ChemCatBio
Chemical Catalysis for Bioenergy

Addressing Rigor and Reproducibility in Thermal, Heterogeneous Catalysis

John West, *Johnson Matthey*
Neil Schweitzer, *Northwestern University*
Rajamani Gounder, *Purdue University*
Robert Rioux, *Penn State University*
January 24, 2024

U.S. DEPARTMENT OF ENERGY | *Office of* ENERGY EFFICIENCY & RENEWABLE ENERGY
BIOENERGY TECHNOLOGIES OFFICE

Resources

- **Website: chemcatbio.org**
  - Tools and capabilities
  - Publications
  - Webinars
  - Interactive technology briefs
- **Tools:**
  - Catalyst Property Database
  - CatCost
  - Surface Phase Explorer
- **Newsletter: The Accelerator**
  - Subscribe

Housekeeping

- Attendees will be in listen-only mode
- Audio connection options:
  - Computer audio
  - Dial in through your phone (best connection)
- Automated closed captions are available
- Use the Q&A panel to ask questions
- Technical difficulties? Contact Erik Ringle through the chat section, lower right of your screen
- Recording will be available at: https://www.chemcatbio.org/webinars.html

Today’s Speakers

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First Speaker

John West

Johnson Matthey

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Johnson Matthey: strong credentials supporting our strategy

Strong brand
206 year history

Technology leadership
#1 or 2
in chosen markets

2022/23 sales[1]
£4,201 million

12,600 employees
worldwide[2]

[1]Sales excluding precious metals [2] As at 31st March 2023

Over 200 years of solving the world's biggest challenges

1817
Precious metal assayer

1918
Powell-Deering method

1974
The world's first emissions control autocatalyst

2002
Hydrogen fuel cell components

2020
Hydrogen electrolyser components

Catalysis

Metals chemistry

Process engineering

1874
Standard kilogramme weight

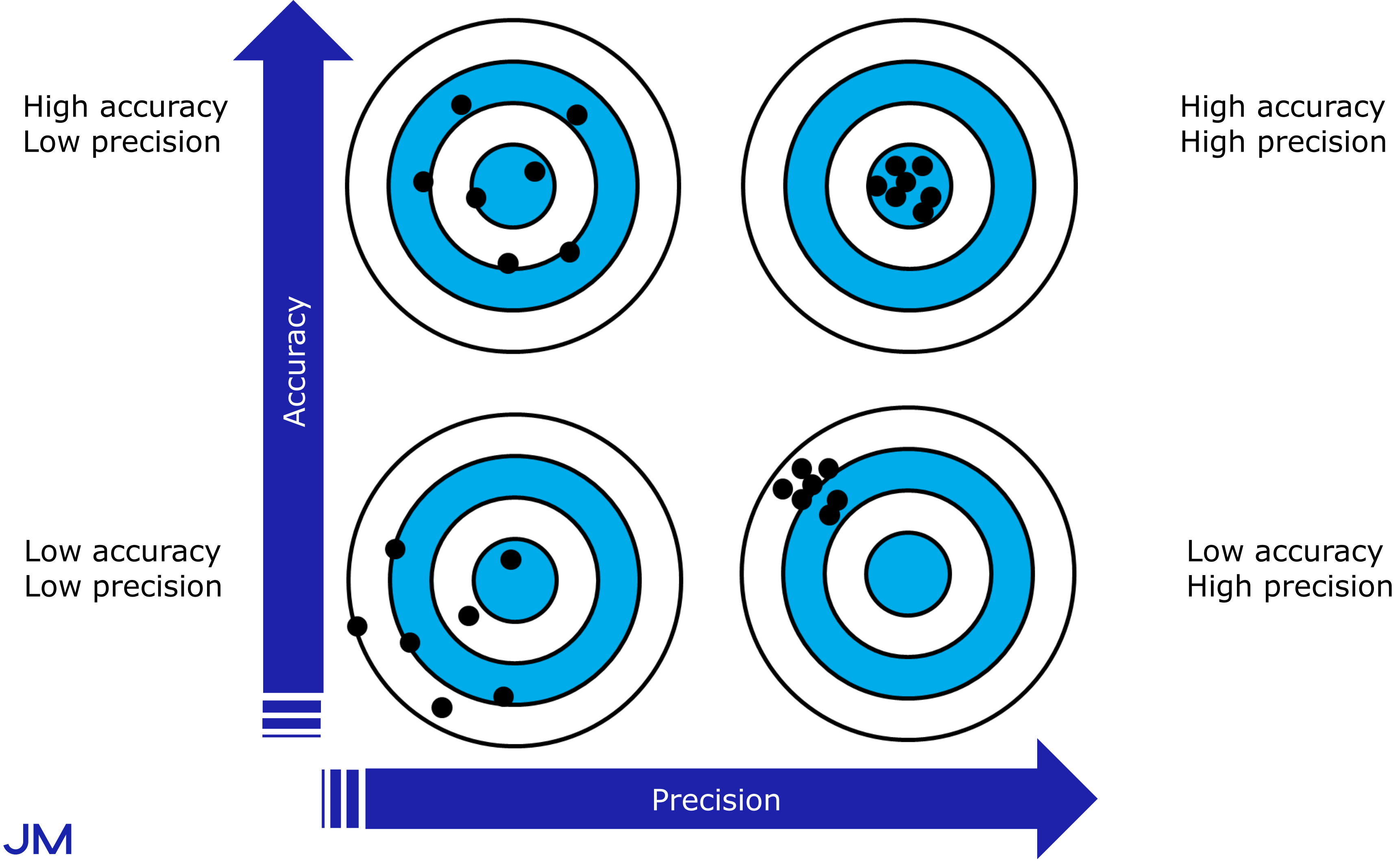
1960
Electrocatalysts for NASA's fuel cells

1983
Platinum group metal circular economy

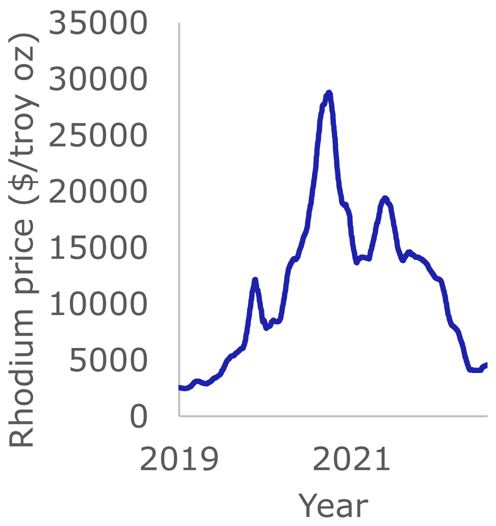
2002
Syngas and chemicals process design and licensing

2018
Our first commercial sustainable fuels plant license

JM

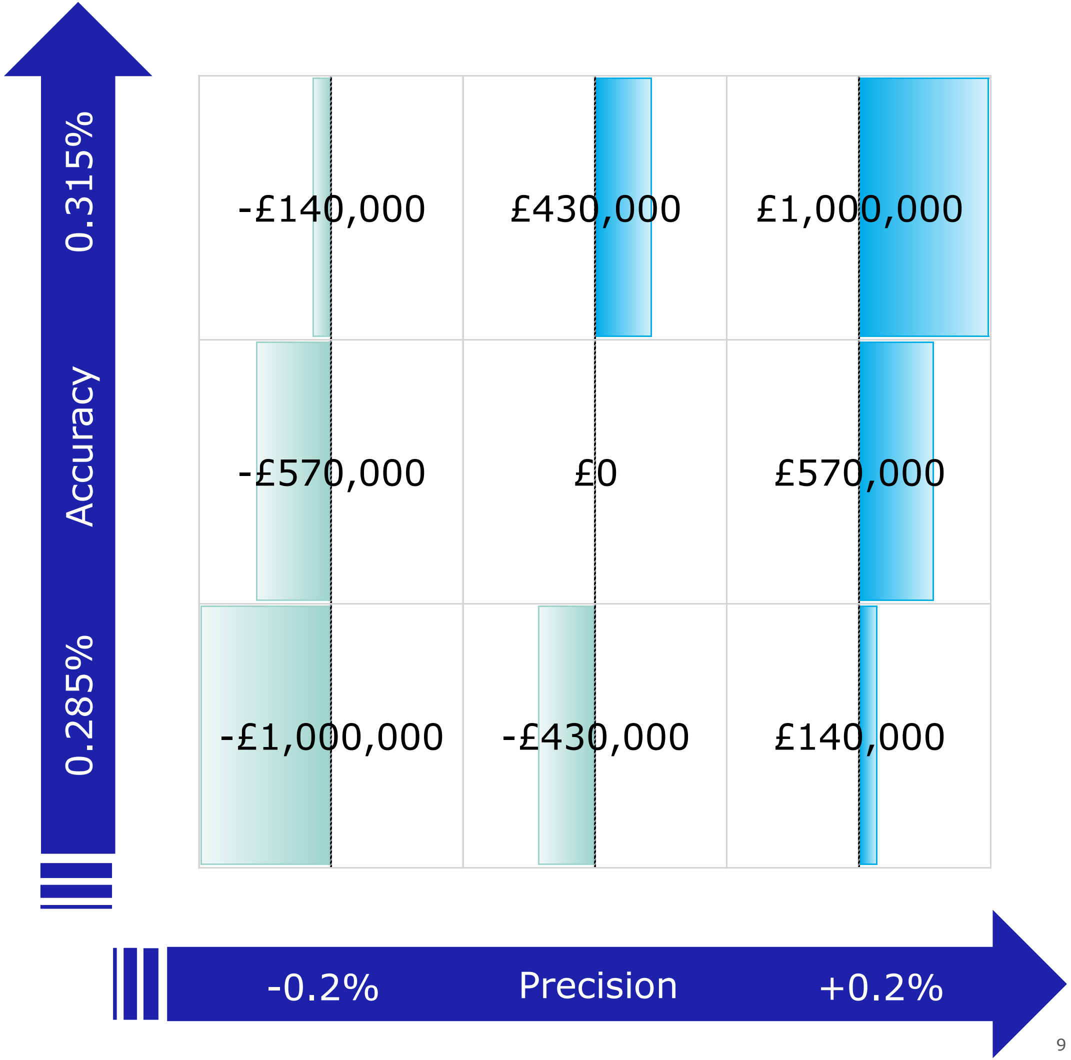

High accuracy
Low precision
High accuracy
High precision
Accuracy
Low accuracy
Low precision
Low accuracy
High precision
Precision

JM



Scenario:

- Target Rhodium loading: 0.3 wt%
- Catalyst loading 3 metric tonnes
  - ~12 drums of catalyst
- Value of Rhodium £8.5 million



Next Speaker

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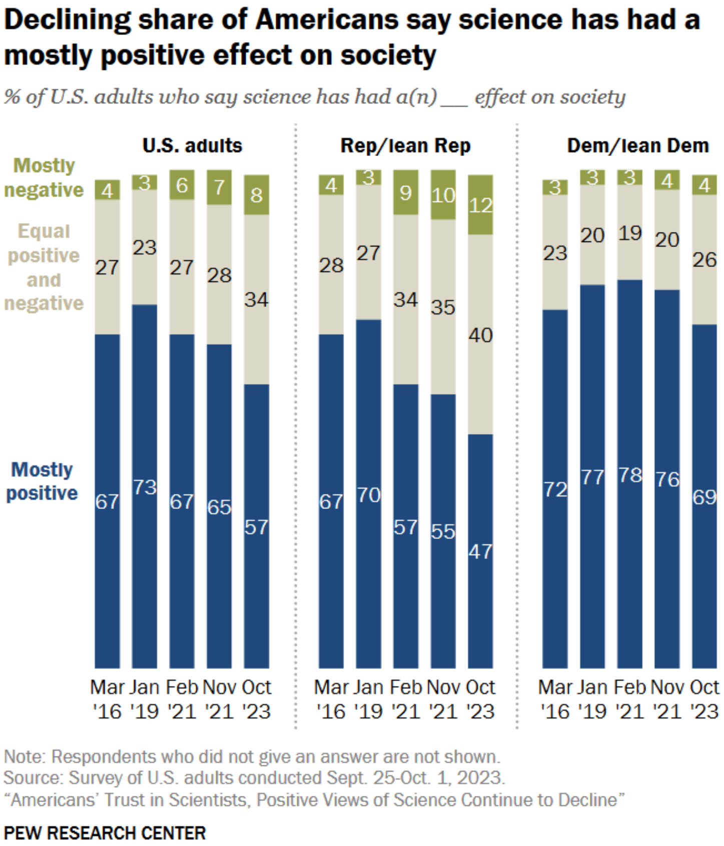
Science Has a Public Perception Problem



- **What drives mistrust in Science?**
  - Influence of political groups/lobbies
  - Public misunderstanding of the scientific method
    - Media coverage of reproducibility issues

A Reproducibility *CRISIS!!!*

Essay

Why Most Published Research Findings Are False

John P. A. Ioannidis

Why Science Is Not Necessarily Self-Correcting

John P. A. Ioannidis

...Research Center, Department of Medicine and Department of ...ford University School of Medicine, and Department ... School of H...

Believe it or not: how much can we rely on published data on potential drug targets?

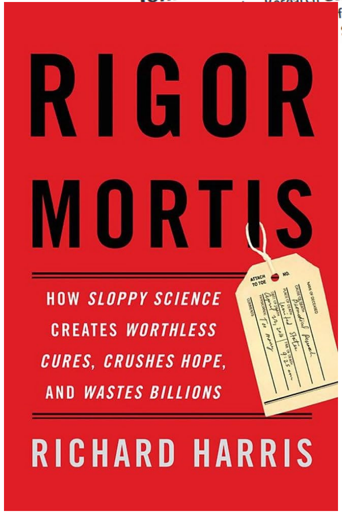
Florian Prinz, Thomas Schlange and Khusru Asadullah

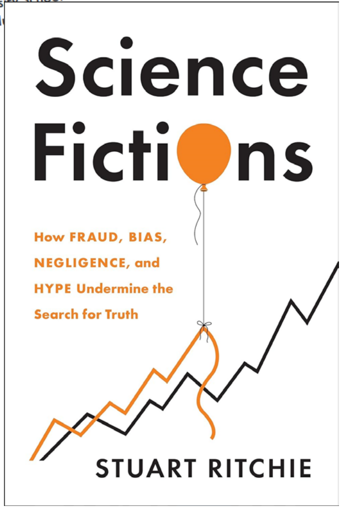
FUTURE PERFECT SCIENCE

Science has been in a "replication crisis" for a decade. Have we learned anything?

Bad papers are still published. But some other things might be getting bett...

By Kelsey Piper | Oct 14, 2020, 12:20pm EDT





WIKIPEDIA The Free Encyclopedia

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Replication crisis

18 languages

Article Talk | Read Edit View history Tools

Contents hide

From Wikipedia, the free encyclopedia

The **replication crisis** (also called the **replicability crisis** and the **reproducibility crisis**) is an ongoing methodological crisis in which the results of many scientific studies are difficult or impossible to reproduce. Because the reproducibility of empirical results is an essential part of the scientific method,[2] such failures undermine the credibility of theories building on them and potentially call into question substantial parts of scientific knowledge.

The replication crisis is frequently discussed in relation to psychology and medicine, where considerable efforts have been undertaken to reinvestigate classic results, to determine whether they are reliable, and if they turn out not to be, the reasons for the failure.[3][4] Data strongly indicates that other natural and social sciences are affected as well.[5]

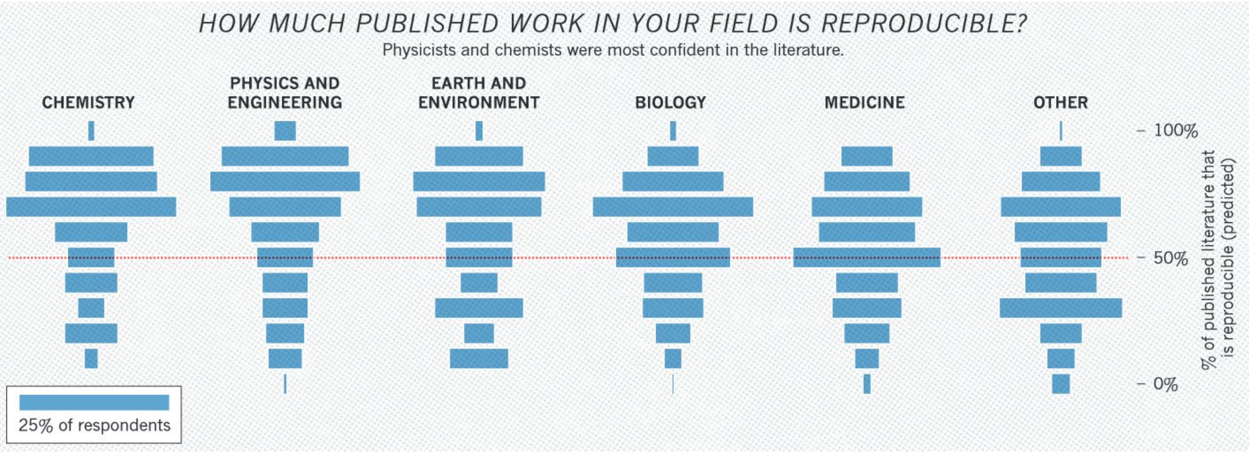
The phrase *replication crisis* was coined in the early 2010s[6] as part of a growing awareness of the problem. Considerations of causes and remedies have given rise to a new scientific discipline, metascience,[7] which uses methods of empirical research to examine empirical research practice.

Considerations about reproducibility fall into two categories. *Reproducibility* in the narrow sense refers to re-examining and validating the analysis of a given set of data. *Replication* refers to repeating the experiment or study to obtain new, independent data with the goal of reaching the same or similar conclusions.

Ioannidis (2005): "Why Most Published Research Findings Are False".[1]

Scientists Tend to Agree…

- *Nature* survey of 1,576 scientists (106 which were chemists)



This is admittedly a small sample size of self-selected participants…

BUT is consistent with the attitude of many researchers I have interacted with

Baker, Monya. "1,500 Scientists Lift the Lid on Reproducibility." Nature 533, no. 7604 (2016): 452–54. https://doi.org/10.1038/533452a.

Why Is Reproducibility Important?

- **It is important for a research community to have reasonable expectations for the *normal variance* of a particular measurement**
  - Interlaboratory studies are used to determine these values
  - Standard practice for institutions like NIST and ASTM


ACS central science

IN FOCUS

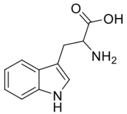
An International Study Evaluating Elemental Analysis

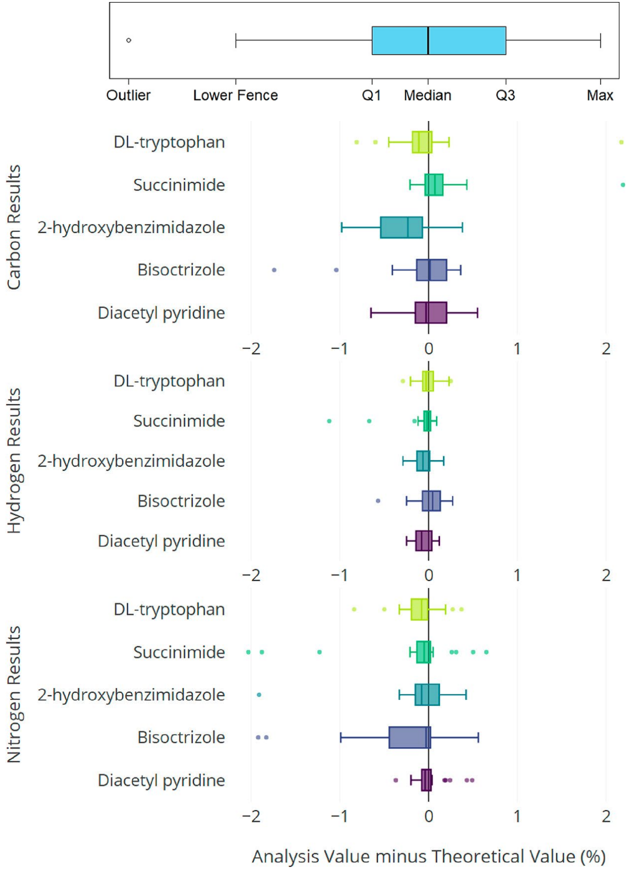
Cite This: ACS Cent. Sci. 2022, 8, 855–863

Rupert E. H. Kuveke, Lachlan Barwise, Yara van Ingen, Kanika Vashisth, Nicholas J. Roberts, Saurabh S. Chitnis, Jason L. Dutton, Caleb D. Martin, Rebecca L. Melen

Examining the literature, we have not been able to determine why ±0.4% was chosen as the standard requirement.

Compound	C (%)	H (%)	N (%)
DL-tryptophan	64.66 (64.69)	5.91 (5.92)	13.62 (13.72)

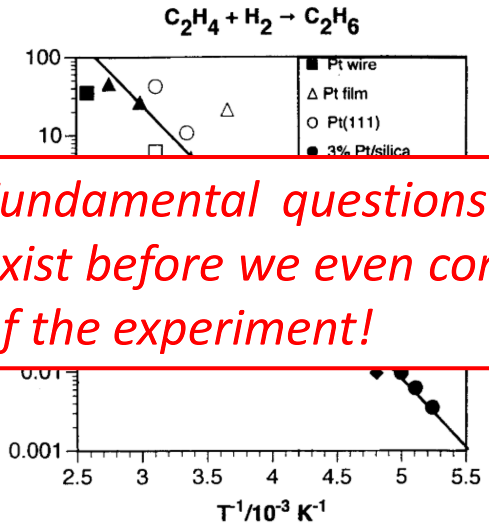




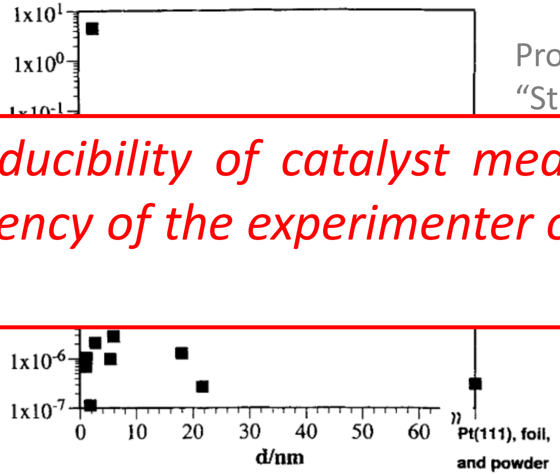
Kuveke, R. E. H.; Barwise, L.; van Ingen, Y.; Vashisth, K.; Roberts, N.; Chitnis, S. S.; Dutton, J. L.; Martin, C. D.; Melen, R. L. An International Study Evaluating Elemental Analysis. ACS Cent. Sci. 2022, 8 (7), 855–863. DOI: 10.1021/acscentsci.2c00325.

Is Data Reproducible in Heterogeneous Catalysis?

- **There have been relatively few studies in reproducibility in catalysis, particularly comparing catalyst rates**
  - Ample reviews reporting data, but there are few analysis of the data
- **What is a reasonable variance for reported catalyst rates? An order of magnitude?**
  - Maybe it depends on the materials and the chemistry?



Ethylene Hydrogenation



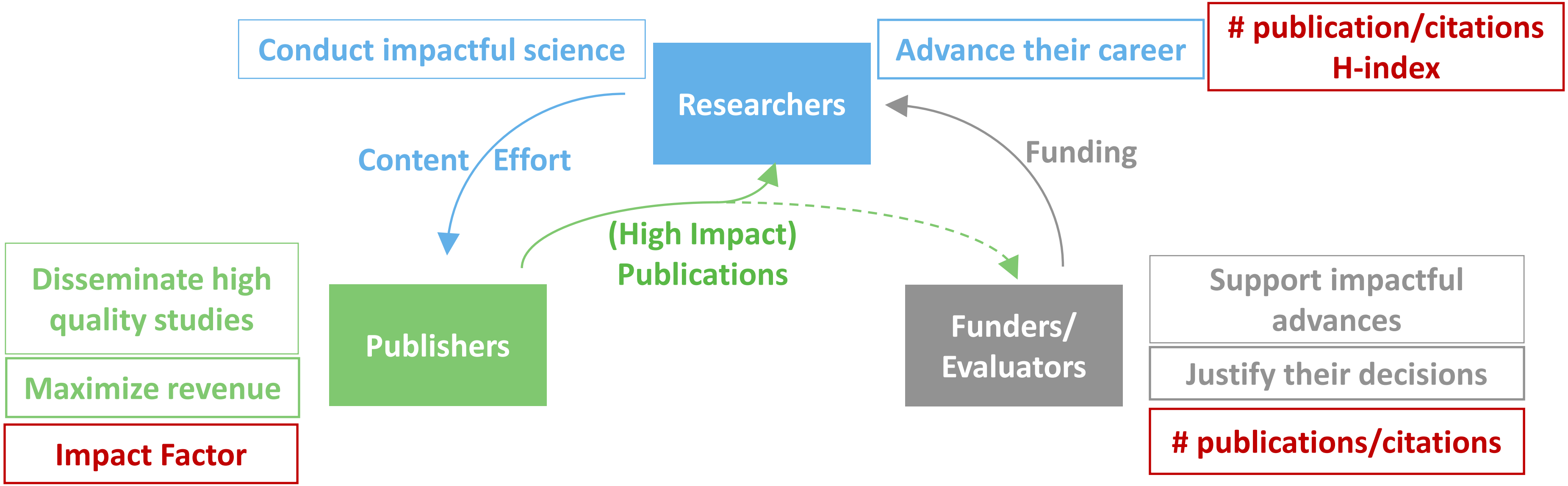
Propane Hydrogenolysis
“Structure Sensitive”

Fundamental questions about the reproducibility of catalyst measurements exist before we even consider the competency of the experimenter or the rigor of the experiment!

RIBEIRO, F. H.; SCHACH VON WITTENAU, A. E.; BARTHOLOMEW, C. H.; SOMORJAI, G. A. Reproducibility of Turnover Rates in Heterogeneous Metal Catalysis: Compilation of Data and Guidelines for Data Analysis. Catalysis Reviews 1997, 39 (1-2), 49–76. DOI: 10.1080/01614949708006468.

Why isn't R+R a priority in science?

- A framework for the research ecosystem:



Citation and publication-based metrics incentivize quantity and haste

Scott, Susannah L., T. Brent Gunnoe, Paolo Fornasiero, and Cathleen M. Crudden. ***"To Err Is Human; To Reproduce Takes Time."***
ACS Catalysis 12, no. 6 (March 18, 2022): 3644–50. https://doi.org/10.1021/acscatal.2c00967.

Planning a Workshop

- **What we know:**
  1. Researchers have little confidence of data reported in the literature
  2. Catalysis is a complex science
     - *Irreproducibility* could stem from any stage of a study: synthesis, storage, characterization, or testing
     - There are lots of unknowns concerning the inherent reproducibility of measurements made in our field
  3. The current research ecosystem incentivizes publishing a lot and publishing quickly!

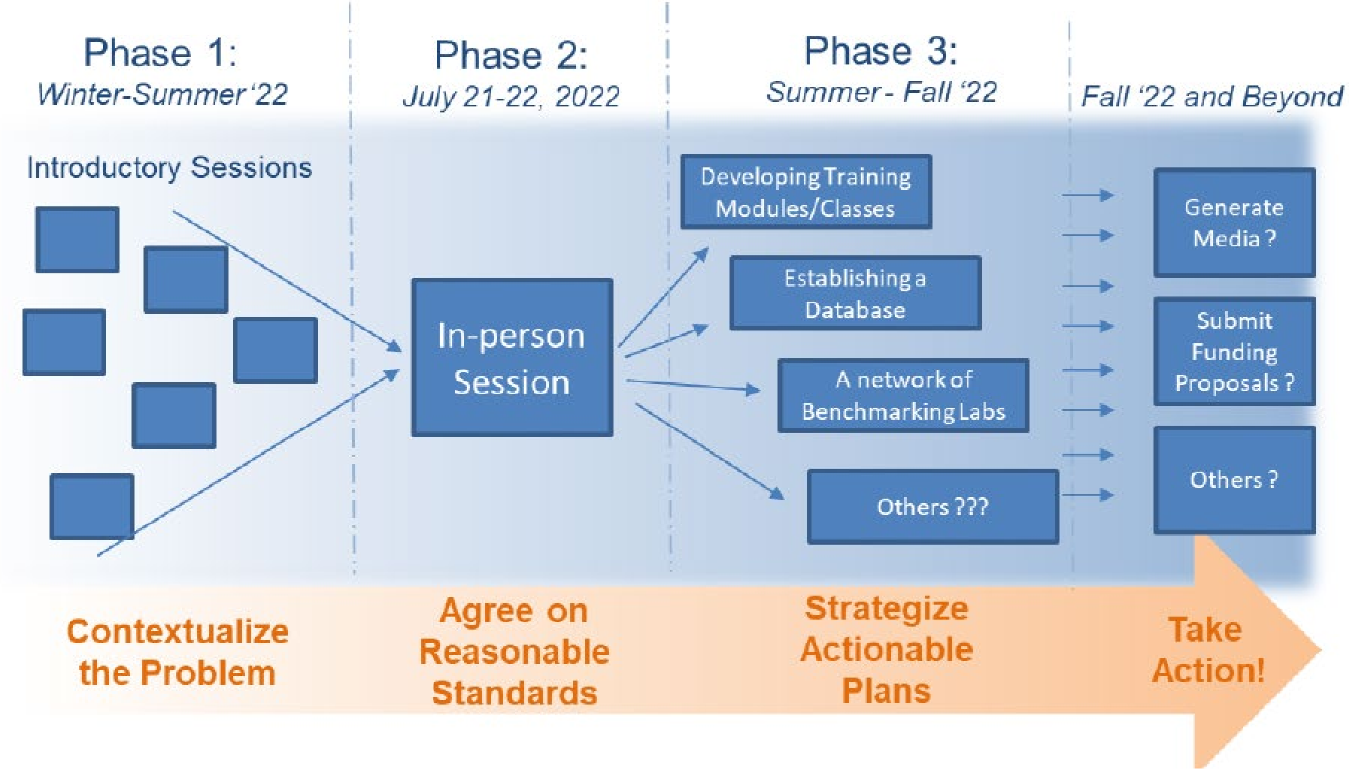
- **How can we make an impact? (with little to no control over publishers or funders)**
  - Our approach was to target two specific groups:

  1. Reviewers – the first line of defense for R+R in scientific literature
  2. New researchers in our field – who need to learn a lot of information in a little amount of time

Next Speaker

Robert Rioux
Penn State University
rmr189@psu.edu

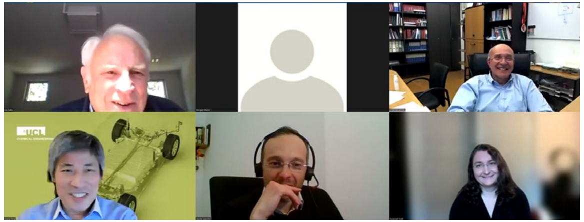
Addressing Rigor and Reproducibility in Heterogeneous, Thermal Catalysis Workshop



Financial support provided by the National Science Foundation, CBET Division, Catalysis Program under Grant No. 2152559 and the U.S. Department of Energy, Office of Science, Basic Energy Sciences under Award No. DE-SC0022918.

Phase I. Setting the Stage. Introducing the Problem. Learning From Other Fields.

- How widespread is the problem?
- What does it mean for data to be reproducible?
- What are possible systemic, institutional, or individual causes?
- How do researchers think about these problems in other fields?
- Can this problem be "solved," and what would even be considered progress?



The Ongoing Battle for More Credible Science: Identifying Interdisciplinary Lessons
Jennifer Tackett, Northwestern University and Editor-in-Chief of Clinical Psychological Science

Lessons Learned From Systematic Studies of Experimental Replication in Adsorption Science
David Sholl, Georgia Institute of Technology, Oak Ridge National Lab, and Editor-in-Chief of AIChE Journal

The Importance of Standard Operation Procedures For Catalysis Research Accelerated By Artificial Intelligence
Annette Trunschke, Fritz Haber Institute of the Max Planck Society

The Data Sea Scrolls
John Kitchin, Carnegie Mellon University

A Unique Journal for the Publication of Reproducible Methods for the Synthesis of Organic Compounds
Rick Danheiser, Massachusetts Institute of Technology and Editor-in-Chief of Organic Syntheses

Panel Discussion with Journal Editors
Moderator: Bruce Gates, UC Davis
Panel: Susannah Scott (ACS Catalysis), Johannes Lercher (Journal of Catalysis), Davide Esposito (Nature Catalysis), Junwang Tang (Chinese Journal of Catalysis)

Phase II. Two-Day Hybrid (In-Person/Virtual) Workshop

- **The immediate outcome of this workshop will be an open access report with:**
- **Technical Content**
  - Best practices for reporting data using common methodologies
  - Recommendations for the use of benchmark materials
- **For New Researchers:**
  - Information currently only available in:
    - Hard-to-search/access literature articles
    - Specialized, expensive textbooks
    - The oral histories of some academic trees
- **For Reviewers:**
  - Serve as a reference for referring authors to best practice resources
  - Help establish consistent guidelines for manuscript/proposal acceptance across different journals and funding agencies.

Day 1 - Standardized Method Reporting
Bulk Synthesis (oxides, zeolites, MOFs, etc.)
Deposition Synthesis (SEA)
Catalyst Testing (Flow, Batch)
Bulk Characterization (XRD, TPx, Physisorption)
Site Characterization (probe molecules, titrations)
Advanced Characterization (XAS, microscopy)

Day 2 - Guidelines for Benchmark Materials
Supported Metal Nanoparticles
Single Atom Metals
Metal Oxides
Zeolites
Metal Organic Frameworks
Bifunctional Materials

Phase II. Workshop Report

Table of Contents

This report is divided among sections that align with the focused topics of breakout group discussions during the in-person workshop.

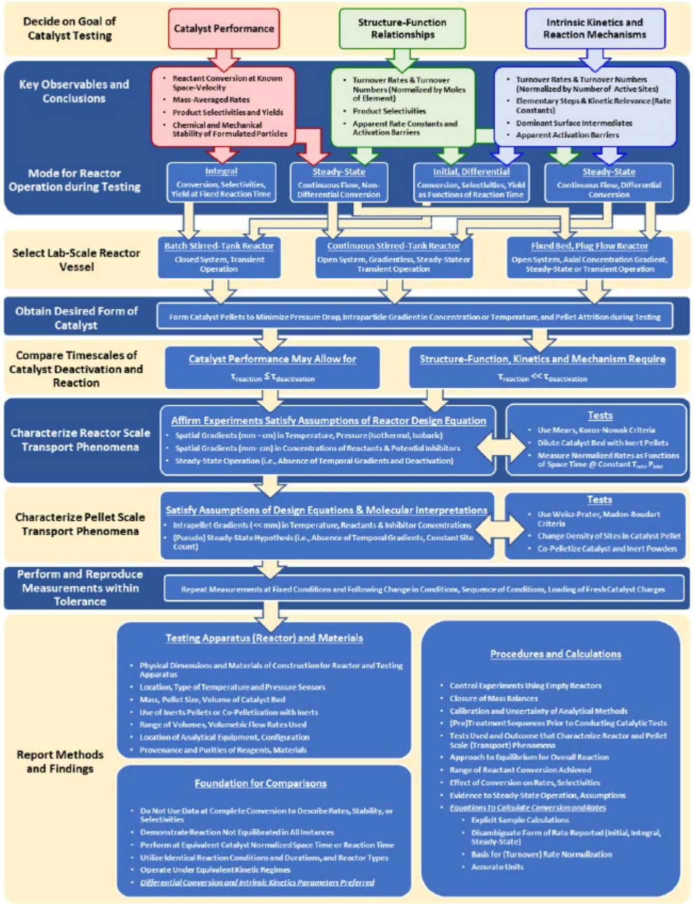
- **As an example:**
- Section 5 focuses on best practices for reactivity testing. The following report template for "Recommendations for Catalyst Testing" includes
  - Common Applications
  - Known Limitations
  - Specific Recommendations for Reporting Data in Literature
  - References for Best Practices

https://www.catalysisrr.org/

https://doi.org/10.5281/zenodo.8029159

Phase II Workshop Report. Catalyst Testing as an Example of Report Layout

Workflow for planning, executing and reporting the results from catalytic testing



- **Catalyst testing typically serves to achieve multiple objectives, which include:**
  - **Catalyst Performance:** Time-on-stream characteristics of ***rates, selectivities and yield***; regenerability.
  - **Structure-Function Relationships:** Quantitative comparisons between ***material descriptors and observed catalytic*** properties.
  - **Determination of Mechanisms and Intrinsic Kinetics:** Identification of the reactive intermediates and intervening elementary steps responsible for consuming reagents and forming products.
- **Technical recommendations to improve rigor and reproducibility**
  - Laboratory reactor selection and design
  - Isothermal and isobaric operation
  - Concentration gradients in reactors
  - Contacting pattern in flow reactors
  - Steady state operation
  - Reproducibility and controls
  - Pellet scale phenomena relevant for catalyst testing
- **Recommendations for reporting results of catalyst testing**
  - Report normalized rates of reaction and not conversion or temperature required to attain a specific conversion
  - Procedures and calculations in reporting of catalyst testing

Next Speaker

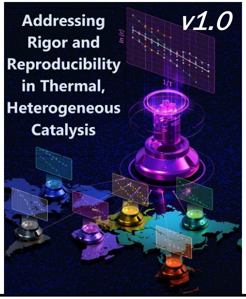
Rajamani Gounder

Purdue University

rgounder@purdue.edu

Future Activities

- Future workshops on topics related to rigor and reproducibility
  - **Thermal, heterogeneous catalysis (v2.0)**
    - Planning to be held in coordination with a major (inter)national meeting
    - Updates to current workshop report
      - Preparing sections on materials, methods, etc. not covered in v1.0
  - **Electrocatalysis**
    - Co-organized by Eric Stuve and Liney Arnadottir
    - **Steering committee:** Jingguang Chen, Suljo Linic, Ezra Clark, Nirala Singh, Kelsey Stoerzinger, Buddie Mullins, and Gregory Jerkiewicz.
    - To be held July 9–11, 2024 at the University of Washington
  - **Homogeneous Catalysis**
    - Organized by Rory Waterman
  - **Machine Learning in Catalysis**
    - Organized by Hongliang Xin, John Kitchin, Nuria López, Neil Schweitzer



Future Activities

- Shorter articles and guides to be published on focused topics

Journal of Catalysis 429 (2024) 115259

Contents lists available at ScienceDirect

Journal of Catalysis

journal homepage: www.elsevier.com/locate/jcat

ELSEVIER

Recommendations to standardize reporting on the synthesis of heterogeneous catalysts

Ana C. Alba-Rubio [a], Phillip Christopher [b], Michelle L. Personick [c], Kara J. Stowers [d,*]

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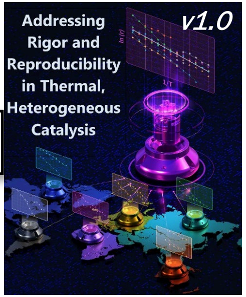
[d] Department of Chemistry and Biochemistry, Brigham Young University, Provo, UT 84604, USA

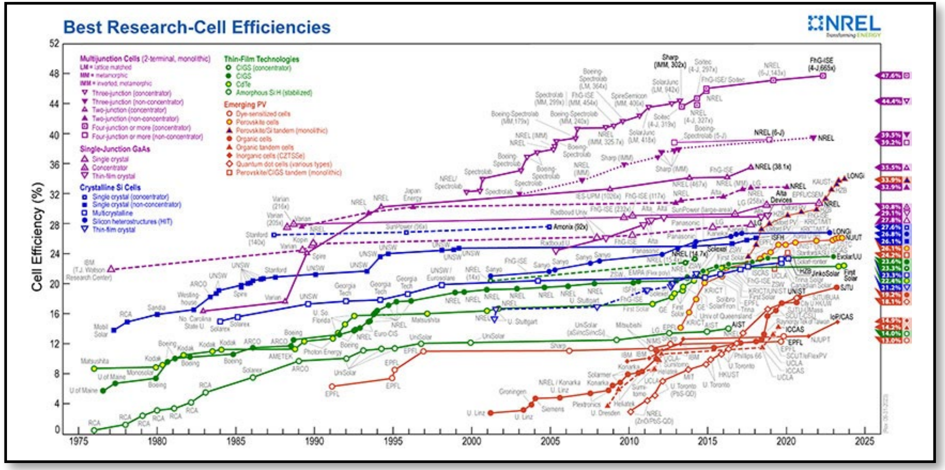
Future Activities



- **Other activities and initiatives for the community to consider**
  - Catalysis-focused Interlaboratory studies (ILS)
  - Mechanisms to make benchmark materials broadly accessible
  - Producing training videos and learning modules

- **Broader issues in community adoption and incentivization**
  - Data storage, formatting, accessibility
  - Journal publications
  - Research proposals

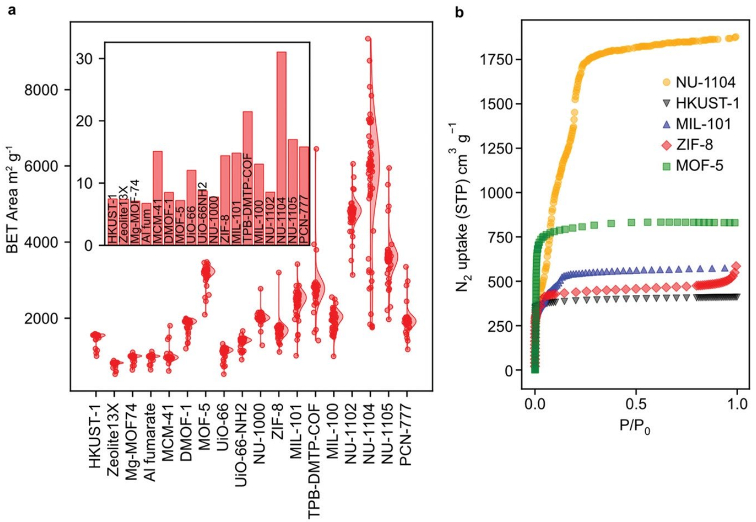
NREL – Photovoltaic Cell Device Performance
https://www.nrel.gov/pv/device-performance.html

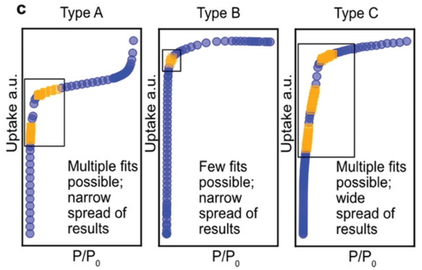


Future Activities

- **Catalysis-focused interlaboratory studies (ILS)**
  - Establish standard properties of benchmark materials
  - Identify sources of measurement of instrumentation error
  - Identify sources of variation (e.g., sample storage, pretreatment)
  - **The community would benefit from ILS**
    - Reaction rate measurements among labs (little precedent of such activities)
    - Case studies for different material classes and chemistries
      - e.g., Bio-feedstock processing often involves bi-functional (metal/acid) materials
      - Develop a benchmark sample (a physical mixture) and rate measurement
    - Methods (e.g., TEM, XAS)
  - Funding and accessibility models to prevent "gatekeeping"

61 different research labs were given isotherm data for 18 different MOF's and asked to calculate the BET surface area





Authors developed a computational algorithm – BET surface identification (BETSI) – to aid researchers

Fairen-Jimenez, D. et. al., "How Reproducible are Surface Areas Calculated from the BET Equation?" Advanced Materials, 2022, 34 (27), e2201502. DOI: 10.1002/adma.202201502.

Future Activities

- **Mechanisms to make benchmark materials broadly accessible**
  - Need routes for the sustainable and reproducible synthesis of (suitable) benchmark materials
    - One reason for the end of the Euro-Pt effort
  - Challenging to produce by a single source
    - A core facility might need to be established and funded to provide this service without passing the cost to the user
  - Alternatively, individual laboratories could use best practices and recipes
    - Organic development of a crowd-sourced database of synthesis, characterization and reaction data
    - Provide opportunities for training new researchers

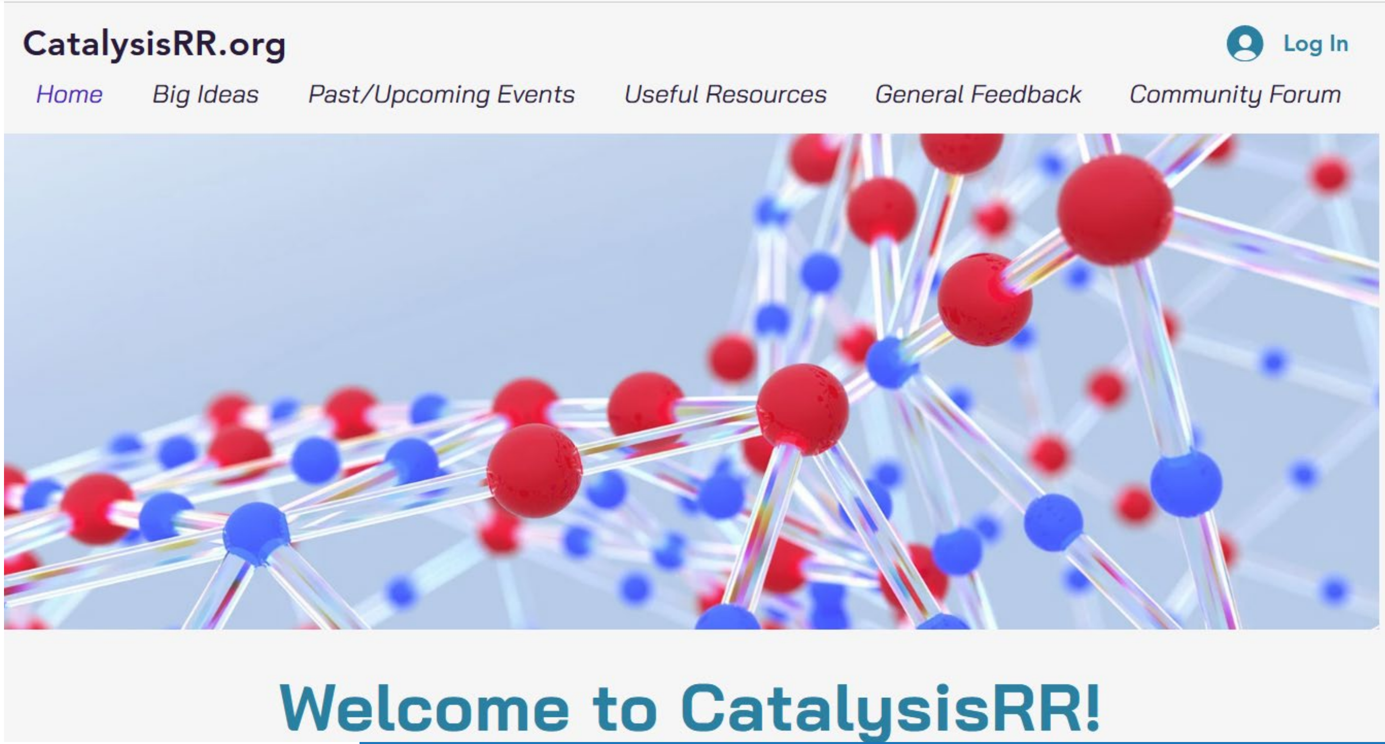
wikipedia.com

Future Activities

- **Producing training videos and learning modules**
  - Videos on synthesis, characterization, testing
    - "Tips and tricks" to improve reproducibility
  - Publications on best practices and techniques
    - "Beginner's Guides"
  - Workshops and short courses
  - Target new researchers (e.g., students) to the field
  - Accessible to diverse researchers to enable bringing in other scientific expertise to catalysis

youtube.com
(intro to chemistry lab techniques)

CatalysisRR.org. The Home of "Addressing Rigor and Reproducibility in Heterogeneous, Thermal Catalysis"



All information presented on past events have been archived on CatalysisRR.org. The greater vision of the effort, useful R&R resources and community R&R outreach are available too!

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BIOENERGY TECHNOLOGIES OFFICE

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Q&A

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