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Does one need to polish electrodes in an eight pattern? Automation provides the answer†

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Automation of electrochemical measurements can accelerate the discovery of new electroactive materials. One of the hurdles to automated electrochemical measurement is the pretreatment of electrodes because mechanical polishing is usually conducted manually. Here we investigate the automation of electrochemical measurements using a robotic arm. We demonstrate automated mechanical polishing using a station with a moving polishing pad and evaluate the effect of different polishing patterns. Our automatic method improved the corroded electrodes, and we found the effect of pattern was not significant, which diverges from the current common belief amongst practitioners that a figure eight pattern is best for pretreatment. This research is a step toward automating electrochemistry experiments without human intervention.

Introduction

Voltammetry techniques such as cyclic voltammetry (CV)¹ and differential pulse voltammetry (DPV)² are ubiquitous in

electrochemical characterization workflows. In typical CV experiments, solid electrodes such as platinum, gold, silver, or carbon are commonly used as the working electrode, and the electrochemical process occurs at their surface. The surface of the solid electrode can be degraded by the adsorption of solution species or the formation of oxide layers.³ Therefore, a clean, flat, well-defined electrode surface is required to minimize non-faradaic processes and give consistent and reproducible results.⁴ Diverse methods, including sonication, (electro-) chemical treatment, and metal-oxide dispersion have been developed to reach precise levels of surface flatness, and the most common method is mechanical polishing. However, polishing is typically a laborious and time-consuming process performed manually.⁵ The disparity between polishing protocols and operator habits introduces significant errors, leading to inconsistent data across reporting sources. Thus, the automation of polishing techniques would reduce the burden of chemists when conducting experiments, while improving the reproducibility of electrochemical experiments. This is further amplified in an end-to-end automated electrochemistry process, where fully automated polishing using robotic systems could unleash the potential of high throughput experimentation (HTE) and eventually help realize a self-driving lab (SDL) for human-off-the-loop electrochemistry research.^{6–8}

In this work, we built a robotic polishing system that combines a robotic-handled polishing station and a portable potentiostat^{*} (Fig. 1). We demonstrated our polishing method with a glassy carbon (GC) electrode. Such electrode type is the most commonly used in electrochemical studies,⁹ especially in experiments requiring high over-potential and wide electrochemical windows. It is reported that the surface of GC gradually deactivated over time possibly because of the adsorption of impurity in the sample solution¹⁰ and the electrochemical corrosion of the electrode surface,¹¹ leading to evolving voltammograms over successive scans. Therefore, proper surface preparation is required for accurate electrochemical measurement, and various polishing protocols with different conditions have been proposed.^{10,12,13} All these features make GC ideal for

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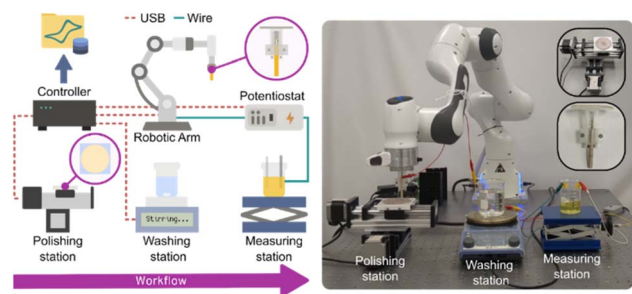


Fig. 1 Automated electrode polishing workflow. The robot moves the electrode between the polishing, washing and measuring stations to automatically activate the electrode by polishing and run cyclic voltammetry measurements using the potentiostat. The complete procedure is conducted without human intervention.

polishing studies. In this paper, we develop an automated polishing system and investigate the effect of polishing pattern, because of the contradictory opinions in the literature. Some literature¹³ recommends circular motions for electrode polishing, while other literature² doesn't recommend a circular shape and suggests using eight-figure motions to minimize potential periodic patterns introduced in circular polishing. The eight-figure protocol is commonly utilized in most laboratories, but no systematic evaluation has been made to the best of our knowledge. With our robotic system, we successfully demonstrated a fully automated electrochemical measurement and electrode polishing workflow. Further evaluation of the polishing conditions suggests that polishing pattern does not significantly affect polishing quality.

Method

We developed and integrated our robotic polishing system with a 7-axis robotic arm, a 2-dimensional polishing station, a washing station, an electrochemistry measuring station, and a mini-potentiostat, all of which were controlled with a single controller to enable end-to-end automation workflow without human intervention. The detail of each component is described in the following sections.

Robot arm

Franka Emika Robot (Franka Emika, Germany) was used as the robot arm. A 3D-printed jig to hold an electrode (Fig. 2) was mounted as an end-effector of the robot arm. The electrode holding part moves vertically and applies a force to an electrode by a spring. The jig was printed by a Form3L 3D printer (Formlabs, USA) and sanded and greased to decrease friction. The design of the jig is available on our GitHub repository (<https://github.com/aspuru-guzik-group/electrode-polishing>).

Polishing station

Initially, we attempted to polish the electrode over a fixed polishing pad similar to human polishing protocols. However, noticeable fluctuations on the Z axis were recorded, which may lead to significant changes in the contact force between the

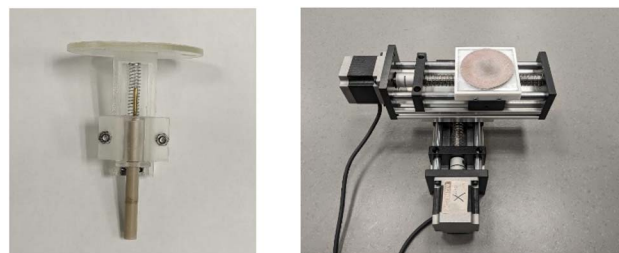


Fig. 2 The 3D-printed electrode jig (left) is attached to the robot hand and applies a stable force to the electrode with the spring. The polishing station (right) is built with two linear actuators. It moves the polishing pad in different patterns.

electrode surface and the polishing surface, possibly causing damage to the electrode. To minimize this error and improve the consistency of polishing experiments, we introduced a polishing station that allows XY movements with a stable Z coordinate, therefore allowing for smooth polishing with stable contact force provided by a 3D printed electrode jig with spring mounted on the robotic arm at a fixed position. The polishing station (Fig. 2) moves an Alumina Polishing Pad (BASi, USA) in two axes by two linear actuators. Alumina suspension (0.05 μm) was dripped on the polishing pad as the abrasive before starting each experiment. EBX1605-100 mm (RATTMMOTOR, China), driven by a Nema23 stepper motor, was used as a linear actuator. Each actuator was controlled by a DM542 2-phase stepper motor driver (Mootea, China). The motor driver receives pulse signals from an Arduino Uno board connected to a robot controller PC *via* a USB cable. A Python interface is provided to communicate with Arduino, driving the polishing station to move the polishing pad in pre-programmed Lissajous patterns,¹⁴ providing diverse polishing types such as linear, circular, and 8-shaped motions. Both Arduino and Python interface codes can be found in the GitHub repository.

Electrochemistry measurement

The quality of the electrode was evaluated by electrochemical measurements. Cyclic voltammetry measurements were performed with a standard 3-electrode system using a portable potentiostat. A planar glassy carbon electrode (BioLogic) was chosen to be studied. Silver wire was chosen as the pseudo-reference and counter electrodes. All measurements were performed on a standard solution containing 0.01 M $\text{K}_4[\text{Fe}(\text{CN})_6]$ analyte, 0.5 M Na_2SO_4 electrolyte and 0.25 M HOAc/NaOAc buffer. In each measurement, five cycles were conducted in an electrochemical window between -1.0 V to 1.0 V at a scan rate of 500 mV s^{-1} . The images of the electrode were taken by a microscopic camera (a2A4096-9gcBAS, Basler) with a $1.000\times$ magnification lens (MC100X, Opto Engineering). For the manual polishing experiment, a trained graduate student conducted polishing with an 8-shape motion. The integral of the CV plot was calculated for the last cycle of measurement.





Fig. 3 Photo of the surface of a glassy carbon electrode before (left) and after (right) corrosion, which was taken during our preliminary experiment. The electrode was applied a 3.3 V potential for 600 seconds inside 0.5 M sodium sulphate solution. Note that a higher voltage (5 V) was used in the actual experiment to cause corrosion quickly and in a controlled manner.

Results

We first evaluated our robotic polishing system by repeating electrochemical measurements for a standard solution with unpolished and polished electrodes. A high voltage (5 V, 30 s) was applied to the electrode to induce corrosion. It is reported that carbon oxidation occurs above 1.5 V. The CV plot changes as a result of increased electrochemical capacitance caused by corrosion,¹¹ and further confirmed by in-house measurement using microphotography (Fig. 3). The polishing was done by repeating the following steps: (i) the robot brings an electrode to a polisher and fixes its position, (ii) the polishing station moves around the electrode for 30 seconds, (iii) the robot brings the electrode to a measurement station, and (iv) surface condition is evaluated using electrochemical methods including CV and cyclic differential pulse voltammetry (CDPV). The process was repeated 10 times. Throughout our experimental process, we evaluated four polishing motions: eight-figure, circular, linear, and complex. They are implemented as Lissajous curves (Fig. 4).

To evaluate the polishing performance, the integrals of the CV plots are plotted against polishing time in Fig. 5 to provide quantitative information to relate the polishing time and the electrode quality. The major contribution to current measured in CV experiments includes electrochemical current, ohmic current, and charging current, with the charging current closely related to the surface properties including roughness and surface oxidation.¹¹ The electrochemical and ohmic currents remain constant between experiments using the same reference solution. Therefore, the integral corresponds to the capacity of the electrode and can be used as a measure of the surface quality.

Discussion

The decreasing CV integration indicates mechanical polishing decreases the capacitance of the electrode surface, which is in line with the long-standing practice of improving electrode surface conditions *via* mechanical polishing. The electrode surface was generally regenerated in 120 seconds of polishing for the damage we applied with over potential. However, a slight improvement could be observed in several cases. In addition to

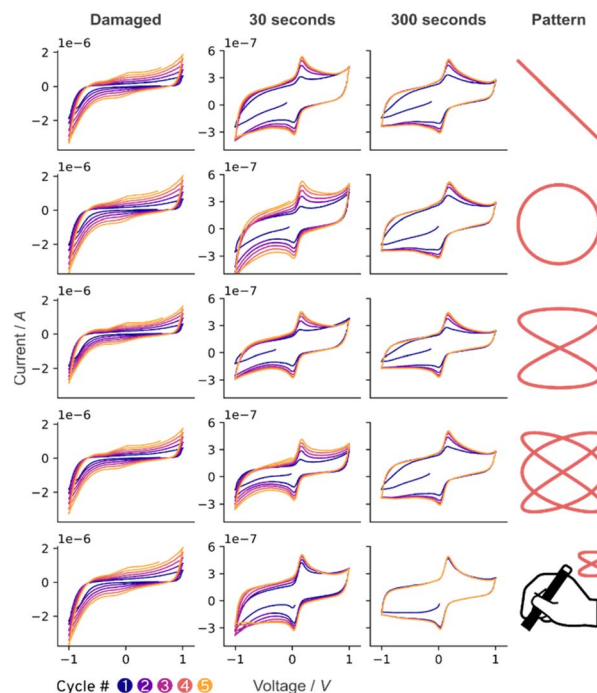


Fig. 4 Cyclic voltammetry plot of damaged and polished electrode. The plots for electrodes damaged by a high voltage, electrodes polished for 30 and 300 seconds are shown. Four different polishing patterns (eight-figure, circular, linear, and complex) were tested for robotic polishing. Manual polishing with an eight-figure shape is shown as a baseline.

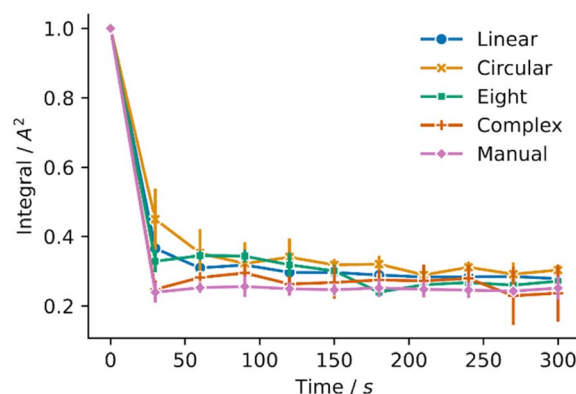


Fig. 5 Overlaid integration of the CV plots over accumulated polishing time. The integrals (the area inside CV plots, normalized by the unpolished value) for different polishing patterns are shown.

polishing duration, the pattern has long been believed to play an essential role in retrieving the best surface conditions. We implemented four mechanical polishing protocols as well as traditional manual polishing. Surprisingly, the four polishing patterns performed similarly within the error range (Fig. 5). This contradicts a long-standing belief that eight-shape polishing outperforms simple patterns such as linear or cyclic motions. However, we need to point out that the robot could apply constant force by using the jig regardless of the polishing



patterns, and it might lead to no significant difference in polishing patterns.

Notably, the performance of robotic polishing was similar to that of manual polishing, with no statistical differences after polishing for 60 s (Fig. 5 pink line). This result indicates our robotic arm system enables the automation of electrochemistry measurement without human intervention. The mechanical polishing was typically conducted by a human, which prevented the automation of the whole measurement pipeline which requires routine refreshment of electrode surfaces for data consistency and quality. With automated polishing, the robot can measure a series of samples with a polished electrode. Noteworthy, in this study we developed a relatively complex polishing system that has two actuators. However, considering our evaluation results that indicate the effect of polishing pattern is not significant, a simpler design might be sufficient for electrode polishing. For example, moving a polishing pad in a circular motion with one motor is a possible design. We also need to note that the first cycle in the CV plots showed a deviation from other cycles for electrodes polished by the robot, while the deviation was negligible for manually polished electrodes. A mechanochemical effect on the electrode surface might cause this. The robot applies more consistent contact forces and geometric positions between the electrode surface and the polishing pad during polishing, leading to a more organized surface state. Another possibility is that a small difference in the evaluation protocol affected the measurements. For example, the electrode was held by a robot during measurement in the robotic polishing evaluation while it was placed in a beaker without a holder in the manual polishing evaluation. We will investigate more about the cause of the difference in the initial cycle by running experiments under more diverse conditions. The changes appreciated during the visual inspection of the electrode suggested a link between the appearance of the electrode and the polishing quality. Still, this connection remains unexplored due to the limited scope of this work.

Automated polishing has potential applications well beyond simple CV measurement. For example, we can investigate electroplating by removing the plated materials from the surface. Studying the corrosion of metal is another potential area of application. This work opened the door to the unexplored areas of automation research.

Conclusions

We constructed an automated electrode polishing system that uses a robotic arm, a polishing station, and an electrode jig. The evaluation experiment on a glassy carbon electrode has shown that the proposed system successfully improves the surface quality of a corroded electrode. The result was similar to manual polishing, and the polishing pattern's effect was insignificant. This result opens the way to fully automated electrochemistry experiments with carbon electrodes. Robotic polishing is more reproducible than manual polishing, which leads to improved reproducibility of overall experiments. Combined with a sample preparation system, the robot can

conduct multiple CV measurements in a row with a polished electrode, which can be applied for the characterization of efficient high-throughput screening of battery molecules.¹⁵ We will further work on end-to-end automation of electrochemical experiments.

Data availability

The code and design files for this paper can be found at <https://github.com/aspuru-guzik-group/electrode-polishing>.

Author contributions

Conceptualization: NY, HH; methodology: NY, GDA, SPG, CY, HH; investigation: NY, HH; visualization: NY, SPG; supervision: HH, AAG; writing—original draft: NY, GDA, HH; writing—review & editing: SPG, CY, AAG.

Conflicts of interest

There are no conflicts to declare.

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