Oxidation

1-Progression in the Oxidation Stability of MXenes

By Soomro, RA (Soomro, Razium A. A.) [1] ; Zhang, P (Zhang, Peng) [1] ; Fan, BM (Fan, Baomin) [2] ; Wei, Y (Wei, Yi) [1] ; Xu, B (Xu, Bin) [1] (provided by Clarivate) Source NANO-MICRO LETTERS Volume 15 Issue 1 DOI 10.1007/s40820-023-01069-7 Article Number 108 Published DEC 2023 Indexed 2023-05-04 Document Type Article

Abstract

MXenes are under the spotlight due to their versatile physicochemical characteristics. Since their discovery in 2011, significant advancements have been achieved in their synthesis and application sectors. However, the spontaneous oxidation of MXenes, which is critical to its processing and product lifespan, has gotten less attention due to its chemical complexity and poorly understood oxidation mechanism. This perspective focuses on the oxidation stability of MXenes and addresses the most recent advancements in understanding and the possible countermeasures to limit the spontaneous oxidation of MXenes. A section is dedicated to the presently accessible methods for monitoring oxidation, with a discussion on the debatable oxidation mechanism and coherently operating factors that contribute to the complexity of MXenes oxidation. The current potential solutions for mitigating MXenes oxidation and the existing challenges are also discussed with prospects to prolong MXene's shelf-life storage and expand their application scope.

Keywords

Author Keywords
MXenesOxidation stabilityAntioxidation strategiesShelf-life of MXenes
Keywords Plus
TITANIUM CARBIDE MXENE2-DIMENSIONAL TI3C2THERMAL-STABILITYMAX PHASEELECTRODESTI2CTX

Oxidation

2-Coupling Photocatalytic Hydrogen Production with Key Oxidation Reactions

---

By Teng, JY (Teng, Jiayan) [1] ; Li, WL (Li, Wenlu) [2] , [3] ; Wei, Z (Wei, Zhen) [1] ; Hao, DR (Hao, Derek) [4] , [5] ; Jing, L (Jing, Lin) [1] ; Liu, YX (Liu, Yuxi) [1] ; Dai, HX (Dai, Hongxing) [1] ; Zhu, YF (Zhu, Yongfa) [2] ; Ma, TY (Ma, Tianyi) [4] , [5] ; Deng, JG (Deng, Jiguang) [1] (provided by Clarivate) **Source ANGEWANDTE CHEMIE-INTERNATIONAL EDITION Volume 63 Issue 50 DOI 10.1002/anie.202416039 Published DEC 9 2024 Early Access OCT 2024 Indexed 2024-11-09 Document Type Review**

Abstract

Hydrogen represents a clean and sustainable energy source with wide applications in fuel cells and hydrogen energy storage systems. Photocatalytic strategies emerge as a green and promising solution for hydrogen production, which still reveals several critical challenges in enhancing the efficiency and stability and improving the whole value. This review systematically elaborates on various coupling approaches for photocatalytic hydrogen production, aiming to improve both efficiency and value through different oxidation half-reactions. Firstly, the fundamental mechanism is discussed for photocatalytic hydrogen production. Then, the advances, challenges, and opportunities are expanded for the coupling of photocatalytic hydrogen production, which focuses on the integration of value-added reactions including O2 production, H2O2 production, biomass conversion, alcohol oxidation, and pollutants treatment. Finally, the challenges and outlook of photocatalytic H2 production technology are analyzed from the aspects of coupling hydrogen production value, photocatalyst design and reaction system construction. This work presents a holistic view of the field, emphasizing the synergistic benefits of coupled reactions and their practical application potential, rather than focusing on catalysts or single reaction systems. This review provides valuable references for the development and application of photocatalytic hydrogen production in energy conversion and environmental conservation through sustainable, eco-friendly and economic pathways.

Keywords

Author Keywords

PhotocatalysisHydrogen ProductionWater SplittingCoupling Reactions

Keywords Plus

GRAPHITIC CARBON NITRIDEQUANTUM EFFICIENCYENERGY-CONVERSIONWATERDRIVENWASTEBIOMASSLIGNINTRANSFORMATIONVALORIZATION

Oxidation

3-Polyoxometalated metal-organic framework superstructure for stable water oxidation

---

By Yue, KH (Yue, Kaihang) [1] , [2] , [3] ; Lu, RH (Lu, Ruihu) [4] ; Gao, MB (Gao, Mingbin) [5] ; Song, F (Song, Fei) [6] ; Dai, Y (Dai, Yao) [1] ; Xia, CF (Xia, Chenfeng) [2] ; Mei, BB (Mei, Bingbao) [6] ; Dong, HL (Dong, Hongliang) [7] ; Qi, RJ (Qi, Ruijuan) [8] ; Zhang, DL (Zhang, Daliang) [9] , [10] ;

(provided by Clarivate) Source SCIENCE Volume 388 Issue 6745 Page 430-436 DOI 10.1126/science.ads1466 Published APR 25 2025 Indexed 2025-07-05 Document Type Article

Abstract

Stable, nonprecious catalysts are vital for large-scale alkaline water electrolysis. Here, we report a grafted superstructure, MOF@POM, formed by self-assembling a metal-organic framework (MOF) with polyoxometalate (POM). In situ electrochemical transformation converts MOF into active metal (oxy)hydroxides to produce a catalyst with a low overpotential of 178 millivolts at 10 milliamperes per square centimeter in alkaline electrolyte. An anion exchange membrane water electrolyzer incorporating this catalyst achieves 3 amperes per square centimeter at 1.78 volts at 80 degrees C and stable operation at 2 amperes per square centimeter for 5140 hours at room temperature. In situ electrochemical spectroscopy and theoretical studies reveal that the synergistic interactions between metal atoms create a fast electron-transfer channel from catalytic iron and cobalt sites, nickel, and tungsten in the polyoxometalate to the electrode, stabilizing the metal sites and preventing dissolution.

Keywords

Keywords Plus

OXYGEN EVOLUTION REACTIONINITIO MOLECULAR-DYNAMICSACTIVITY TRENDSELECTROCATALYSTSEFFICIENTNICKELCOBALTSTABILITYREDUCTION(OXY)HYDROXIDE

Oxidation

4-Carbonaceous materials in structural dimensions for advanced oxidation processes

---

By Wang, YP (Wang, Yunpeng) [1] , [2] ; Liu, Y (Liu, Ya) [1] ; Zhang, HY (Zhang, Huayang) [1] ; Duan, XG (Duan, Xiaoguang) [1] ; Ma, J (Ma, Jun) [2] ; Sun, HQ (Sun, Hongqi) [3] ; Tian, WJ (Tian, Wenjie) [1] ; Wang, SB (Wang, Shaobin) [1] (provided by Clarivate) Source CHEMICAL SOCIETY REVIEWS Volume 54 Issue 5 Page 2436-2482 DOI 10.1039/d4cs00338a Published MAR 3 2025 Early Access FEB 2025 Indexed 2025-02-09 Document Type Review

Abstract

Carbonaceous materials have attracted extensive research and application interests in water treatment owing to their advantageous structural and physicochemical properties. Despite the significant interest and ongoing debates on the mechanisms through which carbonaceous materials facilitate advanced oxidation processes (AOPs), a systematic summary of carbon materials across all dimensions (0D-3D nanocarbon to bulk carbon) in various AOP systems remains absent. Addressing this gap, the current review presents a comprehensive analysis of various carbon/oxidant systems, exploring carbon quantum dots (0D), nanodiamonds (0D), carbon nanotubes (1D), graphene derivatives (2D), nanoporous carbon (3D), and biochar (bulk 3D), across different oxidant systems: persulfates (peroxymonosulfate/peroxydisulfate), ozone, hydrogen peroxide, and high-valent metals (Mn(vii)/Fe(vi)). Our discussion is anchored on the identification of active sites and elucidation of catalytic mechanisms, spanning both radical and nonradical pathways. By dissecting catalysis-related factors such as sp2/sp3 C, defects, and surface functional groups that include heteroatoms and oxygen groups in different carbon configurations, this review aims to provide a holistic understanding of the catalytic nature of different dimensional carbonaceous materials in AOPs. Furthermore, we address current challenges and underscore the potential for optimizing and innovating water treatment methodologies through the strategic application of carbon-based catalysts. Finally, prospects for future investigations and the associated bottlenecks are proposed.

Keywords

Keywords Plus

REDUCED GRAPHENE OXIDEPERSISTENT FREE-RADICALSNITROGEN-DOPED GRAPHENEMETAL-FREE CATALYSTNANOTUBES PERFORMANCE OPTIMIZATIONHYDROGEN-PEROXIDE ACTIVATIONHIERARCHICALLY POROUS CARBONACID ORANGE 7QUANTUM DOTSORGANIC POLLUTANTS

Oxidation

5-Electrochemical hydrogenation and oxidation of organic species involving water

---

By Liu, CB (Liu, Cuibo) [1] ; Chen, FP (Chen, Fanpeng) [1] ; Zhao, BH (Zhao, Bo-Hang) [1] ; Wu, YM (Wu, Yongmeng) [1] ; Zhang, B (Zhang, Bin) [1] , [2] (provided by Clarivate) Source NATURE REVIEWS CHEMISTRY Volume 8 Issue 4 Page 277-293 DOI 10.1038/s41570-024-00589-z Published APR 2024 Early Access MAR 2024 Indexed 2024-04-03 Document Type Review

Abstract

Fossil fuel-driven thermochemical hydrogenation and oxidation using high-pressure H2 and O2 are still popular but energy-intensive CO2-emitting processes. At present, developing renewable energy-powered electrochemical technologies, especially those using clean, safe and easy-to-handle reducing agents and oxidants for organic hydrogenation and oxidation reactions, is urgently needed. Water is an ideal carrier of hydrogen and oxygen. Electrochemistry provides a powerful route to drive water splitting under ambient conditions. Thus, electrochemical hydrogenation and oxidation transformations involving water as the hydrogen source and oxidant, respectively, have been developed to be mild and efficient tools to synthesize organic hydrogenated and oxidized products. In this Review, we highlight the advances in water-participating electrochemical hydrogenation and oxidation reactions of representative organic molecules. Typical electrode materials, performance metrics and key characterization techniques are firstly introduced. General electrocatalyst design principles and controlling the microenvironment for promoting hydrogenation and oxygenation reactions involving water are summarized. Furthermore, paired hydrogenation and oxidation reactions are briefly introduced before finally discussing the challenges and future opportunities of this research field.

The use of water for electrochemical hydrogenation and oxidation of organic species provides a sustainable route for synthesizing chemicals. The electrode types, general electrocatalyst selection principles and interface microenvironment control are elucidated, conducive to designing efficient electrocatalysts and reaction systems.

Keywords

Keywords Plus

ELECTROCATALYTIC HYDROGENATIONELECTRON-TRANSFERSELECTIVE HYDROGENATIONPAIRED ELECTROLYSISREDUCTIONELECTROSYNTHESISCONVERSIONOXYGENTRANSFORMATIONACETYLENE

دانشگاه علوم پزشکی و خدمات بهداشتی درمانی ایران
کتابخانه دانشکده بهداشت

Oxidation

6-Regulation of Oxide Pathway Mechanism for Sustainable Acidic Water Oxidation

---

By Cao, XJ (Cao, Xuejie) [1] ; Qin, HY (Qin, Hongye) [1] ; Zhang, JY (Zhang, Jinyang) [1] ; Chen, XJ (Chen, Xiaojie) [1] ; Jiao, LF (Jiao, Lifang) [1] (provided by Clarivate) Source JOURNAL OF THE AMERICAN CHEMICAL SOCIETY Volume 146 Issue 46 Page 32049-32058 DOI 10.1021/jacs.4c12942
Published NOV 12 2024 Early Access NOV 2024 Indexed 2024-11-20 Document Type Article

Abstract

The advancement of acid-stable oxygen evolution reaction (OER) electrocatalysts is crucial for efficient hydrogen production through proton exchange membrane (PEM) water electrolysis. Unfortunately, the activity of electrocatalysts is constrained by a linear scaling relationship in the adsorbed evolution mechanism, while the lattice-oxygen-mediated mechanism undermines stability. Here, we propose a heterogeneous dual-site oxide pathway mechanism (OPM) that avoids these limitations through direct dioxygen radical coupling. A combination of Lewis acid (Cr) and Ru to form solid solution oxides (Cr x Ru1-x O2) promotes OH adsorption and shortens the dual-site distance, which facilitates the formation of *O radical and promotes the coupling of dioxygen radical, thereby altering the OER mechanism to a Cr-Ru dual-site OPM. The Cr0.6Ru0.4O2 catalyst demonstrates a lower overpotential than that of RuO2 and maintains stable operation for over 350 h in a PEM water electrolyzer at 300 mA cm-2. This mechanism regulation strategy paves the way for an optimal catalytic pathway, essential for large-scale green hydrogen production.

Keywords

Keywords Plus

OXYGENSPECTROSCOPYRUTHENIUMCATALYSTSRU

Oxidation

7-Unlocking Efficient Hydrogen Production: Nucleophilic Oxidation Reactions Coupled with Water Splitting

---

By Wang, P (Wang, Peng) [1] ; Zheng, J (Zheng, Jie) [2] ; Xu, X (Xu, Xue) [1] ; Zhang, YQ (Zhang, Yu-Qing) [2] ; Shi, QF (Shi, Qiao-Fu) [2] ; Wan, Y (Wan, Yong) [1] ; Ramakrishna, S (Ramakrishna, Seeram) [3] ; Zhang, J (Zhang, Jun) [1] ; Zhu, LY (Zhu, Liyang) [4] ; Yokoshima, T (Yokoshima, Tokihiko) [4] ; (provided by Clarivate) **Source ADVANCED MATERIALS Volume 36 Issue 35 DOI 10.1002/adma.202404806 Published AUG 2024 Early Access JUL 2024 Indexed 2024-07-14 Document Type Review**

Abstract

Electrocatalytic water splitting driven by sustainable energy is a clean and promising water-chemical fuel conversion technology for the production of high-purity green hydrogen. However, the sluggish kinetics of anodic oxygen evolution reaction (OER) pose challenges for large-scale hydrogen production, limiting its efficiency and safety. Recently, the anodic OER has been replaced by a nucleophilic oxidation reaction (NOR) with biomass as the substrate and coupled with a hydrogen evolution reaction (HER), which has attracted great interest. Anode NOR offers faster kinetics, generates high-value products, and reduces energy consumption. By coupling NOR with hydrogen evolution reaction, hydrogen production efficiency can be enhanced while yielding high-value oxidation products or degrading pollutants. Therefore, NOR-coupled HER hydrogen production is another new green electrolytic hydrogen production strategy after electrolytic water hydrogen production, which is of great significance for realizing sustainable energy development and global decarbonization. This review explores the potential of nucleophilic oxidation reactions as an alternative to OER and delves into NOR mechanisms, guiding future research in NOR-coupled hydrogen production. It assesses different NOR-coupled production methods, analyzing reaction pathways and catalyst effects. Furthermore, it evaluates the role of electrolyzers in industrialized NOR-coupled hydrogen production and discusses future prospects and challenges. This comprehensive review aims to advance efficient and economical large-scale hydrogen production.

Nucleophilic oxidation (NOR) has great potential as an alternative to OER. NOR offers faster kinetics, generates high-value products, and reduces energy consumption. By coupling NOR with hydrogen evolution reaction, hydrogen production efficiency can be enhanced while yielding valuable by-products or degrading pollutants. Focusing on the reaction mechanisms and reaction paths of different NORs in anodic oxidation reactions. The review is intended to provide theoretical guidance for future research on NOR-coupled HER hydrogen production. image

Keywords

Author Keywords

coupled hydrogen productionelectrocatalysishigh-value productsnucleophilic oxidation reactions

Keywords Plus

ELECTROCATALYTIC 5-HYDROXYMETHYLFURFURAL OXIDATIONEXCHANGE MEMBRANEH-2 PRODUCTIONBIFUNCTIONAL ELECTROCATALYSTSFORMALDEHYDE OXIDATIONUREA ELECTROOXIDATIONNICKEL SITESPERFORMANCEEVOLUTIONPROTON

Oxidation

8-The mechanism of water oxidation using transition metal-based heterogeneous electrocatalysts

---

By Yang, SJ (Yang, Shujiao) [1] ; Liu, XH (Liu, Xiaohan) [1] ; Li, SS (Li, Sisi) [1] ; Yuan, WJ (Yuan, Wenjie) [1] ; Yang, LN (Yang, Luna) [1] ; Wang, T (Wang, Ting) [1] ; Zheng, HQ (Zheng, Haoquan) [1] ; Cao, R (Cao, Rui) [1] ; Zhang, W (Zhang, Wei) [1] (provided by Clarivate) Source CHEMICAL SOCIETY REVIEWS Volume 53 Issue 11 Page 5593-5625 DOI 10.1039/d3cs01031g Published JUN 4 2024 Early Access APR 2024 Indexed 2024-04-29 Document Type Review

Abstract

The water oxidation reaction, a crucial process for solar energy conversion, has garnered significant research attention. Achieving efficient energy conversion requires the development of cost-effective and durable water oxidation catalysts. To design effective catalysts, it is essential to have a fundamental understanding of the reaction mechanisms. This review presents a comprehensive overview of recent advancements in the understanding of the mechanisms of water oxidation using transition metal-based heterogeneous electrocatalysts, including Mn, Fe, Co, Ni, and Cu-based catalysts. It highlights the catalytic mechanisms of different transition metals and emphasizes the importance of monitoring of key intermediates to explore the reaction pathway. In addition, advanced techniques for physical characterization of water oxidation intermediates are also introduced, for the purpose of providing information for establishing reliable methodologies in water oxidation research. The study of transition metal-based water oxidation electrocatalysts is instrumental in providing novel insights into understanding both natural and artificial energy conversion processes.

The oxygen evolution reaction (OER) mechanisms using transition metal-based electrocatalysts are instrumental in providing novel insights into both natural and artificial energy conversion processes.

Keywords

Keywords Plus

O BOND-FORMATIONOXYGEN-EVOLUTION REACTIONX-RAY-ABSORPTIONOXIDE THIN-FILMFE-57 SYNCHROTRON MOSSBAUERSINGLE-ATOM CATALYSTSAMBIENT-PRESSURE XPSIN-SITU FORMATIONPHOTOSYSTEM-IIEVOLVING COMPLEX

Oxidation

9-Hexafluorophosphate additive enables durable seawater oxidation at ampere-level current density

---

By He, X (He, Xun) [1] , [2] , [3] ; Yao, YC (Yao, Yongchao) [1] , [4] ; Zhang, LM (Zhang, Limei) [2] , [4] ; Wang, HF (Wang, Hefeng) [3] ; Tang, H (Tang, Hong) [2] ; Jiang, WL (Jiang, Wenlong) [2] ; Ren, YC (Ren, Yuchun) [2] ; Nan, J (Nan, Jue) [2] ; Luo, YS (Luo, Yongsong) [1] ; Wu, TW (Wu, Tongwei) [2] ;

(provided by Clarivate) Source NATURE COMMUNICATIONS Volume 16 Issue 1 DOI 10.1038/s41467-025-60413-0 Article Number 4998 Published MAY 29 2025 Indexed 2025-06-19 Document Type Article

Abstract

Direct seawater electrolysis at ampere-level current densities, powered by coastal/offshore renewables, is an attractive avenue for sustainable hydrogen production but is undermined by chloride-induced anode degradation. Here we demonstrate the use of hexafluorophosphate (PF6-) as an electrolyte additive to overcome this limitation, achieving prolonged operation for over 5,000 hours at 1 A cm-2 and 2300 hours at 2 A cm-2 using NiFe layered double hydroxide (LDH) as anode. Together with the experimental findings, PF6- can intercalate into LDH interlayers and adsorb onto the electrode surface under an applied electric field, blocking Cl- and stabilizing Fe to prevent segregation. The constant-potential molecular dynamics simulations further reveal the accumulation of high surface concentrations of PF6- on the electrode surface that can effectively exclude Cl-, mitigating corrosion. Our work showcases synchronous interlayer and surface engineering by single non-oxygen anion species to enable Cl- rejection and marks a crucial step forward in seawater electrolysis.

Keywords
Keywords Plus
ELECTROLYSISGREEN

Oxidation

10-Anode-Electrolyte Interfacial Acidity Regulation Enhances Electrocatalytic Performances of Alcohol Oxidations

---

By Huang, BJ (Huang, Bingji) [1] ; Yan, JB (Yan, Jiabiao) [1] ; Li, ZH (Li, Zhenhua) [3] ; Chen, LS (Chen, Lisong) [1] , [4] ; Shi, JL (Shi, Jianlin) [2] (provided by Clarivate) Source ANGEWANDTE CHEMIE-INTERNATIONAL EDITION Volume 63 Issue 40 DOI 10.1002/anie.202409419 Published OCT 1 2024 Early Access SEP 2024 Indexed 2024-09-06 Document Type Article

Abstract

The local acidity at the anode surface during electrolysis is apparently stronger than that in bulk electrolyte due to the deprotonation from the reactant, which leads to the deteriorated electrocatalytic performances and product distributions. Here, an anode-electrolyte interfacial acidity regulation strategy has been proposed to inhibit local acidification at the surface of anode and enhance the electrocatalytic activity and selectivity of anodic reactions. As a proof of the concept, CeO2-x Lewis acid component has been employed as a supporter to load Au nanoparticles to accelerate the diffusion and enrichment of OH- toward the anode surface, so as to accelerate the electrocatalytic alcohol oxidation reaction. As the result, Au/CeO2-x exhibits much enhanced lactic acid selectivity of 81 % and electrochemical activity of 693 mA & sdot;cm-2 current density in glycerol oxidation reaction compared to pure Au. Mechanism investigation reveals that the introduced Lewis acid promotes the mass transport and concentration of OH- on the anode surface, thus promoting the generation of lactic acid through the simultaneous enhancements of Faradaic and non-Faradaic processes. Attractively, the proposed strategy can be used for the electro-oxidation performance enhancements of a variety of alcohols, which thereby provides a new perspective for efficient alcohol electro-oxidations and the corresponding electrocatalyst design.

CeO2-x Lewis acid component has been employed as a supporter to couple with active metal Au nanoparticles to accelerate the diffusion and enrichment of OH- toward the anode surface, which ensures the much enhanced electrochemical activity (current density of 693 mA & sdot;cm-2) and selectivity (81 %) for lactic acid production, so as to accelerate the electrocatalytic oxidation of other alcohols. image

Keywords

Author Keywords

MicroenvironmentLocal acidityAlcohol oxidation reactionsLewis acid oxide

Keywords Plus

QUANTIFICATION

Oxidation

11-Materials design of silicon based ceramic coatings for high temperature oxidation protection

---

By Liu, B (Liu, Bing) [1] ; Sun, J (Sun, Jia) [1] ; Guo, LX (Guo, Lingxiang) [1] ; Shi, HL (Shi, Huilun) [1] ; Feng, GH (Feng, Guanghui) [1] , [2] ; Feldmann, L (Feldmann, Laura) [4] ; Yin, XM (Yin, Xuemin) [1] , [3] ; Riedel, R (Riedel, Ralf) [4] ; Fu, QG (Fu, Qiangang) [1] ; Li, HJ (Li, Hejun) [1] (provided by Clarivate) Source MATERIALS SCIENCE & ENGINEERING R-REPORTS Volume 163 DOI 10.1016/j.mser.2025.100936 Article Number 100936 Published APR 2025 Early Access JAN 2025 Indexed 2025-02-09 Document Type Review

Abstract

To meet the rigorous thermal protection demands of state-of-the-art aircraft and aerospace systems, it is essential to implement comprehensive thermal oxidation shielding for the components exposed to extreme high temperature. Silicon based ceramic coating technology is an efficient way to improve the oxidation resistance of thermal structural materials, such as ceramic matrix composites (CMCs) and carbon/carbon (C/C) composites. Unfortunately, coating cracking under thermomechanical loads results in poor oxidation protection of the composite substrates. Two important perspectives to avoid the coating cracking are reviewed, namely i) crack healing and ii) toughening of the coating. Such two strategies can heal and inhibit the cracking of silicon based ceramic coatings to improve the service life-time. The strategies for the material design are appropriate for the fabrication of advanced silicon based ceramic coatings intended for the upcoming generation thermal structural components. And the outlook development of silicon-based ceramic coating systems is also summarized.

Keywords

Author Keywords
Material designSilicon-based ceramic coatingsOxidationSelf-healing strengtheningToughening
Keywords Plus
THERMAL-SHOCK RESISTANCECOATED C/C COMPOSITESSITU REACTION METHOD1700 DEGREES-CCARBON/CARBON COMPOSITESABLATION RESISTANCEMECHANICAL-PROPERTIESMULLITE WHISKERHFC NANOWIRESSIC COATINGS

Oxidation

12-Plasmonic Near-Infrared-Response S-Scheme ZnO/CuInS$_2$ Photocatalyst for H$_2$O$_2$ Production Coupled with Glycerin Oxidation

---

By Meng, K (Meng, Kai) [1] ; Zhang, JJ (Zhang, Jianjun) [2] ; Cheng, B (Cheng, Bei) [1] ; Ren, XA (Ren, Xingang) [3] ; Xia, ZS (Xia, Zhaosheng) [3] ; Xu, FY (Xu, Feiyan) [2] ; Zhang, LY (Zhang, Liuyang) [2] ; Yu, JG (Yu, Jiaguo) [1] , [2] (provided by Clarivate) **Source ADVANCED MATERIALS Volume 36 Issue 32**
DOI 10.1002/adma.202406460 Published AUG 2024 Early Access JUN 2024 Indexed 2024-06-17
Document Type Article

Abstract

Solar fuel synthesis is intriguing because solar energy is abundant and this method compensates for its intermittency. However, most photocatalysts can only absorb UV-to-visible light, while near-infrared (NIR) light remains unexploited. Surprisingly, the charge transfer between ZnO and CuInS2 quantum dots (QDs) can transform a NIR-inactive ZnO into a NIR-active composite. This strong response is attributed to the increased concentration of free charge carriers in the p-type semiconductor at the interface after the charge migration between ZnO and CuInS2, enhancing the localized surface plasmon resonance (LSPR) effect and the NIR response of CuInS2. As a paradigm, this ZnO/CuInS2 heterojunction is used for H2O2 production coupled with glycerin oxidation and demonstrates supreme performance, corroborating the importance of NIR response and efficient charge transfer. Mechanistic studies through contact potential difference (CPD), Hall effect test, and finite element method (FEM) calculation allow for the direct correlation between the NIR response and charge transfer. This approach bypasses the general light response issues, thereby stepping forward to the ambitious goal of harnessing the entire solar spectrum.

Keywords
Author Keywords
glycerin oxidation reactionlocalized surface plasmon resonancesnear-infrared responsephotocatalytic H2O2 production
Keywords Plus
RESONANCESADSORPTIONCO2

Oxidation

13-Key role of paracrystalline motifs on iridium oxide surfaces for acidic water oxidation

---

By Lu, BZ (Lu, Bingzhang) [1] ; Wahl, C (Wahl, Carolin) [2] , [3] ; dos Reis, R (dos Reis, Roberto) [2] , [3] ; Edgington, J (Edgington, Jane) [1] ; Lu, XK (Lu, Xiao Kun) [1] ; Li, RH (Li, Ruihan) [1] ; Sweers, ME (Sweers, Matthew E.) [2] ; Ruggiero, B (Ruggiero, Brianna) [1] ; Gunasooriya, GTKK (Gunasooriya, G. T. Kasun Kalhara) [4] ; Dravid, V (Dravid, Vinayak) [2] , [3] ; (provided by Clarivate) Source NATURE CATALYSIS Volume 7 Issue 8 Page 868-877 DOI 10.1038/s41929-024-01187-4 Published AUG 2024 Early Access JUL 2024 Indexed 2024-07-16 Document Type Article

Abstract

Water electrolysis using proton exchange membrane technology offers an ideal process for green hydrogen production, but widespread deployment is inhibited by insufficient catalyst activity, stability and affordability. Iridium-based oxides provide the best overall performance for acidic water oxidation, the limiting reaction for this process, but further improvements are impeded by poor understanding of the restructured active catalyst surface that forms under reaction conditions. Here we present a combination of X-ray and electron scattering data that reveals direct evidence for three paracrystalline structural motifs at the restructured surfaces of highly active catalysts (including rutile IrO_2 and perovskite $SrIrO_3$) that have previously been described as amorphous. These insights enable the design of a paracrystalline IrO_x catalyst that is independent of the bulk crystalline support and maintains higher activity, longer stability and more effective use of iridium to promote the production of green hydrogen.

Iridium-based oxides are the most active catalysts for acidic water oxidation, but a complete understanding of their surface reconstruction under operation remains elusive. Now three key paracrystalline structural motifs are identified on the seemingly amorphous reconstructed IrO_x surface.

Keywords

Keywords Plus

OXYGEN EVOLUTION ELECTROCATALYSISTOTAL-ENERGY CALCULATIONSDISSOLUTIONFEATURES

Oxidation

14-Innovations in electrocatalysts, hybrid anodic oxidation, and electrolyzers for enhanced direct seawater electrolysis

---

By Liu, D (Liu, Dong) [1] ; Cai, YH (Cai, Yaohai) [1] ; Wang, X (Wang, Xin) [1] ; Zhuo, YL (Zhuo, Yuling) [1] ; Sui, XL (Sui, Xulei) [1] ; Pan, H (Pan, Hui) [2] ; Wang, ZB (Wang, Zhenbo) [1] , [3] (provided by Clarivate) Source ENERGY & ENVIRONMENTAL SCIENCE Volume 17 Issue 19 Page 6897-6942 DOI 10.1039/d4ee01693a Published OCT 1 2024 Early Access AUG 2024 Indexed 2024-09-10 Document Type Review

Abstract

Seawater electrolysis has emerged as a viable solution for sustainable hydrogen production, leveraging areas abundant in renewable energy but deficient in freshwater. However, the complex composition of seawater presents substantial challenges, such as electrode corrosion and electrolyzer degradation, casting doubt on the viability of direct seawater electrolysis. A thorough review that summarizes the electrochemical challenges, innovative materials, and recent technological advancements in seawater electrolysis is essential yet remains absent. This review provides an in-depth examination of the current state of seawater electrolysis. It highlights the importance of innovative electrocatalysts, cutting-edge electrolyzer technologies, and the integration of hybrid anodic oxidation reactions in enhancing the efficiency of direct seawater electrolysis. Moreover, future directions to enhance the techno-economic viability of this promising field are outlined, aiming to pave the way for the efficient, cost-effective, and sustainable commercialization of direct seawater electrolysis technologies.

This review focuses on the latest developments in direct seawater electrolysis, specifically in electrocatalysts, hybrid anodic oxidation, and electrolyzers, providing a glimpse into the future of environmentally friendly hydrogen generation.

Keywords

Keywords Plus

EFFICIENCY HYDROGEN EVOLUTIONSELECTIVE OXYGEN EVOLUTIONHIGHLY EFFICIENTROBUST ELECTROCATALYSTSBIFUNCTIONAL ELECTROCATALYSTSELECTRONIC-STRUCTURECURRENT-DENSITYH-2 PRODUCTIONPH RANGEALKALINE

Oxidation

15-Activating lattice oxygen in high-entropy LDH for robust and durable water oxidation

---

By Wang, FQ (Wang, Fangqing) [1] , [2] ; Zou, PC (Zou, Peichao) [3] ; Zhang, YY (Zhang, Yangyang) [2] ; Pan, WL (Pan, Wenli) [4] ; Li, Y (Li, Ying) [1] , [2] ; Liang, LM (Liang, Limin) [2] ; Chen, C (Chen, Cong) [2] ; Liu, H (Liu, Hui) [1] , [2] ; Zheng, SJ (Zheng, Shijian) [1] , [2] (provided by Clarivate)

Source NATURE COMMUNICATIONS Volume 14 Issue 1 DOI 10.1038/s41467-023-41706-8 Article Number 6019 Published SEP 27 2023 Indexed 2023-11-27 Document Type Article

Abstract

The oxygen evolution reaction is known to be a kinetic bottleneck for water splitting. Triggering the lattice oxygen oxidation mechanism (LOM) can break the theoretical limit of the conventional adsorbate evolution mechanism and enhance the oxygen evolution reaction kinetics, yet the unsatisfied stability remains a grand challenge. Here, we report a high-entropy MnFeCoNiCu layered double hydroxide decorated with Au single atoms and O vacancies (Au-SA-MnFeCoNiCu LDH), which not only displays a low overpotential of 213 mV at 10 mA cm(-2) and high mass activity of 732.925 A g(-1) at 250 mV overpotential in 1.0 M KOH, but also delivers good stability with 700 h of continuous operation at similar to 100 mA cm(-2). Combining the advanced spectroscopic techniques and density functional theory calculations, it is demonstrated that the synergistic interaction between the incorporated Au single atoms and O vacancies leads to an upshift in the O 2p band and weakens the metal-O bond, thus triggering the LOM, reducing the energy barrier, and boosting the intrinsic activity.

Keywords

Keywords Plus

REDOXCATALYSISPERFORMANCECHALLENGES

Oxidation

16-Locking the lattice oxygen in RuO_2 to stabilize highly active Ru sites in acidic water oxidation

---

By Ping, XY (Ping, Xinyu) [1] ; Liu, YD (Liu, Yongduo) [1] ; Zheng, LX (Zheng, Lixia) [1] ; Song, Y (Song, Yang) [1] ; Guo, L (Guo, Lin) [2] ; Chen, SG (Chen, Siguo) [1] ; Wei, ZD (Wei, Zidong) [1]

(provided by Clarivate) Source NATURE COMMUNICATIONS Volume 15 Issue 1 DOI 10.1038/s41467-024-46815-6 Article Number 2501 Published MAR 20 2024 Indexed 2024-04-10 Document Type Article

Abstract

Ruthenium dioxide is presently the most active catalyst for the oxygen evolution reaction (OER) in acidic media but suffers from severe Ru dissolution resulting from the high covalency of Ru-O bonds triggering lattice oxygen oxidation. Here, we report an interstitial silicon-doping strategy to stabilize the highly active Ru sites of RuO2 while suppressing lattice oxygen oxidation. The representative Si-RuO2-0.1 catalyst exhibits high activity and stability in acid with a negligible degradation rate of similar to 52 mu V h(-1) in an 800 h test and an overpotential of 226 mV at 10 mA cm(-2). Differential electrochemical mass spectrometry (DEMS) results demonstrate that the lattice oxygen oxidation pathway of the Si-RuO2-0.1 was suppressed by similar to 95% compared to that of commercial RuO2, which is highly responsible for the extraordinary stability. This work supplied a unique mentality to guide future developments on Ru-based oxide catalysts' stability in an acidic environment.

Keywords
Keywords Plus
EVOLUTION REACTIONCATALYSTPERFORMANCECHALLENGESSILICAREDOXIR

Oxidation

17-Fe-S dually modulated adsorbate evolution and lattice oxygen compatible mechanism for water oxidation

---

By Luo, X (Luo, Xu) [1] ; Zhao, HY (Zhao, Hongyu) [1] ; Tan, X (Tan, Xin) [2] ; Lin, S (Lin, Sheng) [1] ; Yu, KS (Yu, Kesong) [1] ; Mu, XQ (Mu, Xueqin) [1] ; Tao, ZH (Tao, Zhenhua) [1] ; Ji, PX (Ji, Pengxia) [1] ; Mu, SC (Mu, Shichun) [1] (provided by Clarivate) Source NATURE COMMUNICATIONS Volume 15 Issue 1 DOI 10.1038/s41467-024-52682-y Article Number 8293 Published SEP 27 2024 Indexed 2024-12-27 Document Type Article

Abstract

Simultaneously activating metal and lattice oxygen sites to construct a compatible multi-mechanism catalysis is expected for the oxygen evolution reaction (OER) by providing highly available active sites and mediate catalytic activity/stability, but significant challenges remain. Herein, Fe and S dually modulated NiFe oxyhydroxide (R-NiFeOOH@SO4) is conceived by complete reconstruction of NiMoO4 center dot xH(2)O@Fe,S during OER, and achieves compatible adsorbate evolution mechanism and lattice oxygen oxidation mechanism with simultaneously optimized metal/oxygen sites, as substantiated by in situ spectroscopy/mass spectrometry and chemical probe. Further theoretical analyses reveal that Fe promotes the OER kinetics under adsorbate evolution mechanism, while S excites the lattice oxygen activity under lattice oxygen oxidation mechanism, featuring upshifted O 2p band centers, enlarged d-d Coulomb interaction, weakened metal-oxygen bond and optimized intermediate adsorption free energy. Benefiting from the compatible multi-mechanism, R-NiFeOOH@SO4 only requires overpotentials of 251 +/- 5/291 +/- 1 mV to drive current densities of 100/500 mA cm(-2) in alkaline media, with robust stability for over 300 h. This work provides insights in understanding the OER mechanism to better design high-performance OER catalysts.

Keywords
Keywords Plus
REDOX ACTIVITYGENERATIONCHALLENGESCOHP

Oxidation

18-Regulating socketed geometry of nanoparticles on perovskite oxide supports for enhanced stability in oxidation reactions

---

By Yu, JH (Yu, Jihang) [1] ; Yang, XW (Yang, Xinwei) [1] ; Jia, YY (Jia, Yanyan) [2] ; Wang, ZQ (Wang, Zhi-Qiang) [1] ; Li, WB (Li, Wenbo) [2] ; Jiang, YJ (Jiang, Yongjun) [2] ; Dai, S (Dai, Sheng) [2] ; Zhan, WC (Zhan, Wangcheng) [1] (provided by Clarivate) Source NATURE COMMUNICATIONS Volume 15 Issue 1 DOI 10.1038/s41467-024-54546-x Article Number 10229 Published NOV 26 2024 Indexed 2024-12-01 Document Type Article

Abstract

Heterogeneous catalysts with highly dispersed active particles on supports often face stability challenges during high-temperature industrial applications. The ex-solution strategy, which involves in situ extrusion of metals to form socketed particles, shows potential for addressing this stability issue. However, a deeper understanding of the relationship between the socketed geometry of these partially embedded nanoparticles and their catalytic performance is still lacking. Here, in situ transmission electron microscopy and theoretical calculations are utilized to investigate the oxygen-induced ex-solution process of Pd-doped LaAlO3 with varying concentrations of La vacancies (LaxAl0.9Pd0.1O3-delta). We find that the socketed geometry of Pd-based particles can be tuned by manipulating the levels of La deficiencies in the oxide support, which in turn influences the catalytic performance in high-temperature oxidation reactions. As for the socketed particles, the balance between particle size and outcrop height is crucial for determining the oxidation activity and sinter-resistance behavior. Consequently, the optimized catalyst, La0.8Al0.9Pd0.1O3-delta, exhibits superior catalytic performances, particularly high stability (still working after aging at 1000 degrees C for 50 h) and water resistance in various combustion reactions (e.g., CH4 oxidation and C3H8 oxidation).

Nanoparticles with precisely controlled socketed geometries, created via an ex-solution method, present promising ways to improve the stability of heterogeneous catalysts. Here, the authors demonstrate that adjusting the level of La deficiencies in the oxide support allows tuning of the geometry of Pd-based particles, which in turn influences their catalytic performance in high-temperature oxidation reactions.

Keywords

Keywords Plus

METHANE OXIDATIONPDCATALYSTSURFACEGROWTH

Oxidation

19-Were Persulfate-Based Advanced Oxidation Processes Really Understood? Basic Concepts, Cognitive Biases, and Experimental Details

---

By Hu, XN (Hu, Xiaonan) [1] , [2] ; Zhu, MS (Zhu, Mingshan) [1] (provided by Clarivate) Source ENVIRONMENTAL SCIENCE & TECHNOLOGY Volume 58 Issue 24 Page 10415-10444 DOI 10.1021/acs.est.3c10898 Published JUN 7 2024 Early Access JUN 2024 Indexed 2024-06-16
Document Type Review

Abstract

Persulfate (PS)-based advanced oxidation processes (AOPs) for pollutant removal have attracted extensive interest, but some controversies about the identification of reactive species were usually observed. This critical review aims to comprehensively introduce basic concepts and rectify cognitive biases and appeals to pay more attention to experimental details in PS-AOPs, so as to accurately explore reaction mechanisms. The review scientifically summarizes the character, generation, and identification of different reactive species. It then highlights the complexities about the analysis of electron paramagnetic resonance, the uncertainties about the use of probes and scavengers, and the necessities about the determination of scavenger concentration. The importance of the choice of buffer solution, operating mode, terminator, and filter membrane is also emphasized. Finally, we discuss current challenges and future perspectives to alleviate the misinterpretations toward reactive species and reaction mechanisms in PS-AOPs.

Keywords
Author Keywords
persulfateadvanced oxidationreactive specieselectron paramagnetic resonancescavengers
Keywords Plus
P-HYDROXYBENZOIC ACIDSELECTIVE CIRCUMNEUTRAL DEGRADATION5,5-DIMETHYL-1-PYRROLINE N-OXIDEELECTRON-SPIN-RESONANCEEFFICIENT PEROXYDISULFATE ACTIVATIONKETONE-CATALYZED DECOMPOSITIONRADICAL-INDUCED OXIDATIONZERO-VALENT IRONSINGLET OXYGENHYDROXYL RADICALS

Oxidation

20-Constructing nanoneedle arrays of heterostructured RuO_2-Co_3O_4 with tip-effect-induced enrichment of reactants for enhanced water oxidation

---

By Zhang, X (Zhang, Xu) [1] ; Song, JN (Song, Junnan) [1] ; Sun, TM (Sun, Tongming) [2] ; Wang, MM (Wang, Minmin) [2] ; Zhu, JL (Zhu, Jinli) [2] ; Yu, Y (Yu, Yang) [3] ; Wang, JC (Wang, Jiacheng) [1] (provided by Clarivate) Source CHEMICAL COMMUNICATIONS Volume 61 Issue 48 Page 8723-8726
DOI 10.1039/d5cc01933h Published JUN 10 2025 Early Access MAY 2025 Indexed 2025-05-22
Document Type Article

Abstract

Nanoneedle arrays of heterostructured RuO2-Co3O4 electrocatalysts were constructed, showing improved water oxidation activity and durable stability. The synergy of tip-effect-induced OH- enrichment, superior hydrophilicity, and heterojunction-enhanced electron transfer promotes water oxidation activity.

Oxidation

21-Stabilizing NiFe sites by high-dispersity of nanosized and anionic Cr species toward durable seawater oxidation

---

By Cai, ZW (Cai, Zhengwei) [1] ; Liang, J (Liang, Jie) [2] ; Li, ZX (Li, Zixiao) [2] ; Yan, TY (Yan, Tingyu) [3] , [4] ; Yang, CX (Yang, Chaoxin) [1] ; Sun, SJ (Sun, Shengjun) [1] ; Yue, M (Yue, Meng) [1] ; Liu, XW (Liu, Xuwei) [2] ; Xie, T (Xie, Ting) [2] ; Wang, Y (Wang, Yan) [5] ; (provided by Clarivate) Source NATURE COMMUNICATIONS Volume 15 Issue 1 DOI 10.1038/s41467-024-51130-1 Article Number 6624 Published AUG 5 2024 Indexed 2024-08-18 Document Type Article

Abstract

Electrocatalytic H2 production from seawater, recognized as a promising technology utilizing offshore renewables, faces challenges from chloride-induced reactions and corrosion. Here, We introduce a catalytic surface where OH- dominates over Cl- in adsorption and activation, which is crucial for O2 production. Our NiFe-based anode, enhanced by nearby Cr sites, achieves low overpotentials and selective alkaline seawater oxidation. It outperforms the RuO2 counterpart in terms of lifespan in scaled-up stacks, maintaining stability for over 2500 h in three-electrode tests. Ex situ/in situ analyses reveal that Cr(III) sites enrich OH-, while Cl- is repelled by Cr(VI) sites, both of which are well-dispersed and close to NiFe, enhancing charge transfer and overall electrode performance. Such multiple effects fundamentally boost the activity, selectively, and chemical stability of the NiFe-based electrode. This development marks a significant advance in creating durable, noble-metal-free electrodes for alkaline seawater electrolysis, highlighting the importance of well-distributed catalytic sites.

Developing highly active and stable oxygen evolution electrocatalysts is crucial for enabling large-scale hydrogen production from seawater. Here, authors report a robust O2-producing electrode for alkaline seawater, highlighting the critical role of distributed sites near the catalytic sites.

Keywords

Keywords Plus

LAYERED DOUBLE HYDROXIDESELECTROLYSISCHROMIUM

Oxidation

22-pH-directed polyoxometalate-based supramolecular framework for effectively electrochemical sensing IO3-from glycerol oxidation wastewater

---

By Zhong, BQ (Zhong, Baoqi) [1] ; Liu, JL (Liu, Jinlong) [1] ; Liu, GC (Liu, Guocheng) [1] ; Zhang, Z (Zhang, Zhong) [1] ; Chen, YQ (Chen, Yongqiang) [2] ; Wang, XL (Wang, Xiuli) [1] (provided by Clarivate) **Source JOURNAL OF MOLECULAR STRUCTURE Volume 1332 DOI 10.1016/j.molstruc.2025.141679 Article Number 141679 Published JUN 15 2025 Early Access FEB 2025 Indexed 2025-02-26 Document Type Article**

Abstract

Oxidizing pollutants within the atmosphere or aquatic environments can affect the stability and balance of the entire ecosystem. Therefore, the exploration of rational method for the detection of iodate (IO3-), a notable oxidizing pollutant, is meaningful and obligatory. In this study, a new polyoxometalate-based supramolecular framework [(H2L)3PMo12O40]& sdot;H2O (1) [[HL = 4-(benzimidazole-1-ylmethyl)benzoic acid]] was synthesized from [PMo12O40]3- and the protonated benzimidazole carboxylic acid by environmentally friendly one-pot method, which facilitated by pH adjustment. The detailed characterization of compound 1 was finished by single crystal X-ray diffraction, elemental analysis (EA), infrared (IR) spectroscopy, and X-ray powder diffraction (PXRD). Compound 1 body modified carbon paste electrode (1-CPE) exhibited significant electrocatalytic activity during the electrocatalytic reduction of IO3-, which allowed it to be used for the electrochemical detection of IO3- with robust anti-interference capabilities, stability, and low detection limit (0.427 mu M). 1-CPE also exhibited good detecting effect for the detection of iodate ions within sea water and the waste water from the oxidation reaction of glycerol. This research holds immense significance for the detection of oxidative pollutants and contributes to environmental protection efforts.

Keywords

Author Keywords

PolyoxometalateSupramolecular aggregateIodateElectrochemical detectionChemical wastewater

Keywords Plus

IODATECOORDINATIONREDUCTIONCOMPLEXESCHLORATEBROMATE

Oxidation

23-Anchoring oxidation co-catalyst over CuMn2O4/graphdiyne S-scheme heterojunction to promote eosin-sensitized photocatalytic hydrogen evolution

---

By Yang, C (Yang, Cheng) [1] ; Li, X (Li, Xin) [2] ; Li, M (Li, Mei) [1] ; Liang, GJ (Liang, Guijie) [3] ; Jin, ZL (Jin, Zhiliang) [1] (provided by Clarivate) Source CHINESE JOURNAL OF CATALYSIS Volume 56 Page 88-103 DOI 10.1016/S1872-2067(23)64563-2 Published JAN 2024 Early Access JAN 2024 Indexed 2024-02-10 Document Type Article

Abstract

It is widely acknowledged that efficient charge separation and utilization of photocatalysts are key factors in determing the photocatalytic hydrogen production. Construction of heterojunction has been considered as a promising way to efficiently solve the spatial separation of photogenerated charges. In addition, the introduction of proper cocatalysts can realize the separation of electrons and holes of the photocatalyst and enhance the photocatalytic performance by promoting more carriers to flow to the corresponding active sites. Herein, the S-scheme heterojunction was constructed by introducing graphdiyne into CuMn2O4 for photocatalytic hydrogen evolution. Graphdiyne as a reduction semiconductor and in situ produced Mn2O3 from CuMn2O4 as an oxidation cocatalyst to promote the precisely migration of photogenerated electrons and holes to the corresponding reduction and oxidation sites of photocatalyst. Notably, the photocatalytic performance of the 600-CuMn2O4/GDY-40%(6-CG-40%)could reach 1586.54 mu mol g-1 h-1, which is 13.86 and 21.48 times higher than those of CuMn2O4 (106.73 mu mol g-1 h-1) and graphdiyne (70.57 mu mol g-1 h-1), respectively. Theoretical calculations and experiments results show that both in-situ induced growth of Mn2O3 oxidation co-catalyst and the introduction of graphdiyne to construct S-scheme heterojunction efficiently suppress the severe recombination of photogenerated electron-hole pairs, thus optimizing the photogenerated carrier transfer efficiency, and ultimately leading to the enhanced eosin Y-sensitized photocatalytic hydrogen evolution activity. This work provides a promising method for the construction of oxidation cocatalyst engineered S-scheme heterojunction for solar water splitting.

Keywords

Author Keywords
GraphdiyneOxidation co-catalystCharge separationS-scheme heterojunctionPhotocatalytic hydrogen production
Keywords Plus
CARBONGRAPHDIYNE

Oxidation

24-Cobalt-Doped Ru@RuO$_2$ Core-Shell Heterostructure for Efficient Acidic Water Oxidation in Low-Ru-Loading Proton Exchange Membrane Water Electrolyzers

---

By Chen, JH (Chen, Jinghao) [1] ; Ma, YR (Ma, Yirui) [1] ; Cheng, C (Cheng, Chen) [2] ; Huang, T (Huang, Tao) [1] ; Luo, RH (Luo, Ruihao) [1] ; Xu, JW (Xu, Jingwen) [1] ; Wang, XY (Wang, Xiaoyang) [1] ; Jiang, TL (Jiang, Taoli) [1] ; Liu, HX (Liu, Hongxu) [1] ; Liu, S (Liu, Shuang) [1] ; (provided by Clarivate) Source JOURNAL OF THE AMERICAN CHEMICAL SOCIETY Volume 147 Issue 10 Page 8720-8731 DOI 10.1021/jacs.4c18238 Published FEB 25 2025 Early Access FEB 2025 Indexed 2025-03-05 Document Type Article

Abstract

Proton exchange membrane water electrolysis (PEMWE) is a highly promising hydrogen production technology for enabling a sustainable energy supply. Herein, we synthesize a single-atom Co-doped core-shell heterostructured Ru@RuO2 (Co-Ru@RuO2) catalyst via a combination of ultrafast pulse-heating and calcination methods as an iridium (Ir)-free and durable oxygen evolution reaction (OER) catalyst in acidic conditions. Co-Ru@RuO2 exhibits a low overpotential of 203 mV and excellent stability over a 400 h durability test at 10 mA cm-2. When implemented in industrial PEMWE devices, a current density of 1 A cm-2 is achieved with only 1.58 V under an extremely low catalyst loading of 0.34 mgRu cm-2, which is decreased by 4 to 6 times as compared to other reported Ru-based catalysts. Even at 500 mA cm-2, the PEMWE device could work stably for more than 200 h. Structural characterizations and density functional theory (DFT) calculations reveal that the single-atom Co doping and the core-shell heterostructure of Ru@RuO2 modulate the electronic structure of pristine RuO2, which reduce the energy barriers of OER and improve the stability of surface Ru. This work provides a unique avenue to guide future developments on low-cost PEMWE devices for hydrogen production.

Oxidation

25-Activating lattice oxygen by a defect-engineered Fe_2O_3-CeO_2 nano-heterojunction for efficient electrochemical water oxidation

---

By Huang, QP (Huang, Qiuping) [1] ; Xia, GJ (Xia, Guang-Jie) [2] ; Huang, B (Huang, Bo) [1] ; Xie, DL (Xie, Dongling) [1] ; Wang, JA (Wang, Jianan) [1] ; Wen, D (Wen, Dan) [1] ; Lin, DM (Lin, Dunmin) [1] ; Xu, CG (Xu, Chenggang) [1] ; Gao, L (Gao, Lei) [3] ; Wu, ZD (Wu, Zhenduo) [4] ; (provided by Clarivate)

Source ENERGY & ENVIRONMENTAL SCIENCE Volume 17 Issue 14 Page 5260-5272 DOI 10.1039/d4ee01588f Published JUL 16 2024 Early Access JUN 2024 Indexed 2024-07-02
Document Type Article

Abstract

The sluggish anodic oxygen evolution reaction (OER) is currently the major hinderance for hydrogen production from water splitting. Iron-based materials are promising cost-effective candidates for OER electrocatalysts, however their low intrinsic activity limits their performance. Here we report a defect-engineered Fe2O3-CeO(2)heterojunction with rich oxygen vacancies (Fe2O3@CeO2-OV), exhibiting ultralow overpotential of 172 mV at 10 mA cm(-2) and 317 mV at 1000 mA cm(-2), respectively, alongside superior stability and durability. Advanced characterization and density functional theory calculations demonstrate that defect engineering combined with heterojunction formation activates the lattice oxygen, switching the reaction pathway from the conventional adsorbate evolution mechanism (AEM) to the lattice oxygen mechanism (LOM). The oxygen vacancies are revealed to form preferably on CeO2 and induce not only electronic but also geometric modulation, contributing to strong Fe2O3-CeO2 interfacial interaction and charge transfer from CeO2 to Fe2O3, facilitating the O-2 desorption and boosting the intrinsic activity.

Keywords
Keywords Plus
CATALYSTSSITESALLOY

Oxidation

26-Oxygen Radical Coupling on Short-Range Ordered Ru Atom Arrays Enables Exceptional Activity and Stability for Acidic Water Oxidation

---

By Chang, JW (Chang, Jiangwei) [1] ; Shi, YY (Shi, Yuanyuan) [1] ; Wu, H (Wu, Han) [1] ; Yu, JK (Yu, Jingkun) [1] ; Jing, W (Jing, Wen) [1] ; Wang, SY (Wang, Siyang) [1] ; Waterhouse, GIN (Waterhouse, Geoffrey I. N.) [2] ; Tang, ZY (Tang, Zhiyong) [3] ; Lu, SY (Lu, Siyu) [1] (provided by Clarivate)

Source JOURNAL OF THE AMERICAN CHEMICAL SOCIETY Volume 146 Issue 19 Page 12958-12968
DOI 10.1021/jacs.3c13248 Published MAY 2 2024 Early Access MAY 2024 Indexed 2024-05-31
Document Type Article

Abstract

The discovery of efficient and stable electrocatalysts for oxygen evolution reaction (OER) in acid is vital for the commercialization of the proton-exchange membrane water electrolyzer. In this work, we demonstrate that short-range Ru atom arrays with near-ideal Ru-Ru interatomic distances and a unique Ru-O hybridization state can trigger direct O*-O* radical coupling to form an intermediate O*-O*-Ru configuration during acidic OER without generating OOH* species. Further, the Ru atom arrays suppress the participation of lattice oxygen in the OER and the dissolution of active Ru. Benefiting from these advantages, the as-designed Ru array-Co3O4 electrocatalyst breaks the activity/stability trade-off that plagues RuO2-based electrocatalysts, delivering an excellent OER overpotential of only 160 mV at 10 mA cm(-2) in 0.5 M H2SO4 and outstanding durability during 1500 h operation, representing one of the best acid-stable OER electrocatalysts reported to date. O-18-labeled operando spectroscopic measurements together with theoretical investigations revealed that the short-range Ru atom arrays switched on an oxide path mechanism (OPM) during the OER. Our work not only guides the design of improved acidic OER catalysts but also encourages the pursuit of short-range metal atom array-based electrocatalysts for other electrocatalytic reactions.

Keywords
Keywords Plus
EVOLUTION REACTIONPERFORMANCEMECHANISMCATALYSTSITESREDOX

Oxidation

27-RuO_2-CeO_2 Lattice Matching Strategy Enables Robust Water Oxidation Electrocatalysis in Acidic Media via Two Distinct Oxygen Evolution Mechanisms

---

By Song, HQ (Song, Haoqiang) [1] ; Yong, X (Yong, Xue) [2] ; Waterhouse, GIN (Waterhouse, Geoffrey I. N.) [3] ; Yu, JK (Yu, Jingkun) [1] ; Wang, H (Wang, Hao) [1] ; Cai, JM (Cai, Jinmeng) [1] ; Tang, ZY (Tang, Zhiyong) [4] ; Yang, B (Yang, Bai) [5] ; Chang, JW (Chang, Jiangwei) [1] ; Lu, SY (Lu, Siyu) [1] (provided by Clarivate) Source ACS CATALYSIS Volume 14 Issue 5 Page 3298-3307 DOI 10.1021/acscatal.3c06182 Published FEB 15 2024 Early Access FEB 2024 Indexed 2024-02-28 Document Type Article

Abstract

The discovery of acid-stable and highly active electrocatalysts for the oxygen evolution reaction (OER) is crucial in the quest for high-performance water-splitting technologies. Herein, a heterostructured RuO2-CeO2 electrocatalyst was constructed by using a lattice-matching strategy. The interfacial Ru-O-Ce bridge structure provided a channel for electron transfer between Ru and Ce, creating a lattice stress that distorts the local structure of RuO2. The resulting RuO2-CeO2 catalyst exhibited attractive stability with negligible decay after 1000 h of the OER in 0.5 M H2SO4, along with high activity with an overpotential of only 180 mV at 10 mA cm(-2). In situ attenuated total reflectance surface-enhanced infrared absorption spectroscopy (ATR-SEIRAS), in situ differential electrochemical mass spectrometry (DEMS), and density functional theory (DFT) calculations were used to reveal that the interface and noninterface RuO2 sites enabled an oxide path mechanism (OPM) and the enhanced adsorbate evolution mechanism (AEM-plus), respectively, during the OER. The simultaneous and independent OER pathways accessible by lattice matching guides improved electrocatalyst design for the OER in acidic media.

Keywords

Author Keywords
ruthenium dioxidecerium dioxideacidic wateroxidationoxygen evolution mechanismlattice strain
Keywords Plus
REDUCTIONCATALYSTSTRAINSITES

Oxidation

28-Inhibition of halide oxidation and deprotonation of organic cations with dimethylammonium formate for air-processed p-i-n perovskite solar cells

---

By Meng, HG (Meng, Hongguang) [1] ; Mao, KT (Mao, Kaitian) [1] ; Cai, FC (Cai, Fengchun) [1] ; Zhang, K (Zhang, Kai) [2] ; Yuan, SJ (Yuan, Shaojie) [1] ; Li, TQ (Li, Tieqiang) [1] ; Cao, FF (Cao, Fangfang) [3] ; Su, ZH (Su, Zhenhuang) [4] ; Zhu, ZJ (Zhu, Zhengjie) [1] ; Feng, XY (Feng, Xingyu) [1] ; (provided by Clarivate) Source NATURE ENERGY Volume 9 Issue 5 DOI 10.1038/s41560-024-01471-4 Published MAY 2024 Early Access FEB 2024 Indexed 2024-03-09 Document Type Article

Abstract

The manufacturing of perovskite solar cells under ambient conditions is desirable, yet the efficiency of p-i-n perovskite solar cells fabricated in air still lags behind those made in an inert atmosphere. Here we introduce an ionic pair stabilizer, dimethylammonium formate (DMAFo), into the perovskite precursor solution to prevent the degradation of perovskite precursors. DMAFo inhibits the oxidization of iodide ions and deprotonation of organic cations, improving the crystallinity and reducing defects in the resulting perovskite films. We show the generation of additional p-type defects during ambient air fabrication that suggests the need for improving bulk properties of the perovskite film beyond surface passivation. Upon addition of DMAFo, we demonstrate that the efficiency of inverted p-i-n solar cells based on perovskite layers with 1.53-eV and 1.65-eV bandgaps fabricated under ambient conditions (25-30 degrees C, 35-50% relative humidity) increases by 15-20%. We achieve a certified stabilized efficiency of 24.72% for the 1.53-eV cell, on a par with state-of-the-art counterparts fabricated in an inert atmosphere.

Manufacturing of perovskite solar cells under ambient conditions is desirable. Meng et al. show that dimethylammonium formate suppresses halide oxidation and deprotonation of organic cations, enabling air-processed inverted solar cells with 24.7% efficiency.

Keywords

Keywords Plus

MIXED-ION PEROVSKITESSTABILITYCRYSTALLIZATIONRECOMBINATIONINTERFACES

Oxidation

29-Hollow dodecahedron $K_3PW_{12}O_{40}$/CdS core-shell S-scheme heterojunction for photocatalytic synergistic H_2 evolution and benzyl alcohol oxidation

---

By Sun, LJ (Sun, Lijuan) [1] ; Yu, XH (Yu, Xiaohui) [2] ; Tang, LY (Tang, Liyong) [1] ; Wang, WK (Wang, Weikang) [1] ; Liu, QQ (Liu, Qinqin) [1] (provided by Clarivate) Source CHINESE JOURNAL OF CATALYSIS Volume 52 Page 164-175 DOI 10.1016/S1872-2067(23)64507-3 Published SEP 2023 Early Access OCT 2023 Indexed 2023-12-23 Document Type Article

Abstract

Simultaneous generation of clean energy, H-2, and organic products holds immense potential in the realm of photocatalysis. The S-scheme heterojunction stands out for these dual-function applications due to its robust redox capacity, facilitating both the water reduction reaction and organic oxidation reactions. In this study, a Keggin-type polymetallic oxide, H3PW12O40 hollow dodecahedron (KPW), was synthesized using a hydrothermal approach. Subsequently, cadmium sulfide (CdS) nanoparticles averaging 15 nm in size were integrated in situ onto the KPW shell, resulting in the creation of a core-shell KPW@CdS S-scheme heterojunction. This optimized composite showcased a hydrogen evolution rate of 18.7 mmol g(-1) h(-1), alongside a value-added product, benzaldehyde, with a yield of 17.5 mmol g(-1) h(-1) substantially surpassing the performance of standalone CdS. This S-scheme junction, featuring a pronounced internal electronic field, emerges between the KPW and CdS. It significantly enhances the segregation of photogenerated carriers while preserving formida-ble redox capability. Furthermore, the hollow structure augments light absorption and utility, and the core-shell architecture delivers dual reduction and oxidation sites. As a result, the interplay between the hollow core-shell configuration and the S-scheme mode intensifies the photocatalytic activity. This research provides an innovative approach to crafting hollow S-scheme heterojunc-tions, aiming to optimize photocatalytic redox reactions for effective solar energy utilization.(c) 2023, Dalian Institute of Chemical Physics, Chinese Academy of Sciences. Published by Elsevier B.V. All rights reserved.

Keywords

Author Keywords
S -scheme heterojunctionInternal electric fieldHollow core -shell structurePolyoxometalateBenzyl alcohol oxidation
Keywords Plus
PERFORMANCENITRIDE

Oxidation

30-The isolated Ca-Nx sites in biochar boosting Fe catalyzed Fenton-like oxidation of Tris(2-chloroethyl) phosphate: Properties, mechanisms, and applications

---

By Yang, CY (Yang, Chenyu) [1] , [2] , [3] ; Yao, Q (Yao, Qian) [1] , [2] ; Li, LZ (Li, Liangzhong) [1] , [2] ; Xiao, X (Xiao, Xian) [3] ; Lu, L (Lu, Lun) [1] , [2] ; Liu, C (Liu, Chang) [1] , [2] ; Zhu, CY (Zhu, Chunyou) [1] , [2] ; Zhan, SH (Zhan, Sihui) [4] ; Yuan, HR (Yuan, Haoran) [1] , [2] (provided by Clarivate) Source APPLIED CATALYSIS B-ENVIRONMENT AND ENERGY Volume 366 DOI 10.1016/j.apcatb.2025.125056 Article Number 125056 Published JUN 5 2025 Early Access JAN 2025 Indexed 2025-01-28 Document Type Article

Abstract

The biochar with single Ca atom (1.7 wt%) loaded nanoscale zero-valent iron composites (Fe@Ca-N-C) were prepared and effectively degraded tris(2-chloroethyl) phosphate (TCEP) via Fenton-like oxidation. Systematic experiments and theoretical calculations indicated that the enhanced removal of TCEP was attributed to the synergistic effect of adsorption (Ca-Nx sites) and degradation (Fe sites). Remarkably, the optimal system of 0.1 g L- 1 Fe@Ca-N-C, 1 mM peroxydisulfate (PDS) achieved 99.5 % TCEP degradation (2 mg L- 1) in 30 min and maintained 100 % TCEP removal for 72 h in continuous-flow reaction column. The Ca-Nx sites exhibited high adsorption energy for TCEP (especially the Ca-N1 sites), and the Fe sites (Fe(0) and Fe(II)) activated PDS into HO center dot, which quickly degrades the adsorbed TCEP through C-O bond breaking and C-Cl bond hydroxylation. This study provided insights into single Ca atom utilization and TCEP pollution remediation.

Keywords

Author Keywords

Single Ca atomsTris(2-chloroethyl) phosphateDual-reaction siteAdsorptionFenton-like oxidation

Keywords Plus

ORGANOPHOSPHATE ESTERS

Oxidation

31-Insight into synergy of Mn active sites and spin polarization electrons in Mn-incorporated ZnIn2S4 for boosting photocatalytic hydrogen evolution coupled with benzyl alcohol oxidation

---

By He, ZM (He, Zuming) [1] , [7] ; Xia, YM (Xia, Yongmei) [1] , [3] ; He, G (He, Gang) [1] ; Wang, FH (Wang, Fahui) [1] ; Su, JB (Su, Jiangbin) [1] ; Fareed, H (Fareed, Hasan) [2] ; Fu, XF (Fu, Xiaofei) [3] ; Chen, GH (Chen, Guihua) [4] ; Yang, HP (Yang, Hanpei) [5] ; Zhou, W (Zhou, Wei) [6] (provided by Clarivate) Source CHEMICAL ENGINEERING JOURNAL Volume 506 DOI 10.1016/j.cej.2025.159957 Article Number 159957 Published JAN 15 2025 Early Access JAN 2025 Indexed 2025-02-10 Document Type Article

Abstract
"Spin" being an inherent characteristic of the electrons, can significantly improve the performance of photo- catalysts. In this work, we design and precisely optimize the spin-polarized electrons in ZnIn2S4 (ZIS) system to boost the photocatalytic hydrogen evolution coupled with benzyl alcohol oxidation efficiencies by doping manganese (Mn2+) cations and applying an external magnetic field. The Mn-doped ZIS shows an outstanding photocatalytic hydrogen evolution, and oxidation of benzyl alcohol as compared to the pristine ZIS. Noticeably, applying an external magnetic field further enhances the photocatalytic performance, the optimized Mn0.15-ZIS yields H2 and benzaldehyde production up to 32.75 mmol g-1h-1 and 42.28 mmol g-1h- 1, respectively, which is 13.87 and 12.97-folds higher than that of the pristine ZIS. The Mn0.15-ZIS reaches the highest apparent quantum yield of 18.52 % and 23.60 % at 420 nm for H2 and benzaldehyde production, respectively. These findings are mainly attributed to the spatial synergy of spin polarized electrons, where the Mn sites tend to enrich the holes for benzyl alcohol activation sites. The direct H+ transfer upon benzyl alcohol oxidation at Mn sites facilitates the formation of H2 on adjacent sites with remarkably favorable thermodynamic energy. Moreover, the corresponding mechanisms are systematically investigated by utilizing the magnetic circular dichroism spectroscopy, in situ X-ray photoelectron spectroscopy, transient-state photoluminescence, transient photocurrent, electrochemical impedance spectroscopy, electron paramagnetic resonance measurements, and the density functional theory calculations. This study, exhibits a great potential of manipulating the spin-polarized electrons providing an efficient strategy for enhancing the photocatalytic performance.

Keywords
Author Keywords
PhotocatalysisSpin-polarized electronHydrogen productionExternal magnetic field
Keywords Plus
H-2 EVOLUTIONEFFICIENTHETEROJUNCTION