Supplementary Information for

Advancing Catalysis Research through FAIR Data Principles Implemented in a Local Data Infrastructure - A Case Study of an Automated Test Reactor

Abdulrhman Moshantaf,[1] Michael Wesemann,[1] Simeon Beinlich,[1] Heinz Junkes,[1] Julia Schumann,[1,2] Baris Alkan,[1,3] Pierre Kube,[1] Clara Patricia Marshall,[1] Nils Pfister[1] and Annette Trunschke*[1]

[1]*Fritz-Haber-Institut der Max-Planck-Gesellschaft, Department of Inorganic Chemistry, Faradayweg 4-6, D-14195 Berlin, Germany*

[2]*Consortium FAIRmat, c/o Humboldt-Universität zu Berlin, Zum Großen Windkanal 2, 12489 Berlin, Germany*

[3]*Max-Planck-Institut für Chemische Energiekonversion, Stiftstrasse 34 - 36, 45470 Mülheim an der Ruhr, Germany*

**trunschke@fhi-berlin.mpg.de*

Automation tools and experimental details

AC/CATLAB Archive. The AC-Archive is based on PHP scripts and a MySQL database, which ensures scalability and flexibility.[1] The data is stored on a central storage system (Netapp) that has multiple backups, which increases the protection of the data. The Application Programming Interface (API), which is also programmed in the PHP programming language, is constantly being expanded to improve the programmatic reading and writing of the database.[1]

S3 storage. An S3 storage is used to store experimental data for long-term backup, with each instrument having its own bucket to which all files are uploaded at the end of an experiment; this can also be done automatically via an API provided for the S3 storage.

EPICS. EPICS is an open-source control system that is widely used in large facilities such as synchrotron sources and other beam lines, but can also be used for automation of small laboratory setups.[2, 3] The software can run on any operating system and any platform and provides the necessary tools and utilities to operate, monitor and control experiments. The advantage of EPICS lays in the possibility to connect several computers and input-output controllers (IOCs) based on the server/client model using the channel access protocol. The server, which is normally the IOC, is responsible for controlling the hardware devices and collecting the data. An automatic backup is performed by an archiver appliance instance in the FHI network without interruption (online 24/7). In this way, all the information about the devices and the data of the process variables (PVs) are continuously available via the network. The client, on the other hand, which can be any computer connected on the network, can access these information by reading, writing and monitoring the PVs remotely using EPICS tools.

Using EPICS as a control system allows connection to all types of hardware devices and provides a small database record within the IOC for each PV in the hardware devices. The records can be accessed via the Ethernet using the channel access protocol, which provides the put command to write a value to the record and the get command to read values from the records. To fully automate the writing and reading of values to or from the records, Python scripts can be written. However, to access the EPICS records from Python, some libraries such as the Ophyd library,[4] which offers a hardware abstraction for the devices in Python, are used. The library allows to define the PVs from a certain device as an EPICS signal where its information can be accessed using simple commands like “read” or “set”. The EPICS signals can be grouped into an Ophyd device which can be defined as a class in Python and with that, similar devices can be defined easily as an object of that class just by giving different PV names. In general, Ophyd is associated with Bluesky,[5] where Bluesky manages the experiment and takes advantage of Ophyd’s functions to communicate with the hardware devices and collect their data.

Bluesky is a Python library that provides functions to harmonize the control of an experiment with the collecting of data and metadata in one place.[5] It features real time processing and plotting of the acquired data, and allows implementation of user-defined plans and commands that are triggered automatically and sequentially. A simple plan would be to read a detector value for a specific number of times, but more complex and adaptive plans can also be handled, such as setting target values to the instruments and waiting for a specific condition to occur, while in the meantime reading all data from the detectors.

The most important part of Bluesky is the run engine. This is where the plan is initiated and it manages the workflow of the plan by allowing the plan to be paused, resumed or aborted any time, it is also responsible for streaming the acquired data and metadata from the experiment as JSON dictionaries with a specific structure.[5] The run engine can subscribe to callback functions that can perform, for example, a specific processing task on the collected and streamed data from the run engine in real time and store the results in a specific format. Such a callback function was used in the present automation of a catalytic test reactor to extract the data from the streamed JSON dictionary and putting it in HDF5 and CSV files. When the experiment is finished, the Python script automatically uploads the generated files to the database (AC/CATLAB archive) in two steps. First, the files can be uploaded to a specific S3 bucket for long-term backup storage using the "minio client" API in Python for S3 storage, then an URL link can be generated for the uploaded files to allow the files to be uploaded to the AC/CATLAB archive using the API, which is implemented in the database.

Another backup solution is the EPICS archiver appliance.[3] It can archive the values of PVs for 24 hours even if the experiment is not running, it is very expandable and scalable by adding more resources to it, so it can store a large number of PVs, configuring the archiver or retrieving the stored data is easy using web interfaces or python scripts.

Phoebus. Phoebus is a Java variant of the control system's Studio program that is used to design graphical user interfaces for monitoring and controlling experiments, as it allows easy reading of process variables provided by EPICS via the channel access protocol and place them in drag and drop widgets like text updates, gauges, plots, tables, etc.[6]

Python and Jupyter notebooks. Python is used writing scripts responsible for running experiments and acquiring data, using libraries that provide access to the EPICS PVs within the Python script. This makes it possible to automatically send a set of setpoints to the hardware devices based on the method chosen, and also allows data analysis and conversion to be done within the script functions. Jupyter notebooks are used to write scripts for interactive GUIs that allow the user to enter meta and method data for their experiment. The user can also log into the Jupyter hub, which can be used by anyone at the FHI. The Jupyter hub gives users the ability to write their own Python scripts on the web and run them from anywhere.

Catalyst test reactor. The reactor for rapid tests in ammonia decomposition ("Haber") is a fully enclosed, ventilated system constructed at the Fritz-Haber-Institut (FHI) (schematic representation in Fig. 3 in the main text). In the setup, a vertical reactor oven (HTM Reetz GmbH, type LOBA vertikal) is used to heat a tubular quartz reactor (QSIL GmbH, inner diameter 7 mm, length 400 mm) filled with the catalyst. For the experiment, 41.7 mg of catalyst (particle size of 200-300 µm) is mixed with 83.4 mg of SiC (ESK-SiC Gmbh, 355-400 µm) and placed inside the tube. The catalyst bed is stabilized by quartz wool at both ends of the isothermal zone of the oven (Fig. S1). A thermocouple (Electrotherm, thermoelement type K) connected to a Pico TC-8 (Pico Technology, tech data logger type TC-8) is inserted at the center of the catalyst bed and is used to record and control the reaction temperature. In addition, the oven temperature is monitored using a second thermocouple placed in the oven.

Six mass flow controllers (Bronkhorst, type El-Flow) are connected to the setup to enable gas stream of various gases including central gases of Ar, H_2, O_2 and N_2 (5.0, Westfalen AG) and NH_3 (5.0, Westfalen AG). Downstream the reactor, a thermal conductivity detector (TCD, Xensor, type XEN-5320) is connected and used to monitor hydrogen concentration during the reduction stage in H_2/Ar gas mixtures. To remove water formed during reduction of the catalyst, a molsieve trap (Molsieve 5A, sieve size >355 µm) is inserted before the TCD. A separate gas line is used to stream NH_3. An ammonia detector (IR detector, Rosemount, type Binos 1.2) is used to detect the residual concentration of NH_3. Before entering the detector, the effluent gas from the reactor is diluted with flowing nitrogen in a constant ratio of 225 ml/min N_2 /25 ml/min NH_3. A two-position actuator control module (Vici, Valco Instruments Co. Inc., 4 port 2-pos valve with electric motor) is used to change the gas flow direction and to switch between the detectors. A pressure gauge connected downstream the reactor (Swagelok, 0-10 bar) is used to monitor the pressure drop over the catalyst bed. The NH_3-containing outlet gases are passed through a bottle (polypropylene) filled with water before they enter the exhaust gas. All catalytic tests are performed according to a SOP presented in Fig. 1 b of the main text.

A computer system (Jetway, JBC390F841CA) with 10 serial ports is used to allow communication with the serial devices. This input-output controller (IOC) also has 2 Ethernet ports as it serves as a gateway between the setup and the FHI network. The EPICS control software runs on this computer system and takes control of the hardware devices.

On the desktop computer of the setup (DELL, OPTIPLEX 7020), the graphical user interfaces and the Jupyter notebooks (Python scripts) to control and operate the setup are running. It is also where the data collected and the files generated from the experiment are initially stored.

Catalyst preparation

The Ni catalyst precursor $Ni_xMg_{1-x}O$ (x: 0.034, S36283) was prepared using a computer-controlled co-precipitation in an automatic work station (Mettler Toledo, Optimax 1001, E5). A metal salt solution was prepared dissolving $Mg(NO_3)_2 \cdot 6H_2O$ (126.76 g) and $Ni(NO_3)_2 \cdot 6H_2O$ (3.59 g) in 500 ml H_2O. Another solution of 75 ml of NH_3 (25 %) dissolved in 1000 ml DI H_2O was prepared, and both solutions were dosed at a rate of 10 g/min in 200 mL H_2O at 60 °C and aged for 1h at pH=8.5. The product was filtered and washed three times with 360 mL mqH_2O. The product was centrifugated at 5000 rpm for 15 min, and the solid was dried at 80 °C overnight. 3 g of the solid was calcined at 600 °C for 3 hours with a heating rate of 2 K/min in a rotating furnace (XERION, UTP Carbon). The calcined precursor oxide was pressed at three tons for three minutes and sieved to 200-300 micron.

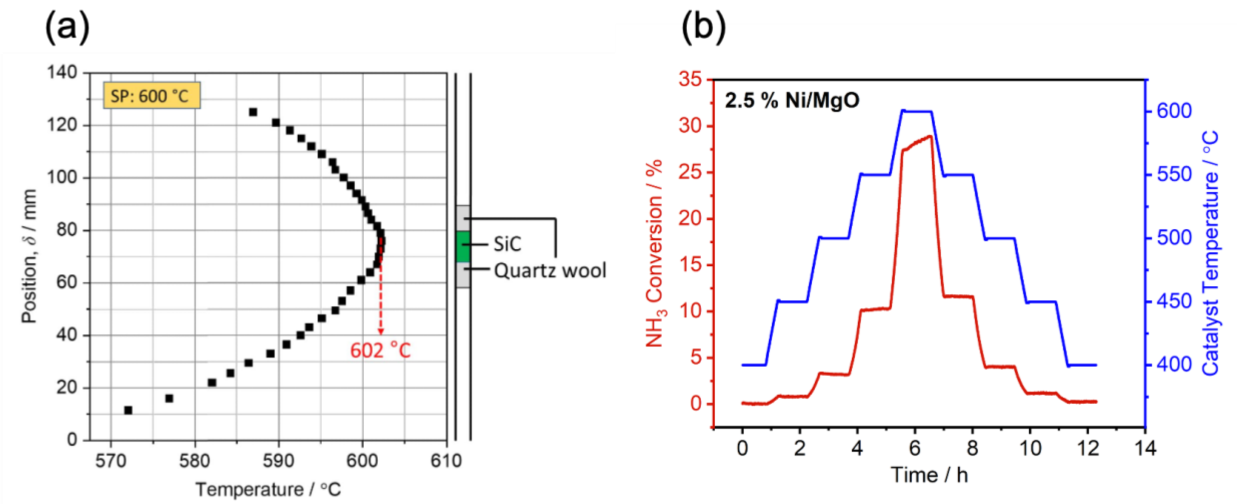

Fig. S1. (a) Temperature profile of the Haber reactor, and (b) ammonia decomposition over a 2.5 % Ni/MgO (S36891) as a function of temperature and time.

Haber Archive v1.0.179 Search: S82 NEW SEARCH MORE TAGS LOGOUT HOME

User: moshantaf Role: user Type: ldap

Metadata

Action	
Id	S82
Descendants:	S83, S84, S85, S86, S87
User	moshantaf
Project	DEFAULT
Access	project
Open Access	
Edit History	SHOW
Date Created	2024-05-24 18:48:03
Date Modified	2024-05-30 13:04:13

Data

Name	Ni2.5%-MgO precursor
Preparator	C. Marshall
Source	FHI
Sample Description	pale green solid
Comment	Solution A was prepared by dissolving Mg(NO3)2 (C150 - 126,76 g) and Ni(NO3)2 (S90 - 3,5911 g) in 500 mL mqH2O. Solution B was prepared by dissolving 75 mL of NH3 (25%) in 1000 mL mqH2O. In the Optimax, A (300 g) and B (300 g) were dosed at constant rate (10 g/min) in 200 mL mqH2O, at 60°C, aged for 1 h. Pumps=70%. Inputs=1A and 2B. The product was filtered once (15A) and then washed 3 times with 360 mL mqH2O, separated by centrifugation as solid was too fine and passing through even the 15A filter, big beakers, 5000 rpm, 15 min. Conductivity last washing= 0,06 mS/cm Dried at 80°C overnight. During the synthesis, the pH stayed around pH=8,5.
Date of Preparation	2022-08-03
Amount of Product	4,9838 g
Yield (%)	
Method of Preparation	precipitation #optimax
Location of Sample	
Special Precautions	
Cross Reference to Literature	
Drying Procedure	Static air cabinet at 80 C
State of Sample	
Elements	Ni, Mg

Json Data


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{
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}
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Files (3)

Id	File	Time	Comment	Size	Action
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818	Experiment 2022-08-03 10-52.xlsx	2024-05-24 18:48:03		638.8 Kb	
819	Experiment 2022-08-03 10-52_Report.pdf	2024-05-24 18:48:04		148.3 Kb	

Linked Entries

Id	Project			Action
S90	DEFAULT	Nickel(II) nitrate hexahydrate	Alfa Aesar	
E88	DEFAULT	Optimax Synthesis Workstation	Marshall	
C1	DEFAULT	Magnesium nitrate hexahydrate (empty)	Magnesiumnitrat Hexahydrat (empty)	
D114	DEFAULT	ATR-IR Measurement S82	C. Marshall	
D115	DEFAULT	XRD of S82	C. Marshall, F. Girgsdies	

Fig. S2. Sample entry of the precipitated catalyst precursor in the example database that has been used in the ammonia decomposition experiment after calcination, pressing and sieving displaying ancestries and descendants of the sample.

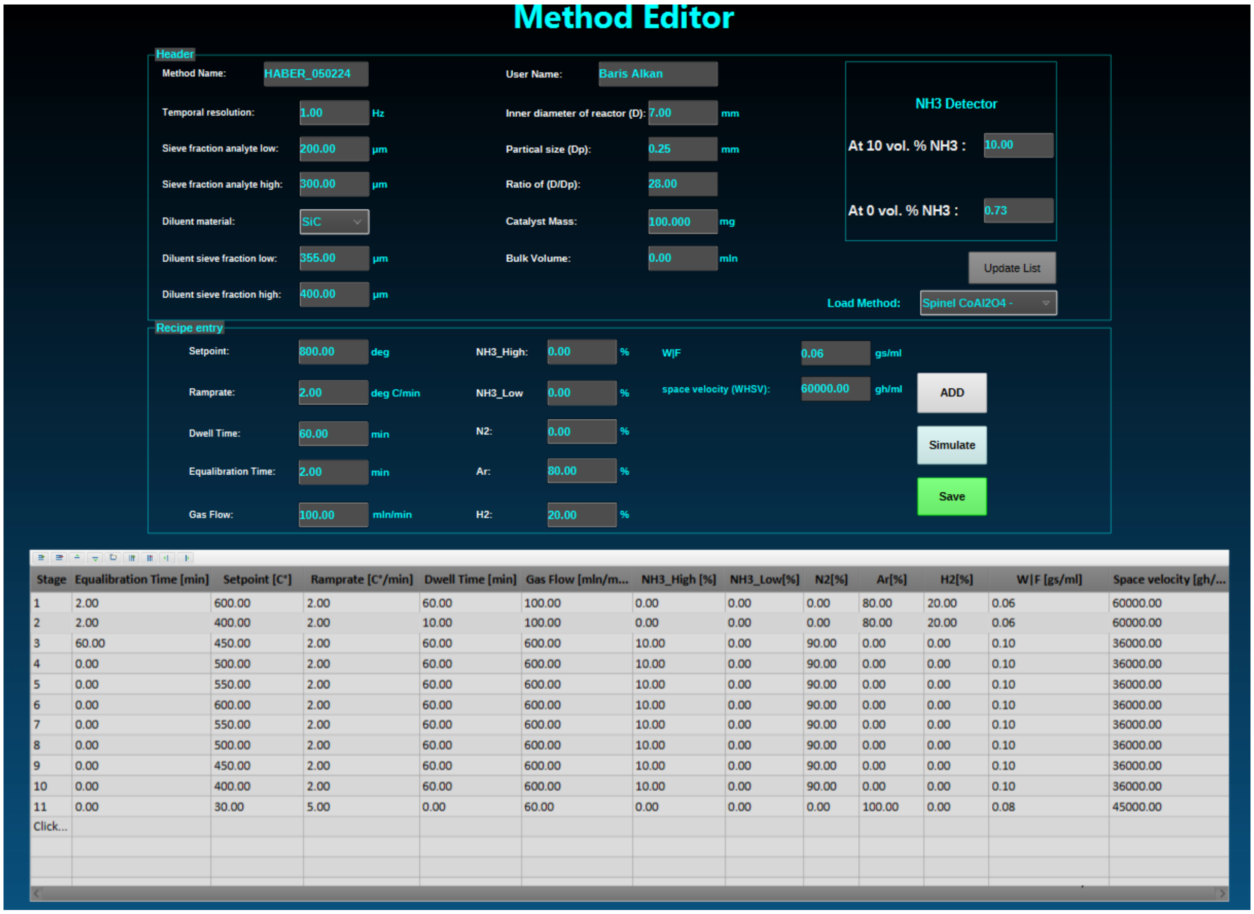


Fig. S3. The method editor GUI for creating new methods and saving them digitally in the database.

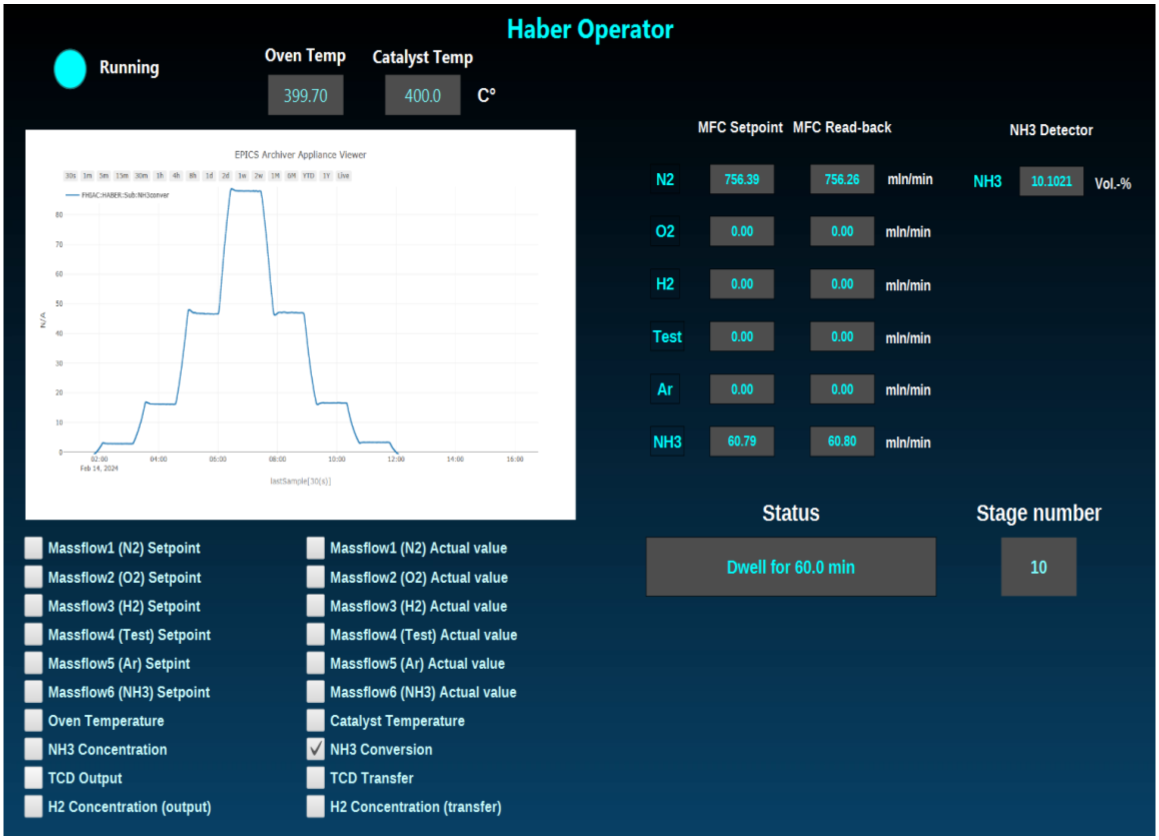


Fig. S4. The operator GUI is a read-only GUI for displaying historical and live plots as well as the status of the experiment and the read-back values from the devices.

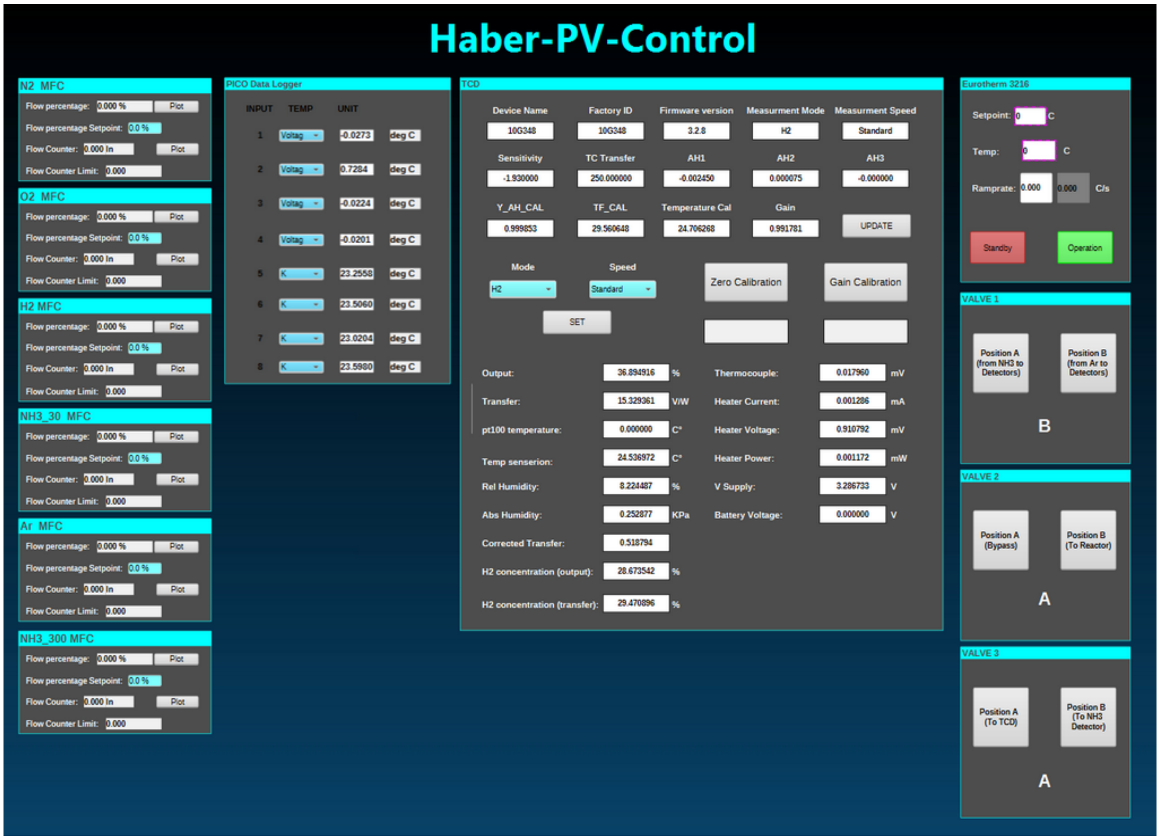
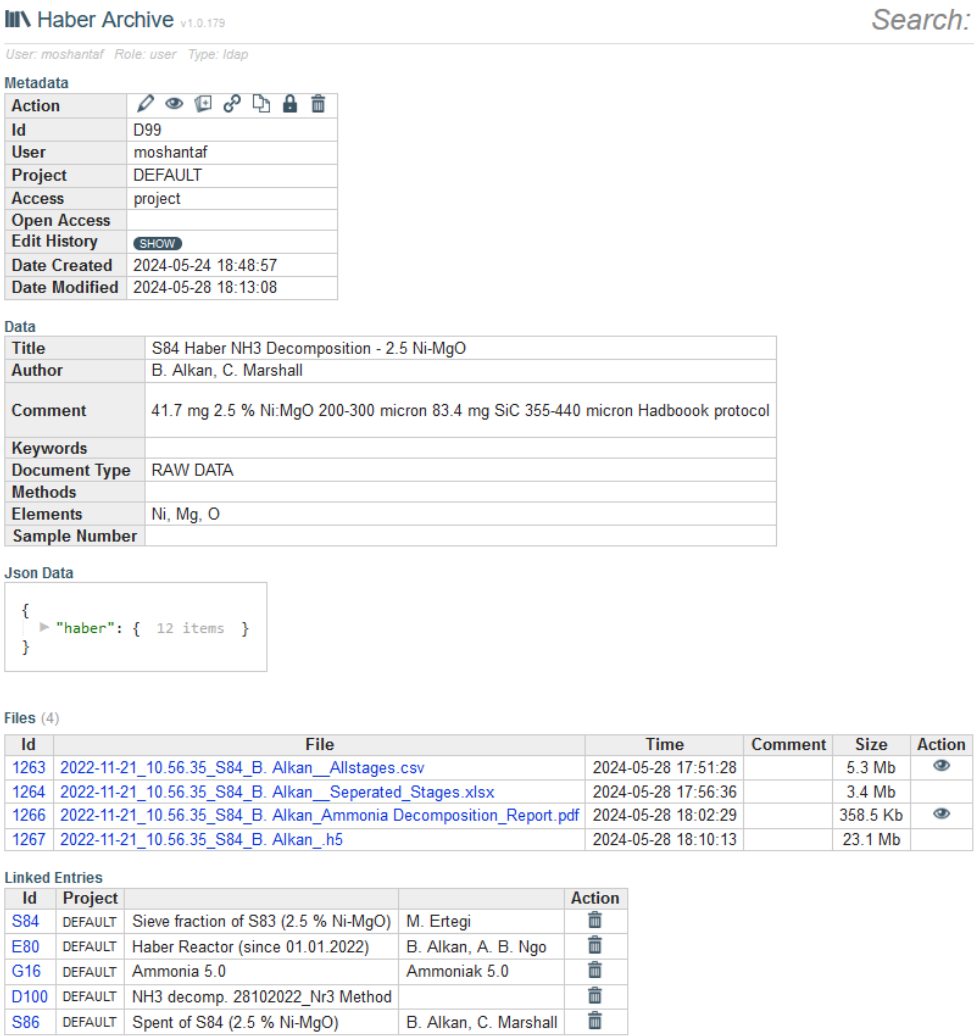


Fig. S5. PV-Control GUI for configuring all connected hardware devices.



Haber Archive v1.0.179 Search:

User: moshantaf Role: user Type: ldap

Metadata

Action	
Id	D99
User	moshantaf
Project	DEFAULT
Access	project
Open Access	
Edit History	SHOW
Date Created	2024-05-24 18:48:57
Date Modified	2024-05-28 18:13:08

Data

Title	S84 Haber NH3 Decomposition - 2.5 Ni-MgO
Author	B. Alkan, C. Marshall
Comment	41.7 mg 2.5 % Ni:MgO 200-300 micron 83.4 mg SiC 355-440 micron Hadboook protocol
Keywords	
Document Type	RAW DATA
Methods	
Elements	Ni, Mg, O
Sample Number	

Json Data

{
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}

Files (4)

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1264	2022-11-21_10.56.35_S84_B. Alkan__Seperated_Stages.xlsx	2024-05-28 17:56:36		3.4 Mb	
1266	2022-11-21_10.56.35_S84_B. Alkan_Ammonia Decomposition_Report.pdf	2024-05-28 18:02:29		358.5 Kb	
1267	2022-11-21_10.56.35_S84_B. Alkan_.h5	2024-05-28 18:10:13		23.1 Mb	

Linked Entries

Id	Project			Action
S84	DEFAULT	Sieve fraction of S83 (2.5 % Ni-MgO)	M. Ertegi	
E80	DEFAULT	Haber Reactor (since 01.01.2022)	B. Alkan, A. B. Ngo	
G16	DEFAULT	Ammonia 5.0	Ammoniak 5.0	
D100	DEFAULT	NH3 decomp. 28102022_Nr3 Method		
S86	DEFAULT	Spent of S84 (2.5 % Ni-MgO)	B. Alkan, C. Marshall	

Fig. S6. Result data entry in the example database generated from Haber after using the catalyst 2.5%Ni-MgO in the experiment; The data entry contains the generated files with the raw data of the experiment, the method data saved as JSON and the pdf report of the experiment; The sample entries of the sieve fraction and the spent catalyst, the equipment (Haber reactor), the gases and the method which has been used are all linked to the result data entry.

References

1. M. Wesemann, AC archive, https://github.com/fhimpg/archive (accessed 2024/05/31).
2. Getting started with EPICS, https://docs.epics-controls.org/en/latest/getting-started/EPICS_Intro.html, (accessed 2024/05/31).
3. EPICS Archiver, https://slacmshankar.github.io/epicsarchiver_docs/details.html, (accessed 2024/05/31).
4. Ophyd Index, https://blueskyproject.io/ophyd/index.html, (accessed 2024/05/31).
5. Bluesky, https://blueskyproject.io/bluesky/, (accessed 2024/05/31).
6. Phoebus, https://github.com/ControlSystemStudio/phoebus, (accessed 2024/06/04).