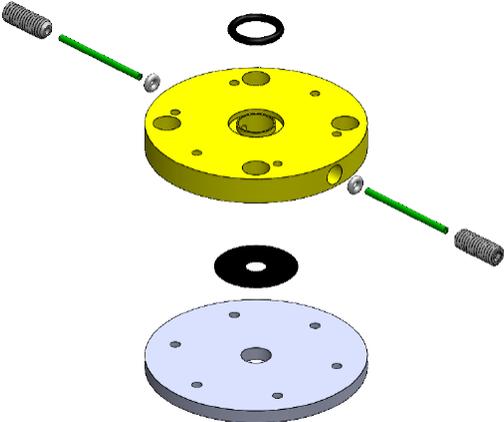
Exploded view of the counter electrode fixture:



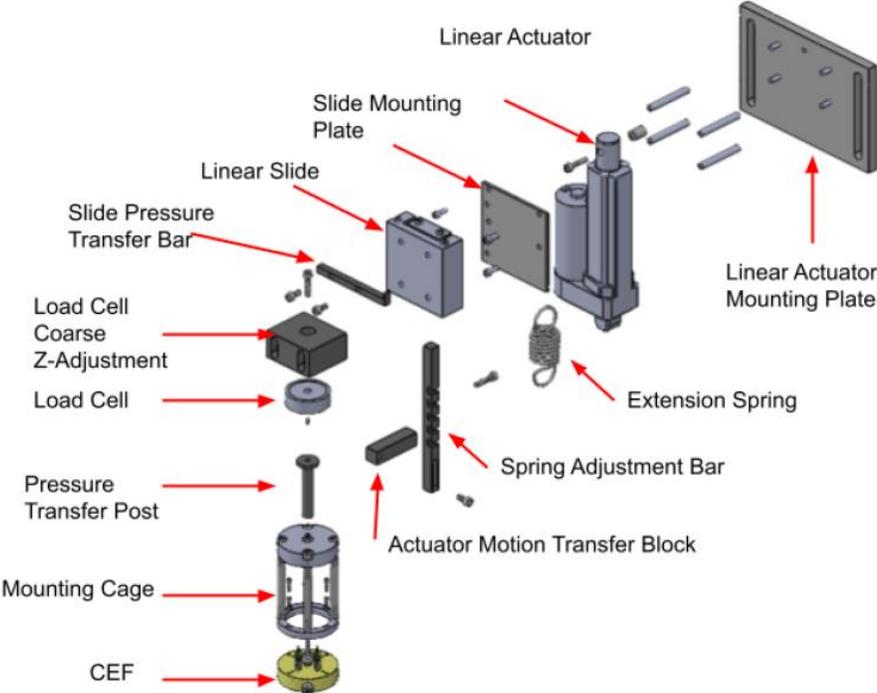
Exploded view of the working electrode fixture:



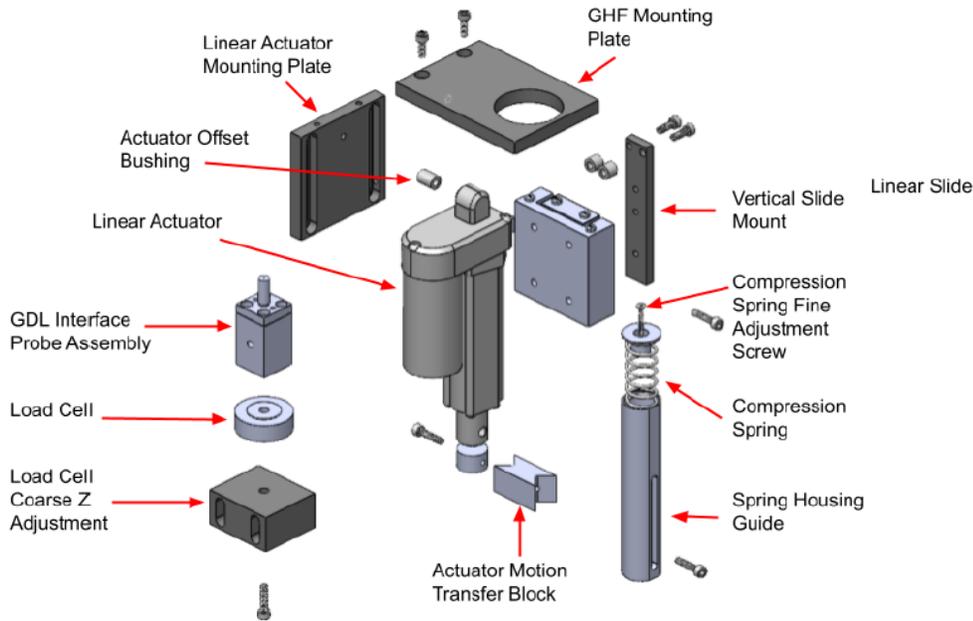
Exploded view of the gas handling fixture:



Exploded view of the actuator for the counter electrode fixture:



Exploded view of the actuator for the gas handling fixture:



References:

1 G. P. Heim, M. A. Bruening, C. B. Musgrave, W. A. Goddard, J. C. Peters and T. Agapie, *Joule*, 2024, **8**, 1–10.