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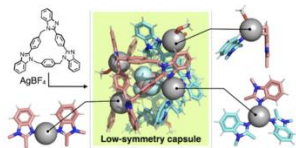
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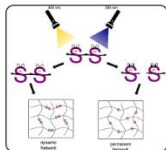
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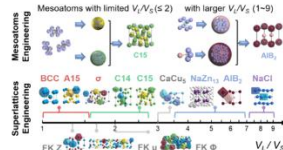
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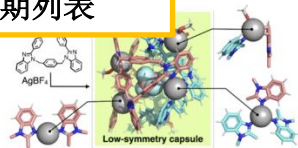
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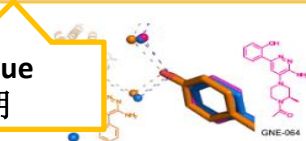
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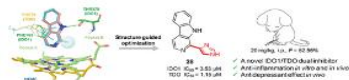
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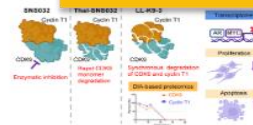
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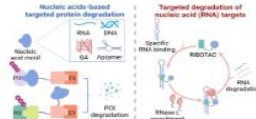
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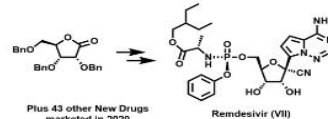
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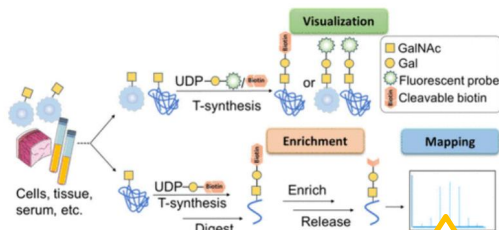
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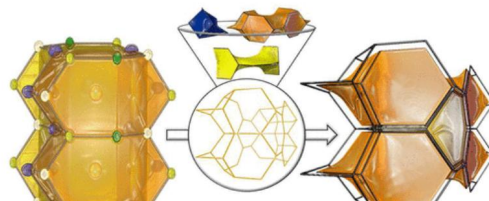
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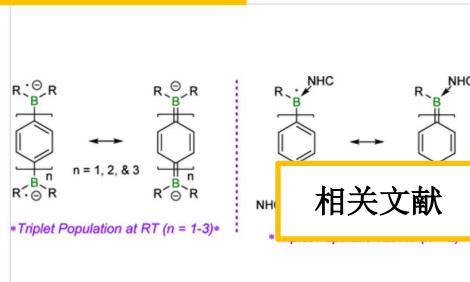
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Herein, we report the syntheses and electronic structures of crystalline dianionic as well as neutral diboron-centered classical diradicaloids as boron analogues of classical Thiele, Chichibabin, and Müller (this only for dianionic diradicaloids) hydrocarbons. These are based on borane radical anion and NHC-stabilized boryl radical spin carriers, respectively. All these dianionic diboron-centered diradicaloids exhibit triplet population at room temperature regardless of the π -conjugated spacer: *p*-phenylene, *p,p'*-biphenylene, or *p,p'*-terphenylene. In the case of neutral diboron-centered diradicaloids, the

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Breaking a Molecular Scaling Relationship Using a
Electrocatalyst for Oxygen Reduction

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Catalysts are essential to the efficient performance of technological systems and all living organisms. However, molecular scaling relationships (1-4) involving trade-offs between thermodynamic and kinetic performance metrics can limit their effectiveness. The Sabatier principle (5,6) indicates that optimal catalysis occurs when the binding between a catalyst and its substrate is "just right" and of intermediary strength. In other words, the interactions should be neither too strong nor too weak; otherwise, the binding of the reactants or desorption of the products will limit the reaction rate. Such trade-offs between thermodynamic and kinetic performance metrics also apply to electrocatalytic reactions, as exemplified in the benchmarking of homogeneous molecular electrocatalysts via comparisons of catalytic Tafel plots (7,8) relating the turnover frequency (TOF) of a molecular catalyst to the overpotential (η) (Figure 1a). In this analysis, TOF is the ratio of moles of product (N_{product}) produced over a set unit of time, in which the catalyst is stable, versus the moles of total catalysts contained within the *reaction-diffusion layer* (N_{cat}), a region near the electrode surface, where the concentration profiles of electro-activated versus nonactivated catalysts differ from their bulk values (eq 1). (8) Correspondingly, η is defined as the difference in absolute value between the applied electrode potential (E_{app}) and the equilibrium potential of the reaction being catalyzed (E_{eq} ; eq 2). (9-11).

$$\text{TOF} = \frac{N_{\text{product}}}{N_{\text{cat}}} = \frac{\text{TOF}_{\text{max}}}{1 + \exp \left[\frac{F}{RT} (E_{\text{app}} - E_{\text{cat}/2}) \right]} \quad (1)$$



Figures



References



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Scaling Relationships
DOI: 10.1038/s4192

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2. Masa, J.; Schuhm
Electrocatalysis. J. S
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Molecular Scaling Relationships for the Electrochemical
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Understanding Catalytic Activity Trends in the Oxygen

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Reversible Spatiotemporal Control of Induced Protein Degradation by Bistable PhotoPROTACS

Patrick Pfaff^{1,†}, Kusal T. G. Samarasinghe^{2,†}, Craig M. Crews^{2,3,4} and Erick M. Carreira¹

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², Department of Molecular, Cell, and Developmental Biology, Yale University, 260 Whitney Avenue, New Haven, CT 06511, United States

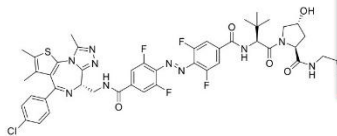
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(2S,4R)-1-((S)-2-(4-(E)-4-(((S)-4-(4-chlorophenyl)-2,3,9-trimethyl-6H-thieno[3,f][1,2,4]triazolo[4,3-a][1,4]diazepin-6-yl)methyl)carbamoyl)-2,6-difluorophenylideneamido)-3,3-dimethylbutanoyl)-4-hydroxy-N-(4-(4-methylthiazol-5-yl)benzyl)pyrrolidine-2-carboxamide (photoPROTAC-1)



JQ-1 amine **18** (10.5 mg, 28.0 μ mol, 1.00 equiv) and acid **54** (21.4 mg, 28.0 μ mol, 1.00 equiv) were dissolved in anhydrous DMF (0.28 mL, 0.1 M). DIPEA (12 μ L, 85 μ mol, 3.00 equiv) and HATU (11.3 mg, 30.0 μ mol, 1.05 equiv) were added to the reaction mixture at room temperature. After 2 hours, the reaction mixture was quenched by addition of sat. aq. NaHCO₃ and the aq. phase was extracted three times with EtOAc. The combined org. layers were washed with brine and dried over sodium sulfate. Residual DMF and tetramethylurea were removed by lyophilization after freezing in a water/dioxane mixture. The crude product was further purified by flash column chromatography (94% EtOAc/4% iPrOH/2% H₂O) to afford **photoPROTAC-1** as an orange oil (16.0 mg, 14.0 μ mol, 51%).

RF = 0.36 (85% EtOAc/10% iPrOH/5% H₂O).

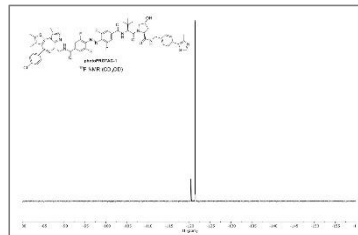
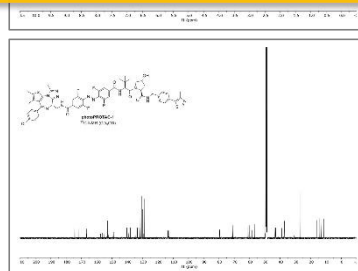
¹H NMR (500 MHz, CD₃OD) δ = 8.87 (s, 1H), 7.70 (dd, *J* = 5.1, 1.6 Hz, 2H), 7.67 (dd, *J* = 5.1, 1.6 Hz, 2H), 7.52 (d, *J* = 8.5 Hz, 2H), 7.48 (d, *J* = 8.5 Hz, 2H), 7.44 – 7.40 (m, 4H), 4.91 (s, 1H), 4.65 – 4.50 (m, 4H), 4.43 (dd, *J* = 13.6, 7.0 Hz, 2H), 4.35 (d, *J* = 15.4 Hz, 1H), 3.98 (d, *J* = 11.0 Hz, 1H), 3.87 (dd, *J* = 11.0, 3.8 Hz, 1H), 2.71 (s, 3H), 2.47 (s, 3H), 2.43 (s, 3H), 2.29 – 2.22 (m, 1H), 2.15 – 2.09 (m, 1H), 1.69 (s, 3H), 1.13 (s, 9H).

¹³C NMR (126 MHz, CD₃OD) δ = 174.4, 172.0, 166.8, 166.7, 166.5, 157.4, 156.1, 155.3, 153.0, 152.2, 149.0, 140.3, 139.2, 138.1, 138.1, 134.3, 133.5, 133.4, 133.3, 133.3, 132.0, 132.0, 131.5, 131.4, 131.3, 130.4, 129.8, 129.0, 113.4, 113.1, 71.1, 60.9, 59.9, 58.2, 56.8, 43.7, 42.9, 39.0, 37.2, 27.1, 15.8, 14.4, 12.9, 11.6.

¹⁹F NMR (471 MHz, CD₃OD) δ = -121.4, -121.5.

IR: 3322, 2925, 28855, 1665, 1533, 1427, 1343, 1243, 1090, 1047, 967, 843.

ESI-HRMS: calcd. for C₅₀H₅₂ClF₄N₁₀O₅S₂ [M+H]⁺ 1108.3135, found 1108.3144.



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COMMUNICATION | October 6, 2023

Three-Component Cross-Electrophile Coupling: Regioselective Electrochemical Dialkylation of Alkenes

Lingxiang Lu, Yi Wang, Wendy Zhang, Wen Zhang, Kimberly A. See, and Song Lin*

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Abstract

The cross-electrophile dialkylation of alkenes enables the formation of two C(sp²)-C(sp³) bonds from readily available starting materials in a single transformation, thereby providing a modular and expedient approach to building structural complexity in organic synthesis. Herein, we exploit the disparate electronic and steric properties of alkyl halides with varying degrees of substitution to accomplish their selective activation and addition to alkenes under electrochemical conditions. This method enables regioselective dialkylation of alkenes without the use of a transition-metal catalyst and provides access to a diverse range of synthetically useful compounds.

Acknowledgments

Financial support was provided by NIGMS (R01GM130928; to S.L.) and NSF Center for Synthetic Organic Electrochemistry (CHE-2002158; to K.A.S.). S.L. is grateful to FMC Corporation for a New Investigator Award and the Camille and Henry Dreyfus Foundation for a Camille Dreyfus Teacher-Scholar Award. K.A.S. acknowledges support from the David and Lucile Packard Fellowship for Science. We thank K. R. Meihaus, L. F. T. Novais, J. I. Martinez Alvarado, and J. Rein for manuscript editing, I. Keresztes and D. Wood for assistance in mass spectrometry data collection and analysis, A. J. Ressler, S. J. Lee, and Z. Lu for assistance in substrate synthesis, and J. Liu for reproducing experiments.

References

This article references 31 other publications.

- (a) Corey, E. J.; Cheng, X.-M. *The Logic of Chemical Synthesis*; Wiley: New York, 1995.
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- (b) Choi, J.; Fu, G. C. Transition Metal-Catalyzed Alkyl-Alkyl Bond Formation: Another Dimension in Cross-Coupling Chemistry. *Science* **2017**, *356*, eaaf7230. DOI: 10.1126/science.aaf7230
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- (b) Luo, Y.-C.; Xu, C.; Zhang, X. Nickel-Catalyzed Dicarbofunctionalization of Alkenes. *Chin. J. Chem.* **2020**, *38*, 1371–1394. DOI: 10.1002/cjoc.202000224

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Abstract

$Z = \text{aryl, boryl, silyl, vinyl, etc.}; X = \text{Br, Cl, OTf}$
 $R^1 = 3'/2' \text{ alkyl}; R^2 = 1' \text{ alkyl, methyl, silyl, gemyl}$

Representative products:

Scheme 1

(A) Redox-neutral alkene dialkylation (refs 3–6)
 $R^1R^2 + R^3-X + R^4-X \xrightarrow[\text{M = Li, Mg, Zn, Sn}]{\text{conditions}} R^1R^2R^3R^4$
 $X = \text{halogen}$

(B) Ni-catalyzed directed reductive dialkylation (ref 9)
 $DO-R^1 + R^2-X \xrightarrow[\text{Mn reductant}]{\text{Ni catalyst}} DO-R^1R^2$
 $DO = \text{amide directing group}; R^1, R^2 = \text{alkyl}; X = \text{redox-active ester, halogen}; Y = \text{halogen}$

(C) Ni-catalyzed nondirected reductive dialkylation (ref 10)
 $Me_2C=CH-R^1 + R^2-X \xrightarrow[\text{Mn reductant}]{\text{Ni catalyst}} Me_2C(R^1)CH_2R^2$



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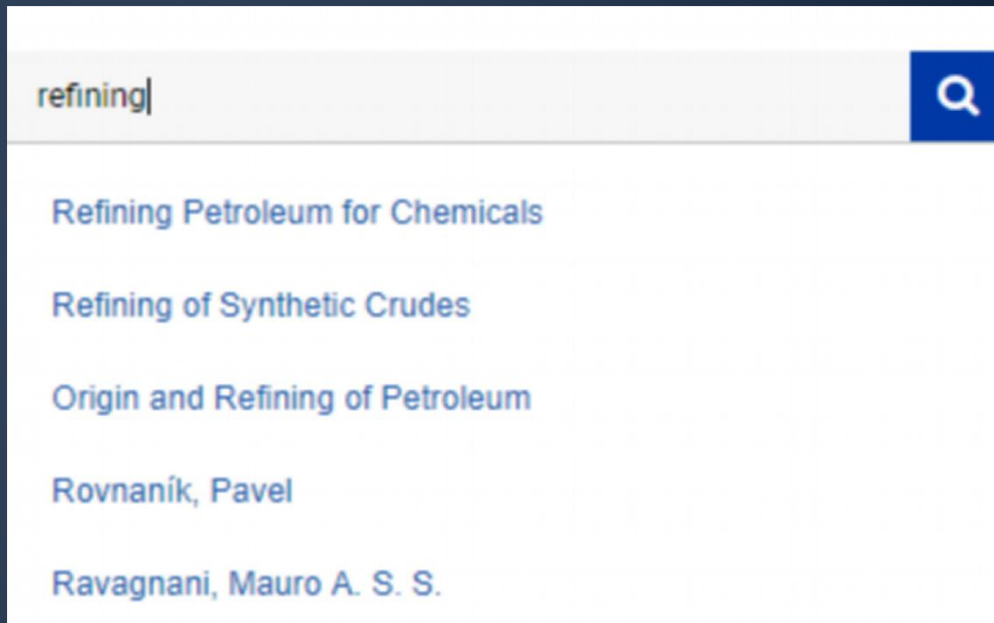
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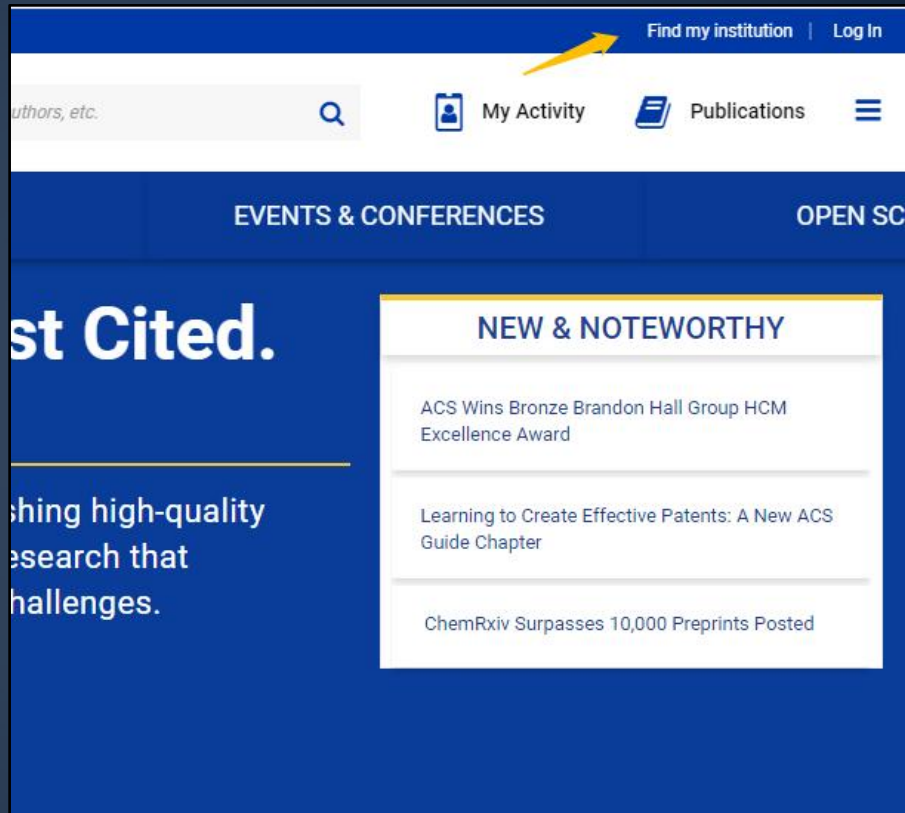


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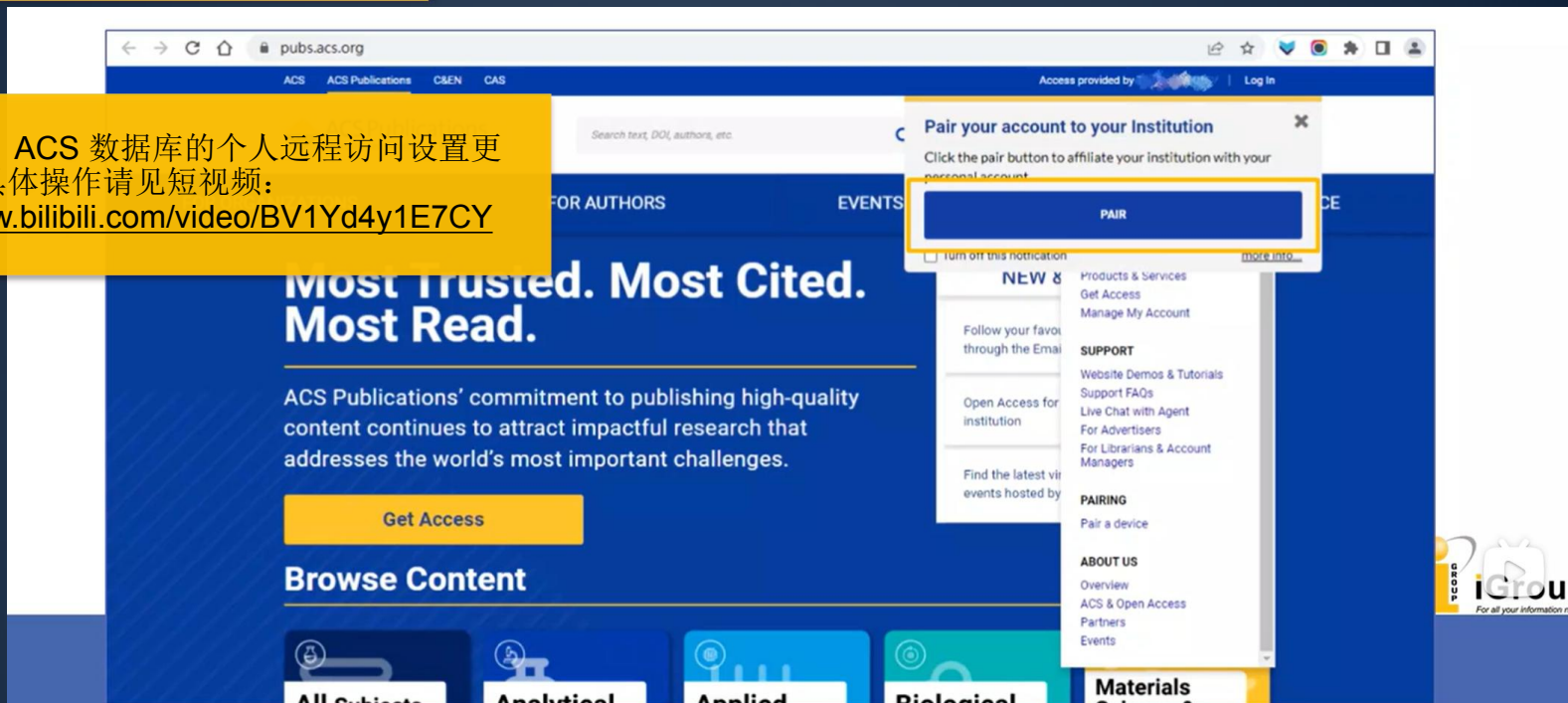


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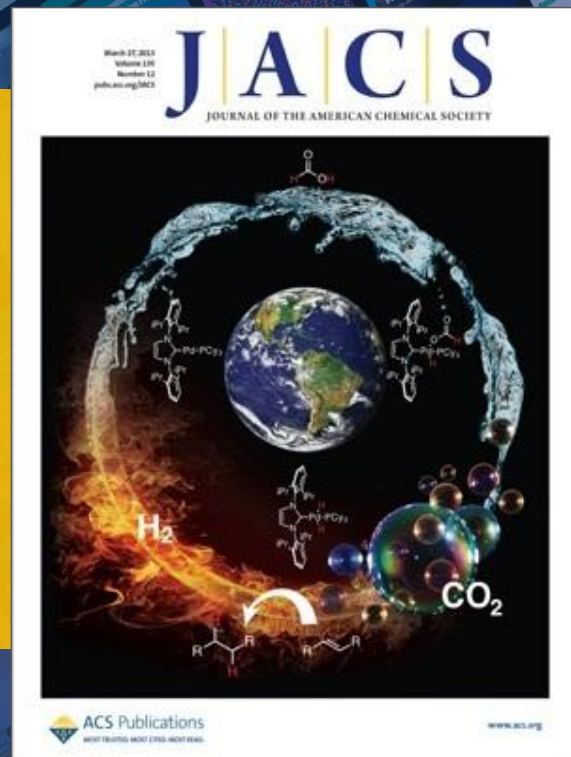
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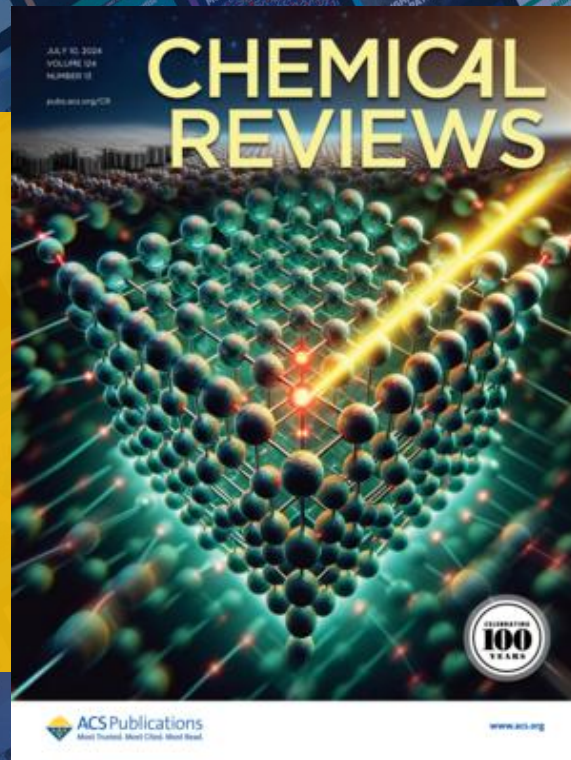
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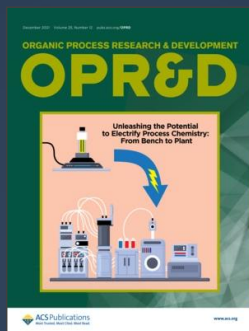
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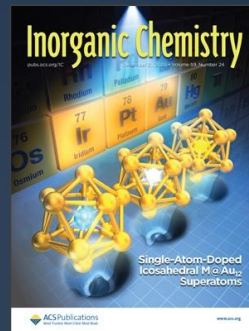
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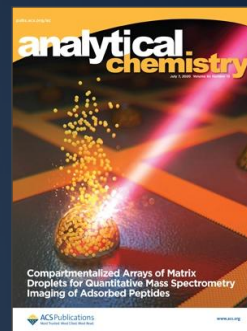
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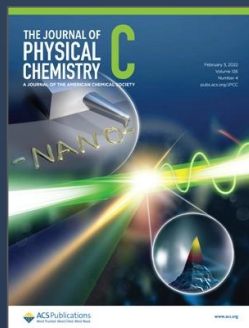
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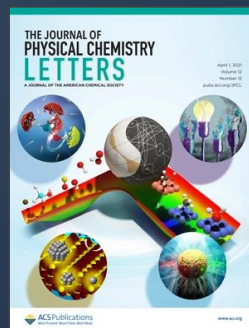
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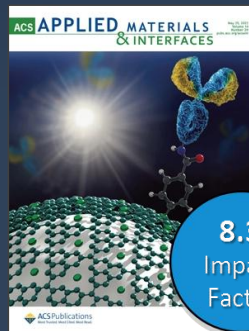
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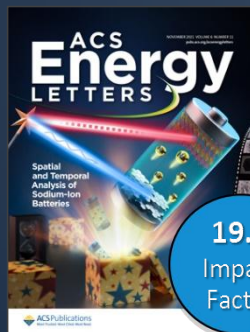
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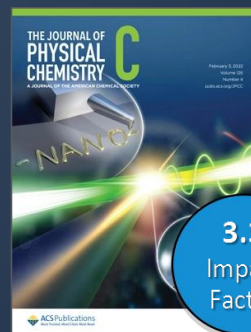
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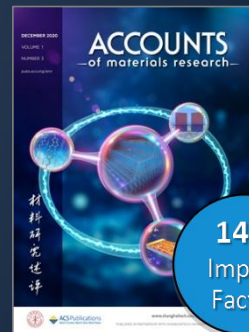
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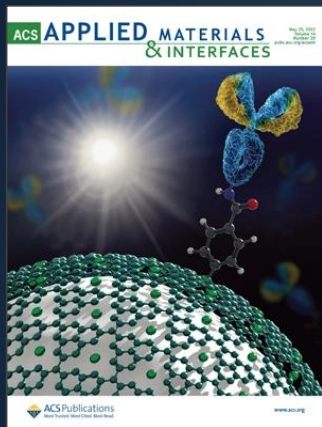
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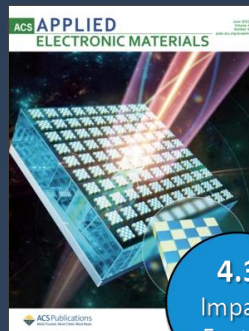


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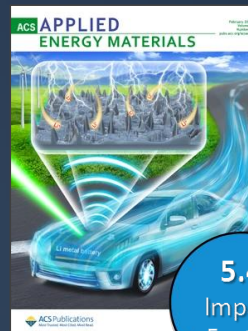
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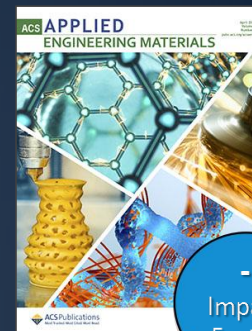
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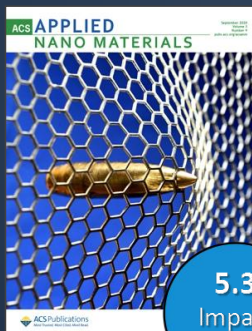
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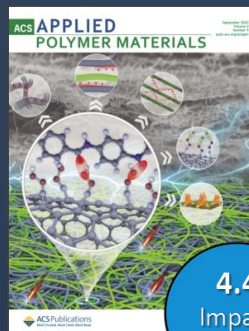
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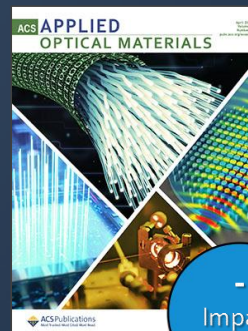
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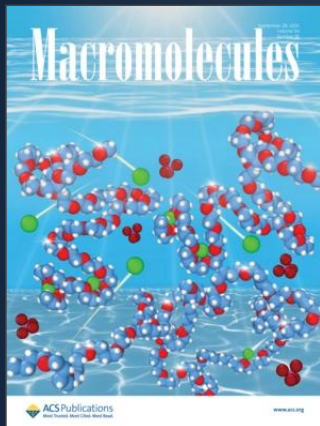
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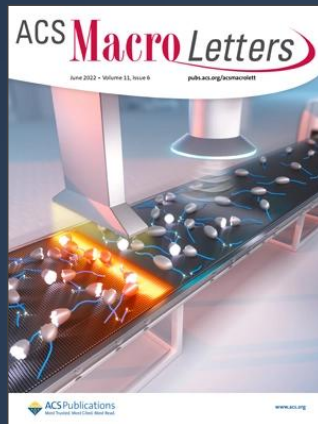
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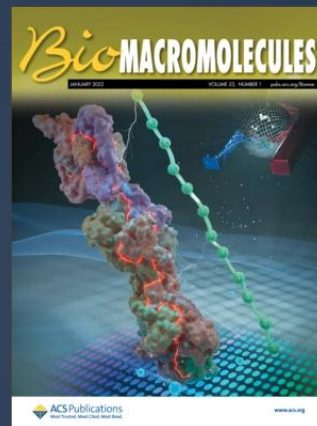
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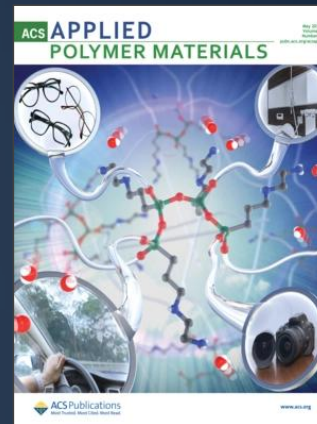
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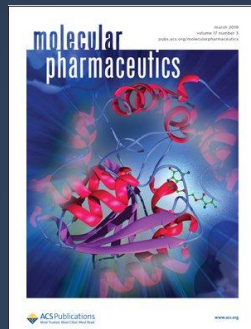
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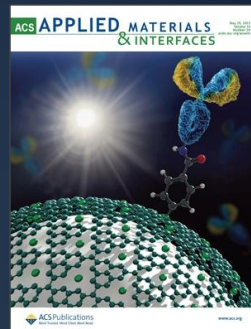
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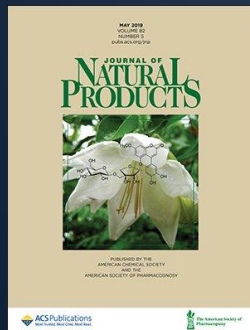
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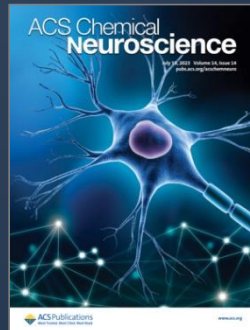
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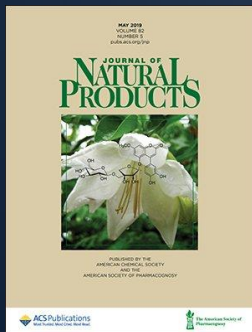
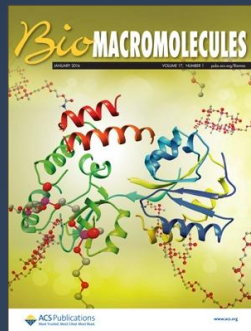
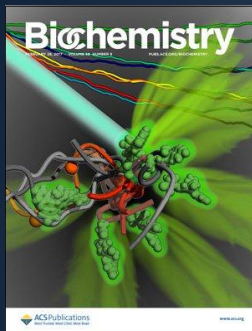


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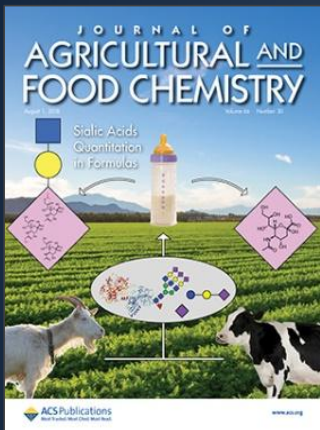
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*Journal of Natural
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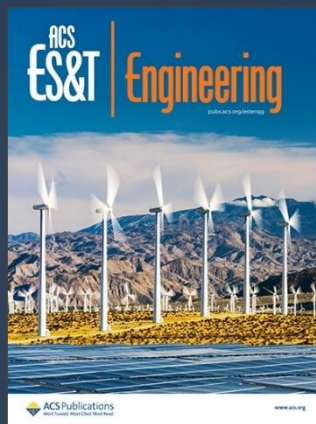
**Environmental
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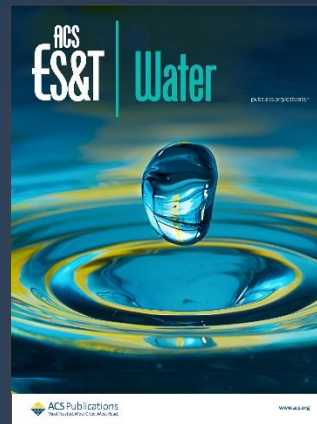
**Environmental
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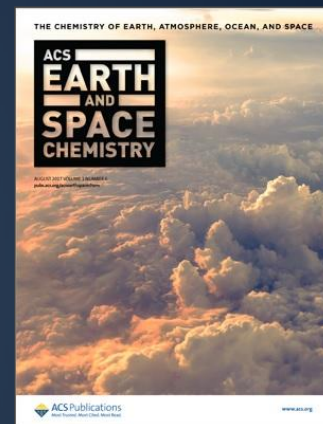
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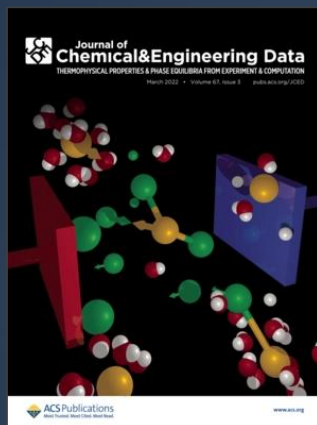
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Energy and Transportation 化工与能源



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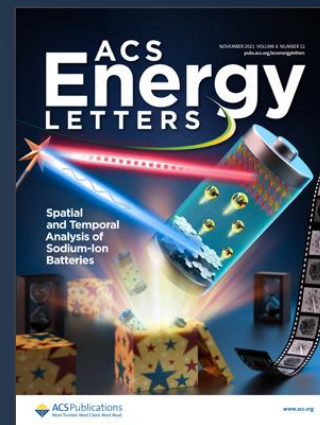
**Journal of Chemical
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Energy & Fuels

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ACS Energy Letters

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Open Access Journals

ACS 对全球开放科学呼声的回应

- 学术出版多年来一直在向开放获取迈进，ACS积极应对全球开放获取以及开放科学的呼声，2020年之前，ACS旗下有两本全OA的期刊。
- 2015年上线的ACS Central Science（不收取发表费用），目标是提升化学作为“核心科学”的关注度，自创刊以来不断发表与其他学科交叉领域杰出的研究成果。
- 2016年上线的ACS Omega，旨在快速发表经过同行评议的研究成果，加快新理念和有潜力的研究的传播，从而推动化学科学的前沿。
- 2021年起上线Au（金）系列期刊，提供覆盖有机、环境、物理等各个领域的全OA期刊。目前JACS Au和ACS Polymers Au已获得首个影响因子。



Open Access Journals

美国化学会旗下的开放获取期刊总共有 **18** 种，分别具有不同的内容和定位。

ACS Central Science, ACS Omega, JACS Au : 跨学科化学期刊

ACS Au Journals 期刊:

- ACS Bio & Med Chem Au
- ACS Engineering Au
- ACS Environmental Au
- ACS Materials Au
- ACS Measurement Science Au
- ACS Nanoscience Au
- ACS Organic & Inorganic Au
- ACS Physical Chemistry Au
- ACS Polymers Au



ACS 与中国的高校/科研机构的合作期刊:

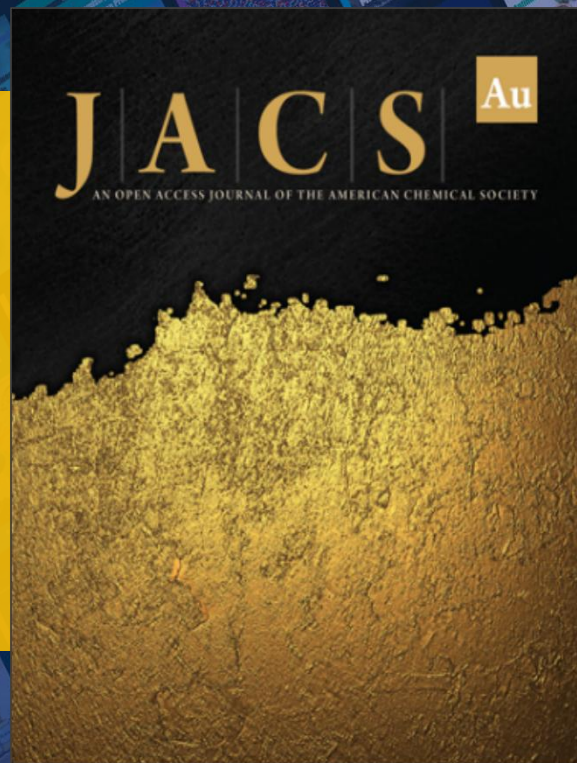
Precision Chemistry 2023 University of Science and Technology of China

Chemical & Biomedical Imaging 2023 Nanjing University

Environment & Health 2023 the Research Center for Eco-Environmental Sciences, CAS

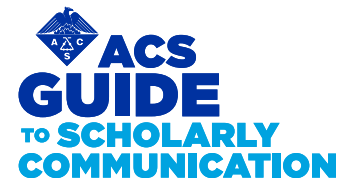
JACS Au——为各个学科推出Au系列OA期刊

- 2021年1月正式出版首期，每期收录10~20篇文章。
- 在涉及化学领域各分支学科的同时，更看重研究的即时影响力。
- 遵循JACS的传统，发表对全球化学群体都具有广泛影响和相关性的研究。
- JACS Au 拥有一支独立于其他期刊的编辑团队。
- 遵循 ACS AuthorChoice政策。



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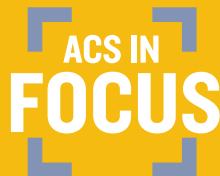
ACS Publications除了拥有丰富的期刊资源外，
还有其它类型的学术资源，例如电子图书、科
研写作指南、C&EN新闻杂志等



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 - Medicinal Chemical Reviews 系列 (2022 - 至今)
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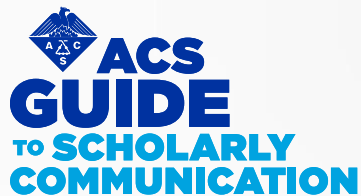
ACS IN FOCUS

<https://pubs.acs.org/series/infocus>

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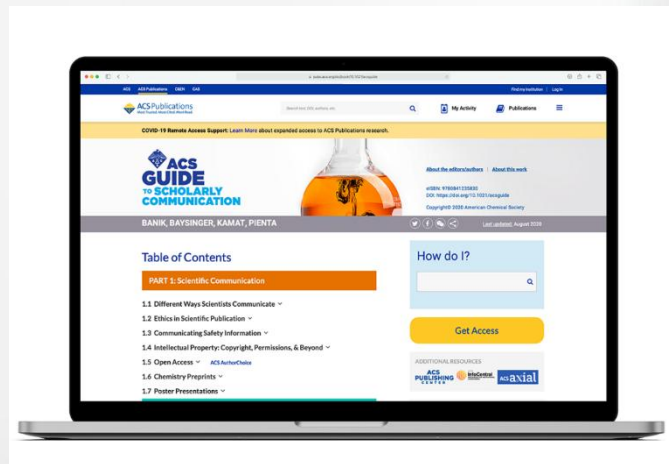


<https://doi.org/10.1021/acsguide>

■ ACS Guide to Scholarly Communication

学术交流指南是一本在线参考工具书，旨在为学生、研究人员、教育工作者和图书馆员提供掌握学术交流所需的指导与建议。

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<https://pubs.acs.org/doi/book/10.1021/acsreagents>

- **ACS Reagent Chemicals** 是一份权威的化学品试剂标准手册。
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- **C&EN Global Enterprise** 是美国化学学会旗下的知名杂志。
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