Cross-Facility Orchestration of Electrochemistry Experiments and Computations

Anees Al-Najjar
alnajjaram@ornl.gov
Computational Sciences and Engineering Division
Oak Ridge National Laboratory
Oak Ridge, TN, USA

Nageswara S. V. Rao
raons@ornl.gov
Computational Sciences and Engineering Division
Oak Ridge National Laboratory
Oak Ridge, TN, USA

Craig A. Bridges
bridgesca@ornl.gov
Chemical Sciences Division
Oak Ridge National Laboratory
Oak Ridge, TN, USA

Sheng Dai
dais@ornl.gov
Chemical Sciences Division
Oak Ridge National Laboratory
Oak Ridge, TN, USA

ABSTRACT

Instrument-computing ecosystems supporting automated electrochemical workflows typically require the integration of disparate instruments such as syringe pump, fraction collector, and potentiostat, all connected to an electrochemical cell. These specialized instruments with custom software and interfaces are not typically designed for network integration and remote automation. We developed a networked ecosystem of these instruments and computing platforms, which includes software to enable automated workflow orchestration from remote computers. Specifically, we developed Python wrappers of APIs and custom Pyro client-server modules to support remote operation of these instruments over the ecosystem network. Herein, we describe a specific workflow for generating and validating voltammogram (I-V) measurements of an electrolyte solution pumped into the electrochemical cell. We demonstrate the orchestration of this workflow which is composed using a Jupyter notebook and executed on a remote computer.

KEYWORDS

instrument-computing ecosystem, science workflow, autonomous chemistry, electrochemical workflow, cyclic voltammetry.

This research is sponsored in part by the INTERSECT Initiative as part of the Laboratory Directed Research and Development Program and in part by RAMSES project of Advanced Scientific Computing Research program, U.S. Department of Energy, and in part by the Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, U.S. Department of Energy, and is performed at Oak Ridge National Laboratory managed by UT-Battelle, LLC for U.S. Department of Energy under Contract No. DE-AC05-00OR22725. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a nonexclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).

SC-W 2023, November 12–17, 2023, Denver, CO, USA

ACM ISBN 979-8-4007-0785-8/23/11.
https://doi.org/10.1145/3624062.3624611

ACM Reference Format:
Anees Al-Najjar, Nageswara S. V. Rao, Craig A. Bridges, and Sheng Dai. 2023. Cross-Facility Orchestration of Electrochemistry Experiments and Computations. In *Workshops of The International Conference on High Performance Computing, Network, Storage, and Analysis (SC-W 2023), November 12–17, 2023, Denver, CO, USA*. ACM, New York, NY, USA, 8 pages. https://doi.org/10.1145/3624062.3624611

1 INTRODUCTION

The instrument-computing ecosystems (ICE) that support science workflows are increasingly being empowered by AI and automated experiments, computations and data services in diverse areas of basic and applied research [10, 13, 18]. In particular, autonomous science experiments and laboratories have received significant attention recently for accelerating the discovery and characterization of new materials, such as electrocatalysts, photovoltaics, quantum dots, light-emitting diodes, and batteries. Examples of these autonomous labs include the Materials Innovation Factory at the University of Liverpool [5], the Autonomous Lab (A-Lab) at Lawrence Berkeley National Laboratory (LBNL) [1], and the Matter Lab at the University of Toronto [6]. The goals for these ICE include the ability to connect self-driving laboratories together, as well as connecting them to high-performance computing and data resources across networks [12, 17]. The software, data services, and networking needed to support the orchestration of science workflows over multi-domain ICE networks are under rapid development, and are driven in part by the specific requirements of their specialized instruments [9].

The electrochemistry ICE are promising for automating significant parts of workflows needed for developing new electrocatalyst designs by combining synthesis and characterization experiments with computations[15]. Indeed, they hold a promise to accelerate the discovery process by coupling the design and analysis codes with automated experiments. They typically require the integration of synthesis and chemical characterization systems, in order to conduct multiple experiments under different conditions, as well as electrochemical testing equipment, as illustrated in Fig. 1. In this paper, we consider the design and implementation such an ICE, and workflows using its electrochemistry workstation consisting of a flow controller, syringe pump, fraction collector, and potentiostat, all connected to an electrochemical cell, shown in Fig. 2. This

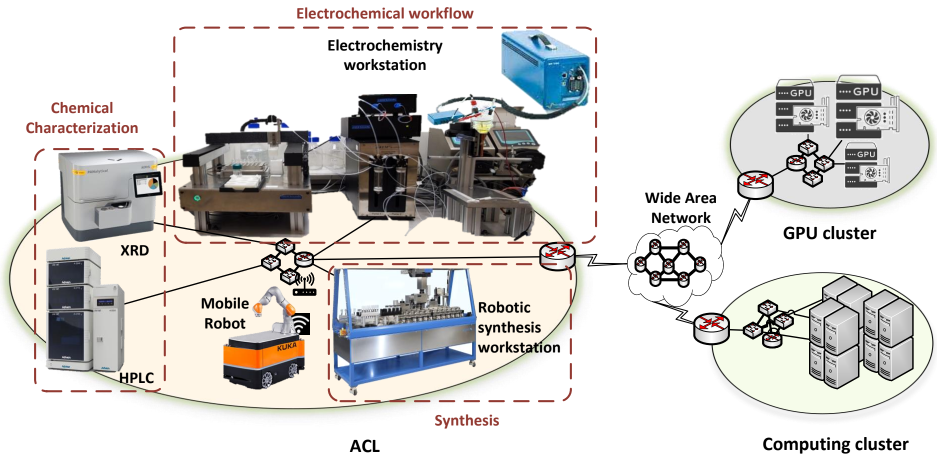


Figure 1: Electrochemistry ICE with specialized instruments and computing systems connected over cross-facility networks.

workstation utilizes an instrument control computer connected to the potentiostat, and J-Kem single-board computer connected to all other instruments. The ICE instruments are typically operated using custom Graphical User Interface (GUI) and software on computers directly connected to them. Specifically, they are not designed for remote automation, which requires network solutions for handling firewalls and security policies. Additionally, these instruments have limited software capabilities for integrating with remote computing platforms over cross-facility networks to steer their experiments and transfer measurements.

In this contribution, we present the design and implementation of network and software solutions for a cross-facility ICE that supports autonomous electrochemical workflows requiring remote experiment steering and real-time analytics on remote computing systems; the developed software is open source and publicly available [1]. This ICE enables electrochemical workflows that adapt system and instrument settings in real-time during multiple rounds of experiments. We develop this ICE from scratch using instruments, computing platforms and networks together with software by: (a) implementing dedicated hub networks that connect instruments to a gateway computer which in turn is connected to the site network, and (b) developing software modules for remote orchestration of instruments of electrochemistry workstation (Fig. 2), which is an integral and important part of this ICE.

We develop Python wrappers for Application Programming Interfaces (APIs) on the instrument computer (acting as a control agent) that enable instrument commands to be executed and integrated into workflow software. We also develop Pyro client-server modules that enable these commands to be executed from remote computers. We utilize file sharing to cross-mount file systems across the ICE, making the measurements collected by the instruments available on remote computing systems. The entire workflow is implemented using a Jupyter notebook on a remote computer to orchestrate the instruments and computations. A similar solution has been previously developed for an ICE with Nion microscopes [10], which was simpler since the microscopes are of single type, already networked, and provide readily usable Python APIs.

We demonstrate a specific workflow that remotely operates the instruments and collects measurements to generate a voltammogram (I-V profile) of a Ferrocene solution that is pumped into the electrochemical cell. The measurements are made available at the remote computing system and used for subsequent analysis, including, a machine learning (ML) method that ensures that I-V measurements are consistent with normal experimental conditions[11].

The organization of this paper is as follows. A brief description of electrochemistry ICE and workflows is provided in Section 2. The network and software designs for the Electrochemistry ICE are described in Section 3. The experimental results are presented in Section 4. Conclusions and future research directions are described in Section 5.

2 ELECTROCHEMISTRY ECOSYSTEM AND WORKFLOWS

Autonomous chemistry laboratories are being developed to accelerate the production and characterization of new materials [1, 6]. The A-Lab [1] is reported to develop inorganic solid-state materials by automating various hardware and software platforms of robots, incorporating chemistry instruments, and software tools utilized for different analyses. The Matter Lab at The University of Toronto[6] is also developing self-driving chemistry experiments utilizing multiple pieces of equipment for synthesis and characterization, such as a robotic synthesis setup from ChemSpeed[15] and a high-performance liquid chromatography-mass spectrometer (HPLC-MS) for characterization, as well as data management and analysis carried out through their ChemOS software. In general, these ICE instruments have limited networking and software support for integrating with remote computing platforms, in particular, for steering experiments and transferring measurements.

The Autonomous Chemistry Laboratory (ACL) under development at Oak Ridge National Laboratory (ORNL) is the main driver of the electrochemistry ICE developed in this paper. It is broadly focused on the synthesis of organic and inorganic compounds in

[1] https://github.com/aneesalnajjar/electrochemistry/

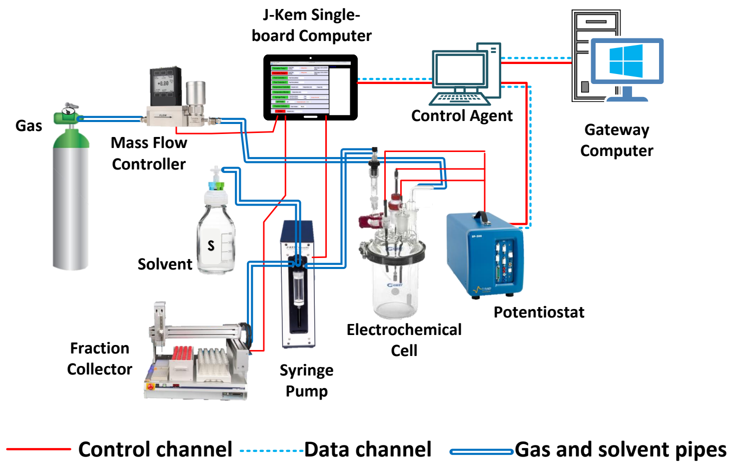


Figure 2: Electrochemistry workstation is an integral part of ICE, consisting of the potentiostat connected to electrochemical cell, which is also connected to fraction collector, syringe pump, and mass flow controller.

the liquid or solid state, and integrates various forms of chemical and electrochemical testing. It comprises multiple science instruments such as HPLC-MS, gas-chromatography-mass spectrometer (GC-MS), and X-ray diffractometer (XRD). The electrochemistry workstation, shown in Fig. 2, is its important part, and its design and implementation to support automated electrochemical workflows are described here.

2.1 Electrochemistry Workstation

The electrochemistry workstation incorporates a variety of instruments for liquid and gas transfer, along with a flexible electrochemical cell designed for electrochemical testing, as shown in Fig. 2. The cell is fed with gas (e.g., argon) from a gas tank via a computer-controlled mass flow controller (MFC). The cell is also connected to syringe pumps that dispense and withdraw liquids. The pumps may be used for filling the cell with solutions, washing the cell, or collecting fractions of liquid. In particular, the fraction collector may collect liquid samples from the cell for later external chemical analysis on any dissolved products that form during testing. The transferred liquids may be pure liquids (solvents), or liquids containing dissolved salts (electrolyte solutions). An initial focus of the workstation is electrocatalytic testing, but the setup is more broadly capable of testing redox active species in liquid electrolytes.

This setup (MFC, syringe pump, and fraction collector) is directly connected and controlled via a computer-based or hardware controller, and the gas and solvent are pumped to the electrochemical cell for conducting electrochemical testing of interest. The reactions performed within the cell are initialized and controlled via a potentiostat. The potentiostat controls the potential of the working electrode with respect to the reference electrode and measures the current flowing through the working electrode, and these measurements are used for generating the voltammograms (I-V profiles).

We utilize ACL's electrochemistry workstation consisting of (i) Bio-Logic SP200 potentiostat controlled using EC-Lab Bio-Logic API or GUI, and (ii) J-Kem single-board computer to control the electrochemical setup of MFC, syringe and peristaltic pumps, temperature controller and monitor, chiller, pH probe and electrode module, and fraction collector, using a set of software modules developed by J-Kem.

The control software of these instruments is installed on a computer-based controller directly connected to the J-Kem single-board computer and the potentiostat via serial ports. The communication between the controller and SP200 potentiostat and J-Kem computer is supported over serial connections for instrument control and measurement transfers.

2.2 Electrochemical Technique: Cyclic Voltammetry

We chose to demonstrate the Cyclic Voltammetry (CV) from a variety of electrochemical experiments possible with the potentiostat. CV is an electrochemical technique in which a potential sweep is applied to a working electrode immersed in the electrolyte solution, while simultaneously measuring the resulting current. The potential is swept between two values at a controlled scan rate, first in the positive direction and then in the negative direction, forming a cycle. The experiment employs a three-electrode system, including a working electrode, a reference electrode, and a counter electrode. During a CV experiment, the potential is varied and redox reactions take place at the electrode surface, producing a current that is proportional to the concentration of electroactive species in the solution. A voltammetry plot of the measured current against the applied potential can reveal a great deal about the analyte, and can be influenced by the rate of the electron transfer reaction(s), the scan rate, and the chemical reactivity of the analyte. Given those dependencies, it is possible under some controlled conditions to gain information about the diffusion of analyte species in solution, the redox potential of materials, the electron transfer rate or reaction rate constant, the reaction mechanism and whether the reaction is reversible under the experimental conditions.

The instruments of the electrochemistry workstation with proprietary software typically force science users to conduct experiments locally and manually transfer measurements to remote computers. This *human-in-the-loop* paradigm constrains the pace of science exploration as it requires significant time and effort. The proposed ICE significantly accelerates this pace by automating instrument steering and measurements transfers to remote computing systems for analysis, as shown in Fig. 1.

3 ICE INFRASTRUCTURE AND SOFTWARE: DESIGN AND IMPLEMENTATION

An autonomous science workflow, in general, may require an ICE that spans science instruments and computing facilities that are geographically dispersed and connected over local and wide-area networks. The electrochemistry ICE in Fig. 1 is an example. We propose its design based on separate control and data channels similar to microscope ICE in [10]. Comparatively, however, it requires more complex network connections and diverse software modules due to the custom nature of electrochemistry instruments and their control software and APIs. Our design utilizes dedicated hub networks to connect instrument computers with different connectivity and firewall requirements to a gateway computer, which in turn is connected to the site network. We implement custom software modules for the instruments of electrochemistry workstation in Fig. 2 for steering and measurements collection to be compatible with Windows and Linux computing platforms. Our solution supports both local and cross-facility instrument control and data transfers as described next.

3.1 Autonomous Electrochemical Testing ICE Design

We propose an ICE architecture to support capabilities to perform autonomous steering and real-time measurement transfers. The proposed design encompasses a science facility where the ACL electrochemistry workstation instruments operate, and a remote computing facility with high-performance resources. The ICE design consists of the following components.

- **Control agents:** ACL instruments are connected to computing systems, called the *control agents*, that host instrument control software interfaces to manage and control the instruments; for instance, Bio-Logic potentiostat is controlled using EC-Lab GUI or Development Package libraries. A control agent can also be associated with a collection of instruments performing specific tasks, and controlled by dedicated controlling hardware/software platforms; in our case, instruments including MFC, syringe and peristaltic pumps, a fraction collector, and others, are all controlled by J-Kem control software.
- **Instrument-based network setup:** Different local hub networks are designed to connect instruments and their controlling platforms to support the integration into ICE network to match the specific connectivity and firewall requirements. In addition, these hub networks are configured to align their security domains with the policies of ICE networks to enable network access.
- **Separate network channels:** The ICE design utilizes separate network channels to transfer control commands and measurements across ICE. We utilize a *control channel* to convey the control messages sent back and forth between instruments and remote computing systems. Meanwhile, the measurements are transferred via a *data channel* specifically designated for this purpose. Various data management solutions can be implemented based on ICE and instruments requirements. The separation of the network channels alleviates the delays of control commands transferred over the shared ICE network.
- **Gateway computer:** The gateway computer is the top-tier control agent in ACL equipped with multiple network interfaces to support the connectivity with instruments control agents over the hub networks. It is utilized to interface the lab instruments with computing facilities across the ICE network.

Only a part of the implemented ICE is used in the workflows described in this paper. It is the electrochemistry workstation consisting of control agent, potentiostat, and J-Kem single-board computer connected to fraction collector and syringe.

3.2 Control Channel: Communications and Software Modules

The control channel communicates the control commands via a dedicated network connection between ACL instruments and local computers as well as remote computers running control and analytics software modules. Its software consists of two types of modules for local steering by computers directly connected to instruments, and remote steering of cross-facility instruments.

3.2.1 Potentiostat local integration: The SP200 potentiostat [2] is directly connected via USB to its control agent (Fig. 2). We utilize Python-based APIs of EC-Lab Developer Package [3] that support potentiostat communications and experiment execution. This control software is *primitive*, and we utilized it to develop more advanced capabilities control modules to provide autonomous instrument steering. They support configuring the potentiostat and CV technique, loading firmware, connecting and disconnecting to potentiostat channels, collecting measurements, and other tasks, such as probing instrument control messages and status.

Scientists are able to utilize the developed potentiostat control modules in automated scripts as an alternative to manual operation using EC-lab software. These modules provide programmable interfaces to support the development of complex potentiostat experiments and analytics using real-time measurements.

3.2.2 J-KEM electrochemical setup local integration: J-Kem control software [4] provides two types of modules that run on a computer directly connected to instruments and a control agent computer connected to it. The back-end modules are embedded on J-Kem single-board computer which is connected via serial ports to a set of electrochemical instruments. The front-end GUI modules are installed and run on the control agent where a science user manually sends control commands to interact with the J-Kem setup. The J-Kem control commands are a set of predefined commands sent over serial channels to the instruments connected with J-Kem single-board computer. Science users use GUI to locally control the setup and implement certain experimental tasks, which has limited capabilities and does not support programmable autonomous workflows.

We developed Python APIs to replace the limited and proprietary J-Kem front-end modules to support programmable and autonomous electrochemical testing tasks. These APIs utilize Pyserial library [8] to configure the communication and transfer control commands between J-Kem setup and the single-board computer.

3.2.3 ACL cross-facility integration: Our programmable interfaces of instrument control commands support local scientific experiment automation. However, to incorporate remote computing systems with powerful CPU/GPU resources for real-time AI-driven electrochemical reaction experiments, the control plane has been expanded and the instrument control commands are transited to remote computing systems across the ecosystem. Pyro library [7] is utilized to support cross-facility real-time autonomous steering of the electrochemical instruments. It performs *server-client* communications among the controlling agents and remote computing systems. We have successfully leveraged this solution before to integrate Nion electron microscope into ORNL science ecosystem and perform AI-driven measurement reconstruction and probe positioning from remote GPU edge systems [10]. In this contribution, we utilize the Pyro solution to wrap and expose the SP200 potentiostat and the J-Kem setup control commands to steer the incorporated instruments through a Jupyter notebook at the remote computing system.

Pyro server modules are embedded in the control agents to wrap the Python objects corresponding to the instrument control commands and expose them to be accessible and executable through the control channel across the ICE. The Pyro daemon turns the Python objects into Pyro objects and registers and publishes them across the ICE network via the control agent IP address and a certain TCP port associated for Pyro communication. These Pyro server objects are connected via associated Pyro client applications executed from remote computing systems across the federation. The skeleton of Pyro client-server communication through the control plane across the electrochemistry ICE is shown in Fig. 3. An instrument control agent contains a number of fabricated Python objects that call native instrument control APIs to perform certain instrument configuration and steering tasks (steps 2 and 3-server side); and these Python objects are wrapped as Pyro objects and exposed for communication across the network (step 1-server side). The Pyro client modules are called as part of the science workflow. They are executed in a Python integrated development environment or a web-based interactive platform, such as Jupyter notebook, on a remote computing system. They communicate with Pyro server objects by determining the communication channel over ICE to the control agent, specified by its IP address and TCP port of Pyro proxy module (step 1-client side).

3.3 Data Channel: Transfers and File Mounts

The data channel supports transferring the data collected on the electrochemistry workstation from their control agents at ACL to the remote computing platforms across the ICE. The transfer occurs during, or after, the local or remote execution of instrument control commands, making the measurements available for (AI-driven) electrochemical testing and autonomous experiment steering. Several data management tools are available for cross-facility data transfer, e.g., GridFTP [16] or Globus [14], yet managing them may not be easy for science users as they require supplementary hardware and software platforms as well as license and credential configurations.

We implemented the data transfer by mounting measurement files to be available at remote computing systems. Our solution is cross-platform as it shares files on Windows systems with Linux platforms across the ICE. Specifically, we set up Common Internet File System (CIFS) technique on the platforms, and users with privileges can access the mounted files on remote computers. This data channel, once setup, is persistent across the ICE.

4 EXPERIMENTAL SETUP AND RESULTS

Experimental demonstrations of a workflow are described in this section using ACL's electrochemistry workstation, involving autonomous remote steering and measurements processing.

4.1 Instruments Integration and Ecosystem Setup

Instruments of ACL workstation are locally integrated on Windows-based control agent which we utilize to extend their access over ICE network. The control agent executes our Python-wrapped APIs to control J-Kem setup and SP200 potentiostat as modules, and they are provided as Pyro server objects that expose the instrument commands over ICE network. The control agent is accessed across ICE from Linux Ubuntu 20.04 NVIDIA DGX workstation (8 GPUs, 80 CPUs, and 250GB RAM) located at K200 computing and data center at ORNL, as shown in Fig. 4.

Several infrastructure parts are established to integrate ICE instruments guided by the design considerations described in 3.1. In particular, we align the facilities' network domains, and open ingress TCP ports on workstation firewalls to enable data and control traffic across ICE networks. Then, we designed and implemented control and data channels to transfer control commands and measurements between the control agent at ACL and DGX workstation at K200. The control channel supports Pyro client-server communication to remotely steer the electrochemistry workstation at ACL, while the data channel is utilized to stream the collected measurements from ACL by remote-mounting control agent's files on DGX using CIFS technique. Once the infrastructure is established, access privileges to Linux workstation (DGX) and Windows control agent are granted to allow users to remotely access and steer their workflows.

4.2 Electrochemical Testing

The testing of electrochemical workflows involves steering the workstation instruments at ACL, and analyzing their measurements at remote computing systems across ORNL. Several experiments are made possible by this setup, such as the analysis of heterogeneous (solid phase) or homogeneous (liquid phase) electrocatalysts, the study of electrolyte stability, the diffusion of species in solution, and others. In our testing, we carry out the reduction and oxidation process of a molecular species in solution as described later. The workflow to control these experiments incorporates i) Python APIs to control J-Kem setup and potentiostat through the control agent at ACL, and ii) Pyro server and client modules deployed on the control agent and DGX, respectively, to remotely steer the instruments.

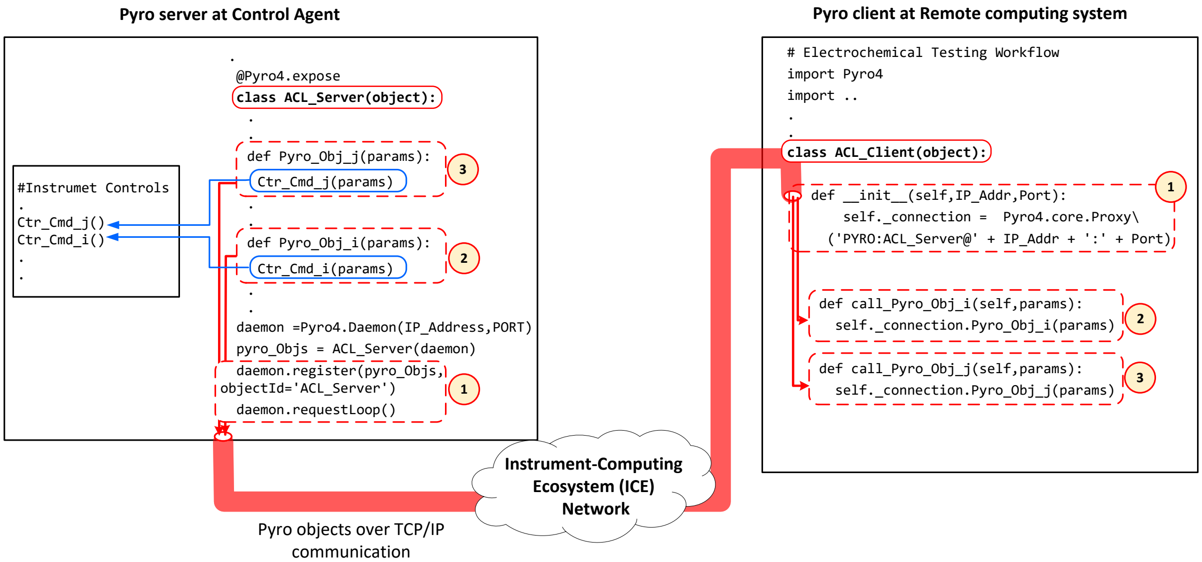


Figure 3: Pyro client-server software architecture between the instrument control agents at ACL and a remote computing system.

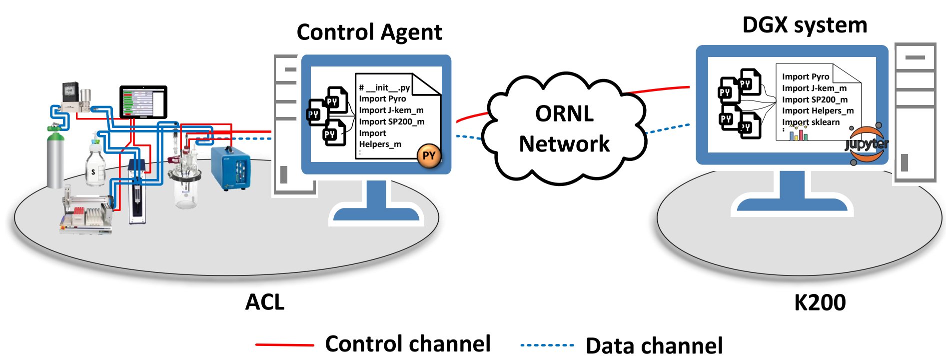
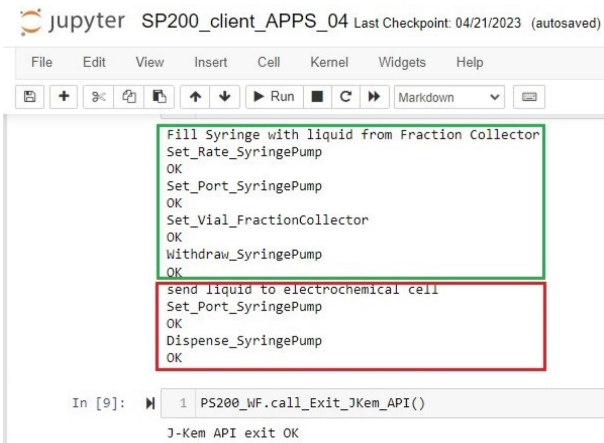


Figure 4: ORNL Electrochemistry ICE with instruments at ACL and DGX system at K200 computing and data facility.

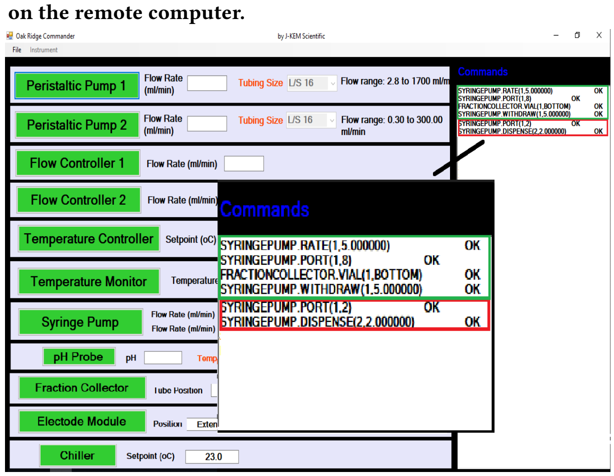
The workflow orchestration is programmed and carried out using a Jupyter notebook on DGX workstation.

Our testing of a cross-facility workflow involves using J-Kem setup to fill the electrochemical cell with a 2 mM solution of the metal complex ferrocene in acetonitrile, with the supporting electrolyte salt tetrabutylammonium triflate at 0.1M concentration. We then electrochemically cycled the molecule between the reduced species ferrocene $[Fe(Cp)_2]$ (where Cp=cyclopentadienyl), and the oxidized species ferrocenium $[Fe(Cp)_2]^+$ in the electrochemical cell by running the CV technique using the potentiostat. Once the experiment is completed, the measurements collected at the control agent are remotely available via files mounted across ICE. Authenticated users on DGX workstation utilize the measurements for analyses and computations. This workflow demonstrates the capability to support autonomous steering and (AI-based) analysis using different hardware platforms across the ICE, namely, Linux-based DGX and Windows-based ACL computers.

The workflow comprises the following real-time tasks: (***A***) establishing the communications using Pyro across ICE between the control agent at ACL and DGX at K200; (***B***) remotely configuring and connecting to J-Kem instrument setup through the single-board computer connected to syringe pump and fraction collector; (***C***) invoking commands via J-Kem setup to fill the electrochemical cell; and (***D***) activating SP200 potentiostat to run and collect I-V measurements. The task D in turn entails multiple steps (listed in Fig. 6a): ***1***) initializing the instrument with system/firmware and connection parameters; ***2***) establishing the connection to potentiostat; ***3***) loading the firmware, ***4,5***) configuring the potentiostat with the CV technique and loading the technique firmware; and ***6,7***) establishing a potentiostat channel and begin acquiring measurements. Once the acquisition finishes, ***8***) the channel is automatically disconnected, and the measurements are made available at DGX through the data channel which are tested in real-time. Lastly,

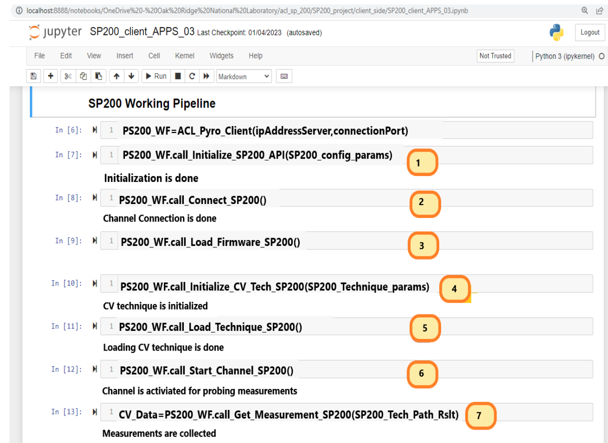


(a) Executing J-Kem control commands using Jupyter notebook on the remote computer.

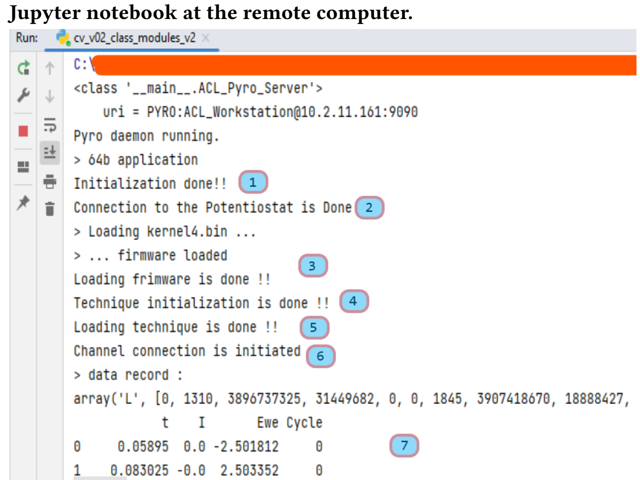


(b) J-Kem single-board computer responses to the remote control commands via the control agent.

Figure 5: Demonstration of remote steering of J-Kem setup using Pyro modules across the electrochemistry ICE at ORNL.



(a) Outputs of executing potentiostat control commands using Jupyter notebook at the remote computer.



(b) Control agent responses to the remote control commands.

Figure 6: Demonstration of remote steering of SP200 potentiostat using Pyro modules across the electrochemistry ICE at ORNL.

when the workflow tasks are completed ***E***, the cross-facility connection is shut down by disconnecting the Pyro communication between the control agent and DGX workstation across ICE.

4.3 Experimental Results

The experimental results demonstrate the execution of remote and real-time tasks in the electrochemical workflow for cyclic voltammetry outlined in Section 4.2. We demonstrated the interoperability of control commands of the instruments controlled by J-Kem single-board computer (Task ***C***) and SP200 potentiostat (Task ***D***) available at ACL. These commands are remotely triggered as a part of the autonomous workflow orchestrated using a Jupyter notebook running on remote DGX system at K200, as shown in Fig. 4. We also demonstrated the real-time analysis of I-V profiles on the DGX after the experiment finishes and the measurement files are made available across the ICE. This part includes the invocation of ML method to ensure that I-V measurements are consistent with normal experimental conditions, or detect abnormal conditions, such as disconnected electrodes or under-filled electrochemical cell. This ML method is custom-designed to extract a feature vector from I-V measurements using the Gaussian process regression (GPR) and classify it using the ensemble of trees (EOT) classifier (details in[11]).

4.3.1 J-Kem: The control commands of J-Kem setup instruments used in this demonstration (syringe pump and fraction collector) are remotely triggered to fill the electrochemical cell with Ferrocene solution, as shown in Fig. 5. The Pyro client modules are executed from the Jupyter notebook on DGX (Fig. 5a) to access objects provided by their peer Pyro servers on the control agent for interacting with the back-end instrument control commands at J-Kem single-board computer and remotely executing them (Fig. 5b). The successful remote steering of J-Kem setup across the electrochemistry ICE at ORNL is validated by receiving an "**OK**" status message after the execution of control commands at the single-board computer, which is provisioned as a part of the autonomous workflow at the remote system, as shown in Fig. 5a.

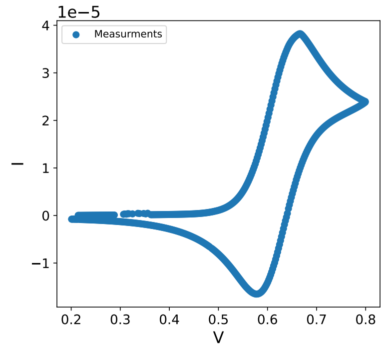


Figure 7: I-V profile generated using Jupyter notebook at DGX.

4.3.2 SP200 Potentiostat: The cross-facility steering of SP200 potentiostat is demonstrated in Fig. 6. The autonomous workflow triggers the Pyro client methods to configure the potentiostat and the CV technique using Jupyter notebook (Fig. 6a), which in turn, communicate with the associated Pyro server objects to execute the instrument control commands, as shown in (Fig. 6b). The generated results are shared, via the mounted file system across the data channel, with DGX workstation, where they are available for real-time computation and analysis.

4.3.3 CV experiments analysis: The voltammogram measurements collected as a result of the execution of CV technique are stored on the control agent, and made available on DGX through the data channel using CIFS file mounting. Fig. 7 shows the resultant I-V plot of the conducted CV experiment after it is completed and the potentiostat channel is disconnected. These measurements are classified as "normal" by the ML method, since they form a continuous curve typical of the tested analyte indicated by the underlying electrochemistry principles. The measurements collected under two abnormal conditions, namely, disconnected electrode and low analyte volume in electrochemical cell, were identified and flagged "abnormal" by the ML classifier (details published in [11]).

5 CONCLUSION AND FUTURE WORK

We presented the design and implementation of networking and software for an ICE to support automated electrochemistry workflows. It required the integration of instruments, such as mass flow controller, syringe pumps, peristaltic pumps, chiller, potentiostat, and others, which are typically not designed for network integration to support remote automation. We developed Python wrappers of APIs and the corresponding custom Pyro client-server modules to support operation of these instruments over ICE network. We described the development of ACL from scratch to constitute an operational electrochemistry ICE. We also described the orchestration of a workflow that uses a part of ACL for generating the I-V profiles of an electrochemically active solution in the electrochemical cell. We demonstrated the composition and orchestration of this workflow using a Jupyter notebook executed on remote computing platforms. This approach of specialized hub networks and software wrappers to compose and remotely orchestrate workflows is generic, and is applicable to more complex scenarios and workflows from other application domains.

Several future research areas can be pursued to expand the ICE and workflows described in this paper. The integration of additional instruments and computing platforms into ACL including mobile robots to transfer materials between different instruments is planned for future work. More comprehensive electrochemical workflows are planned that involve most of ACL instruments, as well as utilize other electrochemical testing techniques supported by the potentiostat. The expansion of the ML method [11] to include additional tasks and cases is of future interest. More generally, it would be of future interest to develop sophisticated AI-driven and real-time electrochemistry workflows. Improving the quality of services and security posture of ICE is a potential target too, particularly, to connect external facilities to ACL. Specifically, these tasks entail investigating and enhancing network delays and throughput, security policies and firewall rules across multi-domain networks connecting the facilities of the ecosystem.

REFERENCES

[1] [n. d.]. A-Lab: Autonomous Lab at LBNL. Available from: https://newscenter.lbl.gov/2023/04/17/meet-the-autonomous-lab-of-the-future/.
[2] [n. d.]. Bio-Logic SP200 Potentiostat. Available from: https://www.biologic.net/products/sp-200/.
[3] [n. d.]. EC-Lab Developer Package. Available from: https://www.biologic.net/support-software/ec-lab-oem-development-package/.
[4] [n. d.]. J-KEM Scientific Inc. Available from: https://www.jkem.com/.
[5] [n. d.]. Materials Innovation Factory. Available from: https://www.liverpool.ac.uk/materials-innovation-factory/.
[6] [n. d.]. Matter Lab. Available from: https://www.matter.toronto.edu/.
[7] [n. d.]. Pyro: Python Remote Objects. Available from: https://pyro4.readthedocs.io/en/stable/.
[8] [n. d.]. pySerial: Python library for serial port operations. Available from: https://pyserial.readthedocs.io/en/latest/index.html.
[9] Anees Al-Najjar, Nageswara SV Rao, Ramanan Sankaran, Helia Zandi, Maxim Mukherjee, Debangshu Ziatdinov, and Craig Bridges. 2023. Cyber Framework for Steering and Measurements Collection Over Instrument-Computing Ecosystems. In *the 9th International Conference on Smart Computing (SMARTCOMP)*. IEEE.
[10] Anees Al-Najjar, Nageswara SV Rao, Ramanan Sankaran, Maxim Ziatdinov, Debangshu Mukherjee, Olga Ovchinnikova, Kevin Roccapriore, Andrew R Lupini, and Sergei V Kalinin. 2022. Enabling autonomous electron microscopy for networked computation and steering. In *2022 IEEE 18th International Conference on e-Science (e-Science)*. IEEE, 267–277.
[11] Anees Al-Najjar, Nageswara S. V. Rao, Craig A. Bridges, and Sheng Dai. 2023. Normality of I-V Measurements Using ML. In *the 19th International Conference on e-Science (e-Science)*. IEEE.
[12] Katerina B Antypas, DJ Bard, Johannes P Blaschke, R Shane Canon, Bjoern Enders, Mallikarjun Arjun Shankar, Suhas Somnath, Dale Stansberry, et al. 2021. Enabling discovery data science through cross-facility workflows. In *2021 IEEE International Conference on Big Data (Big Data)*. IEEE, 3671–3680.
[13] Bjoern Enders, Debbie Bard, Cory Snavely, Lisa Gerhardt, Jason Lee, Becci Totzke, Katie Antypas, et al. 2020. Cross-facility science with the Superfacility Project at LBNL. In *2020 IEEE/ACM 2nd Annual Workshop on Extreme-scale Experiment-in-the-Loop Computing (XLOOP)*. 1–7. https://doi.org/10.1109/XLOOP51963.2020.00006
[14] I. Foster and C. Kesselman. 1997. Globus: A Metacomputing Infrastructure Toolkit. *Intl J. Supercomputer Applications* 11, 2 (1997), 115–128.
[15] Martin Seifrid, Robert Pollice, Andrés Aguilar-Granda, Zamyla Morgan Chan, Kazuhiro Hotta, Cher Tian Ser, Jenya Vestfrid, et al. 2022. Autonomous chemical experiments: Challenges and perspectives on establishing a self-driving lab. *Accounts of Chemical Research* 55, 17 (2022), 2454–2466.
[16] Teragrid [n. d.]. GT 4.0 GridFTP. http://www.globus.org.
[17] Nicholas Tyler, Robert Knop, Deborah Bard, and Peter Nugent. 2022. Cross-Facility Workflows: Case Studies with Active Experiments. In *2022 IEEE/ACM Workshop on Workflows in Support of Large-Scale Science (WORKS)*. IEEE, 68–75.
[18] Rafael Vescovi, Ryan Chard, Nickolaus D Saint, Ben Blaiszik, Jim Pruyne, Tekin Bicer, Alex Lavens, Zhengchun Liu, et al. 2022. Linking scientific instruments and computation: Patterns, technologies, and experiences. *Patterns* 3, 10 (2022).