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Opportunities and challenges for thermally driven hydrogen production using reverse electro dialysis system

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HIGHLIGHTS

- First study of levelized cost of hydrogen by ammonium bicarbonate reverse electro dialysis.
- For economic feasibility, max. membrane cost 20 €/m² and min. membrane life 7 years.
- Waste heat required varies from 480 to 105 kWh/kg_{H₂}.
- In *Present scenario* membrane replacement cost dominates with 44% of total expenses.
- In *Future scenario* cost of heating dictates with 58% of total expenses.

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ABSTRACT

Ongoing and emerging renewable energy technologies mainly produce electric energy and intermittent power. As the energy economy relies on banking energy, there is a rising need for chemically stored energy. We propose heat driven reverse electro dialysis (RED) technology with ammonium bicarbonate (AmB) as salt for producing hydrogen. The study provides the authors' perspective on the commercial feasibility of AmB RED for low grade waste heat (333 K–413 K) to electricity conversion system. This is to our best of knowledge the only existing study to evaluate levelized cost of energy of a RED system for hydrogen production. The economic assessment includes a parametric study, and a scenario analysis of AmB RED system for hydrogen production. The impact of various parameters including membrane cost, membrane lifetime, cost of heating, inter-membrane distance and residence time are studied. The results from the economic study suggests, RED system with membrane cost less than 2.86 €/m², membrane life more than 7 years and a production rate of 1.19 mol/m²/h or more are necessary for RED to be economically competitive with the current renewable technologies for hydrogen production. Further, salt solubility, residence time and inter-membrane distance were found to have impact on levelized cost of hydrogen, LCH. In the present state, use of ammonium bicarbonate in RED system for hydrogen production is uneconomical. This may be attributed to high membrane cost, low (0.72 mol/m²/h) hydrogen production rate and large (1,281,436 m²) membrane area requirements. There are three scenarios presented the *present scenario*, *market scenario* and *future scenario*. From the scenario analysis, it is clear that membrane cost and membrane life in *present scenario* controls the levelized cost of hydrogen. In *market scenario* and *future scenario* the hydrogen production rate (which depends on membrane properties, inter-

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membrane distance etc.), the cost of regeneration system and the cost of heating controls the levelized cost of hydrogen. For a thermally driven RED system to be economically feasible, the membrane cost not more than 20 €/m²; hydrogen production rate of 3.7 mol/m²/h or higher and cost of heating not more than 0.03 €/kWh for low grade waste heat to hydrogen production.

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Introduction

Many large-scale industries for e.g. manufacturing applications dissipate heat at relatively low temperature. In general, this heat is in overabundance, and often cannot be reintegrated entirely on-site or used for district heating. Thus, this heat is rejected to the surroundings [1]. Globally, waste heat in the industrial sector accounts for more than 40% of total energy use, meaning almost half of the energy consumed is wasted as heat to the environment [2]. In Norway, the waste heat potential was estimated to be 19 TWh/a in 2008 [3]. There are various sources of industrial waste heat as shown in the Table 1. Though there is a huge potential for waste heat recovery, the amount of heat varies with temperature as shown in Fig. 1.

The low-grade waste heat is an untapped energy resource generated in industrial plants due to lack of efficient and cost-effective recovery methods. About 300 thousand USD savings was accounted for carbon dioxide equivalent (CO_{2,eq}) emissions from waste heat from industries in UK [5]. In order to mitigate climate change, EU policy 2030 recommended a target to reduce GHG emissions by 40% and improve energy efficiency by 27% in the transportation and industrial sectors. Further, the use of hydrogen in the transportation sector was identified to be an alternative solution. We propose an energy efficient ammonium bicarbonate reverse electro dialysis (AmB-RED) system that utilizes the low grade waste heat (333 K–413 K) to produce hydrogen. Reverse electro dialysis (RED) stack is an electrochemical device that converts chemical energy into electrical energy using concentration gradient across an ion selective membrane. RED is one of the few renewable technologies which is capable of directly producing hydrogen from waste heat.

Cost and some technicalities are major barriers to market penetration of RED technology [6]. Literature survey indicates lack of studies evaluating challenges in market diffusion. The

economic studies report that membrane accounts for almost 40–80% of total investment cost [7–10]. The literature study till date to the best of the authors' knowledge shows quite an uncertainty in estimated membranes costs ranging from 2 €/m² to 640 €/m² as shown in Table 2. Moreover, the membrane life was assumed to be in the range of 7–20 years. While some are optimistic assumptions and rest suppositions, an engineered estimate of membrane cost for levelized cost of energy (LCOE) for RED needs to be addressed.

In this article, we evaluate for the first time the economic feasibility of AmB RED system for hydrogen production with low grade waste heat of (313 K–413 K) as a source. A model with technical and economic aspects was developed to evaluate levelized cost of hydrogen (LCH). The model can be applied to decentralized hydrogen refuelling station. Critical parameters were identified, and their sensitivity was evaluated. We estimate the membrane price for the technology to be economically viable for hydrogen production. In this parametric study we account for five parameters. In the scenario analysis, we propose three scenarios estimating the values of the financial and performance indicators for present state, economically competitive to other clean energy technologies and an optimistic future scenario.

System description

A typical RED cell converts directly the electrochemical potential of salt into electrical energy. The electrochemical potential in the form of concentration gradient across an ion exchange membrane provides the driving force for ions to migrate from the high concentrate channel (HC) to the low concentrate channel (LC). By using alternate permselective membranes, the flow of ions is controlled. This ionic current is converted into electrical current at the electrodes by redox reactions. The ammonium bicarbonate based RED system is divided into a RED stack and a regeneration system as shown in Fig. 2. Similar to a typical reverse electro dialysis system, a concentrate and a dilute solution of ammonium bicarbonate are introduced into the cell in the respective channels. The concentration difference across a cation exchange membrane causes ammonium ions to flow from HC to LC. Similarly, bicarbonate ions flow from the HC to the LC across anion exchange membrane. Mixing of these solutions decreases the concentration of the HC effluent, while the LC increases. This LC effluent is introduced into the regeneration system. The regeneration system includes a stripping column and an absorption column. The low grade waste heat, at 313 K and

Table 1 – Industrial waste heat potential (TWh/a) from different sectors at various temperature range (their share [4]).

Sector	L.T < 373 K	M.T 373–572 K	H.T > 572 K
World	63%	16%	21%
Transportation	7.9 (46%)	–	9.3 (54%)
Industry	3.7 (42%)	1.8 (20%)	12.1 (38%)
Electricity	26.2 (88%)	3.6 (12%)	–
Residential	3.0 (36%)	5.4 (64%)	–
Commercial	2.3 (59%)	0.2 (5%)	1.4 (36%)

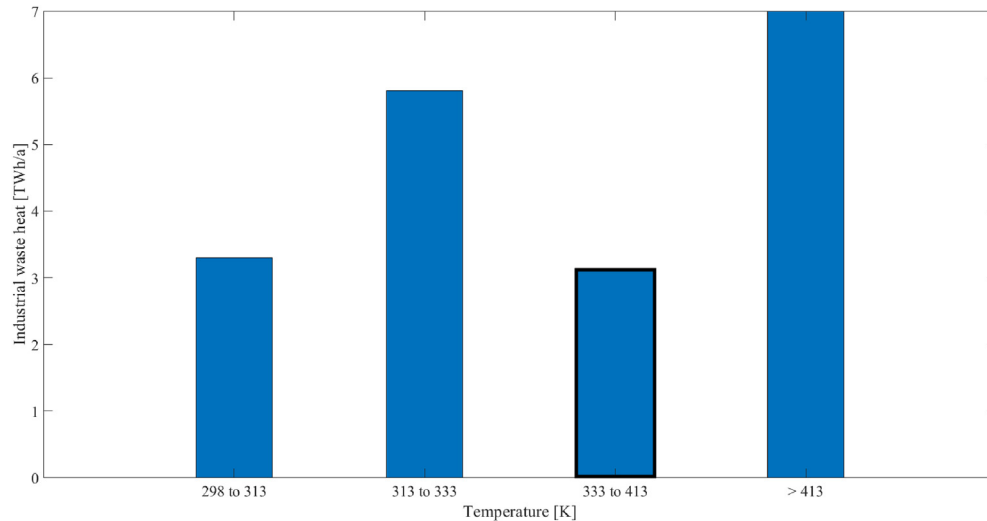


Fig. 1 – Industrial waste heat [TWh/a] in Norway as function of temperature [K].

Table 2 – Summary of previous economic studies on RED for electricity production.

Author	Year	Capacity MW	Lifetime years	C_{mem} €/m ²	t_{mem} years	P_{op} W/m ²	LCOE €/kWh
Turek [11]	2007	–	–	100 ^{a,b}	10	0.46	6.79 ^a
Post [9]	2010	200	40	2	7	2	0.08
Daniilidis [10]	2014	200	25	50	7	2.2	0.72
Weiner [7]	2015	–	20	750 ^{a,b}	–	1.2	6.33 ^a
Bevacqua [12]	2017	100	20	50	20	4.78	0.3

^a Currency in \$.

^b Cost includes membranes, gasket, spacers, electrodes, end plate.

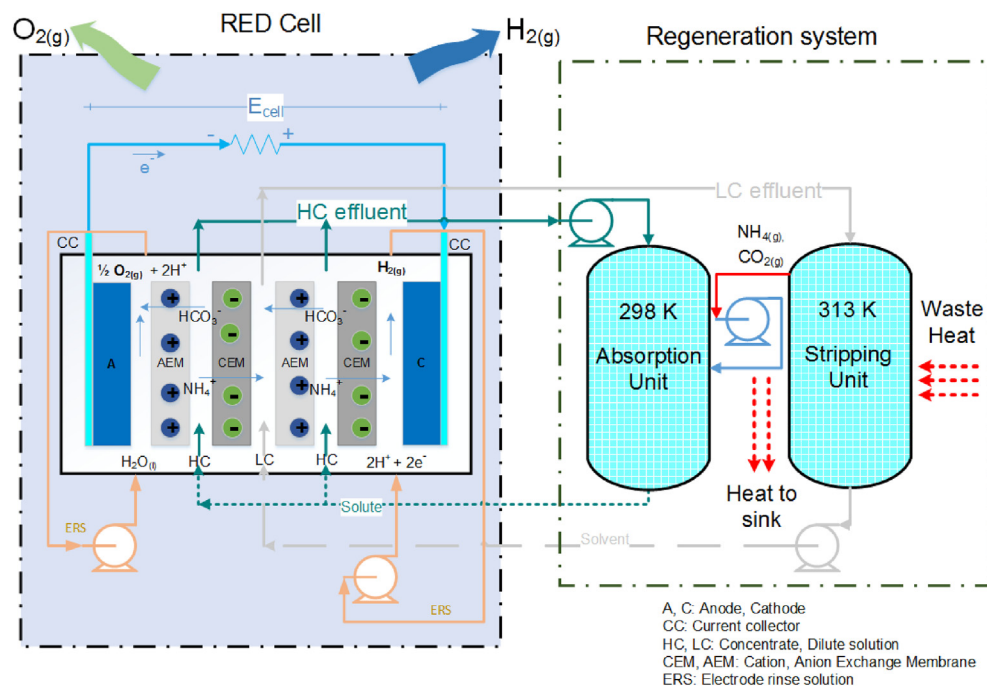
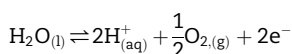


Fig. 2 – Schematic of Reverse Electro-Dialysis (RED) system based on ammonium bicarbonate salt with regeneration system for hydrogen production (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article).

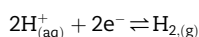
1 atm, heats the LC effluent solution in the stripping column. This heat strips the solution into ammonia and carbon dioxide gases. The stripped gases absorb into the HC effluent in the absorption column at 298 K. The outlets of absorption and stripping column are reintroduced into the RED cell by mixing them with HC and LC inlets.

A typical RED stack consists of $K_4Fe(CN)_6/K_3Fe(CN)_6$ as electrode rinse solution (ERS) with inert electrodes such as Ti coated in Ir–Ru–O [13]. The alternative cost efficient carbon electrode with $FeCl_2/FeCl_3$ as ERS are not used widely due to low conductivity [13,14]. In AmB-RED system, we propose use of 1 M KOH as ERS with Nickel/nickel mesh as electrode for hydrogen evolution reaction (HER) and oxygen evolution reaction (OER) similar to alkaline water electrolyzers, respectively. The redox reactions at the respective electrodes are as follows.

Reactions at Anode:



Reactions at Cathode:



Ammonium bicarbonate has low decomposition temperature of around 60 C at 1 atm that makes it an ideal candidate for low grade waste heat recovery [12,15–18]. Thermolytic salts such as ammonium bicarbonate can access control over solution concentrations. Increase in the salt concentrations increases the osmotic pressure differences which results in higher energy production. An ammonium bicarbonate based RED system can produce osmotic pressure heads equivalent to 380–510 m for higher concentrations (1–1.5 M), which is three to four times higher when compared to seawater–riverwater RED. In addition, use of ammonium bicarbonate salt in RED system avoids the geographic constraints such as coastal regions that are essential for sea/river based RED systems. This eliminates the need for energy intensive pretreatment processes. Further, high solubility in water, and relatively low molecular weight, makes it suitable for RED application. Ammonium bicarbonate production in terms of environmental concern, $CO_{2,eq}$ emissions, production of AmB can be net carbon negative [19]. Moreover, carbon capture can be realized in the oxyfuel process, where oxygen and steam are burnt with natural gas. Recovering the waste heat from such processes, not only as hydrogen but additional oxygen too makes AmB RED to hydrogen a carbon neutral economy. AmB has an additional advantage compared to NaCl solutions because its use reduces electrode overpotentials for both hydrogen evolution reaction (HER) and oxygen reduction reaction (ORR) [20]. Thus, AmB may make these reactions more favourable for energy production than previously achieved using river water and seawater in RED systems.

Methodology

Input data

The economic assessment was performed for a decentralized hydrogen production plant (refuelling station) of capacity 1500 kg/day [21]. Depending on the conversion efficiency, the

waste heat required for such capacity varies from 1 MW to 8 MW. List of input parameters for techno-economic modeling are shown in the Table 3. The dimensions of RED stack are 1 m × 1 m × 2 m (l × w × h) per 10 kW (electric power) [10]. The technical parameters including model equations for membrane resistance and theoretical waste heat for ammonium bicarbonate decomposition are referred from literature [12,22]. It was difficult to extract market price of membranes, due to commercial sensitivity. Hence, the cost estimates were assessed based on the information from the literature (see Table 4).

Assumptions

In this method, inflation rate was moderate and similar to the interest rate. All economic assumptions were based on values taken from literature for European market. In the economic analysis, fuel, insurance and emissions cost, were considered. Ion exchange membranes (IEM) for RED specific applications follows the learning curve and economic trend as Nafion. The levelized cost of hydrogen (LCH) of 3.59 €/kg was considered for market scenario [21]. The operational cost associated to heating including equipment cost heat exchanger was assumed to be 0.01 €/kWh [23].

Table 3 – Input technical and economic data for proposed model.

Parameter	Symbols	Value	Unit
Concentrate solution conc.	c_{hc}	2.0	M
Dilute solution conc.	c_{lc}	0.06	M
Ideal gas constant	R	8.314	J/K/mol
Temperature	T	293	K
Faraday's constant	F	96,485	C/mol
Permselectivity of CEM/AEM	α	0.85	
Diffusivity coefficient (mem)	D_{AmB}	$2 \cdot 10^{-12}$	m^2/s
Inter-membrane distance	δ_{ch}	100	μm
Membrane thickness	δ_{mem}	125	μm
Number of membrane pair	N	10	
Residence time	t_{res}	1	s
Length of channel	l	0.1	m
Width of channel	w	0.1	m
Viscosity of water	μ	$9 \cdot 10^{-4}$	Pa-s
Porosity	ϵ	0.8	
Velocity of dilute sol.	v_{lc}	0.01	m/s
Membrane life time	t_{mem}	4	yr
Plant life time	t	20	yr
Operational hours per year	t_a	8000	h
Pump efficiency	η_{pump}	0.75	
Faradaic efficiency	η_F	0.95	
Production capacity	$m_{H_2}^{cap}$	1500	kg/day
Pumping system cost	C_{pump}	300	€/kW
Membrane cost	C_{mem}	150	€/m ²
Waste heat price	C_{heat}	0.01	€/kWh
Discount factor	r	0.06	
Infrastructure price	C_{infra}	1123	€/m ^{2a}
Regeneration system price	C_{regen}	3400	€/m ^{3/h} ^b
Labour cost	C_{lab}	20%	(% of CAPEX)
RED stack	$C_{RED_{stack}}$	same	€/m ²
		as C_{mem}	

^a Land area.

^b Vol. flow.

Table 4 – Effect of raw material cost and production volume on cost of membrane [28].

Membrane	Production rate m ² /a	C _{raw} \$/m ²	C _{prod} \$/m ²	Total cost \$/m ²	Thickness μm
Nafion	2800	–	–	576	183
	250,000	35	100	–	183
	28,000,000	–	–	62	183
	10,000	–	–	250	25
	600,000,000	–	–	2.8	25
SPEEK	250,000	4.5	50	–	100
NFM	10,000	1 + 0.05	10	–	200 + 25

System model

The system model was subdivided into Amb-RED system, pumping system and regeneration system. In Amb-RED system, electrochemical equations were used to estimate hydrogen produced. Pumping system model deals with hydrodynamics in the cell and related losses. Regeneration system was modelled based on simple mass balances and thermal duty assumed from Bevacqua et al. [12].

Amb-RED system

The objective was to estimate the hydrogen production, and membrane area required. At first, the input data as shown in Table 3 were initialized. Parameters such as conductivity and activity coefficients were estimated. On this basis, cell voltage, resistances of membranes and channels were estimated followed by stack resistance, and stack voltage. Using these values, short circuit current density, peak power current density, and peak power stack voltage were determined. Finally, the moles of hydrogen produced and membrane area required were estimated. The proposed system was mathematically modelled using equations as follows,

- Conductivity of solutions (k): The equivalent conductivity of ammonium bicarbonate depends on concentration and molar conductance which depends on temperature. The conductivity was calculated at constant temperature 293 K using Jones-Dole equation,

$$k = \Lambda \cdot c_i \quad (1)$$

where $\Lambda = \Lambda_0 - \frac{A_\Lambda \cdot \Lambda_i \cdot c_i^{1/2}}{1 + B_\Lambda \cdot c_i^{1/2}} - C_\Lambda \cdot c_i$. Here $A_\Lambda, B_\Lambda, C_\Lambda$ are model parameters used for fitting and referred from Bevacqua et al. [12], c_i is inlet concentration; Λ_0 molar equivalent conductivity of salt at infinite dilution. The conductivity is calculated in mS/cm for both feed solutions at 293 K.

- Activity coefficient of solutions (γ): The activity coefficients depend on molar salt concentration. The linear dependence of activity coefficients on concentration was estimated using ENTRL-RK thermodynamic package in Aspen Plus.

$$\gamma = g_1 \cdot c_i + g_2 \quad (2)$$

here c_i is expressed in mol/l, $g_1 = 0.1366$ l/mol and $g_2 = 1.0007$. These values were estimated at $T = 293$ K [12].

- Unit cell open circuit potential ($E_{u.c.}^{ocp}$): It is the membrane potential [V] for a cell pair (CEM, AEM) with no losses considered. The electrochemical potential difference across an IEM placed between two different concentration solutions can be described using modified Nernst equation,

$$E_{u.c.}^{ocp} = (\alpha_{cem} + \alpha_{aem}) \cdot \frac{R \cdot T}{F} \cdot \ln \left(\frac{\gamma_{hc} \cdot c_{hc}}{\gamma_{lc} \cdot c_{lc}} \right) \quad (3)$$

α is permselectivity of IEMs measured at concentration c_{hc} and c_{lc} at constant temperature 293 K for specific membrane. Here we assume same α for both membranes. F is Faraday constant. T is room temperature. R is ideal gas constant.

- Area specific membrane resistance ($R_{aem/cem}$): It is the ohmic resistance [Ωm^2] of the membranes when immersed in the solution. It is expressed as function of concentration [12].

$$R_{cem} = R_{aem} = r_1 \cdot (c_{lc})^{-0.236} \quad (4)$$

R_{cem}, R_{aem} are area specific membrane resistances for cation and anion exchange membrane, c_{lc} is inlet concentration of low concentrate solution. r_1 is fitting parameter [$\Omega m^2/M$]. It is estimated to be 0.0002 [$\Omega m^2/M$] for *present* and *market* scenario and 0.00004 [$\Omega m^2/M$] for *future* scenario [12]. As concentration lowers below 0.1 M, the contribution to resistance from low concentrate solution dominates.

- Channel ohmic resistance ($R_{lc/hc}$): The resistance (Ωm^2) due to the solution in the channel and spacer geometry. It depends on concentration, and is calculated using molar conductivity of the salt.

$$R_{lc} = \frac{\delta_{lc}}{\varepsilon \cdot k_{lc} \cdot c_{lc}} \quad R_{hc} = \frac{\delta_{hc}}{\varepsilon \cdot k_{hc} \cdot c_{hc}} \quad (5)$$

here δ_{ch} is intermembrane distance in μm , ε is the porosity of spacers, k_{lc} and k_{hc} are conductivities of feed solutions

- Unit cell resistance ($R_{u.c.}$): The cumulative sum of resistance of membranes and channels in a unit cell [Ωm^2].

$$R_{u.c.} = R_{cem} + R_{aem} + R_{lc} + R_{hc} \quad (6)$$

- Current density at peak power ($j_{u.c.}^{pp}$): The peak power current density occurs at potential half the open circuit potential. As the resistance remains constant the current density [A/m^2] at peak power is calculated using Ohm's law,

$$j_{u.c.}^{pp} = \frac{E_{u.c.}^{ocp}}{2 \cdot R_{cell}} \quad (7)$$

- Actual unit cell potential ($E_{u.c.}^{act}$): The potential across the RED unit cell drops due to ohmic resistances in the RED unit cell and is estimated as,

$$E_{u.c.}^{act} = E_{u.c.}^{ocp} - R_{u.c.} \cdot j_{u.c.}^{pp} \quad (8)$$

- Number of unit cells ($N_{u.c.}$): Alkaline electrolyzers operate above reversible potential 1.23 V. The anodic and cathodic overpotentials are estimated to be 300 mV at low current densities [24]. Hence, the required potential for water electrolysis is assumed to be 1.5 V. In order to meet this potential, unit cells are stacked together in series. The minimum number of unit cells to be stacked in series is calculated as,

$$N_{u.c.} = \frac{1.5}{E_{u.c.}^{act}} \quad (9)$$

- Stack open circuit potential (E_{stack}^{act}): The stack open circuit potential [V] drops upon connecting the RED device to external load. Thus the actual voltage across the terminals of the stack is given by,

$$E_{stack}^{act} = N \cdot E_{u.c.}^{act} \quad (10)$$

- RED stack resistance (R_{stack}): The total resistance including unit cells, N and electrodes in a stack.

$$R_{stack} = N \cdot R_{u.c.} + R_{ele} \quad (11)$$

here R_{ele} [Ωm^2] is resistance from electrodes in electrode rinse solution. This resistance value is assumed to be $0.01 \Omega m^2$ [12]. But, its contribution to the performance is not significant for large stacks.

- Hydrogen production rate (\dot{n}_{H_2}): The theoretical moles of hydrogen produced per unit time in the compartment with electrode-electrolyte rinse solution of RED stack.

$$\dot{n}_{H_2} = \frac{j_{u.c.}^{pp} \cdot 3600}{z \cdot F} \cdot \eta_F \quad (12)$$

here $z = 2$ is the ion valence per mole of hydrogen gas, η_F is Faradaic efficiency. η_F signifies that all current density generated is not utilized to make hydrogen, due to system related losses. In a RED system, the main source of loss in Faradaic efficiency is due to ionic short circuiting in feed and drain channels. This loss in Faradaic efficiency is 6% for well designed RED system in comparison with alkaline water electrolyzer where these losses range from 5 to 25% [20,25].

- Membrane area (A_{mem}^{tot}): The total membrane area [m^2] required to produce 1500 kg hydrogen a day ($\dot{m}_{H_2}^{cap} = 0.0174$ kg/s is the required production capacity).

$$A_{mem}^{tot} = \frac{I_{para}^{pp} \cdot N_{u.c.}}{j_{u.c.}^{pp}} \quad (13)$$

I_{para}^{pp} is total current from the stacks in parallel. It is calculated as $I_{para}^{pp} = \frac{\dot{m}_{H_2}^{cap} \cdot 1000 \cdot z \cdot F}{2.016 \cdot \eta_F}$. Where, $\dot{m}_{H_2}^{cap}$ is required hydrogen production capacity [kg/s] and molecular weight of hydrogen is 2.016 g/mol.

Pumping system

Experimentally, a pressure drop is observed over the inlet and outlet of feed water. Hence, to compensate this loss, pumping of feed solutions is essential. The important parameters such as flow rate, pressure drop and pumping power were calculated and are presented in this section.

- Flow rate (Q_{lc}): As suggested in the literature [22,26], the velocity of LC solution influences the double layer resistance and thus the power density. The flow rate [m^3/s] for low concentrate feed solution is estimated as,

$$Q_{lc} = N \cdot v_{lc} \cdot \delta_{ch} \cdot w \cdot \varepsilon \quad (14)$$

here ε is porosity of the spacer, v_{lc} velocity of feed solution in low concentrate channel [m/s], δ_{lc} intermembrane distance [m], w width of channel.

- Hydraulic diameter ($d_{h,lc}$): The spacer filaments obstruct the flow through the channel and this requires additional pumping power. To estimate the influence of spacer filaments, hydraulic diameter of spacer filled channel was determined using following equation,

$$d_{h,lc} = \frac{4\varepsilon}{\frac{2}{\delta_{ch}} + \left((1 - \varepsilon) \cdot \frac{8}{\delta_{ch}} \right)} \quad (15)$$

- Pressure drop (Δp_{lc}): Assuming an ideal case of fully developed laminar flow, the pressure drop (Pa) was estimated using Darcy-Weisbach equation,

$$\Delta p_{lc} = \frac{12 \cdot \mu \cdot l^2}{0.25 \cdot d_h^2 \cdot t_{res}} \quad (16)$$

the μ is dynamic viscosity of water [Pa-s], t_{res} is the residence time [s], l length of channel.

- Pumping power (P_{pump}): The power [W/m^2] required to overcome the hydraulic resistance in pumping the feed solutions through the channels. This strongly depends on the spacer porosity, as it dictates hydraulic radius which influences pressure drop.

$$P_{pump} = \frac{\Delta p \cdot d_{h,lc} \cdot \varepsilon}{t_{res}} + \frac{\Delta p \cdot d_{h,hc} \cdot \varepsilon}{t_{res}} \quad (17)$$

- Reynolds number (Re): It is the ratio of inertial forces to viscous forces within a fluid. The Reynolds number for wide channel corrected for spacer porosity is defined as [27],

$$Re = \frac{\rho \cdot v_{lc} \cdot d_h}{\mu} \quad (18)$$

Mass balances

The concentration difference between the adjacent channels drives the salt flux from high concentrate to low concentrate channel.

- Salt flux (J_{salt}): The total solute transport.

$$J_{\text{salt}} = \frac{j^{pp}}{F} + 2 \cdot \frac{D_{\text{AmB}}(c_{\text{hc}}^{\text{in}} - c_{\text{lc}}^{\text{in}})}{\delta_{\text{mem}}} \quad (19)$$

here δ_{mem} is the thickness of IEMs in operation [m], D_{AmB} diffusivity coefficient of ammonium bicarbonate in membrane [m^2/s]. The first term relates to coulombic (counter ion) transport, while the second term relates to co-ion transport. The factor 2 relates to number of membranes in a unit cell.

Waste heat/Regeneration system

The regeneration system compensates the amount of salt diffused through the IEM from high concentrate channel to low concentrate channel. The regeneration system includes air stripping column and absorption column. The air stripping column decomposes solution from dilute compartment outlet to ammonia gas and carbon dioxide gas at 333 K. The absorption column dissolves the decomposed gases at 298 K in the outlet of concentrate channel.

- Heat required for regeneration (q_{regen}): The total amount of thermal power required to strip Q_{lc} of ammonium bicarbonate salt from LC solution

$$q_{\text{regen}} = \dot{q}_{\text{th}} \cdot Q_{\text{lc}} \cdot 3600 \cdot A_{\text{mem}}^{\text{tot}} \quad (20)$$

\dot{q}_{th} is the specific thermal duty [kWh/m^3] required to decompose ammonium bicarbonate solution into its components $\text{NH}_4(\text{g})$ and $\text{CO}_2(\text{g})$. The value was estimated using relation $q_{\text{th}} = a_1 \cdot e^{a_2 \cdot C_1} - a_3 \cdot C_2^{a_4} + a_5 \cdot C_1^{a_6} \cdot C_2^{a_7}$ from Bevacqua et al. [12] for inlet concentration to stripping column C_1 and outlet concentration C_2 from stripping column. Here a_1 to a_7 are fitting parameters that are function of C_1 , refer to appendix Table 8.

Economic model

Levelized cost of hydrogen (LCH) method assesses the economic feasibility of the proposed process. This method considers annuity factor $\left(\frac{1}{(1+r)^t}\right)$ as shown in equation (21). This method is preferred as it is simple, and provides realistic results. These results of economic assessment of different technologies are easy to compare, transparent, and easy to understand. The disadvantage of using this method is that the distinction in variations in costs and benefits from one year to the next is not possible (same net benefit is applied to every year). Further, the time delay between investment and first year of regular operation are not considered.

$$\text{LCH} = \frac{\sum_{i=1}^{i=t} \frac{\text{CAPEX} + \text{OPEX} + C_{\text{heat}}}{(1+r)^i} + C_{\text{mem, re}}}{\sum_{i=1}^{i=t} \frac{m_{\text{H}_2}}{(1+r)^i}} \quad (21)$$

Capital cost (CAPEX)

The capital cost [€] of the plant was evaluated as CAPEX, using equation (22).

$$\text{CAPEX} = C_{\text{mem}} + C_{\text{REDstack}} + C_{\text{regen}} + C_{\text{pump}} + C_{\text{lab}} + C_{\text{infra}} \quad (22)$$

The capital costs include all the expenses related to purchase and installation (labour and infrastructure) of systems such as RED stack (including membranes), pumping unit,

regeneration unit. The labour cost includes civil work associated with site preparation, process equipment building, offsite services and is assumed to be 20% of capital cost. The cost of regeneration system was normalized to flow rate [m^3/h], and cost of pumping system was normalized to pumping power [kW]. The cost of membranes was normalized per unit membrane area. The cost of membrane was estimated as sum of the production cost and raw material cost. Using analytical approach to investigate the cost of Nafion membranes, Minke et al. [28] suggested that for constant thickness with increase in production rate by four orders of magnitude the total membrane cost decreases by almost one order of magnitude w.r.t the initial cost. With a decrease in thickness by almost 7 times and a further increase in membrane production rate by one order of magnitude, the total membrane cost reduces to one tenth of the present raw material cost [28]. Assuming a learning rate similar to that of Nafion membrane of $25 \mu\text{m}$ thickness, a minimum membrane cost was estimated. The production rate was assumed $60 \times 10^8 \text{ m}^2$ as a RED application requires large membrane areas due to low membrane potential or power density [10].

- Cost of RED stack (C_{REDstack}): It includes cost of electrodes, gaskets, spacers and end plates. It is assumed to be the same cost as that of the membrane.
- Infrastructure cost (C_{infra}): It is the real estate cost and this was calculated using dimensions of RED cell, stack referred from Daniilidis et al. [10].
- Pumping system cost (C_{pump}): The pumping unit consists of two pumps. The cost associated with a pump is c_{pump} [€/kW]. The pumping unit compensates the pressure drop inside a RED stack, the total cost is normalized per unit stack.

$$C_{\text{pump}} = \frac{P_{\text{pump}} \cdot A_{\text{mem}}^{\text{tot}} \cdot c_{\text{pump}}}{1000} \quad (23)$$

- Regeneration system cost (C_{regen}): The specific investment cost for regeneration system is c_{regen} [€/m³/h]. The total cost of the regeneration system is estimated for the required flow rate Q_{lc} from a RED stack.

$$C_{\text{regen}} = \frac{c_{\text{regen}} \cdot Q_{\text{lc}} \cdot A_{\text{mem}}^{\text{tot}} \cdot 3600}{2} \quad (24)$$

Operational and maintenance cost (OPEX)

OPEX [€] includes labour maintenance, service and repairs expenses which is assumed to be 2% of CAPEX. Direct and indirect operational costs such as chemical, emissions to air, supply of water, labour, taxes, administration, and insurance are not included.

$$\text{OPEX} = 0.02 \cdot \text{CAPEX} \quad (25)$$

here t is plant life time [years].

Membrane replacement cost ($C_{\text{mem, re}}$)

The ion exchange membranes have limited lifetime. Hence, these membranes have to be replaced at the end of their lifetime until plant lifetime. The cost associated to membrane replacement is calculated as,

$$C_{mem, re} = \sum_{n=1}^{n=f} \frac{C_{mem}}{(1+r)^{t_{mem} \cdot n}} \quad (26)$$

where C_{mem} and t_{mem} are membrane costs [€/m²] and membrane lifetime [years]. f is the factor that denotes the number of times replacement takes place ($f = t/t_{mem}$).

Waste heat price (C_{heat})

The regeneration system requires heat for producing NH_{3,g} and CO_{2,g} from the LC outlet solution. This heat required by the process can be sourced from various thermal energy intensive industries including cement, paper, biomass, waste incineration etc. Though it is said to be waste heat, a heat exchange system is required to provide the heat at specific temperature. Hence a cost C_{heat} is associated with it. This cost is assumed to be as low as 0.01 €/kWh.

$$C_{heat} = C_{heat} \cdot q_{regen} \cdot t_a \quad (27)$$

t_a is the total number of operational hours per year [h].

Profit

The sum of revenue earned by selling produced hydrogen and operational expenses incurred on annual basis.

$$\text{Profit} = \left(\sum_{i=1}^{i=t} \frac{m_{H_2} \cdot c_{H_2}}{(1+r)^i} \right) - \left(\sum_{i=1}^{i=t} \frac{\text{CAPEX} + \text{OPEX} + C_{heat}}{(1+r)^i} + C_{mem, re} \right) \quad (28)$$

where c_{H_2} is hydrogen selling price [€/kg_{H2}] and assumed to be 3.59 €/kg_{H2} [21].

Scenario case study

The study includes three scenarios based on the cost of membrane, the membrane life time and the hydrogen production rate. In all the scenarios for ammonium bicarbonate solution, the hydrogen production rate was calculated from the corresponding input data as shown in Table 5. In the *present scenario*, the cost and lifetime of membrane were fixed input and LCH was estimated. In the *market competing scenario*, the LCH was assumed the same as estimated by US DOE for hydrogen produced from renewable energy source. In the *market* and *future scenario*, the concentration of feed solutions were optimized to maximize hydrogen production for present properties of membranes refer Table 5 [12]. There are no profits made in *market scenario*, the membrane life was assumed to be 7 years and membrane cost was estimated. Finally, an optimistic *future scenario* was considered, wherein the profits of 10% were made on annual basis, with an increased membrane life of 10 years, and the membrane cost was estimated accordingly. In the *future scenario*, the hydrogen is produced at 3.71 mol/m²/h with increased permselectivity of membranes 0.95 and Faradaic efficiency 0.95, as shown in Table 5. The channel length and width were assumed to be the same for all scenarios. The heating cost for all scenarios was assumed to be 0.01 €/kWh. The Table 6 summarizes all the input and estimated parameters.

Table 5 – Input parameters of AmB RED system for scenario study.

Parameter	present	market	future	Unit
Concentrate solution conc. (C_{hc})	2	2.6	2.6	M
Dilute solution conc. (C_{lc})	0.06	0.05	0.07	M
Permselectivity (α)	0.753	0.85	0.95	–
Inter-membrane distance (δ_{ch})	270	100	100	μm
Residence time (t_{res})	70	60	50	s
Membrane lifetime (t_{mem})	4	7	10	years
Faradaic efficiency (η_F)	0.95	0.95	0.99	–

Table 6 – List of input and output parameters for scenario study.

Scenario	Input	Output
present	C_{mem}, t_{mem}	$\dot{n}_{H_2}, \text{LCH}, \text{Profit}$
market	LCH, t_{mem} , Profit	\dot{n}_{H_2}, C_{mem}
future	Profit, t_{mem}	$\dot{n}_{H_2}, C_{mem}, \text{LCH}$

Results and discussion

Cost of membrane

Fig. 3a shows the influence of membrane cost on the LCH for AmB based RED system for hydrogen production at 0.72 mol/m²/h production rate, and a membrane life, t_{mem} of 4 years. LCH is a linear function of membrane cost, C_{mem} . The LCH of AmB RED system decreases with decrease in membrane cost, C_{mem} . Fig. 3b compares LCH for ammonium bicarbonate RED system in the *market* and the *future* scenarios. The rate of decrease of LCH for *market scenario* is three times as high as *future scenario*, this shows that LCH was sensitive to membrane cost in *market scenario*. The intercepts in Fig. 3b shows the contribution of balance of plant cost to LCH at membrane cost, $C_{mem} = 0$ €/m², and, LCH at this intercepts are below the LCH target set by US DOE. Assessing all the costs in *present scenario*, it was found that the membrane cost influences the most, accounting for almost 40% of CAPEX. This is in agreement with the previous studies [9–11,29]. The minimum predicted cost of membrane was calculated using the learning rate as that of 25 μm Nafion membrane for production rate of 60×10^8 m²/a. The minimum membrane cost was found to be 1.69 €/m². For ammonium bicarbonate RED system, the membrane cost, C_{mem} , must drop below 2.86 and 22.3 €/m² for membrane lifetime t_{mem} of 4 and 10 years at production rate of 1.19 and 3.71 mol/m²/h, respectively, to compete with other clean energy technologies.

Hydrogen production rate

The LCH is predicted for different hydrogen production rates. With increase in production rate, the LCH decreases. With the present membrane cost (150 €/m²) and membrane life of 4 years, the LCH decreased by almost 42% (72.49 €/kg) for increase in production rate from 0.72 mol/m²/h to 1.19 mol/m²/h. This increase in production was due to optimal concentration

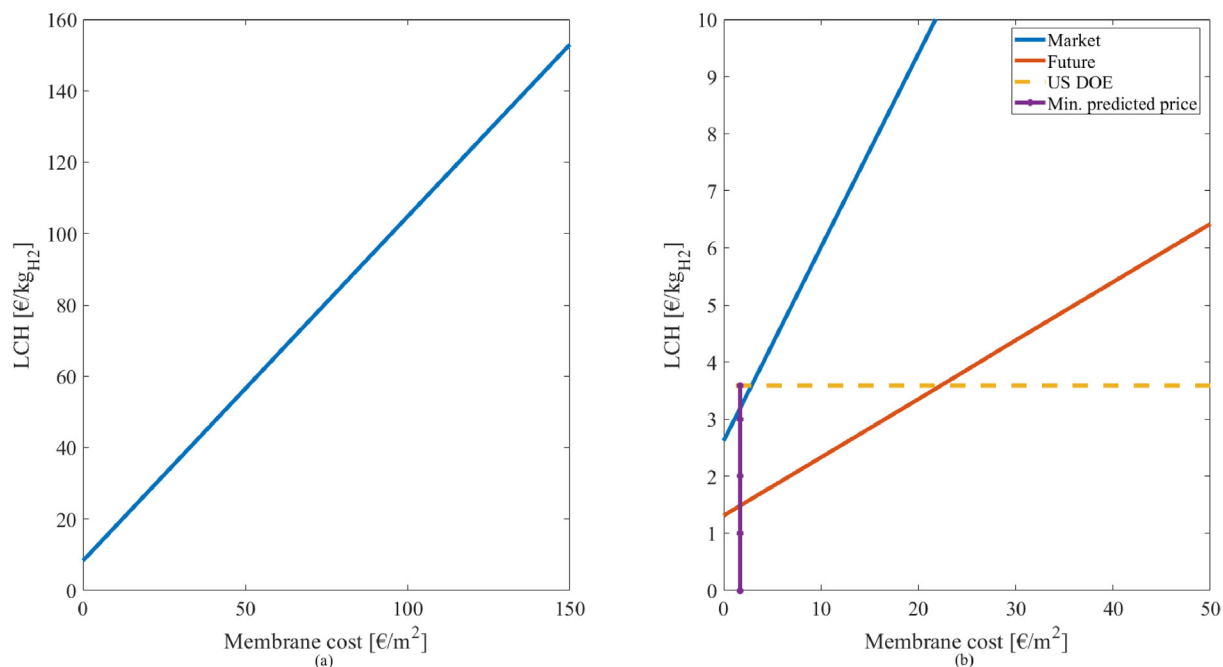


Fig. 3 – a. Influence of cost of membrane on LCH for a membrane lifetime of 4 years in case of present scenario (left). b. Comparison of market, future scenario with US DOE target price, for LCH as function of membrane cost (right).

of feed solutions. The LCH decreased to 23.7 €/kg for further increase in hydrogen production rate to 3.71 mol/m²/h. This increase was a combined result of increase in permselectivity and Faradaic efficiency. The increase in production rate increases the total hydrogen produced in the plant lifetime. Therefore, LCH decreases. Increase in production rate is due to increase in current density, which indirectly relates to concentration ratio. High concentration ratios are achieved with high solubility of salt under the assumption that the electrical conductivity of the solution and permselectivity of membrane remains same. The production rate is a function of peak current density, which in turn depends on Nernst potential. As this Nernst potential relates to ratios of concentration or activity coefficients which are salt properties (solubility etc). The salts that can produce hydrogen at a production rate higher than that of AmB-RED system due to synergetic effect of higher OCP, higher solubility limit and lower resistance needs to be investigated experimentally. However, this is beyond the scope of this work. Hydrogen production rate can be increased by improving membrane properties such as permselectivity, conductance and reducing thickness while maintaining the mechanical strength. Further, the use of optimal combination of electrolytic solutions, electrode material and their geometric configuration can decrease cell overpotential and increase the overall stack potential, which in turn will improve the hydrogen production rate.

Lifetime of membrane

Fig. 4a describes the influence of membrane lifetime at a membrane cost of 150 €/m² for present scenario. With increase in membrane life, the LCH decreases substantially. As the membrane lifetime increases from 3 years to 10 years, the LCH decreases to almost 40% of the initial value in the present

scenario. Fig. 4b shows asymptotic behaviour of t_{mem} required at a hydrogen production rate of 1.19 mol/m²/h to 3.71 mol/m²/h in order to meet a LCH of 3.59 €/kg. The increase in membrane life 7 years or more, show less significant impact on LCH in present and market scenario. Due to the operating conditions, the membrane properties such as permselectivity, membrane resistance etc. can be expected to deteriorate with time. As the performance of the system depends on the membrane properties, corresponding deterioration in the system performance will be observed. The higher the deterioration rate, the quicker the membranes need to be replaced. For best performance, the system is operated at optimum conditions, and to maintain the performance, the membranes need to be replaced on timely basis. Increasing the frequency of replacing a membrane increases membrane replacement cost (contributes 44% of total expenses in present scenario). Thus, the overall trend implies that the lifetime of membrane (indirectly, the degradation rate) of these membranes plays a critical role in minimizing LCH. market scenario becomes economically feasible at membrane life of 7 years while 1 year for future scenario. This shows that increase in membrane life beyond 7 years has relatively low impact on LCH with the case parameters chosen in the study.

Inter-membrane distance

Increase in the inter-membrane distance decreases the actual unit cell potential. The decrease in open circuit potential decreases the peak power current density, which in turn decreases the salt flux. Hence, the theoretical heat required also decreases. On the contrary, the increase in inter-membrane distance cause, an increase in feed solution volume flowing through the channel. This, again, causes an increase in the theoretical heat required to regenerate the ammonium

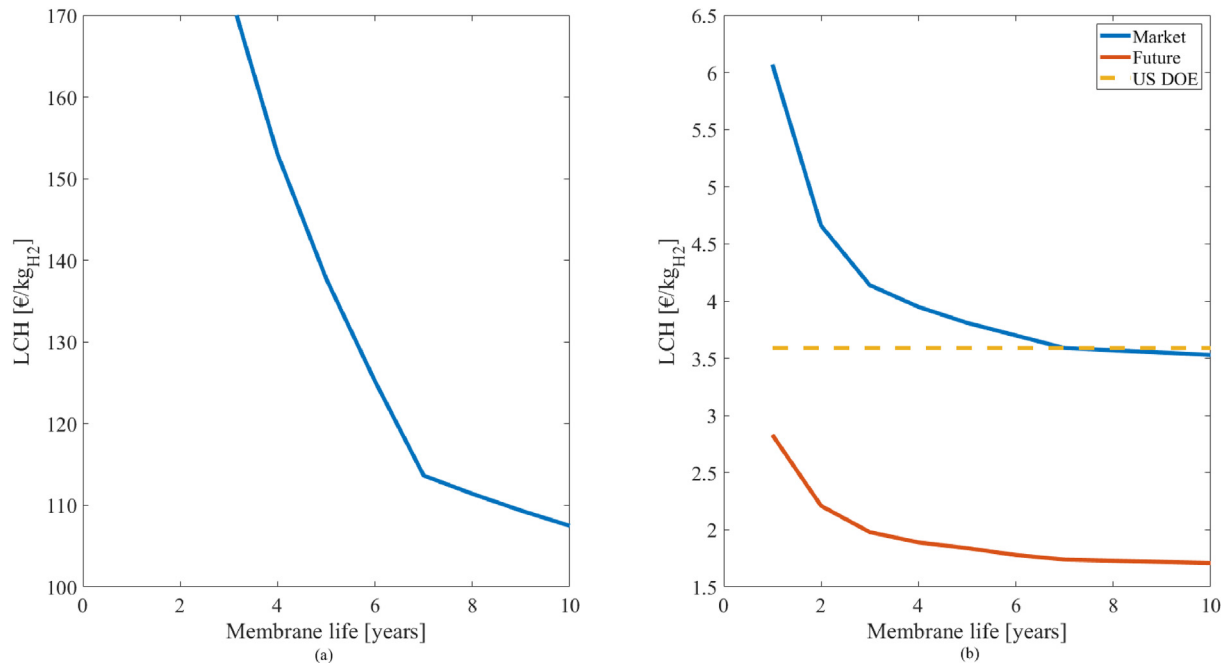


Fig. 4 – a. Influence of membrane life time on LCH for AmB RED present scenario (left). b. Comparison of market and future scenario for membrane lifetime on LCH (right) (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article).

bicarbonate solutions. This increase in the heat requirement nullifies the decrease in heat requirement due to decrease in salt flux. Thus, the net effect observed is an increase in the total heat required.

Moreover, as the increase in inter-membrane distance increases the feed solution flow rate. The increase in flow rate increases the amount of water per gram of salt. Thus, the total heat required to recover same amount of salt increases.

Increased inter-membrane distance has a negative impact on LCH, as shown in Fig. 5a and b. The LCH decreases with decrease in inter-membrane distance. The increase in inter-membrane distance increases the channel resistance. This increase in resistance decreases the open circuit potential and increases number of membrane pairs required. This results in increase LCH. In case of the market scenario, the LCH varies almost linearly with change in inter-membrane distance. In the market scenario and the future scenario the maximum inter-membrane distance to match with the US DOE were found to be 100 μm , and 200 μm from Fig. 5b respectively.

Cost of heating

Fig. 6a and b, shows the influence of cost of heating for the present, market and future scenario. The cost of heating, c_{heat} , has a negative impact on LCH. With increase in c_{heat} , the LCH increases as shown in Fig. 6a and b. In the market and the future scenario, the cost of heating contributes as high as 58% to the total expenses. Hence, it is considered as a critical parameter. From Fig. 6b, the maximum cost of heating was found to be 0.01 and 0.03 €/kWh for market scenario and future scenario respectively. The intercept on y axis shows the LCH for case

with no cost of heating. These values were found to be approximately 148.5, 2.2 and 0.7 €/kg_{H2} for the present, the market and the future scenario respectively.

Scenario analysis

Out of the three scenarios, LCH in present scenario was found to be the highest, with the cost of membrane being the major contributor. Fig. 7, shows contribution of expenses such as membranes, RED stack without membranes, regeneration system, pumping system, infrastructure and labour to the capital expenses. From Fig. 7, in the present scenario, the membrane and RED stack contribute to approximately 80% of CAPEX, this contribution reduces to more than half in the market and future scenario. The labour cost, which includes civil work associated with site preparation, process equipment building, off site services, contributes to 16% of CAPEX in all the scenarios. Pumping system cost increases from present scenario to future scenario but has negligible contribution to LCH in all scenarios. The contribution of infrastructure was of low significance (4.4% max. in the future scenario).

The maximum limit of c_{mem} to achieve LCH of 3.59 €/kg_{H2} for market scenario (2.86 €/m²) is a tenth of c_{mem} in the future scenario). Compared to the present scenario, the contribution of c_{heat} to LCH increased from 3% to 50% and 57% for market and future scenario respectively. This suggests that as the cost of membrane decreases, and the life of membrane increases, the contribution from c_{heat} to the LCH increases (see Table 7).

In order to improve the accuracy and provide more realistic estimates, the following recommendations are given. The value of the heat of regeneration, q_{th} , was taken from a

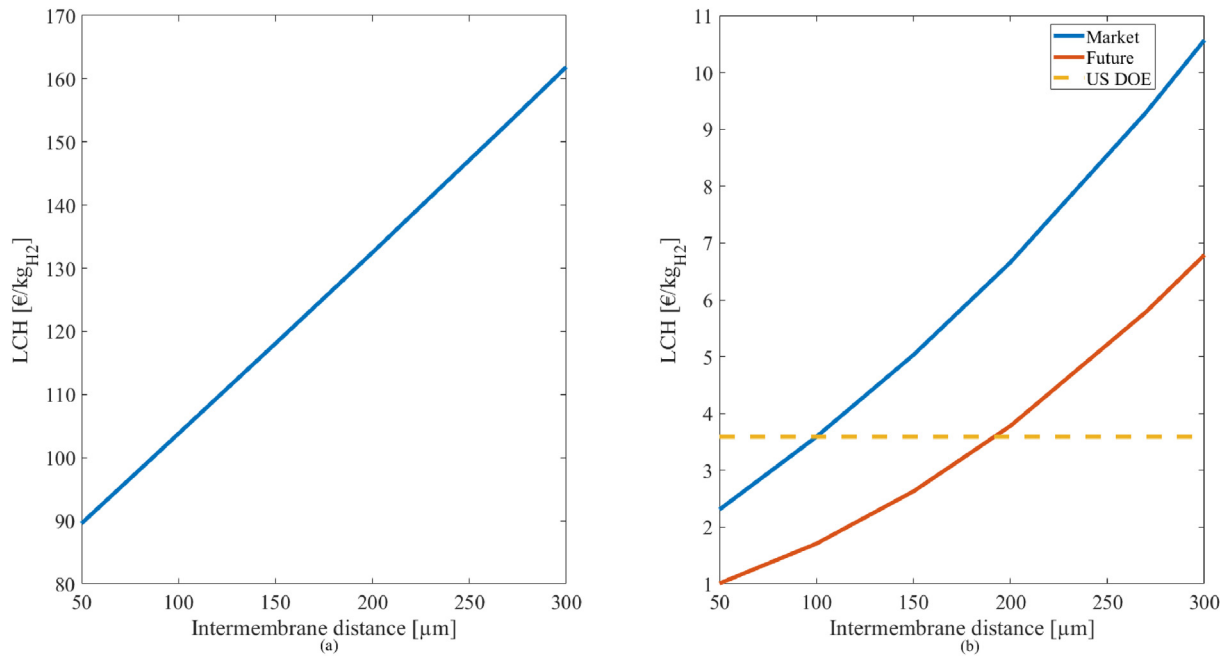


Fig. 5 – a. Influence of inter-membrane distance on LCH for present (left) b. market and future scenario (right).

theoretical study and was not optimized for the present AmB-RED system. Further, to investigate influence of salt type on thermochemical conversion efficiency, η_{th} , modeling and optimization of regeneration system in a process modelling and simulation software is essential. In the economic model, presented simplified LCH method is used to evaluate the specific cost of hydrogen per kg. This method does not include various operational costs and other capital costs such as consultancy and financing costs. Further, the

economic model is developed for a standard capacity of 1500 $\text{kg}_{\text{H}_2}/\text{day}$ and hence no upscaling effects were considered. The use of base cost method proposed by Ulrich et al. [30]. includes these costs and upscaling effect, and thus provides more realistic estimates. The cost of other components in a RED stack were assumed to be the same as the cost of membrane. This may not necessarily be true in case of learning rates. Hence individual component cost estimates are needed. The RED model does not include non-ohmic

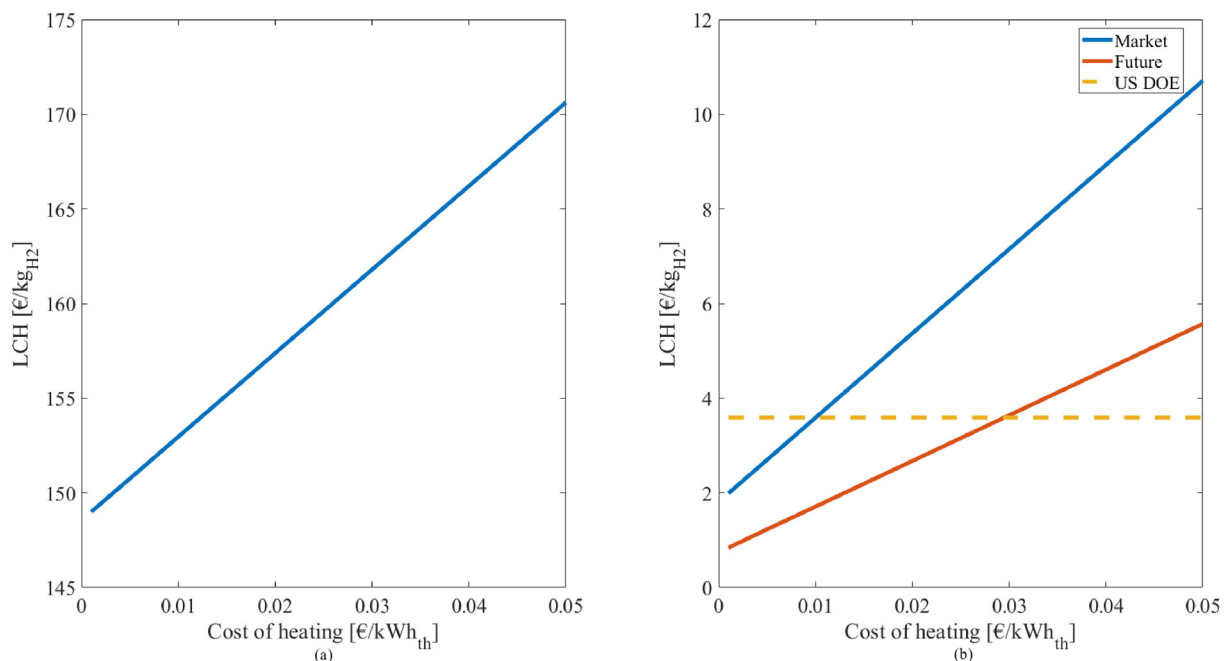


Fig. 6 – a. LCH as a function of cost of heating for present (left) b. market and future scenario (right).

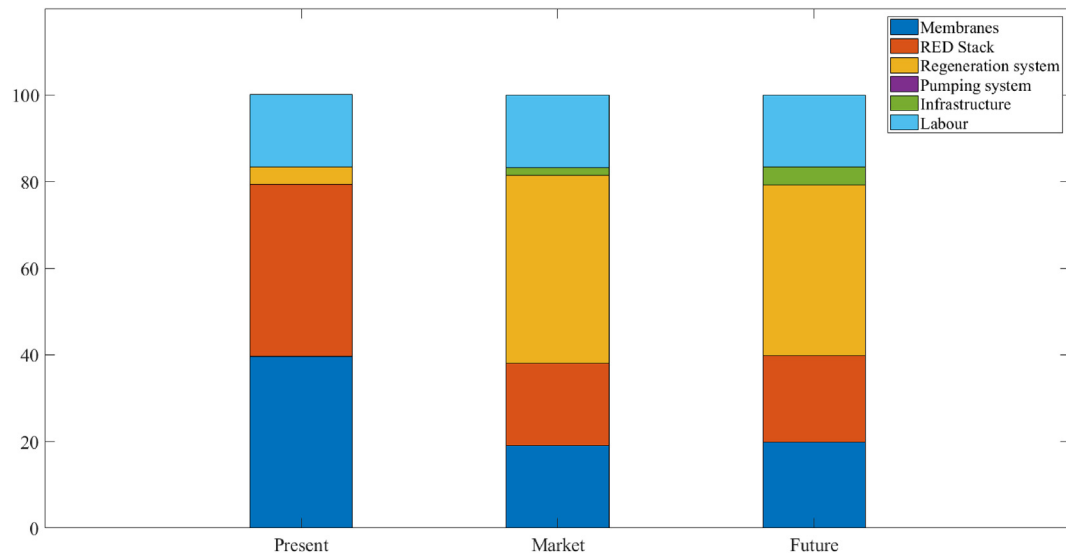


Fig. 7 – Comparison of various system elements in relation to CAPEX in the three scenarios.

Table 7 – Summary of the results from scenario analysis of the techno-economic analysis.

Present Scenario		Market Scenario		Future Scenario	
LCH	152.97 €/kg _{H₂}	LCH	3.59 €/kg _{H₂}	LCH	1.71 €/kg _{H₂}
C_{mem}	150 €/m ²	C_{mem}	2.86 €/m ²	C_{mem}	3.94 €/m ²
t_{mem}	4 yrs	t_{mem}	7 yrs	t_{mem}	10 yrs
\dot{m}_{H_2}	0.72 mol/m ² /h	\dot{m}_{H_2}	1.19 mol/m/h	\dot{m}_{H_2}	3.71 mol/m/h
Waste heat	480 kWh/kg _{H₂}	Waste heat	194 kWh/kg _{H₂}	Waste heat	105 kWh/kg _{H₂}
Re	0.3	Re	0.13	Re	0.16
CAPEX	43%	CAPEX	35%	CAPEX	31%
OPEX	10%	OPEX	8%	OPEX	7%
$C_{mem, re}$	44%	$C_{mem, re}$	7%	$C_{mem, re}$	5%
C_{heat}	3%	C_{heat}	50%	C_{heat}	57%
Profit	-931,378,389 €	Profit	0 €	Profit	11,721,725 €

resistances, they may influence the hydrogen production rate at low Reynolds number [31].

Conclusions

A simplified LCH model is developed for a thermally driven AmB RED system. The model includes capital costs associated with the regeneration system and operation costs for the waste heat. The scenario analysis includes three different cases *present*, *market* and *future*; these scenarios differ in the inlet feed solution concentration, membrane properties, residence time, cost of membrane, membrane lifetime. The scenario analysis demonstrates cost of membrane and membrane lifetime as dictating parameters in *present scenario*; cost of heating and cost of regeneration system, control the levelized cost of hydrogen (LCH) in *market* and *future scenario*. In case of the *present scenario* membrane replacement cost dominates due to high capital cost of membrane (150 €/m²) and limited membrane life of 4 years. While, in *market* and *future scenarios* cost of regeneration system influences with contribution to CAPEX as high as 40%. Except in the case of the

present scenario, cost of heating contributes to almost 57% of total expenses, making it a critical parameter that needs to be optimized to further minimize LCH. Parameters such as inter-membrane distance, has a negative effect on the LCH. A decrease in inter-membrane distance increases pumping power but the decrease in heat required to extract salt dominates. The decrease in inter-membrane distance by one fifth of initial value decreases LCH to as low as 1.35 €/kg_{H₂} in the *market* and 0.83 €/kg_{H₂} in the *future scenario*. In future the cost of membrane can reduce to 1.69 €/m² with increase in production rate, reduction in production cost, raw material cost, and reduction in membrane thickness. It is clear that AmB RED for hydrogen production has economic potential at membrane cost less than 2.86 €/m², and/or, membrane life of 7 years or more. Future efforts must be directed to evaluate performance of potential salts such as lithium bromide that are highly conductive, highly soluble, and may require less heat for recovery. Finally, a thermally driven RED system with membrane cost not exceeding 20 €/m²; hydrogen production rate of 3.7 mol/m²/h or higher and cost of heating not more than 0.03 €/kWh can make an economically feasible solution for low grade waste heat to hydrogen production.

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Appendix

Table 8 – Input values for parameters described in equation (20) as a function of inlet solution concentration for stripping column.

Concentration range [M]	a_1	a_2	a_3	a_4	a_5	a_6	a_7
$0.025 \leq C_1 = 0.1$	12.115	0.26	261.787	0.615	297.46	0.252	0.478
$0.1 < C_1 \leq 0.2$	12.836	1.02	258.324	0.612	260	0.165	0.517
$0.2 < C_1 \leq 0.56$	13.195	0.686	60.592	0.667	55.934	0.687	0.212
$0.56 < C_1 \leq 2$	8.714	0.225	35.796	0.656	45.56	0.758	0.045

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