

Hydrogen or batteries for grid storage? A net energy analysis

Supporting Information

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1 Derivation of ESOI of a regenerative fuel cell in terms of operating parameters

Two relations are useful in deriving equation (10) in the main text. First, equation (12) in the main text states that the quantity of energy that exits the electrolyzer is equal to the quantity of energy provided to the fuel cell. It is restated here.

$$\eta_{\text{lyz}} T_{\text{lyz}} P_{\text{lyz}} = \frac{1}{\eta_{\text{FC}}} T_{\text{FC}} P_{\text{FC}} \quad (\text{S1})$$

This may be rearranged to

$$\frac{P_{\text{lyz}}}{T_{\text{FC}} P_{\text{FC}}} = \frac{1}{\eta_{\text{lyz}} \eta_{\text{FC}} T_{\text{lyz}}} \quad (\text{S2})$$

Second, the energy-to-power ratio R is defined in the main text (Equation 10) as

$$R = \frac{S}{P_{\text{FC}}} \quad (\text{S3})$$

Equation (9) in the main text expresses the ESOI_e ratio of a RHFC system in terms of the operating characteristics of the components (electrolyzer, hydrogen gas storage, and fuel cell).

$$\text{ESOI}_e = \frac{T_{\text{FC}} P_{\text{FC}}}{P_{\text{lyz}} \zeta_{\text{lyz,stack}} \left[\frac{T_{\text{lyz}}}{\tau_{\text{lyz,stack}}} \right] + P_{\text{lyz}} \zeta_{\text{lyz,BOS}} + P_{\text{lyz}} \zeta_{\text{comp}} + S \epsilon_{\text{st}} + P_{\text{FC}} \zeta_{\text{FC,stack}} \left[\frac{T_{\text{FC}}}{\tau_{\text{FC,stack}}} \right] + P_{\text{FC}} \zeta_{\text{FC,BOS}}} \quad (\text{S4})$$

We first collect terms in the denominator

$$\text{ESOI}_e = \frac{T_{\text{FC}} P_{\text{FC}}}{P_{\text{lyz}} \left(\zeta_{\text{lyz,stack}} \left[\frac{T_{\text{lyz}}}{\tau_{\text{lyz,stack}}} \right] + \zeta_{\text{lyz,BOS}} + \zeta_{\text{comp}} \right) + S \epsilon_{\text{st}} + P_{\text{FC}} \left(\zeta_{\text{FC,stack}} \left[\frac{T_{\text{FC}}}{\tau_{\text{FC,stack}}} \right] + \zeta_{\text{FC,BOS}} \right)} \quad (\text{S5})$$

and then divide through by $T_{\text{FC}} P_{\text{FC}}$ to obtain

$$\text{ESOI}_e = \frac{1}{\frac{P_{\text{lyz}}}{T_{\text{FC}} P_{\text{FC}}} \left(\zeta_{\text{lyz,stack}} \left[\frac{T_{\text{lyz}}}{\tau_{\text{lyz,stack}}} \right] + \zeta_{\text{lyz,BOS}} + \zeta_{\text{comp}} \right) + \frac{S}{P_{\text{FC}}} \frac{1}{T_{\text{FC}}} \epsilon_{\text{st}} + \frac{1}{T_{\text{FC}}} \left(\zeta_{\text{FC,stack}} \left[\frac{T_{\text{FC}}}{\tau_{\text{FC,stack}}} \right] + \zeta_{\text{FC,BOS}} \right)} \quad (\text{S6})$$

Substituting equation (S3) gives

$$\text{ESOI}_e = \frac{1}{\frac{P_{\text{lyz}}}{T_{\text{FC}} P_{\text{FC}}} \left(\zeta_{\text{lyz,stack}} \left[\frac{T_{\text{lyz}}}{\tau_{\text{lyz,stack}}} \right] + \zeta_{\text{lyz,BOS}} + \zeta_{\text{comp}} \right) + R \frac{1}{T_{\text{FC}}} \epsilon_{\text{st}} + \frac{1}{T_{\text{FC}}} \left(\zeta_{\text{FC,stack}} \left[\frac{T_{\text{FC}}}{\tau_{\text{FC,stack}}} \right] + \zeta_{\text{FC,BOS}} \right)} \quad (\text{S7})$$

or

$$\text{ESOI}_e = \frac{1}{\frac{P_{\text{lyz}}}{T_{\text{FC}} P_{\text{FC}}} \left(\zeta_{\text{lyz,stack}} \left[\frac{T_{\text{lyz}}}{\tau_{\text{lyz,stack}}} \right] + \zeta_{\text{lyz,BOS}} + \zeta_{\text{comp}} \right) + \frac{1}{T_{\text{FC}}} \left(R \epsilon_{\text{st}} + \zeta_{\text{FC,stack}} \left[\frac{T_{\text{FC}}}{\tau_{\text{FC,stack}}} \right] + \zeta_{\text{FC,BOS}} \right)} \quad (\text{S8})$$

Finally, substituting equation (S2) gives

$$\text{ESOI}_e = \frac{1}{\frac{1}{\eta_{\text{lyz}} \eta_{\text{FC}} T_{\text{lyz}}} \left(\zeta_{\text{lyz,stack}} \left[\frac{T_{\text{lyz}}}{\tau_{\text{lyz,stack}}} \right] + \zeta_{\text{lyz,BOS}} + \zeta_{\text{comp}} \right) + \frac{1}{T_{\text{FC}}} \left(R \epsilon_{\text{st}} + \zeta_{\text{FC,stack}} \left[\frac{T_{\text{FC}}}{\tau_{\text{FC,stack}}} \right] + \zeta_{\text{FC,BOS}} \right)} \quad (\text{S9})$$

2 Alkaline fuel cell life-cycle inventory

Table S1: Alkaline electrolyzer life cycle inventory and energy intensity. Adapted from reference 1.

Material	(g/kW)		
	Low ^a	Med ^b	High ^a
Raney nickel	634	808	981
Raney nickel : Nickel	571 ^c	727 ^c	883 ^c
Raney nickel : Aluminum	571 ^c	727 ^c	882 ^c
Silver	373	475	577
Copper (electrodes)	378	482	585
Copper (frame/sealing)	399	745	1091
Additives	22	18.7	35
PTFE	82	105	127
Plastic (interconnects)	636	1025	1413
Plastic (frame/sealing)	2086	2795	3503
Potassium hydroxide	423	608	792
Electricity (kWh)	9.8 ^d	15.1 ^d	20.3 ^d

^aData from reference 1 except as noted.

^bCalculated from data in reference 1 except as noted.

^cAuthors' calculation. Assumes pre-leaching alloy containing 50% by weight each of nickel and aluminum.²

^dIn kWh/kW.

3 Critical literature review of PEMFC stack energy intensity

In order to determine the energy intensity of PEMFC stacks, we critically reviewed the small and disparate literature on life cycle analysis of PEMFC's (Table S2).³⁻⁶ We compared the life-cycle inventories reported by these four studies, normalized to 1 kW of fuel cell electric power, together with the energy intensity of production for each material (Table S3).

Table S2: Characteristics of PEMFC systems analyzed in life-cycle assessment studies.

	Karakoussis <i>et al.</i> 2000³	Pehnt 2001⁴	Primas 2007⁵	Burnham 2012⁶
Application	transportation	transportation	small-scale cogeneration	transportation
Fuel	methanol	hydrogen or methanol	natural gas	hydrogen
Power	70 (kW) _{el}	275 (kW) _{el}	2 (kW) _{el}	54-101 (kW) _{el}
Balance-of-system contribution to energy intensity	9%	– (balance of system not considered)	88% (includes natural gas reforming)	56%

We encountered several methodological inconsistencies among the published studies. For example, the study by Pehnt⁴ does not report the underlying figures for each component material, but only a final value.

The “Other” category in Table S3 encompasses miscellaneous inputs that were not accounted for in all studies. This includes materials such as carbon black, tetrafluoroethylene, polypropylene, and deionized water. The Primas study⁵ also includes additional process inputs such as transport of components by rail or truck and the energy cost of buildings used for manufacture.

Karakoussis *et al.*³ report quantities for several materials that are approximately 1,000 times larger than values reported by others, on a per-kW basis. For instance, the catalyst loadings are approximately 1,000 times larger than each of the other three studies,⁴⁻⁶ and the carbon paper and Nafion loadings are approximately 1,000 times larger than those reported by Burnham.⁶ These values reported by Karakoussis *et al.* for these three materials most likely reflect an order-of-magnitude error.⁷ We have divided these three values by 1,000 for use in our LCI comparison (Table S3).

To determine the fuel cell embodied energy value, we first took the average, for each individual material, of all available values for the required material quantity per power capacity (g/kW). Separately, we determined the average, for each individual material, of all available values for the energy intensity of production (MJ/g). For each material, we took the product of these two values to find the embodied energy (MJ/kW). The sum of these individual embodied energy values is the total energy intensity (MJ/kW). We computed a total energy intensity for the PEMFC stack value of $570 \text{ (MJ)}_{\text{prim}}/(\text{kW})_{\text{el}}$ ($5.7 \times 10^5 \text{ MJ/MW}$).

Table S3: Primary data and selected values for PEMFC life cycle inventory

Component	Karakoussis et al. 2000 ³			Pehnt 2001 ⁴			Primas 2007 ⁵			Burnham 2012 ⁶			Selected for this study		
	g/kW	MJ/g	E_{emb} (MJ/kW)	g/kW	MJ/g	E_{emb} (MJ/kW)	g/kW	MJ/g	E_{emb} (MJ/kW)	g/kW	MJ/g	E_{emb} (MJ/kW)	g/kW	MJ/g ^a	E_{emb} (MJ/kW)
Pt and Ru	1.04 ^b	197	205	6.25	- ^c	- ^c	0.75	288	216	1.46	147 ^{d,e}	214	1.09	211	229
Carbon paper	62.4 ^b	1.4	87.4	- ^c	- ^c	- ^c	-	-	-	73.2	0.723	52.9	67.8	1.06	72
Nafion sheet	79.8 ^b	0.0143	1.14	- ^c	- ^c	- ^c	-	-	-	20.5	0.185	3.79	50.2	0.0638	3.2
Carbon fiber	231	0.4	92.4	- ^c	- ^c	- ^c	5600 ^e	0.0235 ^e	131	920	0.185	170	575	0.292	168
Aluminum	56.3	0.564	31.7	- ^c	- ^c	- ^c	300	0.176	52.8	340	0.141	47.9	232	0.293	68
Steel	29.2	0.0224	0.654	- ^c	- ^c	- ^c	100	0.0766	7.66	22	0.0465	1.02	50.4	0.0485	2.45
Process energy	-	-	5.85	-	-	- ^c	-	-	12.7	-	-	-	-	-	9.25
Other	-	-	1070	-	-	- ^c	-	-	34.6	-	-	2.06	-	-	18.3
Total			1494			5100^f			455			492			570

^a Average of all values available from primary studies.

^b Corrected value (see main text).

^c Value not disclosed.

^d Economic allocation value. Mass allocation value is 0.46.

^e Outlier value.

^f Alternative value with 90% Pt recycling: 1446

4 Derivation of $[\text{EROI}]_{\text{grid}}$

Table S4: Description of energy intensities and energy return ratios.

Symbol	Quantity	Definition	Note
ε_{gen}	energy intensity of generation	(manufacturing energy for generation facility) per (energy generated over lifetime)	$= \frac{1}{[\text{EROI}]_{\text{gen}}}$
$[\text{EROI}]_{\text{gen}}$	energy return on investment of a generation facility	(energy generated over lifetime) per (manufacturing energy for generation facility)	$= \frac{1}{\varepsilon_{\text{gen}}}$
ε_{st}	energy intensity of storage	(manufacturing energy for storage facility) per (energy dispatched from storage over lifetime)	$= \frac{1}{\text{ESOI}_e}$
ESOI_e	energy stored on invested	(energy dispatched from storage facility over lifetime) per (manufacturing energy for storage facility)	$= \frac{1}{\varepsilon_{\text{st}}}$
$[\text{EROI}]_{\text{grid}}$	aggregate energy return on investment of a storage-equipped grid	(sum of energy dispatched directly from generation and from storage facility over lifetime) per (sum of manufacturing energy for generation and storage facilities)	

We consider a simple grid that contains a generation source and storage facility. The generation source has an EROI of $[\text{EROI}]_{\text{gen}}$, and generates a quantity of energy E_{gen} over its lifetime. A fraction of this energy, ϕ , is overgeneration and cannot be fed directly to the transmission grid. This fraction is either curtailed or diverted to the storage facility, which is characterized by an ESOI ratio and a round-trip storage efficiency η_{st} .

The aggregate EROI of the grid decreases as a result of both curtailment and storage. When generation is curtailed, the overall EROI decreases to $[\text{EROI}]_{\text{curt}} = (1 - \phi)[\text{EROI}]_{\text{gen}}$. When generation is stored, the overall EROI decreases to $[\text{EROI}]_{\text{st}}$, whose value depends on both the efficiency and the net energy balance of the storage facility. To find the value of $[\text{EROI}]_{\text{grid}}$, we analyze the embodied energy of the components and the total quantity of dispatched energy.*

*The following derivation is adapted from the corresponding derivation in Barnhart *et al.* 2013⁸. In the present derivation,

The embodied energy of the generation subsystem is the product of the lifetime generation and the energy intensity:

$$E_{\text{emb,gen}} = E_{\text{gen}} \epsilon_{\text{gen}} \quad (\text{S10})$$

The embodied energy of the storage subsystem is the product of the corresponding quantities for storage, modulated by the fraction of total generation which is diverted to storage, ϕ . This reflects our assumption that the storage facility will be fully utilized.

$$E_{\text{emb,st}} = E_{\text{gen}} \phi \epsilon_{\text{st}} \quad (\text{S11})$$

The total embodied energy for the storage-equipped grid is then

$$E_{\text{emb,total}} = E_{\text{emb,gen}} + E_{\text{emb,st}} \quad (\text{S12})$$

Substituting equations S10 and S11,

$$E_{\text{emb,total}} = E_{\text{gen}} \epsilon_{\text{gen}} + E_{\text{gen}} \phi \epsilon_{\text{st}} \quad (\text{S13})$$

or

$$E_{\text{emb,total}} = E_{\text{gen}} (\epsilon_{\text{gen}} + \phi \epsilon_{\text{st}}) \quad (\text{S14})$$

Finally, we restate $E_{\text{emb,total}}$ in terms of EROI and ESOI, using the identities in Table S4.

$$E_{\text{emb,total}} = E_{\text{gen}} \left(\frac{1}{\text{EROI}_{\text{gen}}} + \frac{\phi}{\text{ESOI}_e} \right) \quad (\text{S15})$$

We now consider the total quantity of energy dispatched from the storage-equipped grid. The energy delivered directly from the generation source (i.e. not diverted to storage) is the total quantity generated reduced by the fraction diverted to storage:

equation (S23) below is revised from its earlier version in reference 8.

$$E_{\text{disp,gen}} = (1 - \phi) E_{\text{gen}} \quad (\text{S16})$$

The quantity of energy dispatched from the storage device (i.e. the total energy out from storage) is the product of the quantity diverted to storage (i.e. the total energy into storage) and the round-trip efficiency of the storage device.

$$E_{\text{disp,st}} = \eta_{\text{st}} \phi E_{\text{gen}} \quad (\text{S17})$$

The total quantity of dispatched energy is the sum

$$E_{\text{disp,total}} = E_{\text{disp,gen}} + E_{\text{disp,st}} \quad (\text{S18})$$

Substituting equations S16 and S17,

$$E_{\text{disp,total}} = (1 - \phi) E_{\text{gen}} + \eta_{\text{st}} \phi E_{\text{gen}} \quad (\text{S19})$$

or

$$E_{\text{disp,total}} = E_{\text{gen}} [(1 - \phi) + \eta_{\text{st}} \phi] \quad (\text{S20})$$

Following the general definition of EROI, we state the aggregate EROI of storage-equipped grid as

$$[\text{EROI}]_{\text{grid}} = \frac{E_{\text{disp,total}}}{E_{\text{emb,total}}} \quad (\text{S21})$$

Substituting equations (S15) and (S20),

$$[\text{EROI}]_{\text{grid}} = \frac{E_{\text{gen}} [(1 - \phi) + \eta_{\text{st}} \phi]}{E_{\text{gen}} \left(\frac{1}{[\text{EROI}]_{\text{gen}}} + \frac{\phi}{\text{ESOI}_e} \right)} \quad (\text{S22})$$

or

$$[\text{EROI}]_{\text{grid}} = \frac{1 - \phi + \eta_{\text{st}} \phi}{\frac{1}{\text{EROI}_{\text{gen}}} + \frac{\phi}{\text{ESOI}_{\text{e}}}} \quad (\text{S23})$$

5 Embodied energy contributions under different RHFC scenarios

Table S5 details the embodied energy contributions of the different RHFC system components under the scenarios listed in Table 2 in the main text.

Table S5: Embodied energy contributions of RHFC components.

Scenario	ESOI _e ratio	Total embodied energy (TJ)	Contribution to total					
			Electrolyzer stack	Electrolyzer BOS	Hydrogen compressor	Hydrogen storage	Fuel cell stack	Fuel cell BOS
Reference case	57	10.0	20%	17%	3%	24%	31%	4%
Efficient fuel cell	75	11.3	19%	15%	1%	23%	40%	4%
Low-Pt fuel cell	61	9.3	23%	18%	1%	27%	28%	4%
Composite cylinder	67	8.4	25%	20%	1%	12%	37%	5%
Durable fuel cell	75	11.3	26%	21%	1%	30%	17%	6%
Durable fuel cell with composite cylinder	100	6.6	35%	28%	1%	14%	15%	8%
Four months of storage	3.8	154	1%	1%	<1%	95%	2%	<1%
Four months of storage with epoxy tank	9.2	64.5	3%	3%	<1%	89%	5%	1%
Four months of storage in underground cavern	77	7.4	28%	23%	1%	<1%	43%	6%

6 Embodied energy of materials in lithium ion batteries

Recent analysis have estimated the life-cycle contribution of the different materials incorporated into Li ion batteries^{9,10} (Table S4).

Table S6: Embodied energy of materials in lithium ion batteries.

Battery type		Notter 2010 ⁹		Majeau-Bettez 2011 ¹⁰			
		Li ₂ Mn ₂ O ₄		LiNi _(1-y-z) Co _(y-z) Mn _z O ₂		LiFePO ₄	
		MJ _{eq} ^b	% of total	kg oil-eq ^c	% of total	kg oil-eq ^c	% of total
Materials involved in charge storage	Anode paste	14	14%	0.029	30%	0.017	28%
	Cathode paste	15	15%	0.0044	4.6%	0.0024	4.0%
	Electrolyte	11 ^d	11% ^d	0.0084	8.7%	0.0053	8.7%
	Separator	5	5%	0.0038	3.9%	0.0024	4.0%
	Subtotal		43%		47%		45%
Other materials	Cell container	–	–	0.025	26%	0.016	26%
	Module and casing ^e	28	27%	0.019	20%	0.012	20%
	Cathode substrate ^f	17	16%	0.0046	4.8%	0.0029	4.8%
	Anode substrate ^g	5.2	5.1%	0.0022	2.3%	0.0028	4.6%
	Other inorganics ^h	9.4	9.0%	–	–	–	–
Subtotal		57%		53%		55%	

^aWe omit the following categories included in the life cycle impact analysis in ref. 10: Battery and components manufacture; Battery use; battery management system; electricity consumed by battery.

^bFunctional unit: MJ_{eq} (vehicle km)⁻¹

^cFunctional unit: kg oil-eq (50 MJ discharge)⁻¹

^dSum of embodied energies for ethylene carbonate and LiPF₆.

^e"Battery pack" in ref. 9; "Module and casing" in ref. 10.

^f"Cathode:Aluminum" in ref. 9; "Substrate of negative electrode" in ref. 10.

^g"Anode:Copper" in ref. 9; "Substrate of positive electrode" in ref. 10.

^hLiF, PCl₅, Mn₂O₃, Li₂O₃, concentrated Li brine.

7 Determination of energy intensity of compressed hydrogen storage in underground salt caverns

To estimate the energy intensity of compressed hydrogen storage in underground salt caverns, we first determined the energy requirement per unit volume for cavern preparation. The values of two key quantities have been estimated for underground compressed air energy storage (CAES). Denholm and Kulcinski estimate the overall energy intensity of cavern preparation for CAES as $16.2 \text{ (GJ)}_{\text{prim}}/(\text{MWh})_{\text{prim}}$.¹¹ For the volumetric energy storage density of CAES, we use an approximate value for the Huntorf, Germany CAES plant of $3 \text{ (kWh)}_{\text{el}}/\text{m}^3$.¹² We assume a grid efficiency of $0.30 \text{ (MWh)}_{\text{el}}/(\text{MWh})_{\text{prim}}$ to compute an energy cost of $1.5 \times 10^{-4} \text{ (MJ)}_{\text{prim}}/\text{m}^3$.

To find the energy content of hydrogen per cubic meter under storage conditions, we assume a pressure range of 50 bars between the fully charged and fully discharged states, and a cavern temperature of 298 K. From the ideal gas law, this provides a volumetric density of $2.0 \times 10^3 \text{ (mol H}_2)/\text{m}^3$, corresponding to a volumetric energy density of $4.8 \times 10^3 \text{ (MJ)}_{\text{LHV}}/\text{m}^3$ for compressed hydrogen storage in the underground cavern.

Then,

$$\epsilon_{\text{st}} = 1.5 \times 10^{-4} \frac{\text{(MJ)}_{\text{prim}}}{\text{m}^3} \times \frac{1}{4.8 \times 10^3} \frac{\text{m}^3}{\text{(MJ)}_{\text{LHV}}} \quad (\text{S24})$$

or $3.0 \times 10^{-7} \text{ (MJ)}_{\text{prim}}/(\text{MJ)}_{\text{LHV}}$. Finally, we use the same value for energy efficiency of storage as our reference case, or 88%. The ESOI_e ratio of the system is computed using Equation 11 in the main text. With all other parameters at their reference values (see Table 2 in the main text), the ESOI_e ratio using subsurface storage in caverns is 77.

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