

**Proposal, design, and cost analysis of a hydrogen production
process from cellulose via supercritical water gasification**

Supplementary Information

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S1. Required Energy

S1-1. Decomposition of cellulose (This study)

Let us determine the energy required by our hydrogen production process. As observed in Section 3-6, the minimum heating value is 28.3 GJ/h. In addition to this, the electricity used in this process is 3.76 GJ/h. Therefore, the amount of energy required for our process can be calculated as $28.3 + 3.76 = 32.1$ GJ/h.

S1-2. Electrolysis of water by wind power (Electrolysis only, experimental)

Let us consider the electrolysis of water when using an electrolysis tank to produce 42 m³/h of hydrogen gas with 200 kW of electricity [S1], so that when 228.7 kmol/h of hydrogen is produced, the volume of hydrogen can be calculated as follows if hydrogen is an ideal gas.

$$\begin{aligned}(\text{Volume of H}_2 \text{ [m}^3\text{/h]}) &= (\text{H}_2 \text{ production [mol/h]}) \times (\text{volume of ideal gas [m}^3\text{/mol]}) \\ &= 228.7 \times 10^3 \times 22.4 \times 0.1^3 \\ &= 5.12 \times 10^3 \text{ m}^3\text{/h}\end{aligned}$$

Since it takes 200 kJ/s per 42 m³/h of hydrogen gas in a commercial electrolysis tank [35], the power [GJ/h] required to produce 5.12 × 10³ m³/h can be calculated as follows.

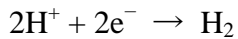
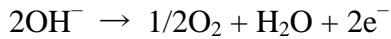
$$\begin{aligned}(\text{Electric power [GJ/h] required}) &= (\text{H}_2 \text{ target production [m}^3\text{/h]}) / (\text{H}_2 \text{ production [m}^3\text{/h] for commercial equipment}) \times \\ &(\text{energy [GJ/h] for commercial equipment}) \\ &= 5123/42 \times 200 \times 10^3 \times 3600 \times 10^{-9} \\ &= 87.8 \text{ GJ/h}\end{aligned}$$

Here, if the conversion efficiency $\eta = 0.20$ [S2] for 87.8 GJ/h of electricity,

$$\begin{aligned}(\text{Required energy [GJ/h]}) &= (\text{Required electrical energy [GJ/h]}) / \eta \\ &= 87.8 / 0.20 = 439.1 \text{ GJ/h}\end{aligned}$$

S1-3. Electrolysis of water by wind power (Electrolysis only, theoretical)

From the energy required for the electrolysis of water, calculate the energetic energy in the electrolysis of water. From the following half-reaction equation [S3], assuming the number of electrons $n = 2$ mol/mol, the potential $E = 2.0$ V [S3], and the conversion efficiency $\eta = 0.20$ [S2],



$$\Delta G = nFE/\eta = (2 \times 96454.56 \times 2.0) / 0.20 = 1929091 \text{ J/mol}$$

Hydrogen production is 228.7 kmol/h, so the amount of energy required per 1 h is

(Energy required for electrolysis of water [GJ/h])

$$= \Delta G \times (\text{H}_2 \text{ production [mol/h]})$$

$$= 1929091 \times 228.7 \times 10^3 \times 10^{-9}$$

$$= 441.2 \text{ GJ/h}$$

S1-4. Electrolysis of water by wind power (Reaction enthalpy)

Calculating the energy required for the electrolysis of water from the enthalpy of water production, 240 kJ/mol [S4],

(Energy required for H₂ production [GJ/h])

$$= (\text{enthalpy of production of H}_2\text{O [GJ/mol]}) \times (\text{H}_2 \text{ production [mol/h]})$$

$$= 240 \times 10^{-6} \times 228.7 \times 10^3$$

$$= 54.89 \text{ GJ/h}$$

S1-5. Steam reforming of methane (w/ CO₂ capture)

Calculate the energy required in the process of steam reforming of methane with carbon dioxide capture. Since the energy required to produce 8.994 t/h of hydrogen is 433.72 MW [S5], we can calculate the energy required to produce 228.7 kmol/h of hydrogen.

(Energy required for H₂ production [GJ/h]) = (Energy required per 1 mol/h of hydrogen [GJ/mol]) × (Hydrogen production [mol/h])

$$= (433.72 \times 3600 \times 10^{-3}) / (8.994 / 2 \times 907 \times 10^3) \times (228.7 \times 10^3)$$

$$= 87.5 \text{ GJ/h}$$

S1-6. Steam reforming of methane (w/o CO₂ capture)

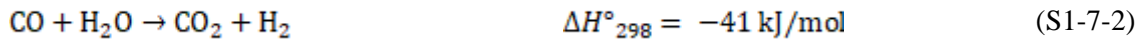
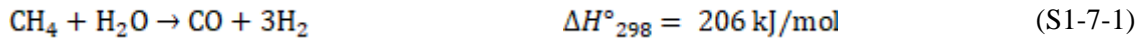
Calculate the energy required for the methane steam reforming process without carbon dioxide capture. Since the energy required to produce 8.994 t/h of hydrogen is 394.77 MW [S5], we can calculate the energy required to produce 228.7 kmol/h of hydrogen.

(Energy required for H₂ production [GJ/h]) = (Energy required per 1 mol/h of hydrogen

$$\begin{aligned}
& [\text{GJ/mol}] \times (\text{Hydrogen production} [\text{mol/h}]) \\
& = (394.77 \times 3600 \times 10^{-3}) / (8.994/2 \times 10^6) \times (228.7 \times 10^3) \\
& = 72.3 \text{ GJ/h}
\end{aligned}$$

S1-7. Steam reforming of methane (Reaction enthalpy)

The energy required for hydrogen production by steam reforming of methane is calculated in terms of reaction enthalpy. The reaction equation for hydrogen production by steam reforming of methane is shown below [S6].



In addition, the temperature-dependent molar heat capacity at constant pressure of each substance extracted from Aspen Plus was used. First, since the reforming reaction of Eq. S1-7-1 takes place at a temperature of 973 K, the heat of reaction for the reforming reaction at a temperature of 973 K is determined as follows:

$$\begin{aligned}
& (\text{Heat of reaction of the reforming reaction at temperature 973 K} [\text{kJ/mol}]) = (\text{standard heat of reaction of the reforming reaction} [\text{kJ/mol}]) + (\text{enthalpy of the standard state reference product at 973 K} [\text{kJ/mol}]) - (\text{enthalpy of the standard state reference reactant at 973 K} [\text{kJ/mol}]) \\
& = 206 + (59.6 + 140) - (36.4 + 24.9) \\
& = 344.3 \text{ kJ/mol}
\end{aligned}$$

First, since the shift reaction in Eq. S1-7-2 takes place at a temperature of 573 K, the heat of reaction for the shift reaction at a temperature of 573 K is determined as follows:

$$\begin{aligned}
& (\text{Heat of reaction of the shift reaction at temperature 573 K} [\text{kJ/mol}]) = (\text{standard heat of reaction of the shift reaction} [\text{kJ/mol}]) + (\text{enthalpy of product at 573 K of standard state reference} [\text{kJ/mol}]) - (\text{enthalpy of reactant at 573 K of standard state reference} [\text{kJ/mol}]) \\
& = -41 + (8.02 + 11.7) - (55.2 + 9.58) \\
& = -86.06 \text{ kJ/mol}
\end{aligned}$$

Hence, the energy required to produce 228.7 kmol/h of hydrogen by steam reforming of methane can be obtained as follows:

$$(\text{Energy required for H}_2 \text{ production} [\text{GJ/h}]) = (\text{Amount of reactants} [\text{mol/h}]) \times (\text{Reaction}$$

$$\begin{aligned}
& \text{heat of reforming reaction [GJ/mol]} + (\text{Amount of reactants [mol/h]} \times (\text{Reaction heat} \\
& \text{of shift reaction [GJ/mol]}) \\
& = (228.7 \times 10^3) / 4 \times 344.3 \times 10^{-6} - (228.7 \times 10^3) / 4 \times 86.06 \times 10^{-6} \\
& = 14.8 \text{ GJ/h}
\end{aligned}$$

Next, find the energy required by the furnace to heat CH₄ and H₂O to 973 K, respectively. The energy required to heat CH₄ and H₂O to 973 K is 36.4 kJ/mol and 24.9 kJ/mol per mol, respectively, and can be calculated as follows.

$$\begin{aligned}
& (\text{Energy required in heating furnace [GJ/h]}) = (\text{Amount of CH}_4 \text{ [mol/h]}) \times (\text{Energy} \\
& \text{required for CH}_4 \text{ per mol [GJ/mol]}) + (\text{Amount of H}_2\text{O [mol/h]}) \times (\text{Energy required for} \\
& \text{CH}_4 \text{ per mol [GJ/mol]}) \\
& = (228.7 \times 10^3) / 4 \times 36.4 \times 10^{-6} + (228.7 \times 10^3) / 4 \times 24.9 \times 10^{-6} \\
& = 2.081 + 1.424 \\
& = 3.51 \text{ GJ/h}
\end{aligned}$$

Hence, the energy required for hydrogen production is $14.8 + 3.51 = 18.3$ GJ/h.

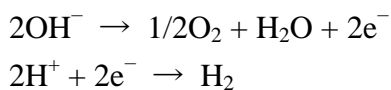
S1-8. Electrolysis of water by electric power system (Electrolysis only, experimental)

From Section S1-2, the electricity required to produce 228.7 kmol/h of hydrogen by electrolysis of water using an electrolysis tank is 87.8 GJ/h. Here, if the conversion efficiency of the power system is $\eta = 0.42$ [S7], the following can be obtained:

$$\begin{aligned}
& (\text{Required amount of energy [GJ/h]}) = (\text{Required amount of electrical energy [GJ/h]}) / \eta \\
& = 87.8 / 0.42 = 209.1 \text{ GJ/h}
\end{aligned}$$

S1-9. Electrolysis of water by electric power system (Electrolysis only, theoretical)

It can be obtained in the same way as in Section S1-3. From the following semi-reaction equation [S3], assuming that the number of electrons $n = 2$ mol/mol, the potential $E = 2.0$ V [S3], and the conversion efficiency of the power system $\eta = 0.42$ [S7],



$$\Delta G = nFE/\eta = (2 \times 96454.56 \times 2.0) / 0.42 = 918614.86 \text{ J/mol}$$

Hydrogen production is 228.7 kmol/h, so the amount of energy required per 1 h is (Energy required for electrolysis of water [GJ/h])

$$\begin{aligned}
&= \Delta G \times (\text{H}_2 \text{ production [mol/h]}) \\
&= 918614.86 \times 228.7 \times 10^3 \times 10^{-9} \\
&= 210.1 \text{ GJ/h}
\end{aligned}$$

S1-10. Electrolysis of water by electric power system (Reaction enthalpy)

Same as Section S1-4.

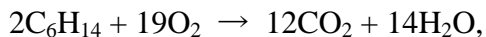
S2. CO₂ Emission

S2-1. Decomposition of cellulose (This study)

Calculate the carbon dioxide emissions from our hydrogen production process. The carbon dioxide emissions from our process consist of the amount of carbon dioxide purged in the final flow shown in Fig. 3-6-1 and Table 3-6-1, and the amount of carbon dioxide emitted when the fuel, hexane, is burned in the furnace. The carbon dioxide purged is the sum of the carbon dioxide emitted in flow numbers 37, 40, and 42 in the final flow diagram in Fig. 3-6-1, which is $89.9 + 45.3 + 0.9 = 136.1$ kmol/h. The energy required by the heating furnace is 28.3 GJ/h. Since the energy required to burn 1 kmol/h of hexane is 2031949 kJ/h based on Aspen HYSYS simulation results, the amount of hexane required is:

$$\begin{aligned}
&(\text{Amount of hexane needed for the furnace [kmol/h]}) = (\text{Energy needed in the furnace [GJ/h]} / (\text{Heat of combustion of hexane [GJ/kmol]}) \\
&= 28.3 / (2031949 \times 10^{-6}) \\
&= 13.9 \text{ kmol/h}
\end{aligned}$$

Hence, since the molar ratio of hexane to carbon dioxide is six from the following reaction equation,



the amount of carbon dioxide emitted can be calculated as follows.

$$\begin{aligned}
&(\text{Amount of carbon dioxide emitted from the furnace [kmol/h]}) = (\text{Amount of hexane burned [kmol/h]}) \times (\text{Molar ratio of hexane to carbon dioxide}) \\
&= 13.93 \times 6 \\
&= 83.6 \text{ kmol/h}
\end{aligned}$$

Since 0.855 pound (lb) of carbon dioxide is emitted per kWh of electricity [S8], the electricity from this process can be converted to carbon dioxide emissions as follows.

(Amount of carbon dioxide converted from electricity [kmol/h]) = (Amount of carbon dioxide per kWh of electricity [kmol/kWh]) × (Power [kW])
 = $(0.855 \times 0.454) / 44.01 \times 1045$
 = 9.22 kmol/h
 Therefore, the amount of carbon dioxide emitted by this process is $136.1 + 83.6 + 9.22$
 = 228.9 kmol/h.

S2-2. Electrolysis of water by wind power (Electrolysis only, experimental)

Consider the carbon dioxide emissions of water electrolysis when using an electrolysis tank: as mentioned in Section S1-2, water electrolysis using an electrolysis tank produces 42 m³/h of hydrogen gas with 200 kW of electricity [S1]. For the production of 228.7 kmol/h of hydrogen, from Section S1-2, it is 5.12×10^3 m³/h, so the required electric power is obtained as follows:

(Electric power required [kW])
 = (H₂ target production [m³/h]) / (H₂ production [m³/h] of commercial equipment) ×
 (Electric power [kW] for commercial equipment)
 = $5123 / 42 \times 200 = 24395$ kW

Hence, because wind power emits 11 g carbon dioxide per kWh [S9],

(Amount of carbon dioxide converted from electricity [kmol/h]) = (Amount of carbon dioxide per kWh of electricity [kmol/kWh]) × (Electricity [kW])
 = $11 \times 10^{-3} / 44.01 \times 24395$
 = 6.097 kmol/h

S2-3. Electrolysis of water by wind power (Electrolysis only, theoretical)

Calculate the carbon dioxide emissions from the electrical energy required for the electrolysis of water by wind power as discussed in Section S1-3. From Section S1-3, assuming that the energy required is 441.2 GJ/h and the conversion efficiency $\eta = 0.42$ [S7], the carbon dioxide emissions can be calculated as follows:

(Carbon dioxide emissions [kmol/h]) = (Amount of carbon dioxide per kWh of electricity [kmol/kWh]) × (Electricity [kW])
 = $11 \times 10^{-3} / 44.01 \times (441.2 \times 0.2 \times 10^6) / 3600$
 = 6.126 kmol/h

S2-4. Electrolysis of water by wind power (Reaction enthalpy)

The carbon dioxide emissions are calculated from the reaction of the enthalpy of water production. From Section S1-4, the energy required for H₂ production is 54.89 GJ/h. Therefore, the carbon dioxide emissions can be calculated as follows:

$$\begin{aligned} \text{(Amount of carbon dioxide converted from electricity [kmol/h])} &= (11 \times 10^{-3} / 44.01) \times \\ & (54.89 \times 10^6 / 3600) \\ &= 3.811 \text{ kmol/h} \end{aligned}$$

S2-5. Steam reforming of methane (w/ CO₂ capture)

Calculate the carbon dioxide emissions in the process of methane steam reforming with carbon dioxide capture. With carbon dioxide capture, 0.0888 kg of carbon dioxide is emitted per 1 Nm³ of hydrogen in the process of methane steam reforming [S5], so the amount of carbon dioxide emitted to produce 41 million Nm³ of hydrogen can be calculated.

$$\begin{aligned} \text{(Amount of CO}_2 \text{ emitted by the methane steam reforming process [kmol/h])} &= (\text{CO}_2 \\ & \text{emissions per Nm}^3 \text{ of H}_2 \text{ [kmol/(Nm}^3\text{)])} \times (\text{H}_2 \text{ production [Nm}^3\text{/h])} \\ &= 0.0888 / 44.01 \times (4.1 \times 10^7 / 8000) \\ &= 10.3 \text{ kmol/h} \end{aligned}$$

S2-6. Steam reforming of methane (w/o CO₂ capture)

Calculate the energy requirements for the methane steam reforming process without CO₂ capture. Without CO₂ capture, the methane steam reforming process emits 0.8091 kg/Nm³ of CO₂ [S5], so the amount of CO₂ emitted to produce 228.7 kmol/h of H₂ can be calculated.

$$\begin{aligned} \text{(Amount of CO}_2 \text{ emitted by the methane steam reforming process [kmol/h])} &= (\text{CO}_2 \\ & \text{emission per Nm}^3 \text{ of H}_2 \text{ [kmol/(Nm}^3\text{)])} \times (\text{H}_2 \text{ production [Nm}^3\text{/h])} \\ &= 0.8091 / 44.01 \times (4.1 \times 10^7 / 8000) \\ &= 94.2 \text{ kmol/h} \end{aligned}$$

S2-7. Steam reforming of methane (Reaction enthalpy)

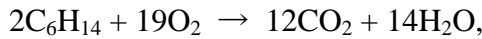
Calculate the CO₂ emission from the reaction in Eqs. S1-7-1 and S1-7-2. The amount of CO₂ emitted in the reaction process of steam reforming of methane with 228.7 kmol/h of H₂ can be calculated in the following way:

$$\begin{aligned}
& \text{(Amount of CO}_2 \text{ emitted by the steam reforming of methane [kmol/h])} = \text{(molar ratio of H}_2 \text{ to CO}_2) \times \text{(H}_2 \text{ production [kmol/h])} \\
& = 1/4 \times 228.7 \\
& = 57.2 \text{ kmol/h}
\end{aligned}$$

The amount of CO₂ emitted from the furnace used in the methane steam reforming process is then calculated. Calculated as in this process, the calculation is as follows.

$$\begin{aligned}
& \text{(Amount of hexane required for the heating furnace [kmol/h])} = \text{(Energy required by the heating furnace [GJ/h])} / \text{((Heat of combustion of hexane [GJ/kmol])} \\
& = 28.3 / (2031949 \times 10^{-6}) \\
& = 1.72 \text{ kmol/h}
\end{aligned}$$

Hence, since the molar ratio of hexane to CO₂ is six from the following reaction equation,



the amount of CO₂ emitted can be calculated as follows:

$$\begin{aligned}
& \text{(Amount of CO}_2 \text{ emitted from the furnace [kmol/h])} = \text{(Amount of hexane burned [kmol/h])} \times \text{(Molar ratio of hexane to CO}_2) \\
& = 1.725 \times 6 \\
& = 10.4 \text{ kmol/h}
\end{aligned}$$

Therefore, the amount of CO₂ emitted in this process is $57.2 + 10.4 = 67.6$ kmol/h.

S2-8. Electrolysis of water by electric power system (Electrolysis only, experimental)

From Section S2-2, the electric power required for the electrolysis of water is 24395 W. If 0.855 lb of CO₂ is emitted to generate 1 kWh of electricity [S8], the CO₂ emissions can be calculated as follows:

$$\begin{aligned}
& \text{(Amount of CO}_2 \text{ converted from electricity [kmol/h])} = \text{(Amount of CO}_2 \text{ per kWh of electricity [kmol/kWh])} \times \text{(Electricity [kW])} \\
& = (0.855 \times 0.454) / 44.01 \times 24395 \\
& = 215 \text{ kmol/h}
\end{aligned}$$

S2-9. Electrolysis of water by electric power system (Electrolysis only, theoretical)

From Section S1-9, the CO₂ emission is calculated based on 210.1 GJ/h of energy required for the electrolysis of water. Assuming a conversion efficiency $\eta = 0.42$ [S7], the following can be calculated:

$$\text{(CO}_2 \text{ emission [kmol/h])} = \text{(Amount of CO}_2 \text{ per kWh of electricity [kmol/kWh])} \times$$

$$\begin{aligned}
& \text{(Electricity [kW])} \\
& = (0.855 \times 0.454) / 44.01 \times 24510 \\
& = 216 \text{ kmol/h}
\end{aligned}$$

S2-10. Electrolysis of water by electric power system (Reaction enthalpy)

Based on the enthalpy of water production from Section S1-4, the energy required for H₂ production is 54.89 GJ/h. Therefore, the carbon dioxide emissions can be calculated as follows:

$$\begin{aligned}
& \text{(Amount of CO}_2 \text{ converted from electricity [kmol/h])} = \text{(Amount of CO}_2 \text{ per kWh of} \\
& \text{electricity [kmol/kWh])} \times \text{(Electricity [kW])} \\
& = (0.855 \times 0.454) / 44.01 \times (54.89 \times 10^6 / 3600) \\
& = 134.5 \text{ kmol/h}
\end{aligned}$$

S3. Economic Cost

S3-1. Decomposition of cellulose (This study)

From Section 3-8, the cost of our hydrogen production process to produce 228.7 kmol/h of hydrogen is \$78.8M per year.

S3-2. Electrolysis of water by wind power (Electrolysis only, experimental)

When an electrolysis tank is used, the cost of water electrolysis can be calculated as follows, assuming CAPEX is \$988/kW and OPEX is \$40/kW [S10].

$$\begin{aligned}
& \text{(Cost of water electrolysis [\$])} = \{(\text{CAPEX per 1kW [$/kW])} + (\text{OPEX per 1kW} \\
& \text{[$/kW])}\} \times \text{(Power [kW])} \\
& = (988 + 40) \times 200 \times 5123 / 42 \\
& = \$25.1\text{M}
\end{aligned}$$

S3-3. Electrolysis of water by wind power (Electrolysis only, theoretical)

Find the cost of electrolysis of water as follows:

$$\text{(Cost required for H}_2 \text{ production [\$])} = \text{(Cost of raw material of water [\$])} + \text{(Cost of electrolysis [\$])}$$

$$= 228.7 \times 8000 \times 18.02 \times 0.00022 + (210.1 \times 10^6 \times 8000 \times 0.1) / 3600$$

$$= \$46.7\text{M}$$

S3-4. Electrolysis of water by wind power (Reaction enthalpy)

Also, calculate the cost of water calculated from the enthalpy of water production.

$$(\text{Cost for H}_2 \text{ production [\$]}) = (\text{Raw material cost of water [\$]}) + (\text{Cost of electrolysis [\$]})$$

$$= 228.7 \times 8000 \times 18.02 \times 0.00022 + (54.89 \times 10^6 \times 8000 \times 0.1) / 3600$$

$$= \$17.7\text{M}$$

S3-5. Steam reforming of methane (w/ CO₂ capture)

For CO₂ absorption, the cost is 16.5 c€/Nm³ (0.18 \$/Nm³)[S5], which can be calculated as follows.

$$(\text{Cost for H}_2 \text{ production [\$]}) = (\text{Water production [Nm}^3]) \times (\text{LCOH [\$/(Nm}^3)])$$

$$= 4.1 \times 10^7 \times 16.5 \times 1.09 \times 10^{-2}$$

$$= \$7.37\text{M}$$

S3-6. Steam reforming of methane (w/o CO₂ capture)

Consider the cost of the methane steam reforming reaction. Since the hydrogen produced in this process is 41 million Nm³, the cost is 11.4 c€/Nm³ (0.12 \$/Nm³)[S5] without considering CO₂ absorption, which can be calculated as follows.

$$(\text{Cost for H}_2 \text{ production [\$]}) = (\text{Water production [Nm}^3]) \times (\text{LCOH [\$/(Nm}^3)])$$

$$= 4.1 \times 10^7 \times 11.4 \times 1.09 \times 10^{-2}$$

$$= \$5.09\text{M}$$

S3-7. Steam reforming of methane (Reaction enthalpy)

In terms of reactions, consider the cost of steam reforming of methane. Assuming that the feedstock cost of methane is 0.823 \$/Nm³[S11], per Nm³ of methane, we obtain:

$$(\text{Cost required for H}_2 \text{ production [\$]}) = (\text{Raw material cost of methane [\$]}) + (\text{Raw material cost of water [\$]}) + (\text{Furnace cost [\$]}) + (\text{Fuel cost of hexane [\$]})$$

$$\begin{aligned}
&= (228.7 \times 10^3 / 4 \times 8000) \times 0.0224 \times 0.823 \\
&+ (228.7 \times 10^3 / 2 \times 8000) \times 18.02 \times 0.00022 \\
&+ 0.1417 \times 10^6 \\
&+ 1.72 \times 86.18 \times 8000 \times 0.23 \\
&= \$8.8521\text{M}
\end{aligned}$$

S3-8. Electrolysis of water by electric power system (Electrolysis only, experimental)

Same as Section S3-2, assuming that the cost of electricity from wind power is the same as the cost of electricity from the power grid.

S3-9. Electrolysis of water by electric power system (Electrolysis only, theoretical)

Same as Section S3-3, assuming that the cost of electricity from wind power is the same as the cost of electricity from the power grid.

S3-10. Electrolysis of water by electric power system (Reaction enthalpy)

Same as Section S3-4, assuming that the cost of electricity from wind power is the same as the cost of electricity from the power grid.

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[S7]

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