

1 Supplementary Information For:
2 Economically Advantageous Pathways for Reducing Greenhouse Gas Emissions from Industrial
3 Hydrogen under Common, Current Economic Conditions
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5 Hoffmann
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39 Model code is attached in the following files: amount_SA_capEx.m, amount_SA_opEx.m,
40 cheapestNextkWhrs.m, define_sens_anal.m, LC_Bat.m, LC_PV.m, LC_SA.m, LCH_model.m,
41 N212_CDtoM.m, optimize_PV.m, plant.m, SB_CapEx.m, and averageYear_shaner.mat
42 **References**
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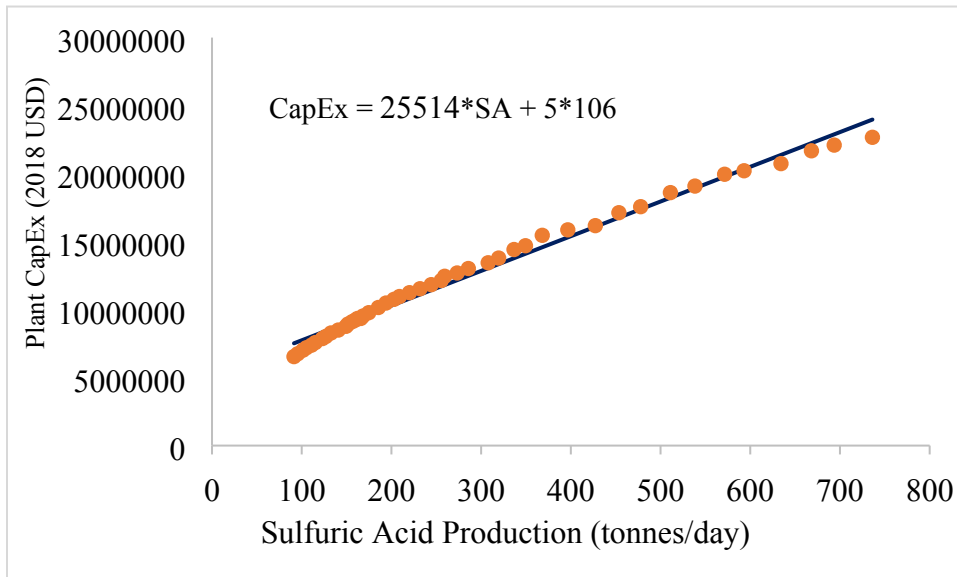
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48 Some relationships below are in 2018 USD, results have been converted into 2020 USD via a standard
49 2% inflation rate for use in the final analysis.

50 **CapEx of Sulfur Furnace**

51 We estimated the CapEx of a sulfur furnace which generates electricity and separates SO₂ from flue gas
52 to be the same as a contact process plant which generates electricity and separates SO₂ from flue gas as
53 well as makes H₂SO₄ thermochemically. We believe that this is a conservative estimate because the
54 contact process has everything that our sulfur burning process needs plus downstream reactors to make
55 oleum from SO₂ and sulfuric acid from oleum. We used a known CapEx and size relationship from
56 Garret, 1989 to estimate the economics of the contact process¹.



57

58 **Fig. S1: CapEx of a sulfuric acid data.** CapEx of contact process plants versus sulfuric acid production
59 rate (orange dotted line), overlaid with the best linear fit (blue solid line)¹.

60 The relationship from the above graph may be found in equation S1. In Equation S1, SA is sulfuric acid
61 production per day in metric tonnes and CapEx is overnight capital expenditure in 2018 USD.

$$62 \text{ CapEx} = 25514 \cdot \text{SA} + 5 \cdot 10^6 \quad (\text{S1})$$

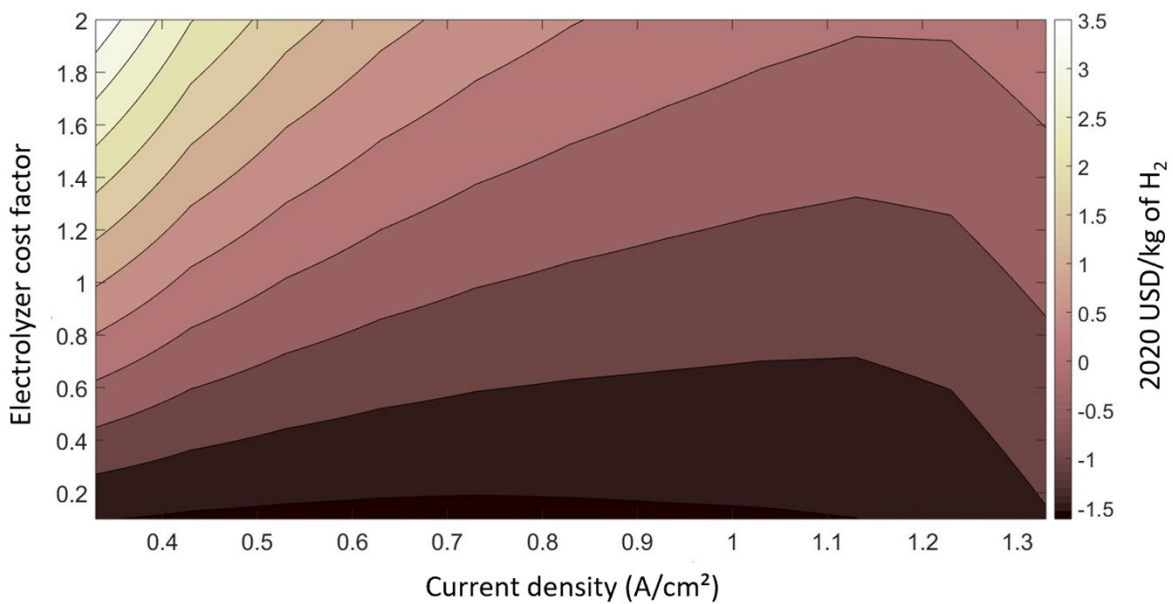
63 A linear fit is conservative because plants tend to get cheaper per unit production with scale.

64 **CapEx and OpEx of Sulfuric Acid Concentration**

65 To determine the cost of electrochemical concentration of sulfuric acid, we modeled a sulfuric acid
66 concentration process using real OpEx and CapEx data for a vacuum concentration process provided via
67 personal correspondence with a senior manager at a Nanjing sulfuric acid vacuum concentration plant
68 owned by Sinopec Nanjing Chemical Industry Co. Ltd (correspondence may be found in the pdf labeled

69 *NanjingCorrespondence_report.pdf*). Data used for CapEx may be found in table S1. This plant
70 concentrates sulfuric acid to 96%.

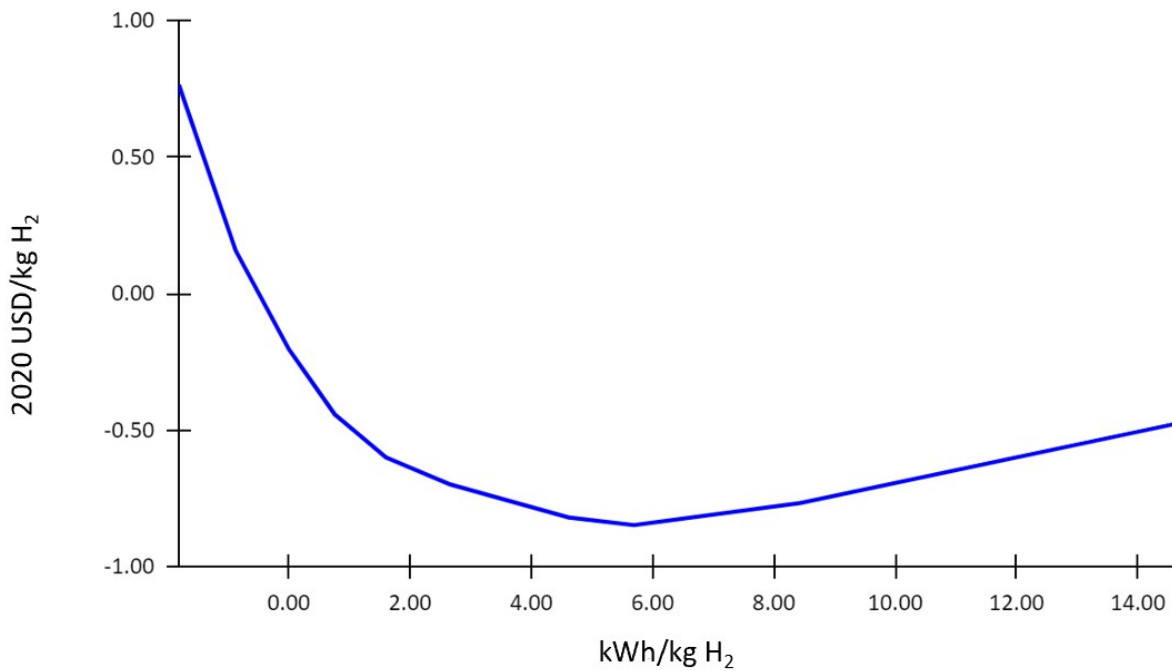
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78 **Fig. S2:** A heatmap of the price of hydrogen for several different operating current densities and
79 electrolyzer CapEx costs. Electrolyzer cost factor is a unitless value which is the multiplication
80 factor on the standard assumed price for an electrolyzer where 0.1 is 10X cheaper than a standard
81 assumption electrolyzer and 2 is 2X more expensive than a standard assumption electrolyzer.
82 Balance of system and catalyst costs per electrolyzer remained constant. Each electrolyzer cost
83 factor corresponds to a different cheapest operating current density.

84



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86

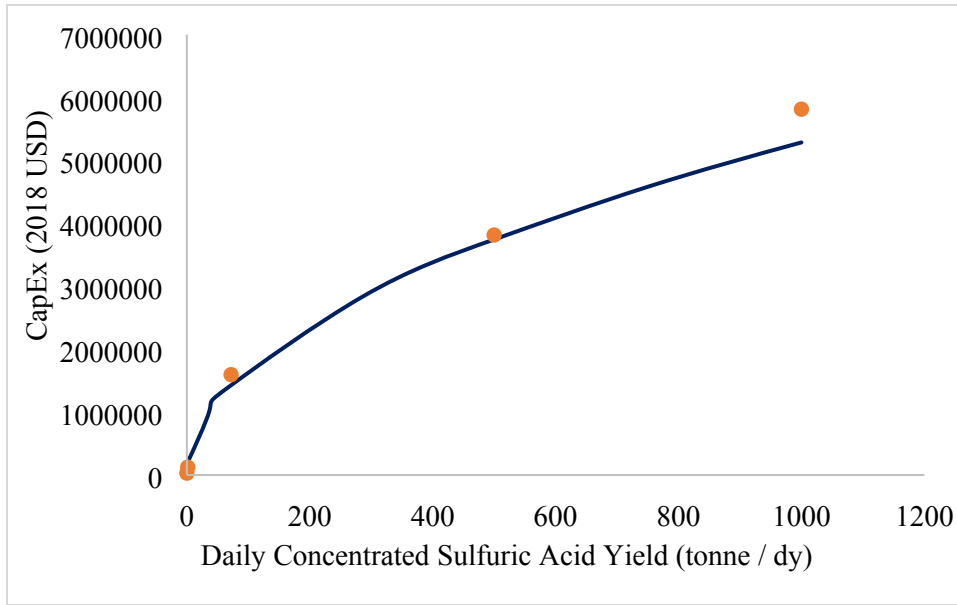
87 **Fig. S3:** Relationship between excess energy required beyond what was available from burning
 88 sulfur assuming a 30% conversion efficiency vs the cost of hydrogen. The primary control on
 89 energy consumption is the operating voltage of the electrolyzer. As operating voltage and current
 90 density (e.g. kWh per kg H₂) decreases, so does energy requirement per kg H₂, but the number of
 91 electrolyzers needed to make a given amount of hydrogen increases.

92

93 **Table S1:** CapEx for sulfuric acid concentration

Plant Size (tonnes H ₂ SO ₄ / dy)	CapEx (2018 USD)
0.018	30,000
1.45	125,000
72	1,595,000
500	3,814,286
1,000	5,814,286

94 Data in table S1 were graphed in fig. S2 to find the empirical relationship in equation S2.



95

96 **Fig. S4. CapEx of Sulfuric Acid Vacuum Concentration Plants.** The best fit relationship between
 97 CapEx and Sulfuric Acid yield for a concentration plant (data from table S1). While this best fit line may
 98 be an underestimate of sulfuric acid concentration CapEx.

99 The relationship between CapEx and OpEx is in Eq. S2 where CapEx is the capital expenditure and SA is
 100 the amount of sulfuric acid produced in tonnes/day.

101
$$\text{CapEx} = 17,000 * \text{SA}^{0.5} \quad (\text{S2})$$

102 OpEx data for this process may be found in table S2 below.

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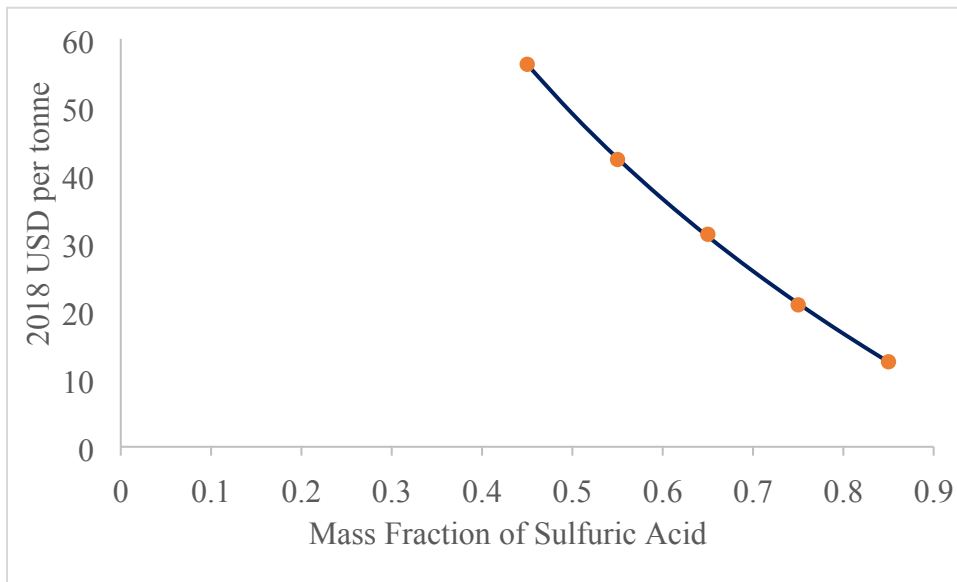
110 **Table S2:** Sulfuric acid concentration OpEx.

Initial Concentration of H ₂ SO ₄ (%)	OpEx (\$ / tonne H ₂ SO ₄)
45	56.25
55	42.25
65	31.25
75	20.88
85	12.50

111

112 Data in table S2 were graphed in fig. S3 to find the empirical relationship in equation S3.

113



114

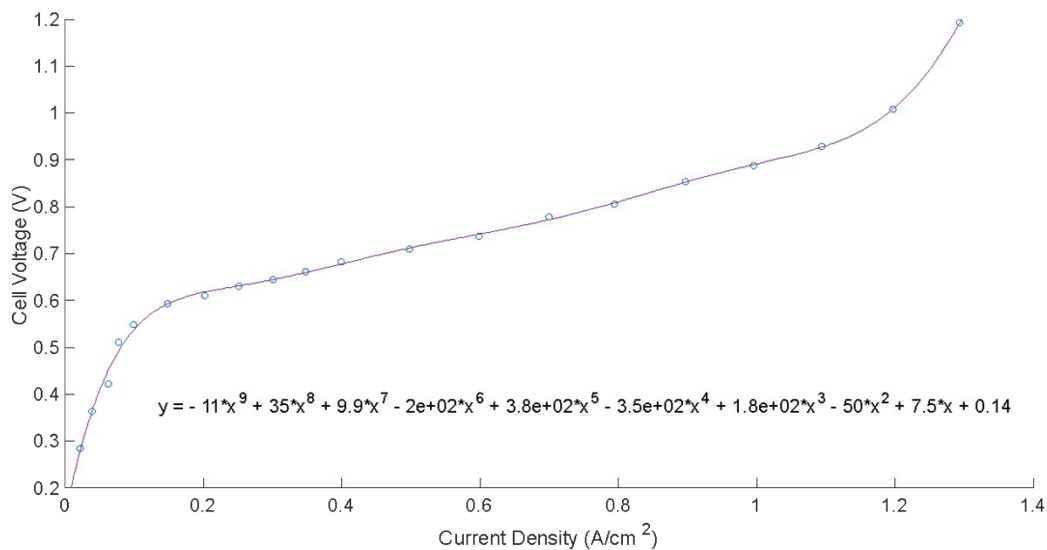
115 **Fig. S5. OpEx of Sulfuric Acid Vacuum Concentration Plants.** OpEx for sulfuric acid concentration
116 from table S2. Sulfuric acid was concentrated from an initial concentration to 96%.

117 The relationship between OpEx and sulfuric acid concentration is in Eq. S3 where OpEx is the OpEx per
118 tonne sulfuric acid and C is the concentration of sulfuric acid in percent mass.

119
$$\text{OpEx} = -1.2443 * C + 118.16 \quad (\text{S3})$$

120 **Voltage and Current relationships**

121 Sulfur and water electrolysis voltage and current density data were fit from real SDE data.



122

123 **Fig. S6. Voltage and Current Relationship for an SDE.** Data interpolation of a real lab scale sulfur
124 depolarization electrolyzer². The interpolated relationship can be found in Eq. S4.

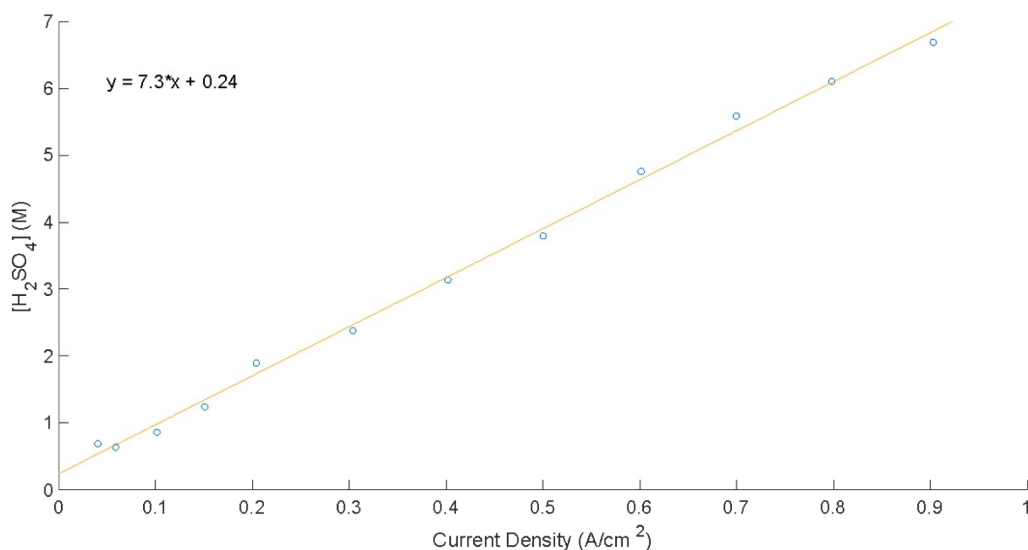
125 The empirical relationship between cell voltage (V) and current density (CD) are in equation S4.

126 $V =$

$$127 - 10.642 \cdot CD^9 + 35.299 \cdot CD^8 + 9.8505 \cdot CD^7 - 200.7 \cdot CD^6 + 379.95 \cdot CD^5 - 346.31 \cdot CD^4 + 175.38 \cdot CD^3 -$$
$$128 49.626 \cdot CD^2 + 7.5476 \cdot CD + 0.13509 \quad (S4)$$

129

130 A voltage shift to account for different starting concentrations of sulfuric acid was used.



131

132

133 **Fig. S7. Graph showing fit of sulfuric acid concentration.** Data interpolation of a real lab scale sulfur
134 depolarization electrolyzer². The interpolated relationship can be found in Eq. S5.

135

136 The empirical relationship between cell sulfuric acid concentration in molar (SA) and current density

137 (CD) may be found in Eq. S6 below. These data were used to determine the concentration of sulfuric acid
138 that was output for a given current density.

$$139 SA = 7.3 \cdot CD + 0.24 \quad (S5)$$

140

141 A voltage shift (ΔV) for different concentrations of sulfuric acid and solubility of SO₂ was applied to the
142 equation based on an empirical relationship of applied potential vs acid concentration derived by Weidner
143 et al, this relationship can be found in Eq. S6 where the shift is cell voltage relative to a water electrolyte
144 is ΔV versus the concentration of sulfuric acid in molar (SA).

145

$$146 \Delta V = 0.00052 \cdot SA^3 - 0.0087 \cdot SA^2 + 0.07 \cdot SA + 0.084 \quad (S6)$$

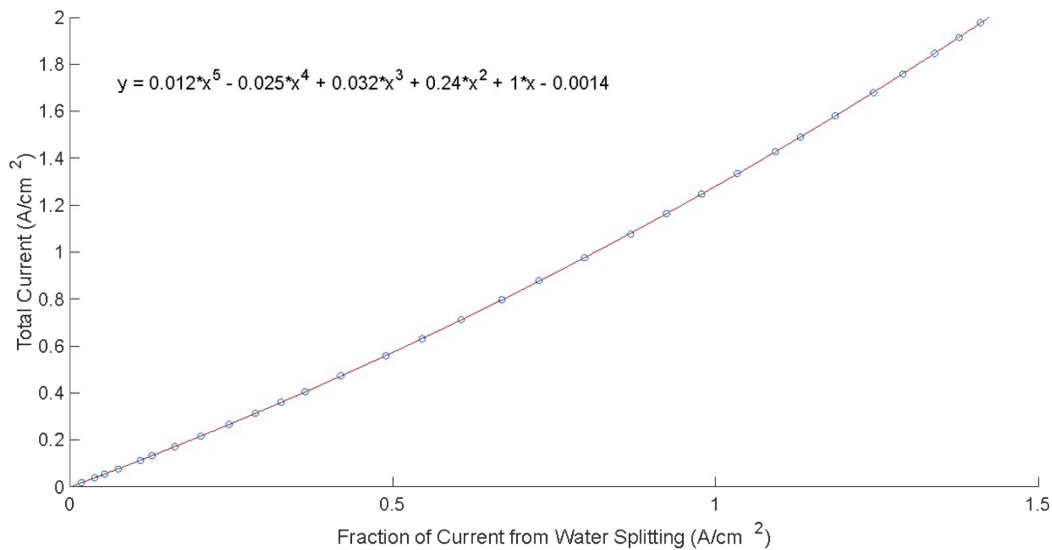
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148 Voltage and Faradaic Efficiency

149

150 For both SE and WE the current density was corrected for faradic efficiency based on voltage from the
151 below graph³.

152



153
154

155 **Fig. S8 Graph showing faradic efficiency for a water electrolysis system.** The fraction of current from
156 water splitting is the total current corrected to have 100% faradaic efficiency.

157

158 **Sensitivity Analysis**

159 **Table S3:** Annotated sensitivity analysis parameters.

160

Variable	Low Value	High Value	Standard Assumption	Discussion
<i>Inputs and Outputs</i>				
H ₂ Produced ^a	1,000	400,000	400,000	1000 kg H ₂ / dy is the H2A forecourt assumption while 400,000 kg H ₂ / dy is the H2A centralized assumption ⁴ .
Buying Price of Sulfur ^c	-0.1	0.45	0.13	This price range was taken from Bloomberg industrial analysis of high and low prices for sulfur. The standard price 2017 West Coast USA contract pricing ⁵ .
Selling Price of Sulfuric Acid ^d	0.05	0.33	0.143	This price range was taken from Bloomberg industrial analysis of high and low prices for sulfuric acid. The standard assumption price was the price for 2017 West Coast USA

				contract pricing ⁵ .
Buying Price of Grid Electricity ^e	0.1	0.3		High and low US energy information administration (EIA) prices. The standard price is the average US industrial price of electricity ⁶ .
Selling Price of Grid Electricity ^e	0.005	0.18	0.07	High and low US energy information administration (EIA) prices ⁶ .
<i>Catalyst Parameters and Components</i>				
SE ^l Catalyst Geometric Current Density ^b	0.33	1.5	1.21	The ranges for SE and WE were taken from real data on either a highly advanced prototype (SE) ² or a commercial prototype (WE) ³ . All values were corrected to 100% faradaic efficiency
WE ^m Catalyst Geometric Current Density ^b	0.33	1.5	1.48	The ranges for SE and WE were taken from real data on either a highly advanced prototype (SE) ² or a commercial unit (WE) ³ . All values were corrected to 100% faradaic efficiency
Voltage Shift ^g	-1.0	1	0	Shifting the voltage for a given current density up or down by a volt represents incredibly large changes in the quality of the catalyst ⁷ .
Catalyst CapEx Cost Coefficient	0.1	1	1	An order of magnitude increase or decrease would be a very large change.
Catalyst Lifetime ⁱ	1	21	7	For the chlor-alkali process, the catalyst lifetime is around 7 years ⁷ . 7 years is also the H2A assumption.

				For the high value we chose 21 years, which, to the knowledge of the authors, has not been attained. This high value would require no membrane replacement during the plant lifetime so therefore it would maximally reduce cost. We believe that showing the maximally impactful value could help scientists understand the maximum potential impact of their research work and therefore motivate their studies. Less than 1 year would be a very unstable catalyst ⁴ .
Catalyst Faradic Efficiency Factor	0.1	10	1	An order of magnitude increase or decrease would be a very large change.
<i>PV Parameters and Components</i>				
PV Nameplate Peak Power ^f	0.1	0.5	0.16	0.16 kW/m ² is a common solar peak power density. 0.5 kW/m ² is a common research goal for PV researchers ⁸ .
PV Balance of Systems Cost Coefficient	0.1	1	1	An order of magnitude increase or decrease would be a very large change.
PV Module Cost Coefficient	0.1	1	1	An order of magnitude increase or decrease would be a very large change.
<i>Battery Components</i>				
Battery Module Cost Coefficient	0.1	1	1	An order of magnitude increase or decrease would be a very large change.
Battery Balance of	0.01	1	1	An order of

Systems Cost Coefficient				magnitude increase or decrease would be a very large change.
<i>Economic and Operating Parameters</i>				
Capacity Factor	0.01	0.97	0.97	A capacity factor of 97% is the H2A assumption ⁴ .
Rate of Return	0.05	0.20	0.12	A 12% rate of return is a high rate of return value that represents a large fraction of equity investment vs debt investment and has been used in previous conservative studies ⁹ .
Carbon Tax ^j	0	1	0.00	0 represents no tax on carbon while 1 represents \$1000/tonne which is very high tax on carbon, most proposed taxes are much lower ¹⁰ .
Carbon Intensity of Grid Electricity ^k	0	1	0.29	0.29 is the carbon intensity of the US grid. 1 kg CO ₂ / kWhr would represent wood burning or very dirty coal based electricity generation ¹¹ .
Initial Sulfuric Acid Concentration ^h	0.0	18.0	10.0	The higher the initial sulfuric acid concentration, the higher the final concentration can be based on the solubility of SO ₂ and the flow rate in demonstrated reactors, 18.0 M H ₂ SO ₄ is 98% sulfuric acid which is the highest grade concentration ¹² .
<i>Electrolyzer Components</i>				
Electrolyzer Balance of Systems Cost Coefficient	0.1	1	1	An order of magnitude increase or decrease would be a

				very large change.
				An order of magnitude increase or decrease would be a very large change.
Electrolyzer Cost Coefficient	0.1	1	1	

161

162 Note. ^a kg / dy ^b A / cm² ^c % / kg ^d \$ / kg ^e \$ / kWhr ^f kW / m² peak power ^g V ^h M ⁱ 7 yrs ^j \$ / kg CO₂ ^k Kg

163 CO₂ / kg kWhr. ^l Sulfur Electrolysis ^m Water Electrolysis.

164

165 CONUS Averages

166 The CONUS spatial average of insolation data introduces limitations because it includes places
167 where solar panels can clearly not be utilized (e.g. National Parks). Additionally, highly sunny
168 places like the US desert Southwest may allow for higher penetrations of solar at a cheaper price
169 of energy while more cloudy places like the east coast of the US may deliver more expensive
170 energy. To address these issues, we varied the cost and efficiency of solar in our sensitivity
171 analysis. Standard assumption values for energy production are shown in table 2.

172

173 Levelized Cost of Hydrogen production via the Steam Methane Reforming and Sulfuric Acid

174 Production via the Contact Process

175

176 CapEx and OpEx data for an SMR plant that produces 341,448 kg/dy H₂ were taken directly from the
177 National Renewable Energy Laboratory H2A model. These numbers were plugged into our LCH equation
178 to determine an LCH from SMR under the same assumptions. The resulting LCH was \$1.25 in 2018
179 USD, ten cents higher than the H2A value. A summary of OpEx and CapEx can be found in Table S4.

180

181 **Table S4:** Tabulated CapEx and OpEx values from the H2A study adjusted to 2018 USD using 2%
182 annual inflation.

183

Plant Parameters		
	Plant Lifetime ^a	40
	Plant Output ^b	341,448
OpEx ^c		
	Variable OpEx	\$9,260,972
	Fixed OpEx	\$65,746,517
	Unplanned Maintenance	\$1,265,773
	Scheduled Maintenance	\$1,289,823
Total OpEx^c		\$77,563,087
CapEx ^d		
	Plant CapEx	\$258,537,342
Total CapEx^d		\$258,537,342

184 Notes: ^ayears ^bkg H₂/day ^c2018 USD/yr ^d2018 USD

185 CapEx and OpEx data were taken from a previous analysis where CapEx was estimated using Fig. S1 and
186 converted into 2018 USD¹³. Wages were converted into an average chemical plant engineer wages in the
187 US and sulfur and sulfuric acid prices and associated laboratory, supervision, and overhead costs were
188 adjusted for consistent assumptions (laboratory costs: 23 % of operating labor, supervision: 20% of

189 operating labor, and plant overheads: 50% operating labor)¹³. We then plugged these CapEx and OpEx
 190 assumptions in eq. 4 from the main text assuming a 12% rate of return. It was found that the levelized cost
 191 of sulfuric acid was \$0.139/kg H₂SO₄ or \$10.36 for the 49 kg H₂SO₄ that would be co-generated with a kg
 192 of H₂ via SMR. Data used for this analysis is shown in table S4 below. All labor costs were scaled by a
 193 ratio of the cost of the 216 tonne/dy H₂SO₄ plant from the reference to the size of the plant we modeled
 194 herein. Consumable materials costs were scaled by the ratio of the produced sulfuric acid. Sulfur prices
 195 were set at \$130/tonne consistent with assumptions in our model.
 196

197 **Table S5:** Tabulated CapEx and OpEx values were adjusted to 2018 USD using 2% annual inflation¹³.
 198

Plant Parameters	
Plant Lifetime ^a	20
Plant Output ^b	9,817
Variable OpEx ^c	
Sulfur (3,200 MTPD)	\$152,106,074
Electricity	\$53,971,581
Heating oil	\$30,487,096
Steam	\$201,524,876
Other Utilities	\$29,160,096
Catalyst	\$2,389,128
Total Variable OpEx^c	\$268,113,977
Fixed OpEx ^c	
Maintenance	\$25,547,094
Operating Labor	\$31,190,845
Laboratory Costs	\$7,173,894
Supervision Cost	\$6,238,169
Plant Overheads	\$15,595,422
Executive Wages	\$925,622
Indirect OpEx ^c	
Indirect OpEX	\$166,892,970
Total OpEx^c	\$253,564,016
Capital Expenditure ^d	
Plant CapEx	\$215,371,424
Catalyst capex	\$40,099,514
Total CapEx^d	\$255,470,938

199 Notes: ^ayears ^btonnes H₂SO₄/day ^c2018 USD/yr ^d2018 USD

200 Comparison to Water Electrolysis

201 We input model conditions from literature into our model and found good agreement with our model and
 202 literature models (Table S6)¹⁴⁻¹⁶.

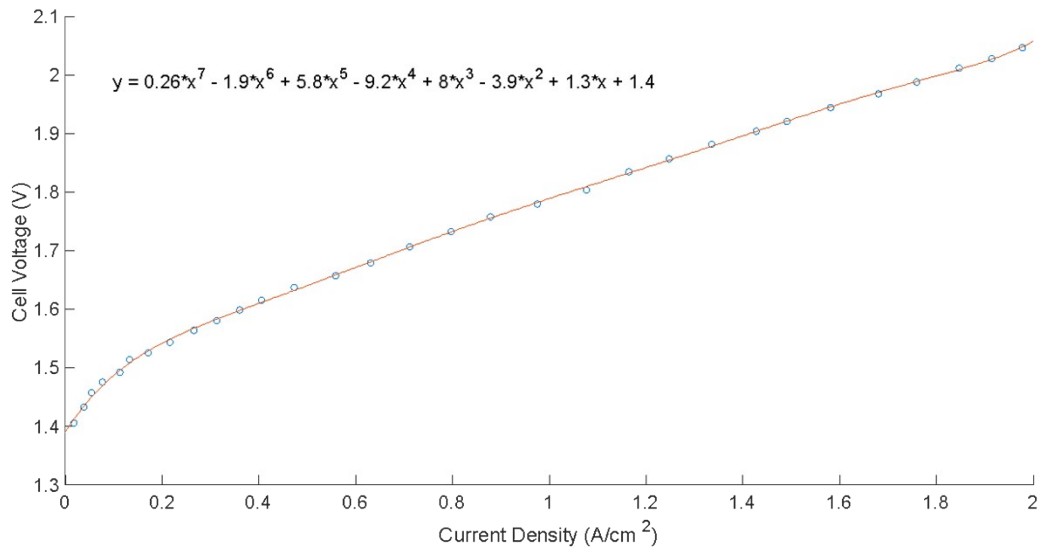
203 **Table S6: Model validation of water electrolysis. Input conditions from other analyses which used**
 204 **regionally specific locations.** CF means Capacity Factor of the plant, r means discount rate, and the
 205 electricity price is the production price of energy less any indicated subsidy.

Condition	LCH from Citation (\$/kg H ₂)	This Paper LCH (\$/kg H ₂)
35% CF, \$0.01/kWh (\$0.023/kWh subsidy), r = 4% ¹⁶	3.23	3.46

99% CF, \$0.024/kWh, r = 10%¹⁵
34% CF, r = 5.75%¹⁴

2.93 3.03
3.60-4.72 3.95-4.94

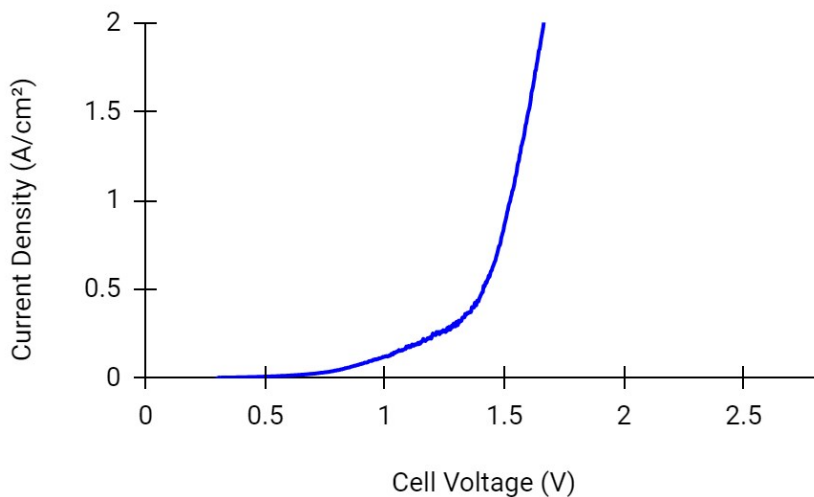
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208 **Fig. S9. Graph of WE Voltage and Current Density Relationship.** Data interpolation of a real
209 industrial scale water electrolyzer³. The interpolated relationship can be found in Eq. S7.

210 The empirical relationship between cell voltage (V) and current density (CD) may be found below.

211 $V = 0.26*CD^7 - 1.9*CD^6 + 5.8*CD^5 - 9.2*CD^4 + 8*CD^3 - 3.9*CD^2 + 1.3*CD + 1.4$ (S7)
212



213

214 **Fig. S10:** Current-voltage curve for a molten sulfur electrolyzer.

215

216 **Key Technical Challenges for SE**

217 *Poor solubility of SO₂ in concentrated sulfuric acid*

218 Reaching the valuable >65% sulfuric acid concentrations purely electrochemically in all demonstrated
219 SDEs known to these authors requires, at some point in the flow cell, dissolution of SO₂ in highly
220 concentrated sulfuric acid^{2,12,17}. The solubility of SO₂ decreases with increased sulfuric acid concentration
221 which leads to increased cell voltages. Engineering controls which increase the activity of SO₂, possibly
222 using a gas diffusion electrode or temperature controls, may further reduce the cost of SE. In this model,
223 demonstrated data was used which accounted for this solubility, however further stability testing may be
224 necessary to determine how important SO₂ concentration is in the long term.

225 *Membrane hydration issues with concentrated sulfuric acid*

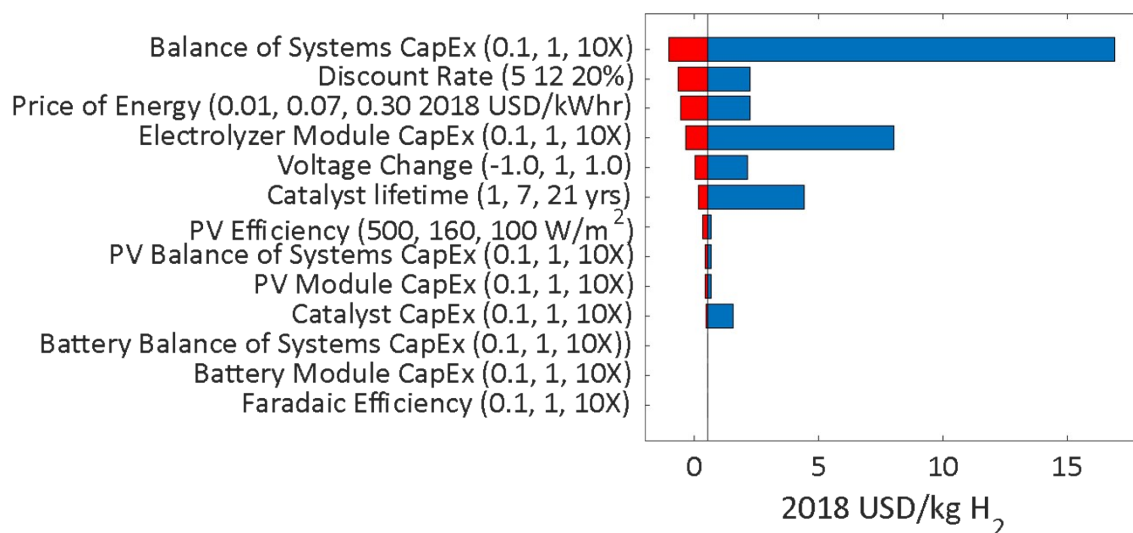
226 When >40% sulfuric acid concentrations exist in the electrolyzer, the membrane which is necessary to
227 prevent wasteful redox shuttling, may dehydrate and therefore become highly resistive. Two strategies
228 have been used in the literature to account for this, one is using membranes that do not depend heavily on
229 hydration (e.g. polybenzimidazole type membranes) or ensuring that the pressure in the catholyte
230 chamber is higher than the pressure in the anolyte chamber thus forcing hydration of the membrane^{18,19}.
231 Production of sulfuric acid in >90% sulfuric acid has been demonstrated in these systems, however, only
232 at very high overpotentials, low current densities, and for short times and more testing and engineering is
233 likely necessary to determine if this is actually feasible therefore we decided to not include the
234 electrochemical production of >90% sulfuric acid in our model²⁰.

235 *Catalyst Fouling with Reduced Sulfur Species*

236 Elemental sulfur may plate on catalysts both reductively and oxidatively. If there is leakage of SO₂ across
237 the membrane, SO₂ may reduce to S_x and block active sites on the cathode. In the anodic chamber, if any
238 (poly)sulfides are present in solution, these may oxidatively plate out on the anode as S_x, again causing
239 fouling²¹. Certainly, membrane integrity and oxygen pressure in the furnace should be controlled
240 precisely to prevent SO₂ in the catholyte and (poly)sulfides in the anolyte. In this analysis we assumed
241 that catalyst fouling was well controlled and catalyst replacement was only necessary once every seven
242 years. No sulfur electrolysis system has been run for this long, however, so we used the chlor-alkali
243 process as a guide. Under these assumptions, our catalyst and membranes were replaced once every seven
244 years.

245 **Sensitivity Analysis of Grid-Assisted Sulfur Electrolysis**

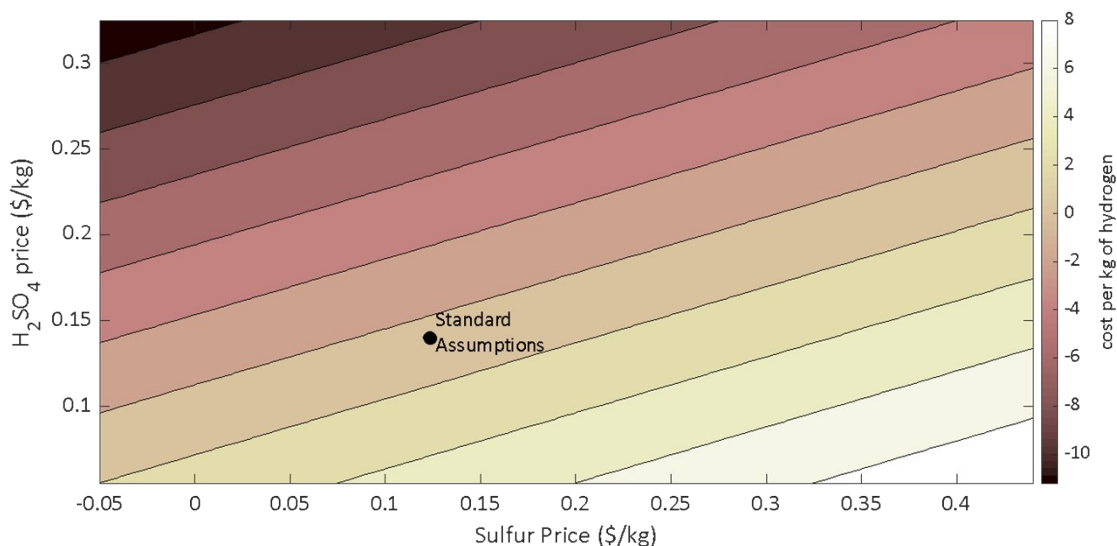
246 Even though grid-assisted SE is cheaper than SMR under standard model assumptions, we applied a
247 sensitivity analysis to understand how technology improvements could impact cost (fig. S11).



248

249 **Fig S11. Sensitivity Analysis of Solar-Only Water Electrolysis.** The parenthetical numbers on the
 250 vertical axis indicate the low, standard, and high assumption for each case.

251



252

253 **Fig. S12. Sulfur and Sulfuric Acid Sensitivity Analysis.** Plot of the price of hydrogen minus a sulfuric
 254 acid credit. Black contours are every \$2. The black circle indicates the price of hydrogen under standard
 255 model assumptions (\$0.52).

256 **Correspondence with Sinopec Nanjing Chemical Company May be found in the pdf labeled:**
 257 **NanjingCorrespondence_report.pdf.**

258 **All code for this model has been uploaded to the repository.**

259

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