Designing an Innovation System to Support Profitable Electro- and Bio-catalytic Carbon Upgrade

Journal:	*Energy & Environmental Science*
Manuscript ID	EE-ART-12-2021-003753.R1
Article Type:	Paper
Date Submitted by the Author:	11-Jan-2022
Complete List of Authors:	Ruttinger, Andrew; Cornell University, Chemical and Biomolecular Engineering Tavakkoli, Sakineh; Johns Hopkins University, School of Advanced and International Studies Shen, Hao; Johns Hopkins University, Chemical and Biomolecular Engineering Wang, Chao; Johns Hopkins University, Chemical and Biomolecular Engineering Jordaan, Sarah; Johns Hopkins University, School of Advanced International Studies; Johns Hopkins University Whiting School of Engineering, Environmental Health and Engineering

Journal Name

ARTICLE TYPE

Cite this: DOI: 00.0000/xxxxxxxxxx

Designing an Innovation System to Support Profitable Electro- and Bio-catalytic Carbon Upgrade†

Andrew W. Ruttinger,[a] Sakineh Tavakkoli,[b] Hao Shen,[c] Chao Wang,[c] and Sarah M. Jordaan*[b]

Received Date
Accepted Date

DOI: 00.0000/xxxxxxxxxx

Carbon capture, utilisation, and storage (CCUS) can mitigate an estimated 14-20% of CO_2 emissions by 2050. We evaluate an integrated electrocatalysis and biocatalysis CCUS pathway, using externally-supplied renewable electricity to convert over 12 $MtCO_2$ to poly(3-hydroxybutyrate) via methanol and formate intermediates. A techno-economic and policy-innovation analysis provides insight into global CCUS innovation. Results suggest that innovation should focus on improving methanol Faradaic efficiency, formate biocatalysis efficiency, electrocatalysis current density, and catalyst performance. Methanol and formate composition influences performance, but higher formate content can improve economics by US$3.8B over a 30 year lifetime. Profitable scenarios hinge on reducing methanol separation costs - through innovation or reduced energy usage - along with cheap renewable electricity and/or carbon pricing. Our evaluation of the global innovation status signifies that coordinated strategies is required to realise emissions cuts from CCUS. Innovation can be spurred by targeted investments and policies that promote emissions reductions.

Introduction

Large-scale carbon capture, utilisation, and storage (CCUS) can play a critical role through our global energy transition to meet the Paris Agreement target of warming well below 2°C above pre-industrial levels.[1,2] With CCUS, an estimated 14-20% of anthropogenic CO_2 emissions, translating to 120-160 metric gigatons of CO_2 ($GtCO_2$), could be avoided by 2050.[3,4] To reach net-zero emissions energy, CCUS is recognized as a solution for even the most difficult-to-decarbonise sectors, namely flexible electricity generation, cement production, and steel production.[5] Importantly, CCUS can be integrated into existing energy systems without new large-scale infrastructure investment.[6] Even with increasing emissions, negative emissions targets become achievable, creating new mitigation pathways and potentially accelerating decarbonisation efforts.[7]

We argue that a comprehensive innovation system approach is required to understand how CCUS can be advanced to the level of widespread diffusion assumed in scenarios compatible with the Paris Agreement. Innovation is anchored in both technology push (*e.g.* research, development, and demonstration (RD&D) investment) and market creation policies (*e.g.* carbon pricing).[8] We focus here on identifying targeted investments to make CCUS competitive, along with the market creation policies that incentivise commercialisation.

CCUS innovation systems should leverage high value markets while decreasing costs. Although carbon capture and storage without utilisation (CCS) is commercially demonstrated technology,[9] low market value of CO_2 and high capture and storage costs inhibit widespread scaling-up in the absence of supporting policy.[10–13] Alternatively, CCUS can offset operation costs through revenue generated from the sale of synthesised products.[14–17] Through displacement of petroleum-based feedstocks at lower cost, market potential may reach US$800B and 7 $GtCO_2$ utilised.[16]

Despite high economic and emissions reduction potential, current techno-economic estimates are nascent, subject to variable technological and market assumptions that make assessment difficult.[18,19] State-of-the-art technologies are often used as baselines despite lack of demonstration in an integrated CCUS system, leading to overly optimistic projections. Policy and macroeconomic drivers are often not considered, despite their documented influence on cost reductions.[8]

To reconcile these challenges with a concrete path towards emissions reduction goals, we develop a framework for the comprehensive techno-economic analysis of early stage CCUS tech-

[a] *Robert F. Smith School of Chemical and Biomolecular Engineering, Cornell University, Ithaca, NY, USA, 14853*
[b] *School of Advanced International Studies, Johns Hopkins University, Washington, DC, USA, 20036; Email: sarahjordaan@jhu.edu*
[c] *Department of Chemical and Biomolecular Engineering, Johns Hopkins University, Baltimore, MD, USA, 21218*
† Electronic Supplementary Information (ESI) available: Spreadsheet with user interface containing economics and design calculations, process emissions calculations, Aspen Plus mass and energy balance, and scenarios evaluated in the present work; detailed information for researchers looking to reproduce the present work, including process description, process assumption, costs, financial assumptions, a user guide for the supplementary spreadsheet, and design and economic calculations. See DOI: 00.0000/00000000.

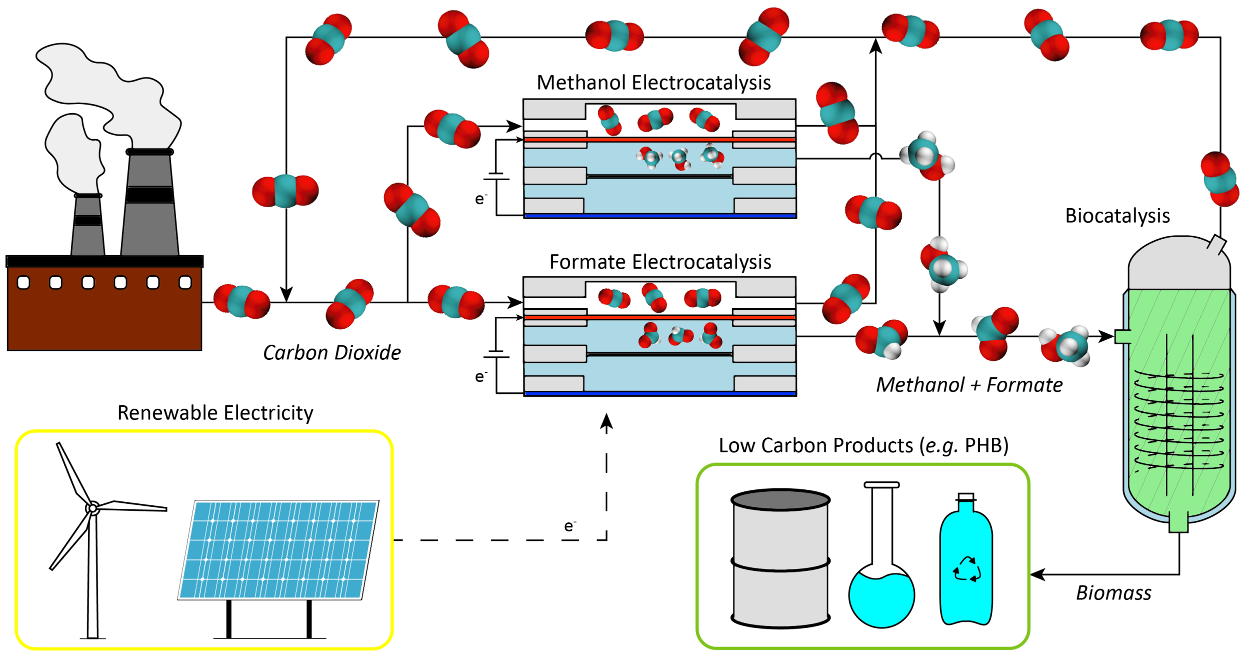

Fig. 1 A scheme of the presented renewably-powered carbon capture, utilisation, and storage process. Here, CO_2 is converted to methanol and formate intermediates during two separate electrocatalysis reactions, followed by conversion of these intermediates to poly(3-hydroxybutyrate) (PHB) and biomass during biocatalysis. Low carbon fuel can also be synthesised through further upgrade of the biomass. More generally, CO_2 can be utilised through this integrated electrocatalysis and biocatalysis pathway, *via* different intermediates, to produce low carbon products such as plastics, chemicals, and fuels.

nologies in the context of the energy innovation system. With annual utilisation potential of 0.2-0.9 $GtCO_2$ by 2050, product synthesis through microalgae pathways offer attractive processes to demonstrate our approach.[20–22] In particular, the integration of electrocatalysis and biocatalysis using microbes engineered to tolerate diverse liquid intermediate mixes can reduce expensive product separation costs in CCUS technologies.[21]

Using this as motivation, we demarcate the constituent and overall pathways of a promising CCUS process: CO_2 upgrade to poly(3-hydroxybutyrate) (PHB) bioplastic *via* methanol and formate intermediates. Taking an innovation system approach, we first identify the technological gap between the current and commercial states of this process. Then, at a level of high technological maturity, we investigate the potential synergies of the co-intermediates to push the process towards commercial profitability. This highlights the critical technological gaps that hinder commercialisation, along with the opportunities through favourable market conditions and process performance. Finally, we position our work in a global context by examining jurisdiction specific indicators that support CCUS from RD&D through commercialisation.

Our examination reflects the integrated nature of innovation, leveraging techno-economic analyses informed by current baselines demonstrated in laboratories to identify RD&D targets and policy levers, which in turn spur technological advancement. We contribute an approach using this technology that is broadly applicable to CCUS innovations. Our global analysis of carbon price, RD&D investment, and infrastructure access in thirty-six jurisdictions – indicators of CCUS maturity - enables scientists and decision-makers to understand and overcome obstacles to commercialisation for early-stage technologies.[23]

Current Reality & Future Potential

Desire to mitigate plastic waste has stimulated bioplastic production, expected to grow to 133 million tonnes in 2024 and largely driven by growth in poly(hydroxyalkanoates).[24] PHB, the simplest poly(hydroxyalkanoate), has properties that make it an attractive candidate to replace polypropylene. Alongside being bio-based and bio-degradable, production is cheaper compared to alternative bioplastics.[25–28] With a current annual production capacity of only 0.1% of polypropylene, scale-up of PHB production to meet the accelerating demand of bioplastics provides an economic opportunity.[26] Although low carbon production of other products such as itaconic acid, succinic acid, and other organic acids are high-value building block materials with growing markets, these products still have limited market sizes compared to low carbon plastics.[24,29] Furthermore, these products do not have the additional benefit of addressing plastic waste on a large scale. Overall, PHB has high market potential with applications ranging from plastic packaging to use in tissue engineering,[30] while advancements in technology, volatile oil prices, and increased environmental awareness create a financial opportunity to shift to PHB-based plastics.[31]

To this end, we assess the commercialisation potential of PHB production from CO_2. While established technology can produce PHB directly from CO_2 using a two-step biological process,[32] production in this approach is slowed by mass transfer of the gaseous carbon to the liquid. Production is further hindered by mass trans-

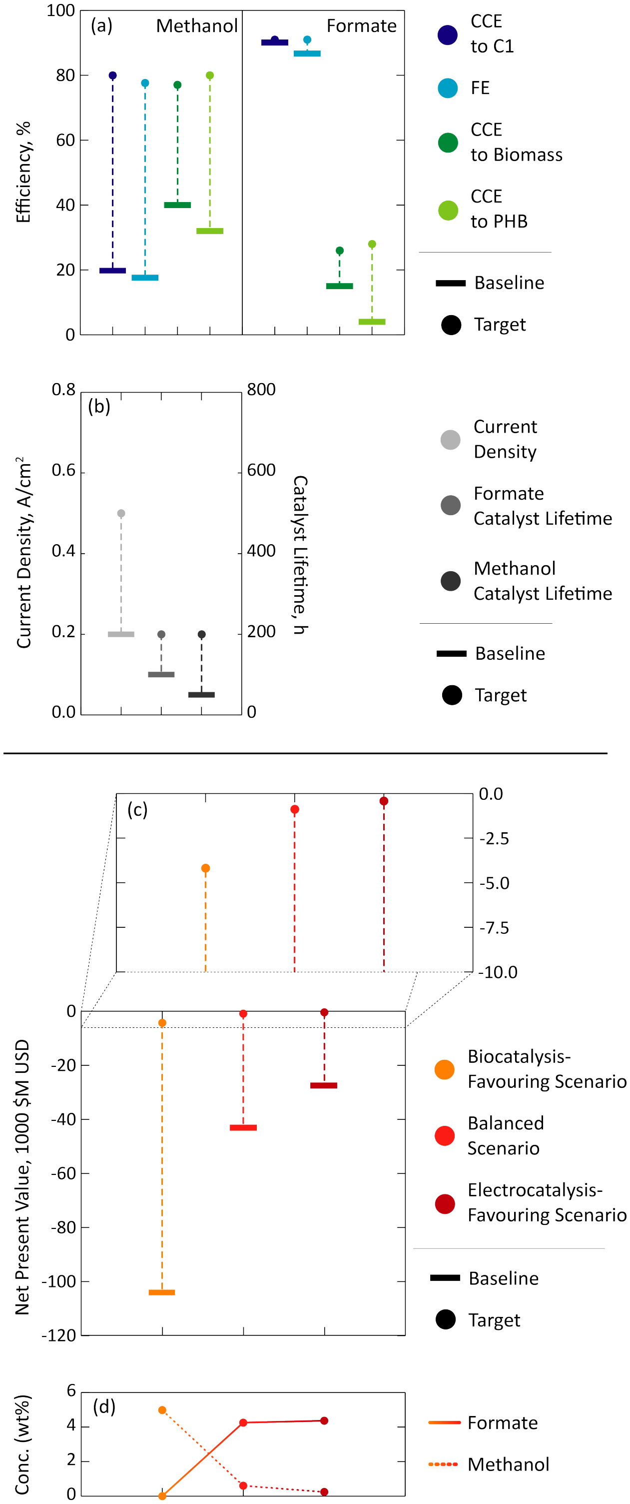

Fig. 2 (a) Difference in the operating efficiencies and (b) operating parameters used for the current technology scenarios (rectangles) and the mature technology scenarios (circles). (c) The difference in net present values for the current technology scenarios (rectangles) and the mature technology scenarios (circles) for three different substrate ratios. An inset is provided for more resolution for the mature technology scenarios. (d) Concentrations of substrates in the target scenarios.

fer of O_2 to the liquid, which is limited to low concentrations to ensure the O_2 and H_2 mixture remains below the explosive limit. For these reasons, as well as the additional costs of gas compression, there are economic challenges associated with this direct CO_2 to PHB pathway. Alternatively, recent research has highlighted the potential of two-step, electrocatalysis and biocatalysis pathways to convert CO_2 to PHB through an intermediate carbon carrier, avoiding the identified barriers of using CO_2 directly.[33,34] These pathways also provide the benefit of process flexibility through the ability to produce PHB from one or more chemical intermediates.

We evaluate such an integrated pathway utilising externally-generated renewable electricity to upgrade CO_2 to PHB *via* methanol and formate co-intermediates, with biomass co-production (Fig. 1).[21,35–38] CO_2 is assumed to be derived from a low cost point source. Methanol and formate serve as a promising chemical intermediate mix due to their high carbon conversion efficiency (CCE) during biocatalysis and electrocatalysis, respectively,[39–41] and their ability to be co-utilized by microbes during biocatalysis.[42] Both are synthesised through CO_2 reduction in separate electrolyzers, mixed together, and fed as substrates for microbial biocatalysis in sequential growth and accumulation bubble column reactors. In each electrolyzer, the CO_2 is selectively reduced based on the catalyst used, with cobalt phthalocyanine and tin oxide catalysing methanol and formate conversion, respectively. Methanol is concentrated up to 5 wt% *via* distillation prior to biocatalysis. PHB is grown and recovered for sale.[30,36,43] Co-produced biomass has value through upgrade using processes like hydrothermal liquefaction for biofuel production.[44,45] Carbon loss is minimised through capture and purification of waste CO_2. A detailed process description, including all process assumptions and operating conditions, are provided in the ESI.

We map the technological maturity to process economics for three different substrate mixtures at an annual commercial production of approximately 100,000 tonnes of PHB. Net present values (NPVs) are estimated for all scenarios using a 30 year economic lifetime in 2019 USD. For this CCUS process, revenue generated from the sale of PHB and biomass, as well as incentives like a carbon price, can offset operating expenses including utilities, feedstocks, maintenance, and labour. This provides an estimate of the potential return on capital investment (*i.e.* profitability) over the plant lifetime. Details on the NPV calculations, costs and financial assumption, and the calculations themselves are found in the ESI.

Scenarios are defined by their stoichiometric methanol-to-formate ratios and named after their relative performance during biocatalysis and electrocatalysis (Fig. 2a), which is specified by the process variables provided in Table S5. This performance trade-off between methanol and formate presents an opportunity to determine intermediate compositions that produce optimal process economics:

- Biocatalysis-favouring Scenario (1:0 methanol:formate): Carbon conversion efficiency is more favourable during biocatalysis due to efficient utilisation of methanol by methantrophic bacteria, relative to formate. Electrocatalysis is inefficient and is energy intensive, requiring six electrons to produce one methanol molecule.

- Balanced Scenario (1:5 methanol:formate): Overall efficiency and energy requirement during electrocatalysis are improved with the synthesis of formate, requiring only two electrons to produce one molecule. Carbon conversion efficiency is lowered during biocatalysis due to poor formate utilisation, leading to a more balanced performance.

- Electrocatalysis-favouring Scenario (1:13 methanol:formate): Carbon conversion efficiency is unfavourable during biocatalysis in the absence of methanol, but electrocatalysis efficiency is greatly improved.

Using this as starting point, we evaluate the economics of each substrate mixture at its current technological state before considering improved economics under some future, mature technological state. A comparison of the relative economics of the constituent electrocatalysis and biocatalysis sub-processes highlights where the gaps between technology performance and economic favourability for each substrate mixture lie, allowing for targeted RD&D to facilitate innovation. In total, process economics of the overall and constituent processes for three substrate mixtures at two technological maturities are evaluated. An additional process which includes hydrothermal liquefaction upgrade of biomass to biofuel is considered in the ESI, but is not included here due to poor process economics.

To complement these techno-economic assessments, we estimate process emissions under all scenarios relative to a business as usual scenario where the CO_2 was emitted at its point source. However, the carbon intensity of PHB production will ultimately depend on the source of the carbon itself. For this reason, we assess the net process emissions under two scenarios. First, we assume CO_2 comes from from a point source. As a result, the overall process is still carbon positive, but the emissions can be lowered if it displaces or avoids higher carbon plastics.[46] Second, we assume CO_2 is sourced from a non-emitting carbon dioxide removal technology. These alternative CO_2 sources such as direct air capture (DAC) and bioenergy CCS (BECCS) could further reduce emissions through negative emission pathways, albeit at higher cost which we estimate at \$100/tCO_2.[12,47] Together these scenarios provide an upper and lower bound of net emissions. Details on the calculation procedure and global emission mitigation potential through product displacement are provided in the ESI.

Economics under all current scenarios suffer from low overall CCE (Fig. 2a), leading to high feedstock throughputs, large equipment requirements, prohibitive energy consumption, and net emissions generated regardless of CO_2 source (ESI). Low current density and short catalyst lifetimes further exacerbate this issue (Fig. 2b). The worst performing process - the biocatalysis-favouring scenario - has a NPV of −104,000 \$M. This is reflective of the current state of CCUS technology, with most projects in the research stage.[18]

Instead of scale-up, focus should be on accelerating technological progression through targeted RD&D investment. Insight from our balanced scenario reveals low efficiency of methanol synthesis during electrocatalysis, coupled with low electrolyzer current density, incurred 68% of capital and 27% of operating expenditure. Low efficiency during biocatalysis creates large methanol requirements upstream, leading to separation costs accounting for 44% of operating expenditure. Additionally, 93% of feedstock costs come from replacement catalysts due to short lifetimes. RD&D efforts focused in these areas create an avenue towards a profitable process (Fig. 2c). Under these mature technology scenarios, NPVs and net emissions range from −4,180 \$M and 6.2-10.0 tCO_2/tPHB for the biocatalysis-favouring scenario to −420 \$M and 0.1-3.9 tCO_2/tPHB for the electrocatalysis-favouring scenario. As reflected here, sourcing CO_2 from DAC reduces net emissions by 3.8 tCO_2/tPHB but correspondingly lowers the NPV by 280 \$M. Improved process efficiencies underlie the dramatic improvements and are reflected in less recirculated carbon, more streamlined expenses, and smaller process equipment (Fig. 3).

A Balancing Act

Juxtaposing our scenarios, the methanol-rich biocatalysis-favouring scenario exhibits a CCE of 62%, over twice the 27% CCE for the formate-rich electrocatalysis-favouring scenario (Fig. 3d-e). While methanol has been demonstrated as a good substrate during biocatalysis, formate is poorly utilised, resulting in high generation of undesirable CO_2.[40] Despite this, economics and emissions analyses suggest higher benefits for the electrocatalysis-favouring scenario. Harnessing this relationship between economics, emissions, and substrate allows us to demonstrate a modular process, adaptive to market risks and uncertainties. Then, reduction of process risk while simultaneously mitigating emissions can encourage private investment.

We perform techno-economic analyses for the constituent electrocatalysis and biocatalysis processes in each scenario (Fig. 4a-c). Our results appraise findings from prior studies that showed electrocatalysis, in general, produces formate at higher selectivity and Faradiac efficiency compared to methanol, while biocatalysis utilizes methanol as a substrate more efficiently than formate.[35,40,48–50] This is further supported by our experimental electrocatalysis studies (ESI). For the biocatalysis-favouring scenario, the biocatalysis subprocess generates 1,920 \$M but is offset by the electrocatalysis subprocess, which loses −7,870 \$M. Costs are driven by high electricity requirements, electrolyzer capital costs, and methanol separation utility consumption. Conversely, the electrocatalysis-favouring scenario loses −3,480 \$M from the biocatalysis subprocess but generates 970 \$M from the electrocatalysis subprocess. Here, electricity and electrolyzer costs decrease in exchange for high formate feedstock requirements. Despite reduced methanol throughput in this scenario, separation costs remain significant, signifying a need for bypassing energy-intensive methanol separation. While there is an economic benefit to utilisation of formate, practical considerations remain such as under-developed formate assimilation routes limiting the use of formate as a viable intermediate.[51] At this stage, RD&D targeted at engineering formate-tolerant microbes is critical for innovation. Alternative viable carbon substrates, including syngas, acetate, and ethanol, could uncover more practical and economic pathways.[14,21] Targeted RD&D towards identification of effective carbon intermediates can broadly stimulate the development of microalgae CCUS pathways.

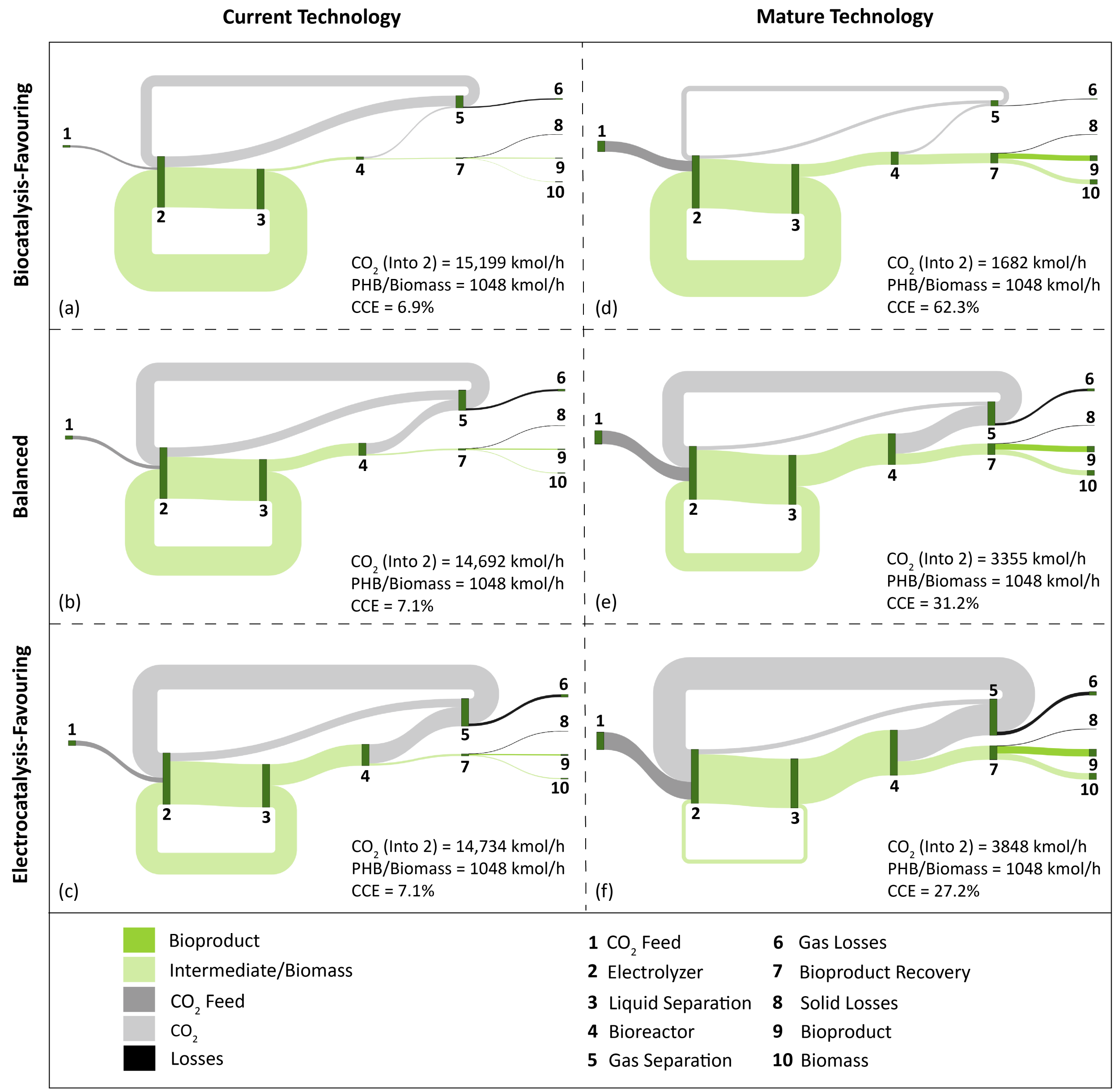

Fig. 3 Sankey diagrams demonstrating the flow of carbon through the process for each technological scenario (left vs. right) and substrate ratio (top vs. middle vs. bottom). Nodes are marked with numbers provided in the legend at the bottom right. The carbon species is denoted by shading provided in the legend at the bottom left. Carbon conversion efficiencies are provided for each scenario. CCE = carbon conversion efficiency.

A Pathway to Commercialisation

By undertaking techno-economic analyses to identify important areas in need of RD&D - such as the integrated process examined here - CCUS innovation can yield mature technologies. At this stage, key parameters, most prominently electricity, steam, and carbon pricing, dictate economic outcome (Table 1 and Fig. 4d-e). For the biocatalysis-favouring scenario, the electrocatalytic conversion of CO_2 to methanol is energy-intensive due to low efficiency coupled with the requirement of six electrons per methanol molecule. As a result, significant electricity input is required which results in high sensitivity to electricity price. High steam demand due to low concentration methanol during separation has a similar effect. Except under extremely optimistic conditions, the biocatalysis-favouring scenario is likely to be unprofitable. However, decreased electricity and steam demands under the balanced and electrocatalysis-favouring scenarios create opportunities for profit. Importantly, carbon pricing has the potential to shift NPVs to positive values, creating an environment where governments can encourage private investment.

Reduced methanol separation cost is imperative for profitability in the presented process and can be complemented by low electricity or high carbon prices (Fig. 5). Namely, the balanced scenario with 50% methanol separation cost and a carbon price over \$75/tCO_2 is profitable. Alternatively, electricity alone can achieve this target if available at \$0.02/kWh. The International Renewable Energy Agency (IRENA) estimates that solar and wind electricity could reach \$0.02/kWh by 2030.[52] The electrocatalysis-

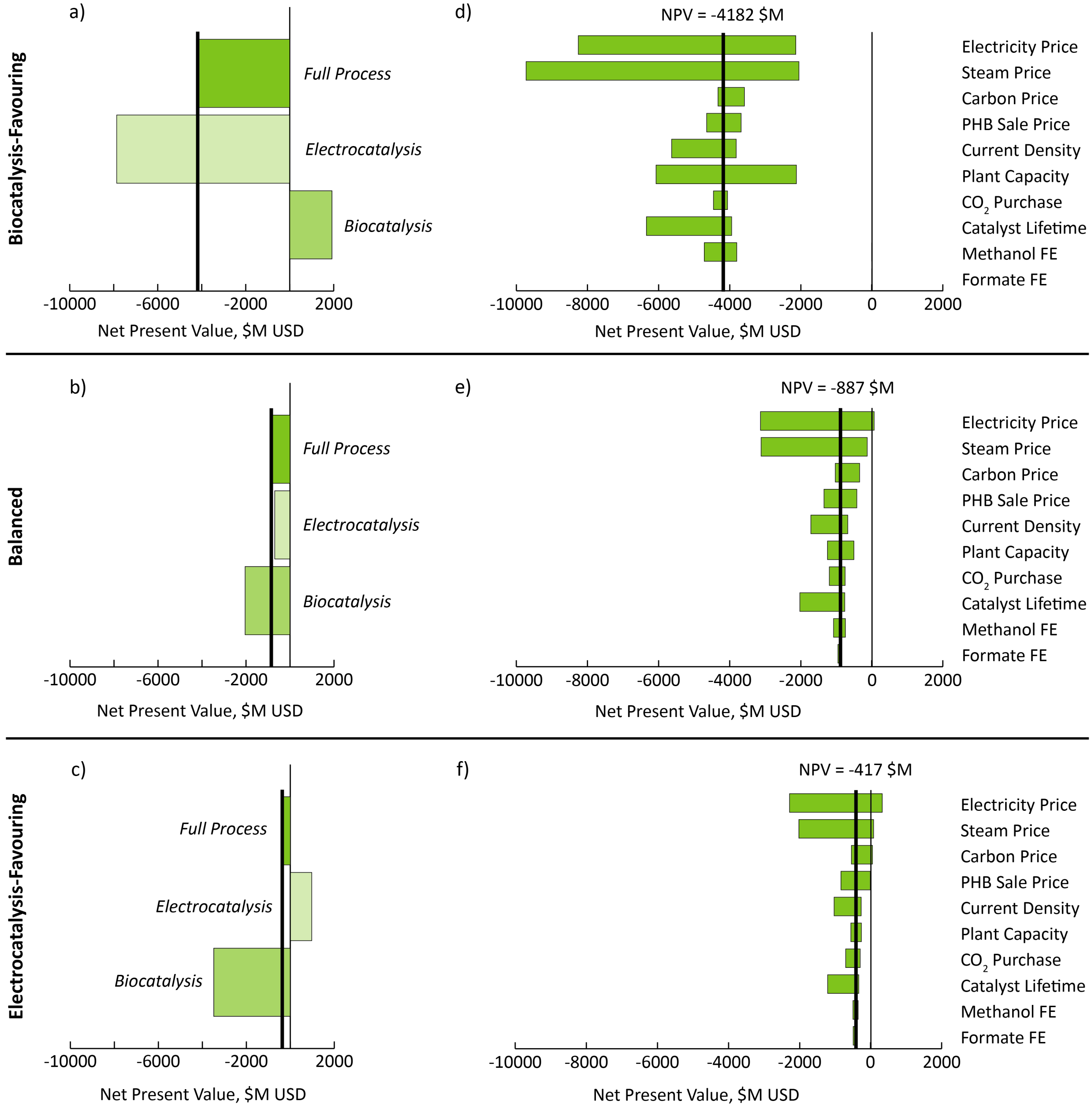

Fig. 4 Left, a-c. Breakdown of the overall NPV for the electrocatalysis and biocatalysis sub-processes for each substrate ratio, compared to the full process. Right, d-f. Sensitivity of the NPV for the full process to key technological and market parameters for each substrate ratio. The substrate ratio for each section is provided in bold on the left of the figure. FE = Faradaic Efficiency.

Table 1 Range of values for sensitivity analysis under mature technological conditions.

Sensitivity Parameters	Worst	Baseline	Best
Electricity Price ($/kWh)	0.05	0.03	0.02
Steam Price ($/kg)	0.05	0.022	0.011
Carbon Price ($/t$CO_2$)	0	40	200
PHB Sale Price ($/tonne)	4000	4482	5000
Current Density (mA/cm^2)	200	500	800
Plant Capacity (tPHB/year)	50,000	102,784	150,000
CO_2 Purchase ($/t$CO_2$)	100	30	0
Catalyst Lifetime (h)	24	200	1000
Methanol Faradaic Efficiency (%)	65	78	90
Formate Faradaic Efficiency (%)	91	97	99

favouring scenario, even without methanol separation cost reductions, can generate a profit with carbon prices above $180/t$CO_2$ or electricity near $0.023/kWh. Profits are also possible under baseline parameters at 57% methanol separation cost.

While distillation of low concentration methanol is energy-intensive, other separation methods also face challenges. Aside from higher costs, they are not universally available, tend to have lower throughputs, and often have hidden energy costs.[53] Distillation also plays a practical role through recovery and recirculation of the electrolyte, avoiding additional feedstock costs. Still, reduction of methanol separation cost is possible. At the U.S. 2019 average natural gas price of $3.91/1000 ft^3,[54] steam gen-

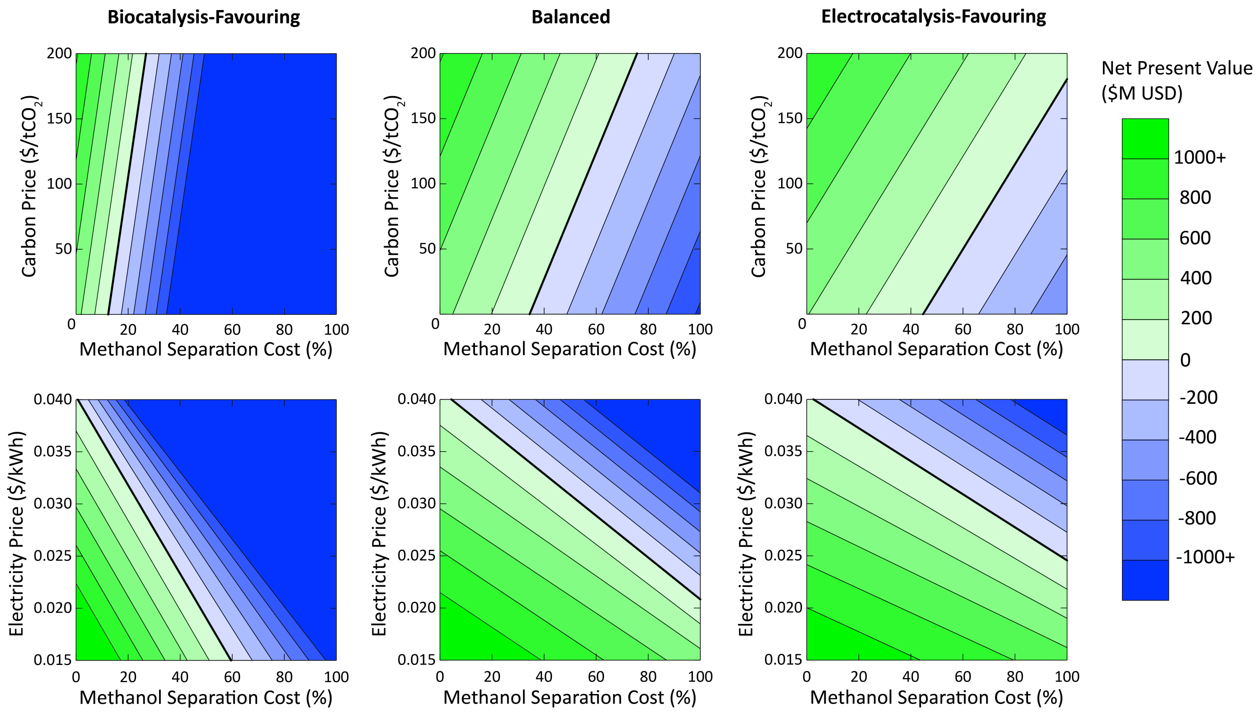

Fig. 5 The NPV of the integrated process as a function of liquid separation cost and carbon price (top) and liquid separation cost and electricity price (bottom). These scenarios are provided under mature technological conditions for each substrate ratio (low-medium-high, left to right). The key for shading is provided in the legend.

erated with natural gas rather than renewable electricity reduces methanol separation costs over 50%. Such a reduction under the electrocatalysis-favouring scenario yields a profit of 80 $M. While this would create an additional 7.4 $tCO_2/tPHB$ in emissions, these short-term economic benefits can encourage CCUS commercialisation while jurisdictions transition to carbon neutral energy sectors. As renewable electricity becomes cheaper, it can replace natural gas as the steam generating utility. Once less energy-intensive separation techniques are demonstrated at scale, methanol separation costs can be further reduced.[53] Ultimately though, development of electrocatalysis processes that can produce and withstand high concentration methanol will preclude the need for methanol separation entirely. Considering this case for the presented process, production of 5 wt% methanol yields profits of 580, 580, and 520 $M for the biocatalysis-favouring, balanced, and electrocatalysis-favouring scenarios, respectively. While the biocatalysis-favouring scenario would still generate net emissions even with CO_2 sourced from DAC, up to 0.4 and 1.1 $tCO_2/tPHB$ would be avoided for the balanced and electrocatalysis-favouring scenarios, respectively, relative to simply emitting the CO_2. The additional cost of DAC would reduce the NPVs to 380, 340, and 270 $M for the biocatalysis-favouring, balanced, and electrocatalysis-favouring scenarios, respectively. Complementary climate initiatives like electrification and hydrogen fuels will further improve these carbon footprints in the future, relative to hydrocarbon-derived plastic. Such favourable economics and carbon footprints emphasize the promise of integrated electrocatalytic and biocatalytic CCUS processes.

Global Policy Innovation & Trends

The eventual commercialisation of CCUS technology, once economically competitive, will depend on jurisdiction-specific policy frameworks and a favourable investment environment. Three enabling mechanisms are key indicators of maturity and progress towards large-scale deployment.[23]

First, carbon prices signal government commitment to reducing emissions.[23] Government policy can drive innovation through market pull that ensures the value of CO_2 is proportional to capture costs.[8] By June 2021, 64 total carbon pricing schemes were implemented or planned for implementation globally.[55] Prices range from $1-$137/tCO_2 and cover 12 $GtCO_2$ equivalent emissions.[55]

Second, CCUS innovation can be enabled through frameworks that provide funding with minimal debt financing.[23] While project risks remain high in the RD&D stages, government investment can advance innovation through technological push.[8,23] Consequently, this push can help lower deployment costs, facilitate sector growth, and meet emissions reduction targets.[1] Globally, to meet the IEA BLUE Map scenario which assumes 145 $GtCO_2$ stored, a total investment of 2.5-3 $T in CCUS is required from 2010-2050.[3]

Third, infrastructure access significantly reduces the barrier of CCUS market diffusion.[23] We argue for the inclusion of infrastructure that supplies low-cost electricity, having demonstrated that cheap electricity can support market competitiveness. With renewable electricity leading growth and solar photovoltaic becoming the cheapest source of electricity in most countries, jurisdictions with low-hydrocarbon electricity generation are posi-

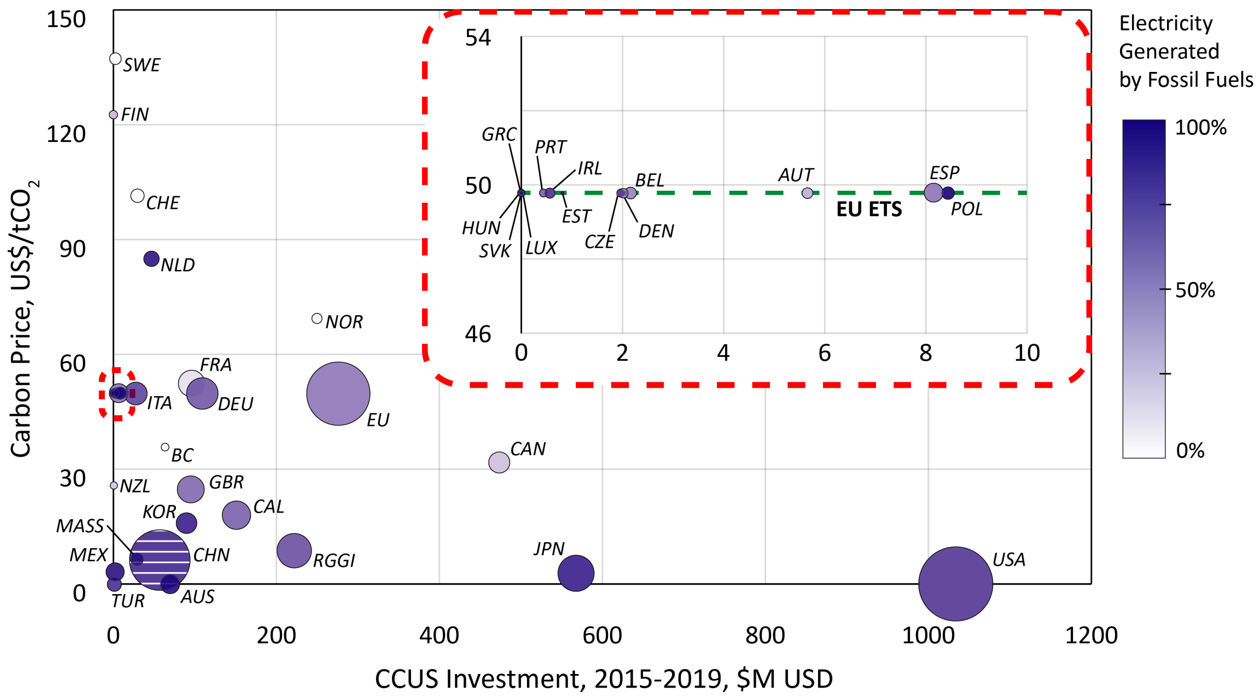

Fig. 6 2021 carbon pricing and CCUS RD&D investment between 2015-2019 are provided on the axes, while bubble intensity denotes the 2018 percent of electricity generated by hydrocarbons. 2019 Gross Domestic Product (GDP) is provided by bubble size to highlight any relationships with the three indicators. CCUS RD&D investment by sub-national jurisdictions was estimated using percent of national GDP. CCUS investment data from China pertains to the period of 2010-2015 due to limited data, indicated by a striped bubble. For countries under the European Union carbon price, the reported carbon price was the higher of the national carbon pricing scheme or European Union carbon price, unless it was explicitly specified that both apply. Carbon prices as of June 2021 were used in our analysis. China also has a national carbon price implemented in 2021 but does not provide details of a price. Instead, the carbon price in Shanghai is used, since it was the highest in the country. Scope of emissions is not considered in this analysis. An inset is provided for data points near the origin. All references for data is provided in the Methods.

tioned at an advantage for CCUS technology.[56]

We examine the global status of CCUS innovation in thirty-six jurisdictions to understand the present commercialisation supports (Fig. 6). Our findings suggest an inverse correlation between CCUS RD&D investment and carbon price. This trend also correlates well with gross domestic product (GDP). Seven of the ten countries with the highest GDPs invested 90 $M or higher in CCUS RD&D, but only France, Germany, and Italy have a carbon price above $40/tCO_2$, recommended by Stiglitz and Stern.[57] None of the five countries with the highest carbon prices had a GDP above 1 $T and only Norway invested above 90 $M. Carbon price tends to correlate with low-hydrocarbon electricity generation. Three of the five jurisdictions with the highest carbon prices have under 2% hydrocarbon usage and another is at 20%. Netherlands, the second largest European producer of natural gas, is the outlier with 78% electricity generation from hydrocarbons. The present reliance on hydrocarbons may change with a national carbon price that began in 2021. Eight of the thirteen jurisdictions that generate over 60% electricity from hydrocarbons have a carbon price below $20/tCO_2$. This includes all three countries with the highest GDPs. Instead, these countries provide strong technological pushes, with United States and Japan providing the highest investment in CCUS technology and China having twenty-two CCUS facilities in development or operation.[58] High global variability among these CCUS indicators emphasizes the importance of evaluating innovation on a country-level to identify gaps in and opportunities for supporting government policy.

Global CCUS Status & Potential

With mature technology and 50% methanol separation costs, our balanced scenario provides a carbon price threshold of $75/tCO_2$ for break-even investment costs. Currently, only three jurisdictions, led by Sweden, meet this threshold. Carbon prices should be increased globally to ensure sufficient market pull and economic incentive for CCUS commercialisation. Ireland, Canada, and Germany are set to exceed this threshold with carbon prices climbing to $117/tCO_2$, $131/tCO_2$, and up to $76/tCO_2$, respectively.[55] While the United States does not have a carbon price, it currently offers a $24/tCO_2$ tax credit for carbon utilisation, rising to $35/tCO_2$ in 2026.[23] When combined with its state level carbon price, California currently provides $42/tCO_2$ in credits. The European Union carbon price should eventually exceed $75/tCO_2$ as its carbon allowances are reduced annually, helping stimulate more investment in low-carbon technologies.[55] The United Kingdom should follow this trajectory, having implemented a carbon price similar to the European Union's, but with a tighter emissions cap.[55] South Korea also has a tighter emission cap as of 2021.[55]

Complementary to carbon pricing, RD&D investment can lower the carbon pricing threshold that makes CCUS technology economically accessible. In this regard, the United States, Japan, and Canada lead with investments of 1,034 $M, 568 $M, and 473 $M from 2015-2019, respectively. However, Norway invests over twice the fraction of their GDP in CCUS than the next best country – Canada – and over twelve times the average, for a total of 250 $M invested. The European Union contributed 276 $M, providing funding benefits for member countries that lack strong national innovation supports. While incomplete data makes assessing true investment from China difficult, they have identified CCUS as an important player in meeting their emissions targets and have pledged investment here during their 13th five-year plan.[58] Regarding access to renewable electricity, Sweden, Switzerland, Norway, and British Columbia have less than 2% hydrocarbon electricity generation. Notably, Canada, with a carbon price set to rise above our $75/tCO_2$ threshold, has less than 20% hydrocarbon electricity generation. While the United States lags with 63% hydrocarbon electricity generation, geographical and state-level diversity provides more favourable areas for CCUS development, including California with 48% hydrocarbon electricity generation. Japan is significantly behind in this area, catalysed by a rapid transition to largely hydrocarbons after the Fukushima nuclear disaster.

To support CCUS as it becomes more competitive, all jurisdictions should increase the scope of emissions coverage in their carbon pricing schemes. Only seventeen schemes account for over 50% of the jurisdiction's emissions and tend to focus on power and industry.[55] In order to monitor global CCUS RD&D progress, jurisdictions should provide more transparent and detailed data to better align RD&D with target investments such as those we identify. India, for example, is a key player in global emissions reduction and shows signs of CCUS RD&D investment, but lacks sufficient data to be included in our analysis.[9] Three of the four leading countries in CCUS investment are notable oil producers, with Japan being the exception. In these cases, RD&D investment may be directed at enhanced oil recovery and should be disaggregated from other utilisation processes.

Conclusions & Recommendations

Reaching our global emissions reduction targets requires unprecedented contributions from all stakeholders, but governments play the decisive role.[56] By contextualising a traditional techno-economic analysis within a policy framework, we provide a novel, broadly applicable approach to characterise the interplay of technology and policy for CCUS innovation. We follow this approach in the present study, assessing an integrated electrocatalysis and biocatalysis CCUS pathway that converts CO_2 to PHB *via* co-utilisation of methanol and formate intermediates. While progress is being made, our analysis demonstrates the wide gap between current and required technology/supporting policy. By addressing identified priority areas, governments can take a more strategic and coordinated innovation systems approach to create a favourable investment environment.

Juxtaposing the process economics using laboratory baselines (current technology) and state-of-the-art performance indicators (mature technology), we find a significant innovation gap that stems from prohibitively low carbon conversion efficiencies at present. Addressing these performance gaps improves the economics to a situation where commercial viability is possible. Therefore, in the short-term, innovation should focus on technological push through targeted CCUS RD&D investment. Priorities for our examined pathway include efficiency of methanol electrocatalysis, methanol separation, and formate biocatalysis. Additional efforts should improve electrocatalysis current density and catalysts.

Under mature conditions, variation of substrate composition provides flexibility to mitigate market risk and develop adaptive processes. Our assessment of different methanol to formate ratios highlights that methanol performance is marred by energy intensive electrocatalysis, while low biocatalysis efficiency hinders formate performance. Despite this trade-off, our assessment shows worse economics for methanol-rich substrate mixes due to the high energy requirement during methanol/water distillation to concentrate the methanol. Only under optimistically low electricity prices or high carbon pricing with reduced methanol separation costs is this methanol pathway profitable. At this stage, efforts to produce high concentration methanol during electrocatalysis that precludes the need for further concentration is the critical step for commercial viability. Formate-rich substrate mixes show improved economics and can be made profitable through favourable carbon pricing and electricity pricing environments. The critical step for this pathway, then, will be the engineering of microbes to tolerate and utilize higher concentrations of formate. These technological improvements lead to reduced PHB production emissions across scenarios, with potential for avoiding upstream emissions. Complementary climate initiatives like electrification and hydrogen fuels can further reduce process emissions. For true negative emission PHB production the CO_2 will need to be sourced using negative emissions technologies.

Analysis of alternative pathways beyond the studied pathway can uncover further opportunities for targeted RD&D investment. Liquid carriers including CO_2-derived Fischer-Tropsch fuels offer an opportunity to reduce emissions in the transportation sector.[37] Organic acids such as itaconic acid and succinic acid are particularly interesting due to their chemical properties, market value, and growing market size.[29] Studies evaluating the production of these chemicals through an integrated process are well-suited to use our analysis as a starting point. While our analysis only provides comprehensive models for CO_2 to methanol/formate to PHB pathways, we provide transparent and detailed design, economics, and life cycle calculations with clear specifications. For this reason, future work can leverage the methods and data to adapt our analysis to models of related integrated CCUS pathways. For instance, future studies could extend our analysis to consider end-of-life PHB emissions and economics for improper disposal (*e.g.* landfill) versus proper disposal (*e.g.* anaerobic digestion, composting). In particular, proper PHB disposal may result in further carbon sequestration and potential negative emissions through production of renewable methane[59] or enhanced carbon intake of the soil.[60,61]

As technological maturity is achieved, governments should focus on market pull factors. Carbon pricing should reflect advancements in technology and should cover emissions from all major sectors to encourage private sector adoption. To improve stability in the sector, carbon prices should be included in larger policy packages with complementary climate and non-climate policy.[62] Ready access to increasingly cheaper solar photovoltaic and wind electricity can help accelerate commercialisation. Ultimately, technological advancement in CCUS, supported by government initiatives, can play an important role in the decarbonisation of our energy sectors and help reduce emissions towards meeting our sub-national, national, and global climate goals.

Methods

Current global carbon prices were reported based on data provided by World Bank.[55] Public investment for research, development, and demonstration in CCUS was estimated based on energy technology data provided by the International Energy Agency.[63] For China, public investment was data was based on a report from their 12th five-year plan.[64] For national jurisdictions and the European Union, World Bank data was used for gross domestic product.[65] For states in the United States and British Columbia, U.S. Energy Information Administration data[66] and Statistics Canada data[67] was used, respectively. U.S. Energy Information Administration international statistics were used for the source of electricity generation for national jurisdictions and the European union.[68] For states in the US and British Columbia, U.S. Energy Information Administration data[69] and Canada Energy Regulator data[70] was used, respectively.

We used ASPEN Plus to develop a rigorous mass and energy balance model that serves the basis of our study.[71] A detailed account of our process, methods, and calculations are provided in the SI.

Data Availability

All data supporting the findings of this study, including data derived from our Aspen Plus model, are available within the article and its supplementary information files. Data for Gross Domestic Product, carbon pricing, RD&D funding, and source of electricity generation is publicly available by the referenced sources in the methods and available from the corresponding author upon reasonable request.

Author Contributions

Andrew W. Ruttinger: Data Curation, Formal Analysis, Investigation, Methodology, Validation, Visualization, Writing - Original Draft, Writing - Review & Editing; **Sakineh Tavakkoli:** Formal Analysis, Investigation, Methodology, Writing - Review & Editing; **Hao Shen:** Investigation; **Chao Wang:** Funding Acquisition, Resources, Supervision, Writing - Review & Editing; **Sarah M. Jordaan:** Conceptualization, Funding Acquisition, Methodology, Resources, Supervision, Writing - Review & Editing.

Conflicts of interest

There are no conflicts to declare.

Acknowledgments

First, we would like to thank our collaborators for providing experimental insight and targets to help guide our analysis. In particular: Prof. Michael J. Betenbaugh, Dr. Pavlo Bohutskyi, and Prof. Marina Kalyuzhnaya for their leadership and direction; Yuxuan Wang and Jack Jenkins for their consultation. We would also like to gratefully acknowledge our industry contacts who provided helpful guidance and estimates for equipment operation. S.M.J. would like to acknowledgment the financial support of the U.S. Department of Energy (EE0008501). A.W.R. acknowledges the financial support of the NSERC Postgraduate Scholarships - Doctoral (PGS-D) award (CGSD3-502578-2017), the Cornell Graduate School, and start-up funding from Johns Hopkins University. A.W.R. would also like to acknowledge the Johns Hopkins Whiting School of Engineering for providing access to ASPEN Plus software for process simulation. Finally, A.W.R. acknowledges the support of his advisor, Professor Paulette Clancy, for providing him with the opportunity to engage in new research through this collaboration.

Notes and references

1 International Energy Agency, *Exploring Clean Energy pathways: The role of CO2 storage*, 2019.
2 V. Masson-Delmotte, P. Zhai, H.-O. Pörtner, D. Roberts, J. Skea, P. Shukla, A. Pirani, W. Moufouma-Okia, C. Péan, R. Pidcock, S. Connors, J. Matthews, Y. Chen, X. Zhou, M. Gomis, E. Lonnoy, T. Maycock, M. Tignor and T. W. (eds.), *Global Warming of 1.5°C. An IPCC Special Report on the impacts of global warming of 1.5°C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the global response to the threat of climate change, sustainable development, and efforts to eradicate poverty*, Ipcc technical report, 2018.
3 E. Levina, S. Bennett and S. McCoy, *Technology Roadmap: Carbon capture and storage*, OECD/IEA, 2013.
4 N. Mac Dowell, P. S. Fennell, N. Shah and G. C. Maitland, *Nature Climate Change*, 2017, **7**, 243–249.
5 S. J. Davis, N. S. Lewis, M. Shaner, S. Aggarwal, D. Arent, I. L. Azevedo, S. M. Benson, T. Bradley, J. Brouwer, Y.-M. Chiang *et al.*, *Science*, 2018, **360**, eaas9793.
6 M. Bui, C. S. Adjiman, A. Bardow, E. J. Anthony, A. Boston, S. Brown, P. S. Fennell, S. Fuss, A. Galindo, L. A. Hackett *et al.*, *Energy & Environmental Science*, 2018, **11**, 1062–1176.
7 C. Pozo, Á. Galán-Martín, D. M. Reiner, N. Mac Dowell and G. Guillén-Gosálbez, *Nature Climate Change*, 2020, 1–7.
8 S. M. Jordaan, E. Romo-Rabago, R. McLeary, L. Reidy, J. Nazari and I. M. Herremans, *Renewable and Sustainable Energy Reviews*, 2017, **78**, 1397–1409.
9 Global CCS Institute, *Global Status of CCS 2019*, Global ccs institute technical report, 2019.
10 H. Naims, *Environmental Science and Pollution Research*, 2016, **23**, 22226–22241.
11 C. Kolster, E. Mechleri, S. Krevor and N. Mac Dowell, *International Journal of Greenhouse Gas Control*, 2017, **58**, 127–141.
12 K. Z. House, A. C. Baclig, M. Ranjan, E. A. van Nierop, J. Wilcox and H. J. Herzog, *Proceedings of the National Academy of Sciences*, 2011, **108**, 20428–20433.
13 E. S. Rubin, J. E. Davison and H. J. Herzog, *International Journal of Greenhouse Gas Control*, 2015, **40**, 378–400.
14 R. G. Grim, Z. Huang, M. T. Guarnieri, J. R. Ferrell, L. Tao and J. A. Schaidle, *Energy & Environmental Science*, 2020, **13**,

472–494.
15 F. M. Baena-Moreno, M. Rodríguez-Galán, F. Vega, B. Alonso-Fariñas, L. F. Vilches Arenas and B. Navarrete, *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects*, 2019, **41**, 1403–1433.
16 Global CO_2 Initiative, *Global Roadmap for Implementing CO2 Utilization*, Global co_2 initiative technical report, 2019.
17 E. I. Koytsoumpa, C. Bergins and E. Kakaras, *The Journal of Supercritical Fluids*, 2018, **132**, 3–16.
18 A. W. Zimmermann and R. Schomäcker, *Energy Technology*, 2017, **5**, 850–860.
19 K. Roh, A. Bardow, D. Bongartz, J. Burre, W. Chung, S. Deutz, D. Han, M. Heßelmann, Y. Kohlhaas, A. König *et al.*, *Green Chemistry*, 2020, **22**, 3842–3859.
20 C. Hepburn, E. Adlen, J. Beddington, E. A. Carter, S. Fuss, N. Mac Dowell, J. C. Minx, P. Smith and C. K. Williams, *Nature*, 2019, **575**, 87–97.
21 P. De Luna, C. Hahn, D. Higgins, S. A. Jaffer, T. F. Jaramillo and E. H. Sargent, *Science*, 2019, **364**, eaav3506.
22 N. J. Claassens, D. Z. Sousa, V. A. M. dos Santos, W. M. de Vos and J. van der Oost, *Nature Reviews Microbiology*, 2016, **14**, 692–706.
23 L. Beck, *Clean Energy*, 2020, **4**, 2–11.
24 R. Carpine, G. Olivieri, K. J. Hellingwerf, A. Pollio and A. Marzocchella, *Processes*, 2020, **8**, 323.
25 K. W. Meereboer, M. Misra and A. K. Mohanty, *Green Chemistry*, 2020, **22**, 5519–5558.
26 Y. S. Mostafa, S. A. Alrumman, S. A. Alamri, K. A. Otaif, M. S. Mostafa and A. M. Alfaify, *Scientific Reports*, 2020, **10**, 1–13.
27 C. Trakunjae, A. Boondaeng, W. Apiwatanapiwat, A. Kosugi, T. Arai, K. Sudesh and P. Vaithanomsat, *Scientific reports*, 2021, **11**, 1–14.
28 R. Sirohi, J. P. Pandey, V. K. Gaur, E. Gnansounou and R. Sindhu, *Bioresource Technology*, 2020, 123536.
29 R. Mazzoli, *Fermentation*, 2021, **7**, 248.
30 K. Rajan, S. Thomas, A. Gopanna and M. Chavali, *Handbook of Ecomaterials; Martínez, LMT, Kharissova, OV, Kharisov, BI, Eds*, 2018, 1–23.
31 P. B. Albuquerque and C. B. Malafaia, *International journal of biological macromolecules*, 2018, **107**, 615–625.
32 L. Garcia-Gonzalez, M. S. I. Mozumder, M. Dubreuil, E. I. Volcke and H. De Wever, *Catalysis Today*, 2015, **257**, 237–245.
33 K. Zhang, Y. Zhou, T. Song and J. Xie, *Energy & Fuels*, 2021, **35**, 15978–15986.
34 E. Vlaeminck, K. Quataert, E. Uitterhaegen, K. De Winter and W. K. Soetaert, *Journal of Biotechnology*, 2021.
35 M. Jouny, W. Luc and F. Jiao, *Industrial & Engineering Chemistry Research*, 2018, **57**, 2165–2177.
36 I. Levett, G. Birkett, N. Davies, A. Bell, A. Langford, B. Laycock, P. Lant and S. Pratt, *Journal of Environmental Chemical Engineering*, 2016, **4**, 3724–3733.
37 J. M. Spurgeon and B. Kumar, *Energy & Environmental Science*, 2018, **11**, 1536–1551.
38 K. Narasimhan, A. C. Cearley, M. S. Gibson and S. J. Welling, *Process for the solvent-based extraction of polyhydroxyalkanoates from biomass*, 2008, US Patent 7,378,266.
39 O. S. Bushuyev, P. De Luna, C. T. Dinh, L. Tao, G. Saur, J. van de Lagemaat, S. O. Kelley and E. H. Sargent, *Joule*, 2018, **2**, 825–832.
40 C. A. Cotton, N. J. Claassens, S. Benito-Vaquerizo and A. Bar-Even, *Current opinion in biotechnology*, 2020, **62**, 168–180.
41 A. D. Nguyen, J. Y. Park, I. Y. Hwang, R. Hamilton, M. G. Kalyuzhnaya, D. Kim and E. Y. Lee, *Metabolic engineering*, 2020, **57**, 1–12.
42 A. D. Nguyen, I. Y. Hwang, O. K. Lee, D. Kim, M. G. Kalyuzhnaya, R. Mariyana, S. Hadiyati, M. S. Kim and E. Y. Lee, *Metabolic engineering*, 2018, **47**, 323–333.
43 S. Price, U. Kuzhiumparambil, M. Pernice and P. J. Ralph, *Journal of Environmental Chemical Engineering*, 2020, 104007.
44 L. Ou, R. Thilakaratne, R. C. Brown and M. M. Wright, *Biomass and Bioenergy*, 2015, **72**, 45–54.
45 S. B. Jones, Y. Zhu, D. B. Anderson, R. T. Hallen, D. C. Elliott, A. J. Schmidt, K. O. Albrecht, T. R. Hart, M. G. Butcher, C. Drennan *et al.*, *Process design and economics for the conversion of algal biomass to hydrocarbons: whole algae hydrothermal liquefaction and upgrading*, Pacific northwest national lab.(pnnl), richland, wa (united states) technical report, 2014.
46 B. Metz, O. Davidson, H. De Coninck, M. Loos and L. Meyer, *IPCC special report on carbon dioxide capture and storage*, Cambridge: Cambridge University Press, 2005.
47 M. Langholtz, I. Busch, A. Kasturi, M. R. Hilliard, J. McFarlane, C. Tsouris, S. Mukherjee, O. A. Omitaomu, S. M. Kotikot, M. R. Allen-Dumas *et al.*, *Land*, 2020, **9**, 299.
48 Y. Chen, A. Vise, W. E. Klein, F. C. Cetinbas, D. J. Myers, W. A. Smith, T. G. Deutsch and K. C. Neyerlin, *ACS Energy Letters*, 2020, **5**, 1825–1833.
49 M.-Y. Lee, K. T. Park, W. Lee, H. Lim, Y. Kwon and S. Kang, *Critical Reviews in Environmental Science and Technology*, 2020, **50**, 769–815.
50 S. M. Jordaan and C. Wang, *Nature Catalysis*, 2021, **4**, 915–920.
51 O. Yishai, S. N. Lindner, J. G. de la Cruz, H. Tenenboim and A. Bar-Even, *Current opinion in chemical biology*, 2016, **35**, 1–9.
52 IRENA, *Future of Solar Photovoltaic: Deployment, Investment, Technology, Grid Integration, and Socio-economic Aspects*, 2019.
53 J. B. Greenblatt, D. J. Miller, J. W. Ager, F. A. Houle and I. D. Sharp, *Joule*, 2018, **2**, 381–420.
54 EIA, *Natural Gas Monthly*, 2020, Based on 2019 average.
55 World Bank, *State and Trends of Carbon Pricing 2021*, 2021.
56 International Energy Agency, *World Energy Outlook 2020*, 2020, `https://www.iea.org/reports/world-energy-outlook-2020`.
57 N. Stern and J. E. Stiglitz, *Report of the high-level commission on carbon prices*, World bank, washington, d.c. technical re-

port, 2017.

58 K. Jiang, P. Ashworth, S. Zhang, X. Liang, Y. Sun and D. Angus, *Renewable and Sustainable Energy Reviews*, 2020, **119**, 109601.

59 N. Benn and D. Zitomer, *Frontiers in Environmental Science*, 2018, **5**, 93.

60 K. Paustian, E. Larson, J. Kent, E. Marx and A. Swan, *Frontiers in Climate*, 2019, **1**, 8.

61 N. E. Tautges, J. L. Chiartas, A. C. Gaudin, A. T. O'Geen, I. Herrera and K. M. Scow, *Global change biology*, 2019, **25**, 3753–3766.

62 C. Bataille, C. Guivarch, S. Hallegatte, J. Rogelj and H. Waisman, *Nature Climate Change*, 2018, **8**, 648–650.

63 International Energy Agency, *Energy Technology RD&D Budget Database*, 2019, data retrieved from Energy technology RD&D, https://doi.org/10.1787/enetech-data-en.

64 Global CCS Institute, *China CCS Deployment Expands*, 2015, https://www.globalccsinstitute.com/news-media/insights/china-ccs-deployment-expands/.

65 World Bank, *GDP (current US$)*, 2021, Based on 2019.

66 US Bureau of Economic Analysis (BEA), *GDP by State*, Retrieved 11 June 2021, https://www.bea.gov/data/gdp/gdp-state.

67 Statistics Canada, *Gross domestic product, expenditure-based, provincial and territorial, annual*, Retrieved 11 June 2021, https://www.150.statcan.gc.ca.

68 EIA, *International Electricity Generation*, 2021, Based on 2018.

69 EIA, *State Electricity Profiles*, 2021, Based on 2018.

70 Canada Energy Regulator, *Canada's Energy Future Data Appendices*, Retrieved 11 June 2021, https://doi.org/10.35002/zjr8-8x75.

71 Aspentech, Aspen Plus, *Aspen Properties V10; Aspen Process Economic Analyzer V10*, 2017.