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Hyping direct seawater electrolysis hinders electrolyzer development

J. Niklas Hausmann,[1,*] Lea R. Winter,[2] M.A. Khan,[3] Menachem Elimelech,[2] Md Golam Kibria,[4] Tobias Sontheimer,[5] and Prashanth W. Menezes[1,6,*]

Jan Niklas Hausmann finished his PhD in 2022 and is currently a postdoc at the CatLab of the Helmholtz-Zentrum Berlin in the group of Prashanth W. Menezes. He is a trained inorganic chemist, and his research focuses on the development of electrocatalysts and structure-activity relations for conventional and hybrid water splitting. Furthermore, he is interested in the techno-economics of these electrocatalytic processes and has recently published an article titled ''Is direct seawater splitting economically meaningful?''

Lea R. Winter is an assistant professor in the Department of Chemical and Environmental Engineering at Yale University. She received a PhD in chemical engineering from Columbia University in 2020. She obtained postdoctoral training as a Nanotechnology Enabled Water Treatment (NEWT) Distinguished Postdoctoral Fellow at Yale in 2020–2022. Her research focuses on electrified processes at the food, energy, water, and climate nexus, including development of sustainable and circularized processes for conversion of CO_2 to chemicals and fuels, green nitrogen fixation to fertilizers and nitrogen-based fuels, and transformation of contaminants in wastewater into useful products while recovering fit-for-purpose water.

M.A. Khan is an assistant professor in the Chemical and Materials Engineering department at the University of Alberta. His research aims to contribute to a sustainable future through the development of innovative technologies that support zero-emission energy carriers like hydrogen and electricity. Dr. Khan's approach integrates techno-economic analysis and life cycle analysis to model energy systems and net-zero transition pathways. His current contributions include advancements in the production of sustainable fuels and chemicals, impacting areas such as steel

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[1] Materials Chemistry Group for Thin-Film Catalysis – CatLab, Helmholtz-Zentrum Berlin für Materialien und Energie, Albert-Einstein-Str. 15, 12489 Berlin, Germany

[2] Department of Chemical and Environmental Engineering, Yale University, New Haven, CT 06520-8286, USA

[3] Department of Chemical and Materials Engineering, University of Alberta, Edmonton, AB, Canada

[4] Department of Chemical and Petroleum Engineering, University of Calgary, Calgary, AB, Canada

[5] Strategy Department of Energy and Information, Helmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner-Platz 1, 14109 Berlin, Germany

[6] Department of Chemistry: Metalorganics and Inorganic Materials, Technische Universität Berlin, Straße des 17. Juni 135, Sekr. C2, 10623 Berlin, Germany

*Correspondence:
niklas.hausmann@helmholtz-berlin.de (J.N.H.),
prashanth.menezes@helmholtz-berlin.de
(P.W.M.)

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production and heavy-duty transportation. He has authored/coauthored 46 scientific articles and 4 government reports and has 6 granted US patents.

Menachem Elimelech is the Sterling Professor of Chemical and Environmental Engineering at Yale University. His research interests include emerging membrane-based technologies at the water-energy nexus, materials for next-generation desalination and water purification membranes, and environmental applications of nanomaterials. Professor Elimelech is a Clarivate Analytics (formerly Thomson Reuters) Highly Cited Researcher. He is a member of the United States National Academy of Engineering and a foreign member of the Chinese Academy of Engineering, the Australian Academy of Technology and Engineering, and the Canadian Academy of Engineering.

Md Golam Kibria is an associate professor at the Department of Chemical and Petroleum Engineering at the University of Calgary. He is the cofounder and CTO of several spin-off companies from the University of Calgary, including O-Two Carbon Inc., CarboMat Inc., and NetZero Hub Inc. Kibria has extensive expertise in electrochemical systems, including water electrolysis and CO_2 electrolysis as well as electrochemical oxidation reactions. Furthermore, he is interested in process modeling and system-level analysis, including techno-economic and life cycle analysis, and has recently published an article titled "Seawater electrolysis for hydrogen production: A solution looking for a problem?"

Tobias Sontheimer heads the Strategy Department for Energy and Information at the Helmholtz-Zentrum Berlin and has been helping to shape the transformation of the energy sector for more than 15 years. In his current role, he is responsible for research and development for green hydrogen technologies and sustainable aviation fuels. From 2014 to 2020, he was chief research manager for the Helmholtz Association's national energy R&D portfolio. Sontheimer studied physics at RWTH Aachen University and Harvard University and completed his doctorate in the field of renewable energy technologies as a scholarship holder of SCHOTT AG.

Prashanth W. Menezes is head of the Materials Chemistry Group for Thin-Film Catalysis at the CatLab of the Helmholtz-Zentrum Berlin and leads the Inorganic Materials Group at Technische Universität Berlin. He received his PhD from the Max Planck Institute for Chemical Physics of Solids in Dresden, after which he moved to Technische Universität München and then to Technische Universität Berlin to work on energy catalysis. His research focuses on the design, development, and dynamic structural understanding of novel functional precatalysts in heterogeneous catalysis, especially for applications in redox oxygen catalysis, (photo)electrocatalytic water splitting, and electrochemical redox reactions.

To limit global warming to less than 1.5°C, global greenhouse gas emissions must be reduced by 45% by 2030 and reach net zero by 2050.[1] In the transition to net-zero emissions, low-carbon hydrogen will play a vital role in the decarbonization of hard-to-abate sectors such as the chemical industry, long-term energy storage, aviation, and shipping.[1,2] Low-carbon hydrogen can be produced by water electrolysis using carbon-neutral electricity. The US Department of Energy set an ambitious hydrogen price of USD $1 per kg until 2031 as a goal, and for its realization, electrolyzers must become cheaper, more efficient, and more durable. Academic research plays a central role in achieving these goals, and researchers have a responsibility to pursue societally relevant aims due to the publicly funded nature of academic research. With the urgent need to decarbonize our energy systems and address climate change, we must consider realigning our financial and workforce resources toward developing practical solutions to reach net zero by 2050.

Direct seawater electrolysis (DSE, or direct seawater splitting) is a technology that uses seawater (>96% of Earth's water) instead of ultrapure water to produce hydrogen. In this article, the term DSE is also employed for approaches using seawater with additives, e.g., alkali hydroxides or buffers. DSE is promoted as a process enabling the cheap production of hydrogen fuel from abundant seawater,[3–5] e.g., in arid coastal areas with low-cost renewable power. This hydrogen can be globally transported and then used as a chemical feedstock or to provide energy and salt-free water when oxidized.[3–5] Following these superficial arguments, DSE seems to be a holy grail that can potentially contribute to solving two humanitarian crises: climate change and water scarcity. Consequently, the number of publications on DSE has been growing exponentially (more than 500 in 2023; see Note S1), and DSE reports receive outstanding public interest and media coverage (Altmetric scores above 500).[4] However, commercially available water purification and water electrolysis can already robustly produce

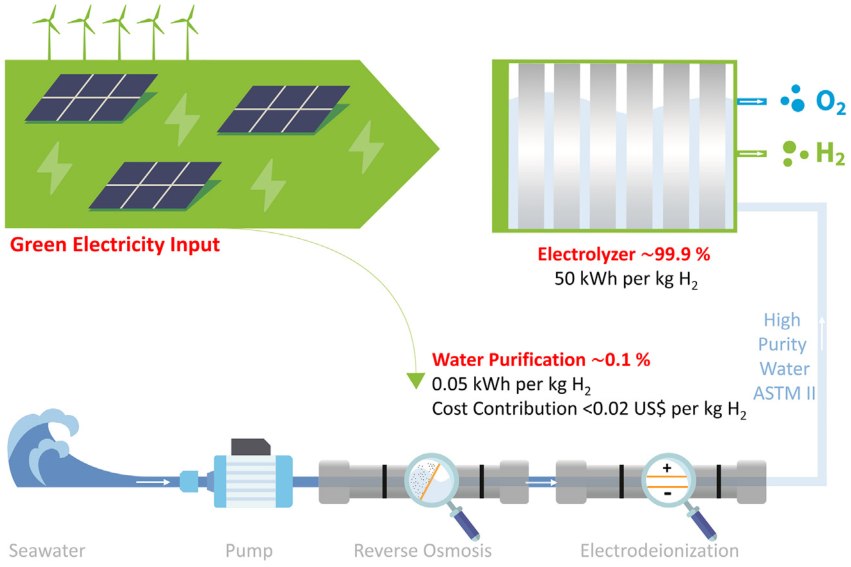

Figure 1. Comparison of the energy and overall costs of seawater purification and electrolysis

Hydrogen production using seawater with commercially available, mature technologies. The water source is insignificant, as (sea)water purification to ASTM Type II water adds insignificant costs (including energy, maintenance, and capital costs) to the overall process (see Note S3). The thickness of the green arrow tails pointing to the electrolyzer and to the water purification system is proportional to their respective electricity consumptions.

hydrogen from seawater, and there are no significant technological, economic, or environmental incentives for the highly demanding development of DSE.[6–8] Thus, we anticipate that DSE cannot contribute to reaching net zero until 2050, and focusing on its demanding development misspends limited funding resources.

Mature technologies can produce hydrogen from seawater without DSE

Current electrolyzers use purified water (American Society for Testing and Materials [ASTM] Type II, <1 μS/cm; see Note S2), as it is beneficial for an electrolyzer's lifetime and performance. The purified water is obtained through the treatment of a given water source, such as fresh water, seawater, brackish water, or industrial or household wastewater.[6–8] Globally, the most-applied water purification technology is reverse osmosis, where the feed water source is pumped through a water-selective membrane to achieve over 99% removal of total dissolved solids. Reverse osmosis can be followed by ion-exchange resins or electrodeionization units to produce the required higher water purity.[7] Therefore, with fully established, commercially available technologies for water purification and electrolysis, hydrogen can be made from a wide variety of water sources, including seawater.[7] Thermodynamically, seawater purification accounts for only 0.03% of the total energy required for producing hydrogen from seawater through this coupled approach (salinity of 3.5 g per L, 0.0135 kWh per kg H_2 for the water purification and 33.3 kWh per kg H_2 for the electrolysis).[6] For commercially available technologies, seawater purification to ultrapure water has a negligible influence on the total energy consumption associated with electrolysis (~0.1% of total energy; see Note S3). Since energy consumption is the main cost driver for both technologies, water purification also has a negligible contribution to the overall costs, including maintenance and capital costs (<USD \$0.02 per kg H_2; see Note S3). This comparison is illustrated in Figure 1. Therefore, eliminating the water purification step using DSE would provide essentially no improvements to the energy and cost requirements for making hydrogen from seawater.

DSE involves enormous technical challenges

While the development of DSE can lead to no significant advantages, it involves tremendous challenges. Seawater contains an enormous variety of impurities, including inorganic salts of all of Earth's elements, charged and uncharged small organic molecules, dissolved natural organic matter, living organisms, and dissolved gases.[6] These chemical species affect all parts of the electrolyzer, including the electrodes, ion exchange membrane or diaphragm, catalysts, and the balance of plant. For example, specific already-reported harmful interactions include[6,9,10] (1) biofouling; (2) formation of toxic, environmentally harmful, and corrosive hypochlorite and chlorine gas; (3) blocking of the diaphragm or ion exchange membrane by charged impurities; (4) degradation of the membrane by iron; (5) blocking of catalyst active sites by ions; (6) dissolution of catalysts by organic species with coordinating functional groups; (7) adhesion to all surfaces by fluoride; (8) poisoning of the hydrogen purity by preexisting gases or electrogenerated gases; (9) deposition of colloids such as aluminosilicate clays; (10) deposition of aliphatic organic compounds; and (11) decreasing faradaic efficiency due to side reactions. Furthermore, every pipe, seal, device, or electrolyzer part is threatened by corrosion and biofouling through seawater exposure, decreasing lifetime and increasing materials requirements and, thus, capital and maintenance costs. Additionally, due to the unpredictable local and seasonal occurrences of these diverse impurities, electrolyzers would need to be built to withstand these constantly changing and uncertain conditions. Overcoming all of these challenges will require substantial investment and research time, and it is questionable whether direct seawater electrolyzers can ever achieve robustness and lifetime anywhere near those of high-purity-water-fed electrolyzers.

Real advancements for green hydrogen should focus on improving high-purity-water-fed electrolyzers

The scientific community must use public research funds earmarked for green hydrogen and climate change goals to focus on improving electrolyzer technology that can make hydrogen more affordable. Thus, electrolyzers must be optimized toward parameters that significantly affect the real practical barriers for hydrogen production, which are (1) energy efficiency, (2) lifetime, (3) capital costs, (4) resource availability, (5) operational flexibility (e.g., start-up time and minimal and maximal power consumption), (6) recyclability, (7) maintenance, (8) hydrogen purity, and (9) hydrogen pressure. These parameters can be improved by optimizing the balance of plant, operating conditions (i.e., temperature and current density), catalysts, diaphragm or membrane separators, and electrolytes. Using seawater as the electrolyte inhibits precise electrolyte optimization, which has been shown to be crucial for efficiency and lifetime.[9,11] It would be miraculous if seawater were the optimal electrolyte for all of these parameters, and we note again that seawater varies depending on location and season. Furthermore, optimizing one parameter involves trade-offs for other design goals. For example, increasing catalyst loading leads to a better lifetime but will increase capital costs and could negatively impact the availability of critical metal resources. Focusing on the complexities involved in DSE (which, under the best circumstances, can only result in negligible improvements to cost and durability) will significantly detract from advancing the high-purity-water-fed electrolyzer design needs.

Demonstrated DSE technologies are unsuitable for green hydrogen production

In this section, we discuss two prominent DSE approaches to show the consequences of a focus on DSE instead of on innovations that improve significant performance metrics. In the first approach, water-selective membranes are applied around the electrolysis stacks to supply pure water to the electrolyzer from surrounding seawater.[3] In contrast to reverse osmosis, no pump is required, as different driving forces, such as osmotic pressure, are utilized. The most prominent example of DSE using a water-selective membrane was published in 2022.[3] News outlets described it as a "groundbreaking achievement" and "milestone" that avoids "cumbersome, land-intensive, and costly" desalination, and it was listed as one of the top ten scientific advances of 2022 by China's Ministry of Science and Technology.[12] From our point of view, this approach is inventive and marks a substantial step forward in the development of DSE. However, this does not change the fact that DSE lacks real-world potential and consumes funding resources that are limited and designated for societal impact. The water-selective membrane DSE approach uses almost 2 m^2 of a hydrophobic, porous, polytetrafluoroethylene-based, waterproof, breathable membrane between the seawater and the electrolyzer, which is filled with 30 wt % aqueous KOH. These 2 m^2 membranes provide around 11 L of purified water for the electrolyzer per day. In comparison, a 2 m^2 membrane in a conventional single-pass reverse osmosis setup can provide approximately 5,000 L of purified water daily.[13] Thus, the DSE approach uses substantially more membrane material, increasing the capital costs and environmental footprint compared to a conventional electrolyzer with a reverse osmosis unit.

There are numerous other drawbacks to the water-selective membrane electrolyzer approach. Notably, the system design and conditions are strongly restricted, as water migration and consumption must be balanced to maintain the optimal electrolyte concentration. Thus, the temperature and current density cannot be changed easily. In addition, the membrane can operate only at up to 7.5 bar, making the production of pressurized hydrogen infeasible. Further, the large polytetrafluoroethylene-based membrane area in contact with seawater is prone to biofouling. We also note that platinum is used for the alkaline electrolyzer, and most significantly, the device's energy consumption (2.3 V at 400 mA cm^{-2}) is 25% more than that of state-of-the-art electrolyzers (<1.8 V at 400–2,000 mA cm^{-2}).[2,9,14] The lower efficiency is likely caused by the restrictions in the reaction conditions (e.g., temperature) caused by the integrated membrane design. These substantial disadvantages are enormously larger than adding an inexpensive water desalination and purification step, which increases the energy consumption by a negligible amount, e.g., from 50 kWh per kg H_2 to, at most, 50.05 kWh per kg H_2.

The other approach that received even more public interest is a report from 2023.[4] Therein, a Lewis acid layer is introduced to the catalyst to produce hydroxyl anions locally, and the noble-metal-free electrochemistry setup is directly exposed to seawater. The approach is brilliant and inventive, considering the tremendous challenges of DSE. However, the performance parameters reveal its real-world incompatibility. The electrolyzer deactivates in the first few hours, even at 25°C, by more than 100 mV, reaching a current density of 500 mA cm^{-2} at 2.15 V. Thus, this electrolyzer requires around 20% more energy than a conventional proton-exchange membrane electrolyzer (<1.8 V at 400–2,000 mA cm^{-2}).[2,9,14] Furthermore, even when it is operated at a constant current density, its hydrogen evolution reaction faradaic efficiency is only 93%, compared to around 99% in commercial high-purity

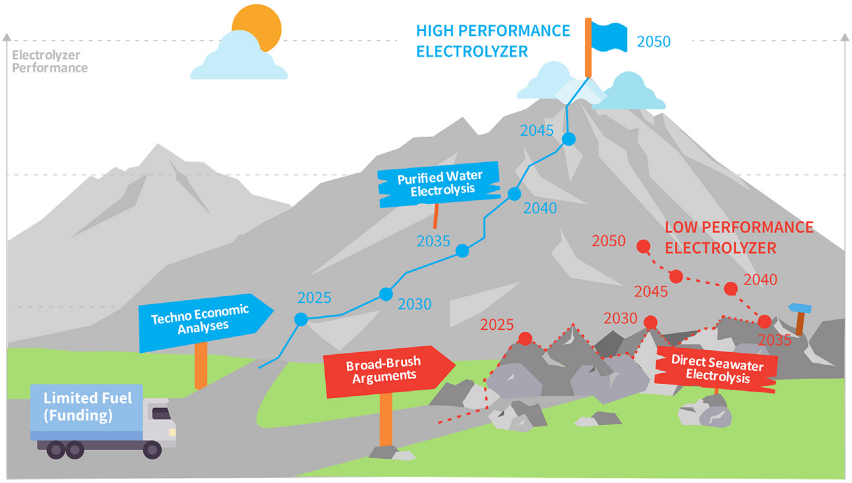

Figure 2. Focusing on the development of direct seawater electrolysis misspends limited funding resources

We stand at a crossroads with two pathways for the development of electrolyzers. The red path leads into the boulder field of direct seawater splitting and follows the "broad-brush arguments" in favor of direct seawater electrolysis and partly misspends limited resources, such as funding or workforce (represented as the truck's fuel), on the development of direct seawater electrolysis (500 publications in 2023; see Note S1), a technology that techno-economic analyses (see Note S4) have clearly shown to have no significant potential to improve electrolyzer performance (represented as altitude). The development of direct seawater electrolysis is symbolized as a field of boulders leading to no increase in altitude. The blue path follows the "techno-economic analyses" and focuses resources on improving parameters relevant to electrolyzer performance and final green hydrogen price. Thus, it omits the misspending on DSE, leading to better electrolyzer performance in 2050.

electrolyzers,[15] resulting in another 7% energy-to-hydrogen-efficiency decrease. These disadvantages are immense compared to saving ~0.1% of the energy and <USD $0.02 per kg H_2 of the overall costs by adding a water purification unit, and they show how electrolyzer design with a focus on DSE leads to the development of technologies with diminished real-world applicability.

Public resources must be spent following techno-economic analyses and fundamental thermodynamic laws

We have limited resources to develop electrolyzers, and those public resources must be spent toward achieving societal goals following logical guidance provided by techno-economic analyses and fundamental laws of thermodynamics (i.e., seawater desalination requires 0.03% of the total energy required for water splitting). The choices between this path and one that pursues DSE are represented in Figure 2. Academic research does not always have to lead to immediate real-life applications, and we do not intend to stop blue-skies research if it is not promoted as a real-world solution, as is often done for DSE. However, if we spend substantial resources pursuing the current DSE hype (500 publications in 2023, with exponential growth; see Note S1), we will miss opportunities for attaining electrolyzers with improved lifetime, capital cost, and efficiency.

Misinformation fuels the DSE hype

Even though adding a seawater purification step before electrolysis has a negligible influence on the cost of hydrogen (Figure 1), the hype surrounding the development of DSE continues. This hype is fueled by ignoring the straightforward techno-economic details and focusing on misleading broad-brush arguments (Figure 2). For example, the leading author of the aforementioned DSE report using a Lewis acid layer claimed that "we have split natural seawater into oxygen and hydrogen with nearly 100% efficiency."[16] Obviously, such a statement creates tremendous public interest and can lead to increased attention and funding for DSE. However, this claim is, at best, strongly misleading, as their device efficiency was only around 53%, compared to up to 79% if separate water purification and high-performing commercially available water electrolysis were used (see Note S5 for the efficiency calculations).[2,9,14] Furthermore, it is stated in the aforementioned report using a water-selective membrane electrolyzer approach that water purification "requires additional energy input, making it economically less attractive."[3] However, the additional energy input is ~0.1% and, thus, economically insignificant. Moreover, DSE scientists constantly claim that large-scale hydrogen production would threaten freshwater supplies, while the opposite is true, as shifting from our current fossil-fuel-based system to a hydrogen economy would substantially reduce our water demand, as has been discussed intensively elsewhere.[6,7,17] Furthermore, fresh water is not scarce when electricity and seawater are available, as it can easily be obtained through reverse osmosis for a price of approximately USD $0.001 per liter.[6–8]

Other recurring statements in favor of DSE assert that desalination plants have high capital costs or make the electrolysis system less flexible.[3,5] However, the capital costs of the purification system add only around USD $0.005 to the overall hydrogen cost per kg (see Note S3), and the purified water can easily be stored in tanks, making the system flexible. These statements from electrochemists and materials scientists working on DSE likely originate from a lack of knowledge of desalination technologies. We agree that developing new materials and electrodes for more impurity-resistant electrolyzers is meaningful, as this can extend electrolyzer lifetime and might slightly reduce the substantial balance of plant costs (see Note S6 for the cost contributions of water purification to the balance of plant).[6,9,10,14] Furthermore, advancements that enable the use of water with slightly lower purity than ASTM Type II water could eliminate several polishing steps, simplifying water treatment prior to electrolysis. In this regard, a single-pass reverse osmosis step can already remove >99% of seawater impurities and comes with almost no additional energy and capital costs, space requirements, or flexibility loss with respect to electrolysis. We anticipate that this commentary will close the knowledge and communication gap between research communities with different scientific backgrounds that contribute to the advancement of green hydrogen, as its authors combine techno-economic, electrochemical, materials, and water purification expertise.

Conclusion

Seawater purification prior to electrolysis is inexpensive and straightforward. Thus, DSE remains a solution looking for a problem.[8] We do not suggest that DSE devices cannot be developed, and we acknowledge the high scientific quality of reports on the topic. However, we assert that the techno-economic disadvantages outweigh the minimal possible savings from DSE. In this regard, to put it into an electrochemist's perspective, omitting seawater purification brings, at most, the energy-saving equivalent of an overpotential reduction by 0.002 V. Simultaneously, DSE faces tremendous challenges, and designing an electrolyzer with a focus on meeting those leads to restriction (e.g., in electrolyte or membrane design) and comes with trade-offs for real-world-relevant application metrics (e.g., lifetime, capital costs, and energy-to-hydrogen efficiency). Thus, while DSE contributes to blue-skies research, extending the DSE hype through promoting it as a real-world solution misspends the limited resources needed to develop effective decarbonization technologies. Scientists, journalists, and the public have been presented with misleading arguments in favor of DSE, which we have systematically exposed with this commentary. The conclusions presented herein are also in line with the reports of the most reputable agencies, such as the International Energy Agency, which states that "[water desalination by reverse osmosis] has only a minor impact on the total costs of water electrolysis, increasing total hydrogen production costs by USD \$0.01–0.02/kg H_2,"[2] and the International Renewable Energy Agency, which asserts, "Water use is no barrier to scaling up electrolysis. Even in places with water stress, seawater desalination can be used with limited penalties on cost [USD \$0.01/kg H_2] or efficiency."[18] We anticipate that this commentary will show editors, funding agencies, electrochemists, and materials scientists that vague arguments in support of DSE clash with the results of careful techno-economic analyses and that focusing efforts on developing DSE in the name of mitigating climate change can actually impede decarbonization efforts.

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at https://doi.org/10.1016/j.joule.2024.07.005.

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DECLARATION OF INTERESTS

The authors declare no competing interests.

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Zinc-anode reversibility and capacity inflection as an evaluation criterion

Zhuoxi Wu,[1] Yu Wang,[1,*] and Chunyi Zhi[1,2,3,4,*]

Zhuoxi Wu is currently a PhD student at the Department of Materials Science and Engineering in City University of Hong Kong, under the supervision of Professor Chunyi Zhi. His current research mainly focuses on zinc-anode reversibility improvement and electrolyte modification of zinc-ion battery.

Yu Wang is a postdoc in the Department of Materials Science and Engineering at City University of Hong Kong in Professor Chunyi Zhi's group. Her current research mainly focuses on advanced aqueous zinc-ion batteries and the design of advanced metallic anodes. Dr. Wang obtained her bachelor's and master's degree in chemistry from Harbin Institute of Technology in 2012 and 2014. She obtained her PhD from the Chinese University of Hong Kong and mainly focuses on Li-air and aqueous Li-ion batteries.

Chunyi Zhi obtained a PhD in condensed matter physics from the Institute of Physics, Chinese Academy of Sciences. After 2 years of being a postdoc at the National Institute for Materials Science (NIMS) in Japan, he was promoted to ICYS researcher, researcher (faculty), and senior researcher (permanent position) in NIMS. Dr. Zhi is now a chair professor at Department of Materials Science and Engineering in City University of Hong Kong. Dr. Zhi has extensive experience in aqueous electrolyte batteries and zinc ion batteries.

Introduction

Studies on zinc-ion batteries (ZIBs) are typically conducted under controlled conditions, and researchers have attempted to report reversibility of zinc anodes by optimizing the experimental conditions to obtain high Coulombic efficiency (CE).[1–5] Although CE is widely used to evaluate the reversibility of zinc anodes, primarily because electrochemical tests do not differentiate between electrons provided by main or side reactions, the statistics of CE may be biased.[6–8] Therefore, a precise method is required for evaluating the reversibility of zinc anodes in ZIBs. In this study, we used Zn||Cu half-cells as a platform to explore the reversibility patterns of a zinc anode and its CE in terms of rate, areal capacity, electrolyte dosage, and zinc utilization ratio (ZUR) (Note S1; Figures S1 and S2).

Zinc-anode reversibility under different conditions

Rate

In ZIBs, zinc deposition is affected by irreversible side reactions, including chemical and electrochemical side reactions. Chemical side reactions are spontaneous and time dependent. When a battery is operated for a long duration, these irreversible side reactions accumulate. Electrochemical side reactions are irreversible reactions affected by the electric field inside the battery. At high current density, or high rate, these reactions accumulate with each cell cycle. At a low rate, chemical side reactions are the main factor influencing the performance of batteries. By contrast, at a high rate, the time required for a single cycle is shorter, and electrochemical side reactions are predominant.

Figure 1A depicts the average cycle life and CE of a battery operated at different rate with an areal capacity of 3 mAh cm^{-2} (Note S1; Figure S1). When the rate is increased from 0.05 to 1 C, the number of cycles also increases. However, when the rate exceeds 1 C, the cycle life of the battery starts to

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[1] Department of Materials Science and Engineering, City University of Hong Kong, Kowloon 999077, Hong Kong SAR, China

[2] Hong Kong Institute for Advanced Study, City University of Hong Kong, Kowloon 999077, Hong Kong SAR, China

[3] Hong Kong Institute for Clean Energy, City University of Hong Kong, Kowloon 999077, Hong Kong SAR, China

[4] Centre for Advanced Nuclear Safety and Sustainable Development, City University of Hong Kong, Kowloon 999077, Hong Kong SAR, China

*Correspondence: yuwang@cityu.edu.hk (Y.W.), cy.zhi@cityu.edu.hk (C.Z.)

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