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Using Salinity Gradients as an Energy Source to Produce Hydrogen

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Abstract

Only renewable energy sources should be used to produce "green" hydrogen. Salinity gradient energy, which can be obtained by reversibly mixing solutions, offers several advantages over other renewable energy sources. Salinity gradient energy sources have high potential and energy density, and are stable. Using natural solutions has minimal adverse environmental impact. Membrane methods for converting salinity gradient energy have received the most development. Pilot industrial plants using osmotic semipermeable membranes and ion-selective membranes have been created. However, the technical characteristics of currently produced membranes and their high cost make salinity gradient energy uncompetitive compared to other renewable energy sources. Direct conversion of salinity gradient energy into electrical energy is possible through reverse electrodialysis in an electrodialysis battery consisting of alternating anion- and cation-exchange membranes. The directed movement of ions through ion-selective membranes creates a potential difference across the electrodes located in the outer compartments of the battery. Voltage is applied from the electrodes of the electrodialysis battery to an external load. When inert electrodes are used, electrolysis by the solution occurs on them, lowering the battery's potential. The products of electrolysis can be utilized. Thus, the hydrogen produced during electrolysis serves as a secondary energy source. Its use allows for greater energy recovery by converting the energy of salinity gradients, increasing efficiency.

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Introduction

Salinity gradient energy (SGE) refers to the energy that can be converted into other forms of energy through the reversible mixing of solutions with different concentrations. The processes involved in converting this energy are the reverse of those used in water desalination and demineralization. River estuaries are considered a primary source of salinity gradient energy. Energy can also be obtained through the reversible mixing of concentrated brines from highly saline bodies of water, underground deposits, or industrial waste with fresh or seawater. The global potential of SGE is very high. The energy density of SGE sources is higher than that of other energy sources. The energy that can be obtained by mixing river water and seawater is equivalent to that produced by a dam 240 meters high at the river's mouth, since the osmotic pressure of seawater with a salinity of 35‰ is 24 atmospheres. The osmotic pressure of concentrated brines is even higher; for instance, the osmotic pressure of Dead Sea brine is 500 atmospheres. Unlike other renewable energy sources, SGE generation is relatively stable, as the solutions can be stored and their flow rates regulated. SGE conversion plants have a minimal environmental impact. The solutions resulting from the mixing of seawater and fresh water within these plants are indistinguishable from seawater diluted by river water under natural conditions. When using brines from hypersaline bodies of water or underground deposits, the high-concentration discharge solutions can be returned to the original brine source.

A drawback of using salinity gradient energy (SGE) is the slow rate of the reversible solution-mixing processes, which necessitates large surface areas for the mixing to occur. These interfaces can be the boundaries separating different phases of the solutions, such as liquid-gas or liquid-ice. Materials that expand and contract through the periodic adsorption and desorption of solutions on their surfaces may also be employed; such expansion and contraction generates mechanical energy. Methods for converting SGE using membranes have seen the most significant development, leading to the creation of pilot-scale industrial installations based on these techniques.

Osmotic semipermeable membranes allow water molecules to pass through but do not allow salts to pass through. The direction of this transfer depends on the ratio of the difference in osmotic and hydraulic pressures of the solutions. In the method proposed by Sidney Loeb [1]-known as pressure-retarded osmosis, water passes through an osmotic semipermeable membrane from a solution with low concentration and pressure into a solution with high concentration and pressure. This process occurs if the difference in hydraulic pressures between the solutions is lower than the difference in osmotic pressures. Energy is generated when this additional amount of water is passed through a hydraulic turbine or hydraulic motor.

Ion-exchange membranes are membranes that allow ions of a single charge to pass through them: cation-exchange membranes allow cations to pass, anion-exchange membranes allow anions to pass, and they do not allow water to pass. An electrodialysis cell consists of a stack of alternating cation- and anion-exchange membranes sandwiched between electrodes. Such electrodialysis cells are used for water desalination and concentration of solutions when voltage is applied to the electrodes. However, if solutions of different concentrations are supplied to the alternating chambers formed by pairs of membranes, ions will pass from the chambers with concentrated solutions to the adjacent chambers with diluted solutions: cations through cation-exchange membranes, anions through anion-exchange membranes. This directed ion transfer leads to the emergence of a potential difference at the electrodes. The direct conversion of SGE into electrical energy is carried out. In 1952, the German chemist Georg Manecke proposed using an electrodialysis battery for energy storage [2,3]. When an external voltage is applied to the electrodes of a battery filled with a solution, a concentration difference occurs between adjacent chambers. The reverse transfer of ions generates electricity. In 1954, the British scientist Pattl proposed using a similar battery to produce energy by mixing fresh and salt water [4]. The power of the battery he tested was 0.015 watts. This method of SGE conversion is called the reverse electrodialysis method.

A pilot-scale osmotic power plant utilizing river and sea water was launched by the Norwegian company Statkraft on November 24, 2009, in Tofte, Norway. The design capacity of this prototype is 10 kW [5]. However, on January 20, 2012, Statkraft halted work on the project due to the plant's lack of economic viability. The facility

achieved a power density of 1 W per square meter of membrane surface, whereas economic viability requires 5 W/m² at a membrane cost comparable to that of standard osmotic membranes. Although some experimental membranes achieve 10 W/m², no such membranes are currently available commercially [6]. Higher power output per unit of membrane area can be achieved by using concentrated brines instead of seawater.

A pilot electrodialysis plant with a capacity of 50 kW was commissioned on November 26, 2014, at the Afsluitdijk dam in the Netherlands. The plant is fed by fresh water from the part of the Zuiderzee bay cut off by the dam—which formed the freshwater Lake IJsselmeer—and by water from the North Sea. The facility demonstrated the project's technical feasibility under real-world conditions. Based on the plant's performance, the estimated Levelized Cost of Electricity (LCoE) is €0.11 per kWh for the initial 100 MW of capacity, with a projected drop to €0.05 per kWh after 2040 [7].

Currently, the use of salinity gradient energy (SGE) is not competitive with traditional or other renewable energy sources, primarily because osmotic and electrodialysis systems require large-surface-area membranes, yet the technical and economic performance of existing membranes remains inadequate. While large-scale production of membranes for water desalination and solution concentration is well-established, there is a specific need for membranes designed for energy generation. Furthermore, improvements are required in SGE conversion processes and the design of power generation systems. Producing chemical products alongside electricity would enhance the economic viability of SGE conversion plants.

Electrode Systems in Batteries for Reverse Electrodialysis

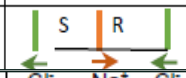
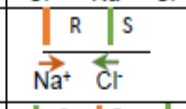
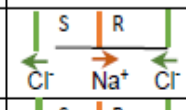
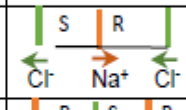
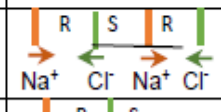
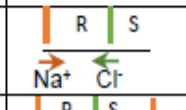
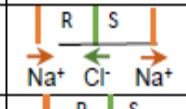
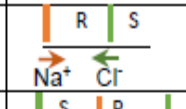
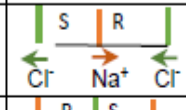
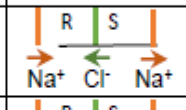
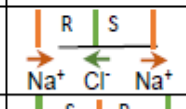
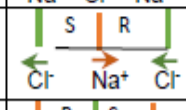
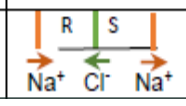
Electrode systems in batteries for reverse electrodialysis can be of four types [8].

1. Electrodes that participate in the reaction. Clampitt and Kiviat proposed a concentration cell using silver/silver-chloride electrodes to generate energy from the concentration difference between solutions [9]. Chloride ion exchange with the solutions occurred at the surface of these electrodes. To regenerate the electrodes and continue power generation, the positions of the solutions were swapped.
2. Inert electrodes that do not participate in the reaction itself but facilitate gas evolution—hydrogen at the cathode and chlorine or oxygen at the anode, depending on the solutions contacting the electrode. This reaction causes a drop in the voltage generated by the battery and consumes a portion of the generated electrical energy.
3. Inert electrodes used with a redox solution pair. A commonly used pair involves iron ions (Fe^{2+}/Fe^{3+}). At the anode, Fe^{2+} is oxidized to Fe^{3+}, while at the cathode, Fe^{3+} is reduced to Fe^{2+}. Solutions containing iron ions circulate between the electrode chambers. Since no chemical reaction occurs, power losses in the battery are minimal.
4. Use of capacitive electrodes. Anions are adsorbed on one electrode, and cations on the other. In a study by Vermaas et al., activated carbon on a Ti/Pt mesh was used for this purpose [10]. Once the electrodes become saturated, the process is reversed by switching the solution feed.
5. The electrode systems described in the literature, along with their anodic and cathodic reactions, are presented in Veerman's table [11], supplemented by us (Table 1).

Hydrogen Production

In recent years, interest in hydrogen energy has grown. However, hydrogen is a secondary energy carrier; its production requires the consumption of other energy resources—specifically traditional fuels such as coal, oil, and gas, or electricity from nuclear power plants and renewable energy sources. Currently, the cheapest method of producing hydrogen is from natural gas, but this process results in carbon dioxide emissions. Since hydrogen is chosen as a fuel for environmental reasons, its production should ideally be free of harmful emissions. Hydrogen produced via water electrolysis powered by renewable energy sources best meets this criterion. The most common types of electrolyzers include alkaline water electrolyzers and solid polymer electrolyte electrolyzers [12].

Table 1: Electrode systems described in the literature.

№	Anode, reaction	Anolyte	Repeating cells	Catholyte	Cathode, reaction	Author Overall reaction
1	Cu	$CuSO_4$		$CuSO_4$	Cu	Pattle (1954, 1955)
	$Cu \rightarrow Cu^{2+}+2e$				$Cu^{2+}+2e \rightarrow Cu$	No
2	Metal $2Cl^- \rightarrow Cl_2+2e$	Sea water		Sea water	Metal $2H_2O+2e \rightarrow H_2+2OH^-$	Weinstein/Leitz (1976) $2H_2O+2Cl^- \rightarrow H_2+2OH^-+Cl_2$
3	Ag/AgCl $Ag+Cl \rightarrow AgCl+e$	NaCl		NaCl	Ag/AgCl $AgCl+2e \rightarrow Ag+Cl^-$	Audinos (1983, 1992) No
4	Zn $Zn \rightarrow Zn^{2+}+2e$	$ZnCl_2$		$ZnCl_2$	Zn $Zn^{2+}+2e \rightarrow Zn$	Jagur-Grodzinski/ Kramer (1986) No
5	C/Pt $H_2O \rightarrow {}^1/_2O_2+2H^++2e$ и $2Cl^- \rightarrow Cl_2+2e$	NaCl		NaCl	C/Pt, air ${}^1/_2O_2+2H^++2e \rightarrow H_2O$	Jagur-Grodzinski/ Kramer (1986), anolyte and catholyte are separate
6	Ti/Pt $2Cl^- \rightarrow Cl_2+2e$	Sea water		Sea water	Ti/Pt $2H_2O+2e \rightarrow H_2+2OH^-$	Knyazhev (1987) $2H_2O+2Cl^- \rightarrow H_2+2OH^-+Cl_2$
7	Ti/Pt $H_2O \rightarrow {}^1/_2O_2+2H^++2e$	Na_2SO_4		Na_2SO_4	Ti/Pt $2H_2O+2e \rightarrow H_2+2OH^-$	Turek (2007, 2008) $H_2O \rightarrow {}^1/_2O_2+H_2$
8	C $2Cl^- \rightarrow Cl_2+2e$	Sea water		Sea water	C $H_2O+2e \rightarrow H_2+2OH^-$	Suda (2007) $2H_2O+2Cl^- \rightarrow H_2+2OH^-+Cl_2$
9	Ti/RuIr $Fe^{2+} \rightarrow Fe^{3+}+e$	Fe^{2+}/Fe^{3+} NaCl/ HCl		Fe^{2+}/Fe^{3+} NaCl/HCl	Ti/RuIr $Fe^{3+}+e \rightarrow Fe^{2+}$	Veerman et al. (2010) No
10	Ti/RuIr $Fe(CN)_6^{4+} \rightarrow Fe(CN)_6^{3-}+e$	$Fe(CN)_6^{3-}/Fe(CN)_6^{4-}$ NaCl		$Fe(CN)_6^{3-}/Fe(CN)_6^{4-}$ NaCl	Ti/RuIr $Fe(CN)_6^{3-}+e \rightarrow Fe(CN)_6^{4-}$	Veerman et al. (2010) No
11	Ti/RuIr $2Cl^- \rightarrow Cl_2+2e$	NaCl		NaCl	Ti/RuIr $2H_2O+2e \rightarrow H_2+2OH^-$	Veerman et al. (2009) $2H_2O+2Cl^- \rightarrow H_2+2OH^-+Cl_2$
12	Graphite $Fe^{2+} \rightarrow Fe^{3+}+e$	Fe^{2+}/Fe^{3+} NaCl/ HCl		Fe^{2+}/Fe^{3+} NaCl/HCl	Graphite $Fe^{3+}+e \rightarrow Fe^{2+}$	Veerman et al. (2010) No
13	Graphite $Fe(CN)_6^{4+} \rightarrow Fe(CN)_6^{3-}+e$	$Fe(CN)_6^{3-}/Fe(CN)_6^{4-}$ NaCl		$Fe(CN)_6^{3-}/Fe(CN)_6^{4-}$ NaCl	Graphite $Fe(CN)_6^{3-}+e \rightarrow Fe(CN)_6^{4-}$	Veerman et al. (2010) No

Symbols in the table:

- anion-exchange membrane

- cation-exchange membrane

R- River Water

Osmotic and reverse electrodialysis systems for salinity gradient energy conversion can be used to power electrolyzers. For instance, Zhao investigated a multi-stage reverse electrodialysis system powering a solid polymer electrolyte electrolyzer [13].

However, water electrolysis can occur within the electrodialysis stack itself. As noted above, during reverse electrodialysis with inert electrodes, gas evolution takes place at the electrodes; this consumes a portion of the energy and reduces the stack's electromotive force (EMF). If sodium chloride solutions or seawater are used, hydrogen and chlorine gases are evolved at the electrodes. Such a stack is illustrated in Figure 1; it can be used to produce these gases, thereby yielding a useful outcome instead of incurring energy losses. An analysis of the

technical and economic characteristics of such a reverse electrodialysis stack is presented in [14]. It was found that when the system is fed with seawater and fresh water, current densities are significantly lower than those in industrial electrolyzers. This issue can be partially mitigated by using a concentrated solution and seawater. Nevertheless, to achieve competitiveness with currently employed technologies, it is necessary to produce thinner and highly selective membranes.

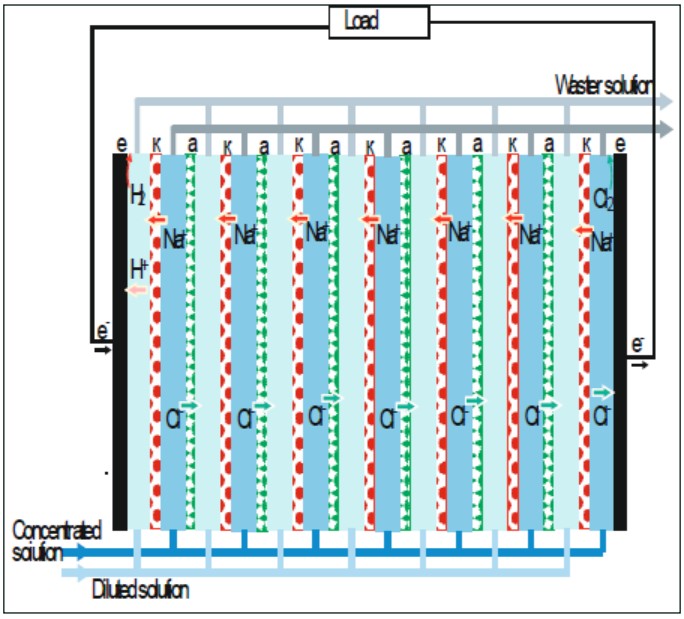


Figure 1: Electrodialysis battery. Reverse electrodialysis and electrolysis with sodium chloride solutions. The figure shows: a and k – anion- and cation-exchange membranes, respectively, e – electrodes.

But if chlorine is not needed for further use, it cannot be discharged into the environment, and its disposal is difficult, then special solutions are supplied to the electrode chambers. Electrolysis of these solutions occurs. Figure 2 illustrates a configuration involving the recirculation of sodium hydroxide solution between the electrode chambers. The solutions are recirculated by peristaltic pumps to maintain a stable concentration. Which will reduce the leakage of electric current between the electrode chambers through the supply channels of electrode solutions. Hydrogen is produced in the same way as in an aqueous-alkaline electrolyzer. Oxygen can be accumulated or released into the environment. If the electrodes are simply short-circuited, the entire SGE, minus losses in the battery, will be consumed for gas production.

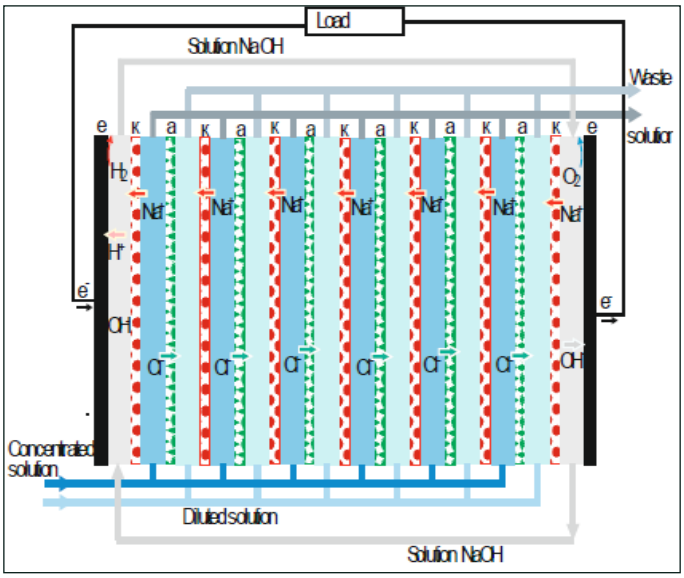


Figure 2: Electrodialysis battery for electrolysis of sodium hydroxide solutions. The figure shows: a and k – anion- and cation-exchange membranes, respectively, e – electrodes.

A study on hydrogen production via electrolysis in a reverse electrodialysis unit, along with a techno-economic analysis, was conducted in [15]. The battery consisted of 20 membrane cells. Titanium mesh substrates coated with platinum and iridium oxides served as the anode and cathode. A 0.5 M aqueous HCl solution was used as the catholyte, while a 0.5 M aqueous NaOH solution served as the anolyte. Tests were performed using a dilute NaCl solution with a concentration of 1 g/L (river water) and two different concentrate concentrations: 35 g/L NaCl (seawater) and 300 g/L NaCl. To increase the current and, consequently, the hydrogen production rate, operating modes involving connection to an external voltage source were tested.

Within the scope of this work, higher current densities were achieved than in other studies using similar technology, and consequently, higher hydrogen productivity. However, compared to modern electrolyzers, the current density is lower. Nevertheless, despite the low productivity, a minimum price of 1.2 euros per kg of hydrogen can be achieved due to the energy of salinity gradients when using concentrated brine. This figure can be improved by optimizing the processes and the installation.

Testing of an Electrodialysis Unit and Analysis of its Operation

We previously created an experimental electrodialysis unit based on a water desalination unit (Fig. 3) [16, 17]. It was tested under real-world conditions using fresh water from an artesian well and seawater from a bay as solutions. The electrodialysis battery was assembled from 22 heterogeneous anion-exchange membranes MA-40 and 21 heterogeneous cation-exchange membranes MK-40, sandwiched between electrodes made of platinum-coated titanium. The area of a single membrane was 0.165 m². The membranes are separated by polypropylene working and rotating frames; a turbulator made of corrugated perforated vinyl plastic is placed inside the frames. Diluted solutions were prepared in a 610-liter tank with fresh water by adding seawater or by supplying seawater directly into the channel with fresh water. As the concentration of the diluted solution increased, the battery's EMF and internal resistance decreased.

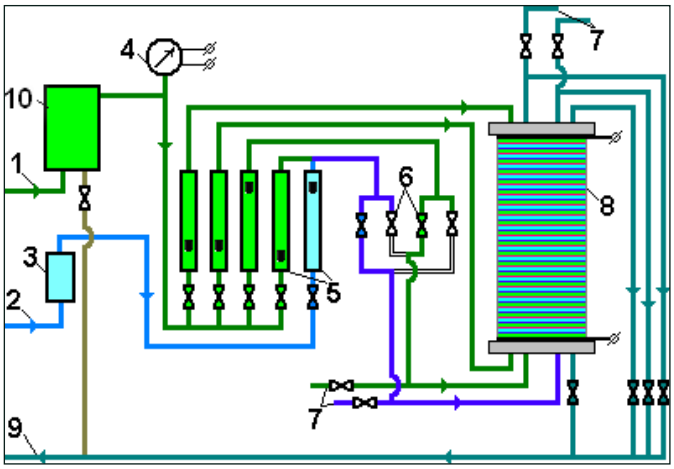


Figure 3: Experimental electrodialysis unit: 1 – seawater supply pipeline, 2 – fresh water supply pipeline. 3 - filter, 4 – electric contact pressure gauge, 5 - rotameters, 6 – valves, 7 – taps for sampling solutions, 8 – electrodialysis battery, 9 - pipelines for draining solutions.

Three years of testing the setup confirmed the feasibility of using reverse electrodialysis for energy production, but the resulting performance parameters were quite low. This was due to the high surface resistance of the membranes used (approximately 10 Ω/cm²) and the large 2 mm gap between them, which resulted in high resistance within the solution compartments. In the electrode chambers of the battery, electrolysis of seawater occurred on the electrodes, as insoluble titanium platinum electrodes were used. This resulted in a decrease in the battery's electromotive force. However, the hydrogen obtained during electrolysis can be used to generate energy. The total power of the battery, taking into account the electrical power and the power from the obtained hydrogen, will be higher. Let us estimate the total power that can be obtained from this battery.

The hydrogen consumption rate J_H (in kg/s) released at the cathode can be obtained from the equation:

$$J_H = \frac{Im}{nF}, \qquad (1)$$

where I is the electric current (A), m is the molar mass of hydrogen, m = 2 g/mol= 0.002 kg/mol), n is the number of electrons in the electrode reaction (n = 2), and F is the Faraday constant (F = 96485.332 C/mol).

Electric power dissipated in the load resistance:

$$W_e = I^2 R_L = (\frac{V_b}{R_b + R_L})^2 R_L \,, \quad (2)$$

where *RL* is the load resistance (ohms) and *Rb* is the internal resistance of the battery:

$$W_H = QJ_H = \frac{QIm}{nF} \,, \quad (3)$$

where *Q* the specific heat of combustion of hydrogen is 140,000 kJ/kg.

Total battery power:

$$W_g = W_e + W_H = I(IR_L + \frac{Qm}{nF}), \quad (4)$$

The dependencies of the electrical, hydrogen, and total power of the battery on the load resistance for one of the test modes, calculated using equation (4), are shown in Fig. 4.

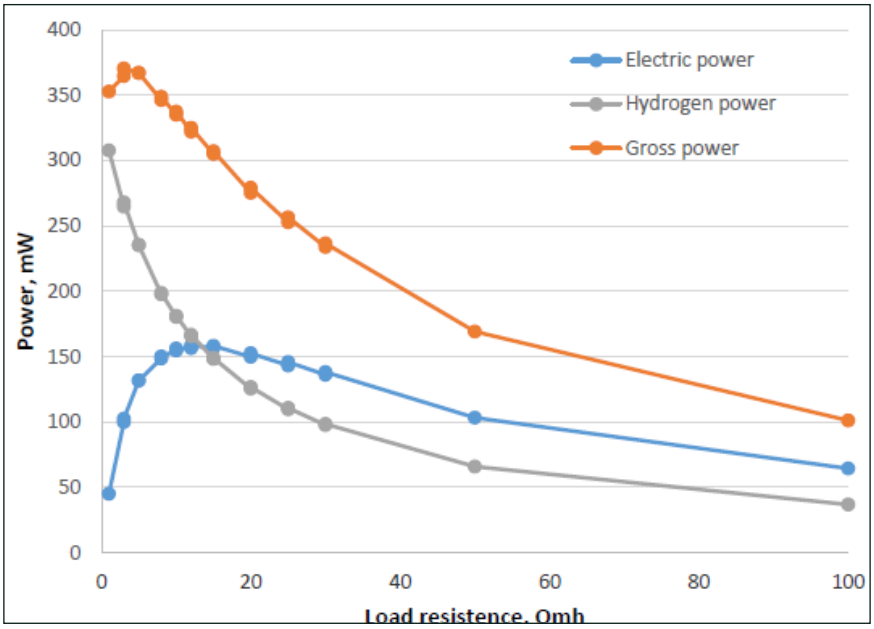


Figure 4: Dependence of the battery's electrical, hydrogen, and total power on load resistance; seawater salinity 33‰, diluted solution salinity 0.34‰.

Based on the relationships shown in Fig. 4, it is evident that hydrogen power makes a significant contribution to the total power of the battery.
Electric current in the circuit according to Ohm's law:

$$I = \frac{V_b}{R_L + R_b} \,, \quad (5)$$

Total battery power from equations (4) and (5):

$$W_g = (\frac{V_b}{R_b + R_L})^2 R_L + \frac{V_b Qm}{nF(R_L + R_b)} \,, \quad (6)$$

Differentiating this equation with respect to the load resistance, equating the derivative to zero, and substituting the value of the constants, we find that the total power will be maximum at:

$$R_L = R_b \frac{1-\frac{Qm}{nFV_b}}{1+\frac{Qm}{nFV_b}} = R_b \frac{1-\frac{1.45}{V_b}}{1+\frac{1.45}{V_b}} \quad (7)$$

The electrical power dissipated across the load resistance is maximized when $RL = Rb$ [16]. The battery EMF increases linearly with the number of membrane pairs in the battery. Equation (7) shows that as the battery EMF increases, the load resistance value optimal for total power output tends toward the battery's internal resistance, while Equation (6) indicates that the share of hydrogen-derived power in the total battery power output decreases. Therefore, to increase hydrogen production, it is advisable to subdivide a large stack of membranes using intermediate electrodes, with hydrogen evolving on one side and chlorine or oxygen on the other.

Conclusion

Salinity gradient energy sources offer high potential and energy density compared to other renewable energy sources. However, converting this energy requires large surface areas of expensive membranes, which currently renders the technology uncompetitive against other renewables. Producing membranes with superior technical and economic characteristics, alongside improvements to conversion systems and processes, will help overcome this challenge. Furthermore, utilizing reverse electrodialysis batteries to produce hydrogen in addition to electricity will enhance the overall efficiency of salinity gradient energy conversion.

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