eurizon
European network
for developing new horizons for RIs

Project information

Project full title	European network for developing new horizons for RIs
Project acronym	EURIZON
Grant agreement no.	871072
Instrument	Research and Innovation Action (RIA)
Duration	01/02/2020 – 31/01/2024
Website	https://www.eurizon-project.eu/

Deliverable information

Deliverable no.	D4.22 (D102)
Deliverable title	Report on the 1st International workshop
Deliverable responsible	ESRF
Related Workpackage/Task	WP4 / Task 4.6
Type (*e. g.* Report; other)	Report
Author(s)	M. Nentwich, D. Novikov, O. Seeck, M. Krisch
Dissemination level	Public
Document Version	1
Date	31 January 2023 (month 36)
Download page	https://www.eurizon-project.eu/

Document information

Version no.	**Date**	**Author(s)**	**Comment**
1	31 Jan. 2023	M. Nentwich, D. Novikov, O. Seeck, M. Krisch	–

Contents

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 871072.

Report on the 1st International Workshop

1 Abstract

X-ray Absorption Spectroscopy (XAS) is one of the most widespread techniques at synchrotron X-ray facilities with applications across many scientific disciplines with an increased interest by industry, most notably in catalysis, battery research, and material science. This class of experiments requires complex sample environments for *in-situ* and *in operando* studies, a high level of automation, mail-in services, remote access capacities, and automated data analysis. We will develop a *Conceptual Design Report* (CDR) for a generic automated XAS beamline for catalysis research that describes the principles of all the above requirements. As a starting point we organized a 1st workshop in order to gather the desired requirements from the user community as well as existing solutions of different facilities.

2 Motivation for Highly Automated X-ray Absorption Spectroscopy Experiments

Catalysis is one of the key technologies for tackling major challenges within the current energy policy targeting energy sovereignty, reduction of the dependence on fossil and nuclear fuels, and increased utilization of sustainable and renewable fuels. In general, investigating a catalyst implies analyzing the relationships between the structure of a functioning catalyst and its activity under realistic conditions (operando data) [1]. The synthesis of the catalyst material could involve different synthesis routes and starting materials (synthesis data). The structural properties are determined before, during, and after the reaction (characterization data). The reaction itself is characterized by the reaction data and derived quantities (performance data), which serve as input for modeling and simulations, which are the basis for the design of catalytic reactors and processes [1]. The investigation of catalytic processes facilitates efficient conversion processes among all kinds of application areas:

- "energy" (distribution and storage of energy in the form of chemical energy carriers) [2, 3],
- "green chemistry" (to close the carbon cycle) [4, 5],
- "earth & environment" (bio-catalysis as part of biotechnology) [6, 7], and
- "air & space transportation" (synthetic fuels) [8, 9].

The technically most important elements for catalytic experiments are among the transition metals and lanthanides. In order to study the underlying processes and, thus, changes in short and long range order within the (crystal) structure, spectroscopic experiments are perfectly suited. ***X-ray Absorption Spectroscopy*** (XAS) analyses the energy dependent intensity changes in the vicinity of an absorption edge. The element under investigation determines, which electronic transitions are possible and, thus, which absorption edges exist. The local environment determines significant details of the energy dependent intensity, especially for transitions concerning the outer most shells (K and L edges), see Fig. 1.

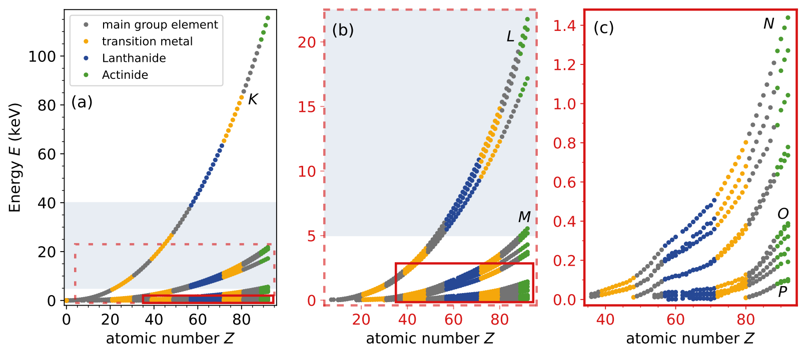

Figure 1: Energies of different absorption edges in dependence of the elements up to atomic number 92, according to X-ray Data Booklet [10]. The gray area highlights the energy range covered by a typical hard X-ray spectroscopy beamline.

As briefly mentioned, catalytic experiments typically depend on numerous factors. Currently, the complete experimental coverage of this enormous parameter space is highly time consuming and requires a vast degree of manual work. An increase in the degree of **automatization** would mean a reduced effort for exploring the parameter space in a brute force manner. Further, the automatization would be accompanied with the standardized of the data management to facilitate the FAIR principles (findability, accessibility, interoperability, and reuse of digital assets) [11]. The consortium NFDI4Cat is working on realizing those principles within the German catalyst community [1]. The principles feature the machine-accessibility, which, on the one hand, facilitates the structured analysis of own data, and on the other hand the possibility to compare with data of other scientists.
Based on highly standardized data, **machine learning** and artificial intelligence could reduce the reasonable parameter space while measuring. In turn, this will enhance the data quality and the sample throughput.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 871072.

In general, these improvements will increase the attractiveness to industry and non-expert users and, thus, speed-up the innovative process within Germany. Further, an increase of remote and mail-in experiments will be facilitated.

3 Preparation of the 1st Workshop

In a first workshop, the state of the art of automated and remote *in-situ* XAS experiments shall be collected in order to gather the requirements from the LEAPS facilities and industry.
Originally, the 1st Workshop was planned as a satellite meeting of the 5th *LEAPS Plenary Meeting* (26th to 28th 2022 October at the Paul Scherrer Institute in Villigen, Switzerland). These plans had to be revised to meet the actual date of the project restart. We decided to use the *DESY Photon Science Users' Meeting* (26th to 27th January 2023) as new host for the workshop [12].
This rearrangement offers the advantages of reaching a large part of the LEAPS community (DESY, XFEL, HZB, and others), who regularly participate in the DESY Users' Meeting and of multiple synergies with existing projects with complementary objectives. Those projects are

- ROCK-IT (Remote, Operando Controlled, Knowledge-driven, IT-based):
  joint Helmholtz activity to evaluate and develop all necessary tools for the automation and remote-access also of more complicated *in-situ* and operando XAS experiments. The methods developed with ROCK-IT should be extendable to other methods within Helmholtz.
- PETRA IV (upgrade of the existing PETRA III synchrotron towards 4th generation):
  development of an high-resolution 3D X-ray microscope for chemical and physical processes (*e. g.* during catalysis, within batteries, or microchips); with focus on high automation, especially for application-oriented industrial research [13].
- DAPHNE (DAta from PHoton and Neutron Experiments):
  Facilitating FAIR (Findable, Accessible, Interoperable, and Re-usable) data from photon and neutron experiments for more efficient scientific work and higher knowledge gain from data; with focus on recording and managing metadata, *e. g.* electronic logbooks and (meta)databases [14].

In consultation with the organizers of the regular XAS satellite workshop of the DESY User's Meeting (Wolfgang Caliebe from P64 and Edmund Welter P65), we decided to dedicate a micro-symposium of the 2023 satellite workshop to the subject of automation, see also Sec. 5. Most of the necessary steps on realizing automation and remote-access are not exclusively hot topics for catalytic XAS experiments, but for most research. Therefore, we decided that also presentations not covering the complete scope will be welcome.
In order to learn the basics and to understand the overall challenges of data storage at large research facilities, Dr. M. Nentwich was happy to attend the DAPHNE4NFDI work meeting from 5th to 7th December 2022 (https://events.hifis.net/event/569/). This network provided her with contacts to potential speakers for the satellite workshop at the DESY Users' Meeting, not only covering the topic of data storage but also of including technical components into the beamline control, which is also key knowledge for highly synchronized experiments.

4 Conclusion

Table 1 gives an overview of the speakers and their presented subjects during the dedicated 2 h-long microsymposium [12]. Dr. M. Nentwich, member of task 4.6, selected and invited the speakers and chaired the session. Nominally, 71 people attended the XAS workshop either in-person or online.
The complete XAS workshop and especially the automation session gave fascinating insights into the aspects to consider for an automated *in-situ* XAS beamline and the corresponding challenges. Generally, the speakers and participants emphasized that the full automatization of *in-situ* experiments is a very challenging and complex task. They proposed the realization via an intermediate step of fully automatizing standard XAS experiments (including basics as changing the energy towards a new absorption edge). They emphasized that many users (including industry) would strongly benefit from those improvements. This task can draw on experience of automated lab experiments. The full automatization of *in-situ* XAS experiments is especially challenging for catalysis, *e. g.* because of the handling of gases. Additionally, it is typical for user groups to only trust their own equipment and to mistrust external equipment with respect to purity of gases (talk by M. Görlitz about *Technical Developments at P64*). However, a working example for *in-situ* experiments on battery cells was briefly introduced [15].

Table 1: List of the speakers and their subjects during the micro-symposium *Automated in-situ/operando experiments at PETRA III and PETRA IV XAFS beamlines* of the DESY Users' Meeting [12].

Speaker	Institution	Title
A. Schökel	P02.1, DESY	Automatization of synchrotron experiments – present and future
K. Kiefer	Helmholtz-Zentrum Berlin	SECoP – the Sample Environment Communication Protocol
P. Weidler	Karlsruhe Insitute for Technology	Automation: first steps and beyond
S. Paripsa	University of Wuppertal	XAS reference database under DAPHNE4NFDI
B. Hinrichsen	BASF	Relevanz von Automatisierung in der Industrie
J. Timoschenko	Fritz-Haber-Institute, Berlin	Tracking the evolution of heterogeneous structures in working catalysts using machine learning

Figure 2: Group photo of the in-person participants of the XAS satellite workshop.

5 Acknowledgements

The authors are indebted to Dr. Wolfgang Caliebe and Dr. Edmund Welter (P64 and P65 at DESY, respectively) valuable discussions and useful recommendations on possible speakers for the workshop. Additionally, we thank all members of the project team as well as the speakers and participants of the workshop.

References

[1] DECHEMA e. V. Homepage of NFDI (Nationale Forschungsdateninfrastruktur) for Catalysis related Science. https://nfdi4cat.org, 6 2022.

[2] M. Wu, G. Zhang, H. Tong, X. Liu, L. Du, N. Chen, J. Wang, T. Sun, T. Regier, and Sun. S. Cobalt (II) oxide nanosheets with rich oxygen vacancies as highly efficient bifunctional catalysts for ultra-stable rechargeable Zn-air flow battery. *Nano Energy*, 79:105409, 2021. 10.1016/j.nanoen.2020.105409.

[3] E. Marini, D. O. De Souza, G. Aquilanti, M. Liebert, F. Rossi, and B. Bozzini. Operando XAS of a Bifunctional Gas Diffusion Electrode for Zn-Air Batteries under Realistic Application Conditions. *Applied Sciences*, 11:11672, 2021. 10.3390/app112411672.

[4] B. Yang, H. Li, Z. Zhang, K. Xiao, M. Yang, F. Zhang, M. Wang, X. Guo, Q. Li, W. Fu, R. Si, L. Wang, and H. Chen. Nickel dual-atom catalysts for the selective electrocatalytic debromination of tribromoacetic acid as a green chemistry process. *Chemical Engineering Journal*, 427:131719, 2022. 10.1016/j.cej.2021.131719.

[5] M. Guo, M. J. Gray, H. Job, C. Alvarez-Vasco, S. Subramaniam, X. Zhang, L. Kovarik, V. Murugesan, S. Phillips, and K. K. Ramasamy. Uncovering the active sites and demonstrating stable catalyst for the cost-effective conversion of ethanol to 1-butanol. *Green Chemistry*, 23:8030, 2021. 10.1039/D1GC01979A.

[6] Y. Injongkol, P. Khemthong, N. Yodsin, Y. Wongnongwa, N. Sosa, S. Youngjan, T. Butburee, B. Rungtaweevoranit, S. Kiatphuengporn, J. Wittayakun, F. Roessner, and S. Jungsuttiwong. Combined in situ XAS and DFT studies on the role of Pt in zeolite-supported metal catalysts for selective n-hexane isomerization. *Fuel*, 314:123099, 2022. 10.1016/j.fuel.2021.123099.

[7] L. J. A. Macedo, A. A. E. Santo, G. C. Sedenho, A. Hassan, R. M. Iost, G. T. Feliciano, and F. N. Crespilho. Three-dimensional catalysis and the efficient bioelectrocatalysis beyond surface chemistry. *Journal of Catalysis*, 401:200, 2021. 10.1016/j.jcat.2021.07.022.

[8] M. Chen, D. A: Cullen, S. Karakalos, X. Lu, J. Cui, A. J. Kropf, H. Mistry, K. He, D. J. Myers, and G. Wu. Single Atomic Iron Site Catalysts via Benign Aqueous Synthesis for Durability Improvement in Proton Exchange Membrane Fuel Cells. *Journal of the Electrochemical Society*, 168:044501, 2021. 10.1149/1945-7111/abf014.

[9] J. Kang, Q.-Y. Fan, W. Zhou, Q. Zhang, S. He, L. Yue, Y. Tang, L. Nguyen, X. Yu, Y. You, H. Chang, X. Liu, L. Chen, Y. Liu, F. Tao, J. CHeng, and Y. Wang. Iridium boosts the selectivity and stability of cobalt catalysts for syngas to liquid fuels. *Chem*, 8:1050, 2022. 10.1016/j.chempr.2021.12.016.

[10] A. C. Thompson, D. Lindau, I. Attwood, Y. Liu, E. Gullikson, P. Pianetta, M. Howells, A. Robinson, K.-J. Kim, J. Scofield, J. Kirz, J. Underwood, J. Kortright, G. Williams, and H. Winick. *X-Ray Data Booklet*. Lawrence Berkeley National Laboratory, 2009.

[11] Go FAIR. Homepage of Go FAIR. https://www.go-fair.org/, 1 2022.

[12] Deutsches Elektronen-Synchrotron DESY. DESY Photon Science Users' Meeting – Overview Satellite Meetings. https://indico.desy.de/event/36974/page/4290-overview-satellite-meetings, 01 2023.

[13] Deutsches Elektronen-Synchrotron DESY. PETRA IV, New Dimensions – Partner for research and industry. https://petra4.desy.de/petra_iv/users_and_industry/index_eng.html, 11 2022.

[14] DAPHNE4NFDI. DAPHNE4NFDI Homepage (Data for Photon and Neutron Experiments for Nationale Forschungsdateninfrastruktur). https://www.daphne4nfdi.de/english/index.php, 9 2022.

[15] Z. Wu, W. K. Pang, L. Chen, B. Johannessen, and Z. Guo. In Situ Synchrotron X-Ray Absorption Spectroscopy Studies of Anode Materials for Rechargeable Batteries. *Batteries & Supercaps*, 4:1547, 2021. 10.1002/batt.202100006.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 871072.

Supplement

SATELLITE WORKSHOP - Photon Science

DESY.

X-Ray Absorption Spectroscopy today and perspectives for future PETRA III and IV beamlines

Thursday, 26. January 2023 , Bldg.3
BAH I + II

Organizers: W. Caliebe, E. Welter, M. Nentwich (DESY)

PROGRAMME		
11:00	Status of P64	A. Kalinko
11:15	Technical Developments	M. Goerlitz
11:30	Status of P65	E. Welter
11:45	Status of XEOL @P65	S. Levcenco
12:00	Laser Pump X-ray Probe at P64	M. Naumova
12:15	Research with the Autoclave	M. Borchert
12:30	***LUNCH***	
	Plans for PETRA IV	
13:30	Plans for P63/MatSciBL	W. Caliebe
13:45	Plans for (time-resolved) XAS/XES	A. Kalinko
14:00	Plans for Analytical XAS	E. Welter
14:15	Discussion	
14:30	Coffee break	
	Beamline Automation	
14:45	Automatization of synchrotron experiments - present and future	Alexander Schökel
15:05	SECoP - the Sample Environment Communication Protocol	Klaus Kiefer (Via Zoom)
15:25	TBD	Peter Weidler
15:45	XAS reference database under DAPHNE4NFDI	Sebastian Paripsa
16:05	Relevanz von Automatisierung in der Industrie	Bernd Hinrichsen
16:25	TBD	Janis Timoshenko
16:45	Discussion	
17:00	**End of Meeting**	

Figure 3: Program of the Satellite Workshop as in the Draft program of the Satellite.

Title: **X-Ray Absorption Spectroscopy today and perspectives for future PETRA III and IV beamlines** **Draft programme**
Organizer: W. Caliebe, E. Welter, M. Nentwich (DESY)
Venue: Bldg. 3, BAH I+II
Duration: 11.00 - 17.00h
Type: Hybrid
Short description:
This year's XAFS satellite meeting focuses on the status of PETRA III XAFS beamlines as well as on upcoming and ongoing projects. One session is intended to give an overview about the status of the two currently operating XAFS beamlines P64 and P65 and present some resent work by different user groups. A second session will present the current status of the planning for XAFS beamlines at PETRA IV and of the upcoming XAFS/XRD beamline P63. The third session is dedicated to two projects which aim at the implementation of automated in-situ/operando experiments at the existing PETRA III and future PETRA IV XAFS beamlines. Their main objective is basically to enable mail-in experiments in catalysis and related fields and to make them a normal mode of operation for industrial and academic users in future years. These projects are driven by the experience during the Corona pandemic and by the wish to lower the activation barrier for industrial users of synchrotron radiation.
Present and future users are strongly encouraged to participate in this workshop and to discuss their options with the beamline staff.

Figure 4: Abstract of the Satellite Meeting as in [12].

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 871072.