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Prevention of Chloride Corrosion in Direct Seawater Electrolysis

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Abstract

Hydrogen is emerging as a green, renewable and zero-emission energy carrier. Seawater is emerging as renewable feed because of freshwater scarcity but it requires prior desalination. The energy intensive desalination can be avoided using direct sea water electrolysis. Direct sea water electrolysis poses various challenges of electrode corrosion, poisoning and competitive side reactions. Most of these challenges are posed by the high concentration of chloride ions present in seawater. This review focuses on the challenges caused by the presence of chloride ions and other species on the direct seawater electrolysis. It also discusses the different strategies utilized to prevent chloride harmful effects. Finally, the future directions are discussed to give a roadmap for future research directions in this field.

Keywords: Direct seawater electrolysis, Chloride corrosion, Oxygen evolution reaction (OER), Chlorine evolution reaction (ClER), Electrocatalyst selectivity, Overpotential reduction

Introduction

Hydrogen is considered as one of the unparalleled fuels in terms of its renewability, sustainability, greenness and emission-free nature. The world is aiming to replace all fossil fuel-based energy systems with green hydrogen [1]. There are multiple ways for green hydrogen production including photocatalytic, thermos-catalytic, biological hydrogen production, photoelectrochemical and electrocatalytic hydrogen production [2-6]. Out of all these methods, electrolytic hydrogen production is the most employed one by integrating it with green electricity from hydroelectric, wind or solar power plants [7]. The electrolytic hydrogen production from the water splitting has already achieved very high efficiencies in some electrolysis technologies [8]. All of the currently mature electrolysis technologies require the use of desalinated or freshwater for electrolytic splitting [9]. This dependence of electrolytic systems on scarce freshwater limit their usefulness. Currently the focus has been on the utilization of seawater for electrolytic hydrogen production as being the largest water reservoir making around 97% of all earth water resources [10]. But unfortunately, the utilization of seawater in electrolyzer require prior desalination consuming a lot of energy making the overall process again energetically inefficient [11]. So, currently a lot of focus is being laid on the development of such electrolysis technologies which could directly utilize seawater for hydrogen production without desalination [12]. This technology saves a lot of energy making the overall process economical, but it also face various challenges due to the presence of various pollutants in seawater.

Challenges of Direct Seawater Electrolysis

Direct Seawater Electrolysis (DSE), despite being the most efficient form of electrolytic hydrogen production faces a lot of challenges limiting its usefulness. Seawater contain various impurities including different anionic species like chloride and bromide, cationic species like calcium and magnesium ions, various microbes and suspended solids [13]. The feeding of this seawater directly to electrolyser poses various problems including the corrosion of electrodes and electrolyser, the clogging of adsorp-

tion sites and the poisoning of catalytic active sites [14, 15].

The ionic species present in seawater react with the electrodes and electrolyser materials causing their corrosion [16]. Sometimes the calcium and magnesium ions precipitated as their hydroxides near the active sites due to local pH changes clogging the active sites [17]. Similarly, microbes may also deposit on the electrodes either due to surface activity or due to redox conversion causing the clogging of catalytic surface. The greatest challenge in direct seawater electrolysis is posed by the chloride ions because of their very high concentration (0.5M) in seawater and very close oxidation potential to water [18].

Oxygen Evolution Reaction & Chlorine Evolution Reaction (Thermodynamics vs Kinetics)

As the concentration of chloride ions is very high in seawater, so, while electrolysis, the chloride ions get attracted towards the anode and compete with the oxygen evolution reaction (OER). There is a difference of around 480mV in the oxidation potentials of water and chloride ions to form O2 or Cl2 under nearly neutral electrolytic conditions [19]. It makes the OER thermodynamically more favorable than chlorine evolution reaction (ClER) under normal conditions, but the main problem arises due to the kinetics of both of these reactions. The oxidation of water to form oxygen is 4 electron process which requires more time making it kinetically less favorable than the ClER which is a 2 electron faster process. The 4 electron OER requires a lot of overpotential reducing the potential window between oxidation potentials of water and chloride.

Methods for Prevention of Chloride Corrosion

The main focus of the development of electrocatalysts for direct seawater electrolysis is the increase of electivity of OER over ClER. Various strategies are being implemented to achieve this goal. The main strategies for this selectivity involve the reduction of overpotential required for OER, the blocking and the repulsion of chloride ions from the electrocatalyst.

Reduction of OER Overpotential

The overpotential required for OER is reduced by developing the catalysts which provide better stability for the reaction intermediates by providing suitable adsorption sites with nearly zero adsorption energy. For this purpose, the OER electrocatalysts are developed by tuning the binding energies of different sites on their surfaces but different reaction intermediates. The catalysts with required properties are developed by combining, compositing, and doping different materials to get suitable adsorption energies.

The strategies used to prevent chloride ions from reaching the electrocatalysts either involves the steric blocking of the chloride ions or the electrostatic or common ion effect-based repulsions.

Chloride Blocking

The steric blocking of chloride ions from reaching the anode is a quite effective strategy for a stable direct seawater electrolysis system. This strategy involves the development of such layers on the surface of OER catalysts which does not allow the chloride ions to pass through them due to slightly larger size as compared to hydroxides. In a study, defective antifluorite MnO2 layer was electrodeposited on RuO2 catalyst and it was observed that it protected the catalysts from chloride ions sterically blocking them. This system was used for electrolysis of unbuffered seawater with 100% FE for OER for 100 hours [20].

Chloride Repulsion

The chloride ions are also prevented from reaching the anode by repulsion either by electrostatic effect or by common ion effect. The catalysts are developed which have Lewis acidic species on their surface which preferentially adsorb hydroxide ions making the surface negatively charged causing the repulsion of chloride ions [21]. In a study, Zhou et al. developed an ultrathin layer of MoO3 on the surface of the CoO electrocatalyst which protected the catalysts by preferentially adsorbing hydroxides [22]. In another study by Cai et al., an OER catalyst with Cr2O3 was utilized for seawater electrolysis. This chromium oxide adsorb hydroxide ions ultimately increasing their concentration near catalysts [23]. Some studies indicate that the intercalation of different anions between the layers of 2D catalysts also have repulsive effects on chloride ions, increasing the selectivity for hydroxide ions. Various ionic species including sulphate, carbonate, oxalate, citrate, phosphate, and hexaflurophosphate etc. were introduced into the interlayers of 2D materials and almost all more or less provided the chloride repulsion [24-28]. This intercalation of anionic species is considered one of the most effective strategies providing stability up to 1000 hours under various compositions and media.

Future Directions

Despite various studies conducted on direct seawater electrolysis, it is still far from practical. There is a lot of space in various areas of direct seawater electrolysers. Research should be conducted on the development of OER electrocatalysts providing oxygen evolution at even lower overpotentials. There is also a need to develop multifunctional coating which could provide selectivity without affecting the conductivity. Self-healing and adaptive electrode surfaces should be developed to work in seawater. Currently, the more focus is on the protection of electrodes from chloride so there is also a need for the studies on the prevention of other species found in seawater like calcium magnesium ions and organics which limit the efficiencies of direct seawater electrolyzers. There is also a need for the testing of developed devices in real seawater and the systems should be developed accordingly. There should also be the integration of hydrogen production research with AI and interdisciplinary collaboration between different fields of electrochemistry, surface, material science and ocean sciences for the development of better catalysts and electrolyzer designs to get better performance.

Conclusion

Hydrogen energy, being the fuel of the future, is progressing day by day; but the freshwater scarcity is a prevailing challenge in this field. This issue can be addressed by using the approach of direct seawater electrolysis. Direct seawater electrolysis currently suffers from many limitations including the instability of the device, corrosion, catalytic poisoning and competing side reactions like ClER. Different approaches have been utilized to address these issues including the efficiency enhancement of OER catalyst by lowering the overpotential and increasing the selectivity of the OER catalyst by chloride ion repulsion. The chloride ions repulsion strategies including electrostatic repulsion by

anionic intercalation, common ion effect-based repulsion and steric repulsion were tried. Despite various great efforts, DSE is far from commercialization due to overall instability of the system due to one reason or the other. There is a need to explore more stable, selective and efficient systems by integrating different disciplines of chemistry, material science and AI to get such materials and system designs which could provide better performance.

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