



Hydrogen production from industrial wastewaters: An integrated reverse electrodialysis - Water electrolysis energy system

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ABSTRACT

This work presents a novel approach combining reverse electrodialysis (RED) and alkaline polymer electrolyte water electrolysis (APWEL) for renewable hydrogen production. APWEL is fuelled by salinity gradient power (SGP) extracted from sulfate (SO_4^{2-})-rich industrial wastewater. The performance of a pilot-scale RED unit (200 cells, active area: $31.5 \times 63.5 \text{ cm}^2$), using salt solutions mimicking sulfate-rich waste streams ($0.01\text{--}0.3 \text{ M Na}_2\text{SO}_4$), was evaluated. An open circuit voltage (OCV) of 12.3 V, a maximum power density of $0.22 \text{ W/m}^2\text{MP}$ (MP: membrane pair) and internal area resistance of $43.2 \Omega\text{cm}^2/\text{cell}$ were recorded by using $0.01 \text{ M}/0.3 \text{ M Na}_2\text{SO}_4$ solutions at 35°C . The APWEL stack (6 cells, active area: $5 \times 5 \text{ cm}^2$), equipped with Ni foam electrodes and heterogeneous anion-selective membranes, was tested with varying concentrations of liquid electrolyte ($0.85\text{--}2.5 \text{ M KOH}$) and varying temperatures ($28\text{--}48^\circ\text{C}$). The APWEL stack attained a maximum current density of 110 mA/m^2 at 1.85 V/cell (i.e. 11 V per stack), 2.5 M KOH and 48°C . Under these conditions, the integrated system exhibited a maximum hydrogen production rate of $50 \text{ cm}^3/\text{h}\cdot\text{cm}^2$. This study opens up a new perspective on renewable hydrogen production fuelled by non-intermittent SGP from SO_4^{2-} -rich industrial effluents.

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1. Introduction

Hydrogen is a clean and versatile energy carrier for the future. It can be produced from water, natural gas, biomass and various other sources. Over the last decade, hydrogen production using water electrolysis has found acceptability due to the simplicity of this technology and the possibility to produce hydrogen of high purity. Moreover, water electrolysis is a flexible process driven by renewable energy resources, such as solar and wind power. More recently, the use of salinity gradient power (SGP) generated by reverse electrodialysis (RED) as a non-intermittent power source to fuel water electrolyzers has also been explored as an interesting alternative for renewable hydrogen production (Tufa et al., 2016, 2017). In RED, cation-exchange membranes (CEMs) and anion-exchange membranes (AEMs) are alternately aligned to create a low-concentration compartment (LCC) and a high-concentration

compartment (HCC) which are fed with solutions of low and high concentrations, respectively. A scheme of RED is shown in Fig. 1. The transport of ions occurs through ion-exchange membranes from HCC to LCC solutions driven by the electrochemical potential difference. Electricity is generated by a redox reaction evolving over electrodes placed at the ends of the membrane pile. RED technology is mostly investigated at lab-scale (Farrell et al., 2017), while studies of large-scale RED systems for SGP generation are relatively rare. A more recent demonstration of a pilot-scale RED system involved the testing of a stack equipped with 125 cell pairs ($44 \times 44 \text{ cm}^2$) and using brine and brackish water from salt works (Tedesco et al., 2016; Tufa et al., 2018).

Alkaline water electrolysis represents a mature process for hydrogen production combining the advantages of robustness and relatively low capital and operating costs. However, it has several limitations, such as inadequately optimized separator, low process efficiency and lack of suitability for intermittent power operations (Chanda et al., 2015). The latest development in alkaline water electrolysis is alkaline polymer electrolyte water electrolysis

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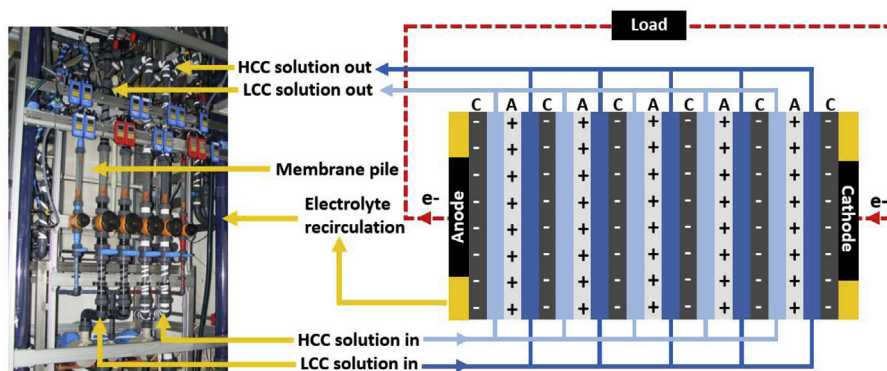


Fig. 1. Picture and scheme of the pilot-scale RED set-up (HCC: high-concentration compartment; LCC: low-concentration compartment. C: Cation-exchange membrane, A: Anion-exchange membrane. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(APWEL), which employs a solid polymer electrolyte based on AEMs (Hnát et al., 2011; Xiao et al., 2012). Such a design has the advantage of low gas cross-over, higher flexibility, and suitability for scale-up and operability at high pressure, allowing a simplified system with lower costs. In fact, most APWEL studies focus on single-cell designs rather than multiple-cell designs; the latter allow a better understanding of system performance on a large scale. Therefore, the present study uses scaled-up RED and APWEL systems in contrast to the set-ups used in most of the previous studies.

Industrial waste streams discharged into the ocean have SGP potential of up to 18 GW (Logan and Elimelech, 2012). However, the performance of RED using waste streams from various sources (Dil et al., 2017a; Dil et al., 2017b; Luo et al., 2017; Mehrabi and Alipanahpour Dil, 2017) other than NaCl-based salt solutions is a poorly investigated topic. Recent attempts have focused on the application of hybrid RED/electrodialysis systems for power generation using phenol-containing wastewaters (Luo et al., 2017), and wastewater from a fish canning factory and a sewage treatment plant (Di Salvo et al., 2017). Meanwhile, hydrogen production from waste resources is a rapidly growing field. In particular, biological hydrogen production from industrial wastewaters containing organic matter is regarded as a promising strategy for renewable hydrogen production; however, the drawback of this process is the low yield (Hatzell et al., 2014). On the other hand, given an electrolyzer's energy consumption of 53–70 kWh/kg (Levene et al., 2007), efficient exploitation of SGP produced from waste streams to fuel water electrolyzers would produce an annual yield of up to 3 M tons of hydrogen. Therefore, indirect production of hydrogen from industrial waste streams by water electrolyzers may be a viable alternative to other renewable hydrogen technologies.

Most industrial waste streams are rich in SO_4^{2-} which can be converted into renewable energy and hydrogen. SO_4^{2-} -rich waste streams are usually obtained from salt lake brines, mining processes and industrial waste streams from sewage treatment plants, tanneries and rechargeable battery manufacturing processes. In most cases, industrial wastewaters contain sulfate concentrations from 1 to 40 g/l (0.01–0.4 M) (Siles et al., 2010), which is far above the permitted limit (0.75–1.5 g/L) of discharge to surface water bodies (Act, 2003). A high content of SO_4^{2-} brine with concentrations of up to 1 M can be obtained from membrane treatment of industrial wastewater (Quist-Jensen et al., 2017). Therefore, SO_4^{2-} -rich industrial wastewaters can potentially be exploited for SGP generation and subsequent hydrogen production in the logic of the circular economy, waste-to-energy (WtE) and power-to-gas (P2G) (Götz et al., 2016).

To our knowledge, no attempt has been made to produce SGP

from SO_4^{2-} -rich industrial effluents with subsequent use as fuel for hydrogen production by water electrolysis. In the present study, a novel approach is investigated, combining SGP RED and APWEL systems for hydrogen production driven by non-intermittent energy generated from SO_4^{2-} -rich industrial waste streams. The process is conceptually illustrated in Fig. 2. Two key objectives were set. Firstly, a pilot-scale RED unit was optimized for SGP generation using industrial waste streams: the performance of the RED unit was evaluated in terms of voltage and power density at varying flow velocity and temperature of waste streams. Next, a laboratory-scale APWEL system, scaled-up 6-fold compared to most cases studied using single-cell designs (Ju et al., 2018; Tufa et al., 2016), was tested for potential hydrogen production driven by SGP: the hydrogen production rate was evaluated at varying electrolyte concentrations and temperatures to identify optimal operating conditions. The ultimate goal was to demonstrate the possibility of converting the electrochemical potential of industrial waste streams into clean energy and hydrogen by an integrated RED-APWEL energy system.

2. Materials and methods

2.1. Pilot-scale reverse electro dialysis stack

A commercial pilot-scale EDR-III/500/0.8 unit (MEGA a.s., Czech Republic) was adapted to a RED configuration in co-flow mode. A picture of the pilot-scale RED used in the present study is shown in Fig. 1. The stack consisted of 200 cell pairs with alternatively aligned

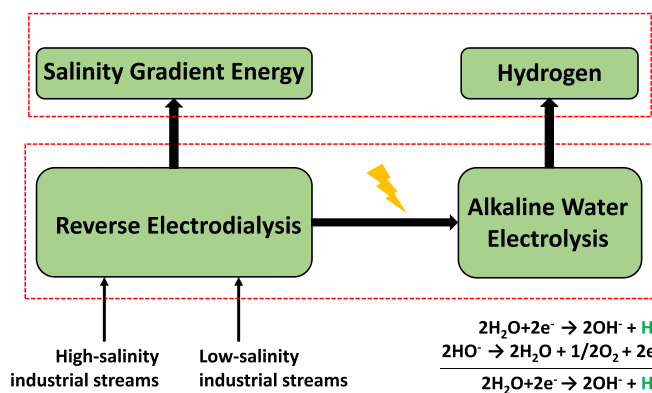


Fig. 2. Conceptual illustration of renewable hydrogen production by an integrated reverse electro dialysis and alkaline polymer electrolyte water electrolysis energy system.

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