Roadmap

A US perspective on closing the carbon cycle to defossilize difficult-to-electrify segments of our economy

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Abstract

Electrification to reduce or eliminate greenhouse gas emissions is essential to mitigate climate change. However, a substantial portion of our manufacturing and transportation infrastructure will be difficult to electrify and/or will continue to use carbon as a key component, including areas in aviation, heavy-duty and marine transportation, and the chemical industry. In this Roadmap, we explore how multidisciplinary approaches will enable us to close the carbon cycle and create a circular economy by defossilizing these difficult-to-electrify areas and those that will continue to need carbon. We discuss two approaches for this: developing carbon alternatives and improving our ability to reuse carbon, enabled by separations. Furthermore, we posit that co-design and use-driven fundamental science are essential to reach aggressive greenhouse gas reduction targets.

Sections

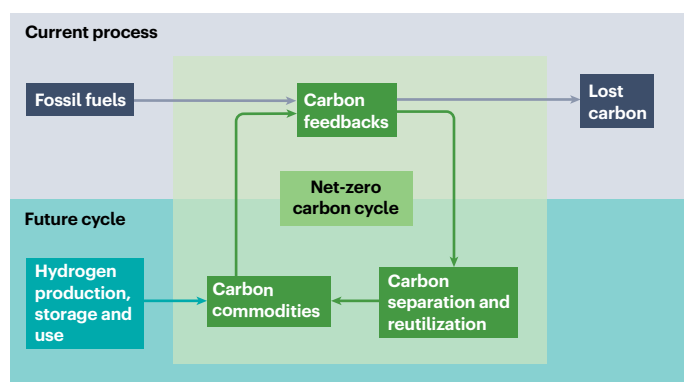✉e-mail: Wendy.shaw@pnnl.gov; kidderm@ornl.gov; srbare@slac.stanford.edu; delferro@anl.gov; morrisj@ameslab.gov; fmtoma@lbl.gov; ssenanay@bnl.gov

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Introduction

The use of fossil resources as a source of both carbon and energy has led to a substantial rise in the standard of living across the globe. However, our current energy and materials infrastructure has also caused a rapid increase in the atmospheric carbon dioxide (CO_2) concentration[1]. The Intergovernmental Panel on Climate Change (IPCC)[2,3] suggests this rise has already led to more frequent and intense weather events, negative environmental and ecosystem impacts, and reduced food and water security[4–9]. The present and looming climate consequences of continued CO_2 emissions[4,10–12], limited clean water resources[13–16], dispersal of plastics in the environment[17,18] an pervasiveness of mismanaged municipal wastes[19–21] have led to an increasing emphasis on decarbonizing our economy and infrastructure, and the design and aspiration of a circular economy[22]. Electrifying with carbon-free energy sources will be a critical component of decarbonization[23,24]. However, several segments of our economy, including the manufacturing of chemicals and polymeric materials, will continue to need carbon. In addition, segments of our transportation economy[25] will also be difficult to electrify due to the size and weight of the batteries that would be needed, including aviation[26–28], long-haul, heavy-duty[29], and marine transportation[30]. Together, these 'hard-to-electrify' segments contribute to ~20% of the overall US greenhouse gas (GHG) emissions (see Fig. 1a, in which the areas not covered by parallel lines represent the areas difficult to electrify[31–33]). Although efforts are being made to decarbonize parts of these segments, they are unlikely to completely transition from carbon[34]. We can reduce carbon use by increasing efficiency and reducing waste, but that approach alone will not be sufficient to achieve net-zero CO_2 emissions. We therefore posit that defossilization[35], or removing fossil fuels while still using carbon in our economy, is a critical part of achieving net-zero CO_2 emissions[35] for difficult-to-electrify sectors.

Defossilization of difficult-to-electrify sectors can help create a circular carbon economy (Fig. 1b) in one of two ways. The carbon can either be replaced with non-carbon containing alternative such as clean hydrogen[36], or fossil carbon can be replaced with non-fossil sources of carbon such as CO_2[37,38], agricultural and forestry residues and other forms of biomass (biomass)[39,40], food waste[41], polymer waste[42–45], and biogenic methane (CH_4)[46], in conditions in which, effectively, waste becomes a feedstock[47–52]. Ideally, each carbon atom would be reused multiple times, reducing the need to extract fossil fuels and creating a circular economy that would allow us to move towards net-zero CO_2 emissions in these segments of our economy[53]. If circularity could be implemented with 100% efficiency, fossil fuels would no longer be needed. This scenario is unlikely to materialize, certainly in the near term. However, focusing on areas in which reusing carbon is achievable will begin moving the needle to net-zero CO_2 emissions and may provide a foundation upon which defossilization can be achieved in the areas with the greatest impact. These efforts will move us towards 'closing the carbon cycle', achieving net-zero CO_2 emissions in segments of our economy[38,54] that cannot be easily electrified. Although fully utilizing non-fossil sources of carbon is the ultimate goal, in the intervening time, using fossil-derived waste, such as polymers or fossil-derived CH_4, is an important step in developing the science to close the carbon cycle.

Unlike other recent Reviews and Perspectives, which focus on electrification to reduce CO_2 emissions[54–59], the scope of this Roadmap will intentionally focus on the gaps in knowledge that must be filled in areas of our economy that will not easily be electrified[60–62]. These knowledge gaps are outlined, along with the technological advances required for success. These advances are organized around the science and technology needed to, first, provide alternatives to carbon, and second, reuse non-fossil carbon in areas in which it is necessary to keep carbon in play. Non-fossil carbon reuse will be enabled in part by the development of reactive and energy-efficient separations. We close with a vision for the future utilizing this innovative science, along with considerations for metrics and analysis, as well as environmental and social justice for the approach to be sustainably responsible.

Additional resources that provide topical insight in related areas can be found in IPCC reports[1,3,5] as guidelines to policy makers, the Department of Energy (DOE), Basic Energy Sciences Basic Research Roundtable reports[63–65], the National Academies[66,67], energy agencies or consultant companies[68,69], and broad scientific reviews[70–73]. To enable readers to understand the science and technology challenges and requirements, we have carefully chosen the two central issues identified, namely, replacing carbon or reusing non-fossil carbon, on which this Roadmap will focus. Specifically outside of the described scope, and expertly detailed elsewhere, are CO_2 sequestration[1,38,74–77], the redesign of polymers for improved recyclability[78–80] or biodegradability[71,72], low-carbon intensity carbon-based fuels[73], materials designed for reuse[78,79,81,82], alternatives to current polymers[79,83,84], mechanical separation of waste[85–87], fusion power[88,89], biological solutions[90–93], increasing the availability of renewable energy[94–97], improvements to the grid infrastructure[89,98–102], net-negative emissions[23,25], crop design[103–107], and fertilizers[103,104]. Finally, the reader should be aware that this Roadmap represents a USA-based perspective, and additional considerations may be necessary based on the geographical needs and resource availability of other countries.

Fundamental science opportunities

To achieve defossilization in difficult-to-electrify segments of our economy, we must accelerate the pace of discovery of transformative technologies. Continuing to proceed with today's 15–20 year 'discover, design, development and deployment' cycle will not allow us to meet the aggressive 2030 and 2050 US targets[24,108,109] needed to slow, stop or even reverse climate impacts. We must be innovative in how we approach the development of these fundamental advances, necessarily using a different approach to translating between fundamental and applied science compared to the approach used today. In the following sections, we will discuss the fundamental advances that need the most focus to accelerate discovery.

Alternatives to carbon-based fuels

Both H_2 and ammonia (NH_3) have the potential to be low-carbon-intensity or carbon-free fuels that can reduce our carbon footprint, provided renewables are used to generate them. For example, researchers are making advances in using H_2 for small aircraft, and NH_3 is under consideration as a fuel for the maritime industry[30,110,111]. H_2 and/or H_2 carriers may also be considered as viable long-term, grid-scale energy storage media. This could mitigate the temporal oscillations of renewable energy, providing further opportunities to reduce carbon emissions.

Ammonia

For difficult-to-electrify sectors, NH_3 is being considered as an alternative to carbon-based fuels. The maritime industry is particularly focused on using NH_3 fuels through both direct combustion in specially designed 2-cycle engines and in fuel cells[112–115]. There are advantages of using NH_3 as a fuel compared with H_2, including an energy density about 30% higher than liquid H_2 and storage requirements that are less stringent than those for H_2 (NH_3 is already used as a refrigerant). In addition to direct combustion, for transportation, NH_3 can be oxidized directly in fuel cells to release electrons (and protons)[116–119], providing
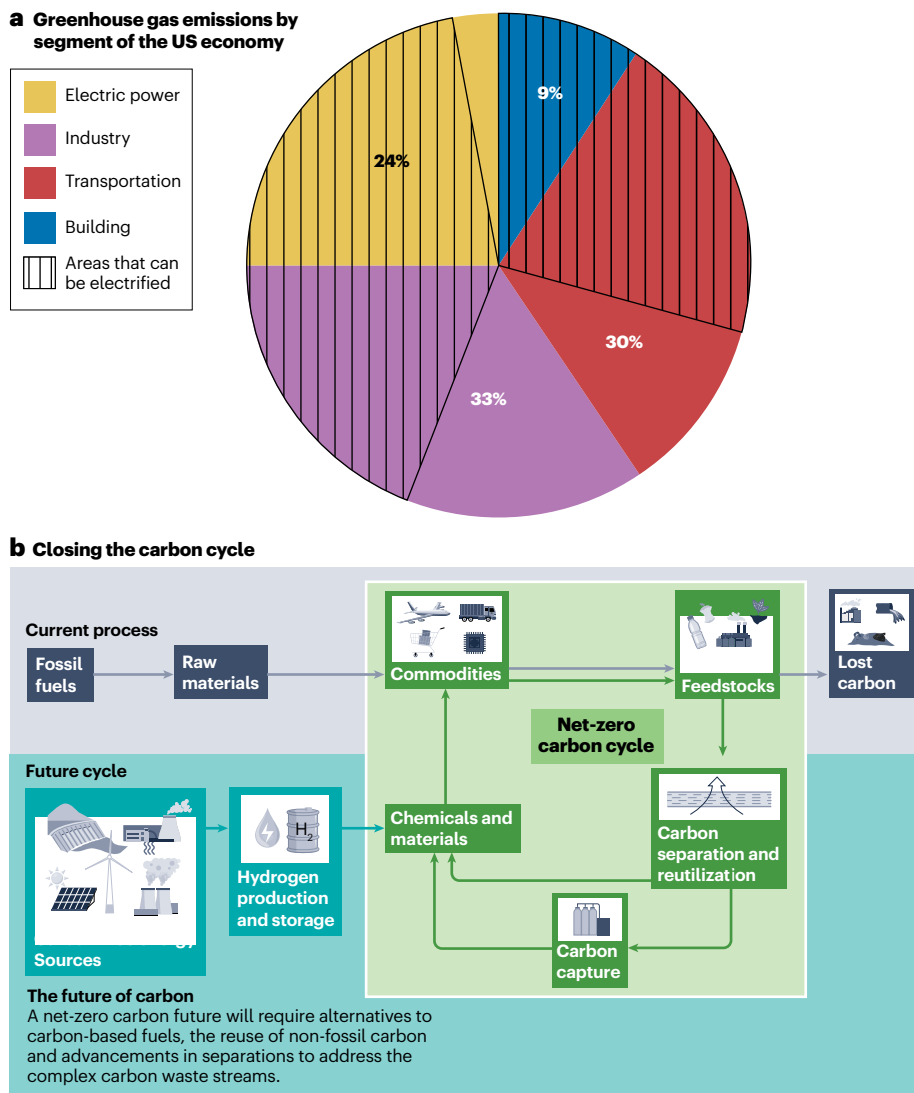
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a Greenhouse gas emissions by segment of the US economy

Legend:
- Electric power
- Industry
- Transportation
- Building
- Areas that can be electrified

9%, 24%, 33%, 30%

b Closing the carbon cycle

Current process: Fossil fuels → Raw materials → Commodities → Feedstocks → Lost carbon

Net-zero carbon cycle

Future cycle: Sources → Hydrogen production and storage → Chemicals and materials → Commodities; Feedstocks → Carbon separation and reutilization → Carbon capture → Chemicals and materials

The future of carbon
A net-zero carbon future will require alternatives to carbon-based fuels, the reuse of non-fossil carbon and advancements in separations to address the complex carbon waste streams.

Fig. 1 | Closing the carbon cycle to remove CO_2 greenhouse gas (GHG) emissions. a, Chart showing the percentage of GHGs coming from different segments of the US economy. The four major sectors are electric power (yellow), buildings (blue), industry (purple) and transportation (red). The portions of these segments that are marked by parallel black lines indicate areas that can be electrified. The segments that will be difficult to electrify are shown as solid colours. Industry, electric power, and transportation sectors have opportunities to decarbonize and contribute to about 20% of the current GHGs, driving the need to achieve net-zero carbon for these areas as well. To generate what we feel is a conservative estimate of ~20% GHG reduction, we extracted data from tables 18 and 19 in refs. [31] and [32], respectively, and included 100% from truck, freight rail, international shipping and air transportation; 50% for lubricants, pipeline fuel, domestic shipping and military use; 100% from agriculture; 50% from construction and mining from non-manufacturing; and 50% of all emissions from refining, food processing, paper products, bulk chemicals, cement and lime, iron and steel, wood products, and plastics from industry. We used the 50% figure in cases in which heat and power are ~50% of the current carbon intensity. See ref. [33] for additional details. **b**, The future of net-zero CO_2 emissions for hard-to-electrify industries involves moving away from fossil fuels and our current linear use of raw materials (top). It focuses on providing alternatives to carbon-based fuels (that is, H_2) and reusing available 'above-ground' carbon, such as traditional waste sources: CO_2, biomass, food waste, polymers, and biogenic or stranded CH_4. Energy-efficient separations must be achieved to allow the use of more complex carbon feedstocks without increasing CO_2 emissions from inefficient separations. Moving to the proposed carbon cycle of the future would build upon renewable energy sources, such as wind and solar, as well as clean H_2, which can be used both for keeping carbon in play to serve as energy carriers and for long-duration energy storage.

advantages in energy efficiency with respect to combustion. A recent technoeconomic analysis demonstrated that direct ammonia fuel cells can be cost-competitive with carbon-based fuels[118].

In addition, NH_3 can be used as a hydrogen storage medium[120], subsequently being catalytically decomposed to H_2 and N_2. This technology is currently being developed and commercialized to provide H_2 for fuel cell applications[121,122].

Although the long-established Haber–Bosch process catalytically reduces N_2 to NH_3 on a massive worldwide scale, a substantial effort has been devoted to achieving NH_3 synthesis electrochemically[123–125],

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ideally with much lower energy input and greatly reduced environmental impact. Molecular catalysts have been studied in detail[126,127]. Although important advances have been reported in the design of heterogeneous catalysts, further improvements are needed to lower the overpotential, increase the Faradaic efficiency, improve selectivity and minimize competitive H_2 production[128].

Despite the promise of NH_3 fuels, there are still limitations to consider. With respect to GHG emissions, the byproducts of the combustion of NH_3 at high temperatures, N_xO and NO_x, are substantially more potent greenhouse gases than CO_2. Notably, N_2O has almost 300 times the global warming impact of a similar weight of CO_2[129]. There are also concerns that the use of NH_3 could disrupt the nitrogen cycle, promoting eutrophication and air pollution[130]. Should we find a way to capture the byproducts of combustion, there are additional equity and justice concerns. For instance, making NH_3 for fertilizer already consumes between 1% and 3% of the world's energy, which prevents the poorest countries gaining adequate access to it[131-133]. Furthermore, adoption of NH_3 fuels on a large scale in the maritime industries would require the use of substantial renewable energy resources to avoid adding further carbon emissions, given the large amount of energy needed to make it. Additionally, storage of large quantities of NH_3 in port communities would introduce considerable safety concerns[115,134].

Hydrogen

Another carbon-free alternative fuel is H_2. The versatility of H_2 ensures that it will play a key role in reducing carbon emissions in both industrial and energy sectors. The US DOE has made a series of investments to overcome the challenges of commercial H_2 adoption. These include the H_2@Scale[135] initiative to address the large-scale production, storage and utilization of H_2 and the DOE Hydrogen Shot, which aims to reduce the production cost of renewably generated H_2 by 80% to US\$1 kg^{-1} H_2 (ref. [136]). The challenges to produce H_2 have been covered extensively by others, and we refer the interested reader to these documents[63,137,138].

An area in which additional research is needed is to address the costs for storage and transport of H_2. The current cost for transport is US\$10 kg^{-1} H_2, an order of magnitude greater than the targeted production cost[139,140]. There are several current options, but none of them viable on a large scale. For instance, H_2 can be transported in pipelines, but there are currently only 1,600 miles of H_2 pipelines in operation within the USA, and the infrastructure cost is high[141]. H_2 can be transported as a 10% mixture in current natural gas pipelines without degrading the pipeline, but H_2 may require a separation step for use[138,142]. Tube trailers can be used for transporting limited amounts of compressed gaseous H_2 (250 kg at 200 bar). Multi-layered vacuum-insulated double-walled vessels can transport 4000 kg of liquid H_2; however, liquefaction is an energy-intensive process with about 35% efficiency[138]. Although these forms of H_2 transport are helpful in the short term, new infrastructure is needed in the mid-to-long term if H_2 is to play a greater role in reducing carbon emissions.

As renewable power resources are developed in the short term, the electricity generated should be used directly to replace power provided by fossil sources to reduce GHG emissions. However, as excess renewable energy becomes available, novel approaches to store energy for longer durations need to be developed[143] to achieve the US 2050 goals[24,144]. By 2050, the Energy Information Administration predicts that there will be an excess of wind and solar resources in the USA and a need to store between 35,000 and 200,000 GWh of energy daily[145]. At these large scales, because power and energy are decoupled, storing energy in the form of H_2 is expected to be more economical than batteries, because adding additional storage (energy) only needs relatively inexpensive tanks.

Hydrogen carriers for energy storage and transportation

Liquid organic hydrogen carriers (LOHCs) are molecules that store energy (H_2) in chemical bonds[146] and are derived from carbon feedstocks, such as methylcyclohexane from toluene[147], methanol and formic acid, which have promising pathways from CO_2, or perhydrobenzyltoluene from benzyltoluene[148]. A key differentiator between LOHCs and storage of hydrogen in metals, as hydrides, or carbon sorbents, such as metal–organic frameworks (MOFs), is the need for catalysts to activate C–H, N–H and O–H bonds[149]. Although catalysts and catalytic reactors add complexity compared with metal hydrides and carbon sorbents, LOHCs have the enormous advantage of decoupling power and energy to enable large-scale energy storage (GWh) and long-duration energy storage[150] (>100 h). However, both life cycle analysis[151] (LCA; to show where there is a reduced carbon footprint) and techno-economic analysis[152,153] (TEA; to show which carbon feedstocks are economically viable) should be used to focus the research on H_2 carrier development.

An additional advantage of LOHCs is that the current infrastructure, including pipelines, shipping and rail, for transporting liquids nationally or internationally would only need moderate modifications to transport LOHCs. H_2 carriers can also be used to provide energy in difficult-to-electrify sectors, such as heavy-duty, long-haul transportation[154].

Over the next decade, research dedicated towards enhancing the round-trip efficiency of LOHCs is needed as a step towards realizing the large-scale storage and transport of H_2[155-157]. The round-trip efficiency metric is defined as the combined efficiencies of water electrolysis to produce H_2, the catalytic efficiency of adding and releasing H_2 from the storage molecules, and the fuel-cell efficiency to generate electricity from H_2. Therefore, highly selective, stable catalysts need to be developed that operate under mild conditions to achieve economical round-trip efficiency, and also over many cycles to avoid the irreversible formation of side products[149]. Current commercial processes for the release of H_2 from LOHC use precious metal catalysts, but these approaches typically require temperatures in the 250–300 °C range[158,159]. Owing to the large endothermic nature of H_2 release from the homocyclic alkanes, there is growing interest in the study of LOHCs that can release H_2 at lower temperatures. For example, the dehydrogenative coupling of alcohols, glycols and amino-alcohols[160-166] releases H_2 at lower temperatures (~200 °C) than the homocyclic LOHCs. These studies are at lower technology readiness levels (TRLs) than the homocyclic organic carriers, and more research is needed to understand the efficiency of catalytic hydrogenation and dehydrogenation under realistic conditions, where the LOHC is not diluted with an inert solvent.

Beyond the conventional LOHCs described above, CO_2 offers promise for storing hydrogen in chemical bonds. Captured CO_2, converted to formic acid or aqueous formate salts[167-170] can be a carbon-neutral energy carrier, given that the initial capture removes CO_2 from circulation[171]. In Fig. 2, a variety of feedstocks are compared based on their enthalpy of combustion (energy to burn) and energy needed to convert them to feedstocks (enthalpy of formation). It is notable that formic acid is formed from CO_2 in a mildly exothermic reaction[172]. However, the loss of entropy associated with the production of formic acid from CO_2 and H_2 results in an unfavourable Gibbs
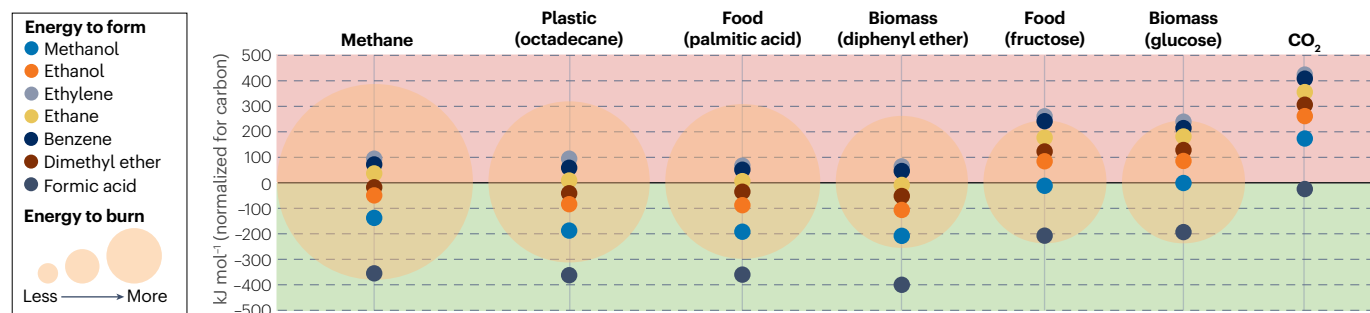
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Fig. 2 | The energetics of various feedstocks and of their conversion.
A diagram capturing the overall energetics of the carbon streams discussed in this Roadmap to aid in evaluating the most energetically useful carbon streams. The smaller dots represent the difference in enthalpy of formation (kJ mol^{-1}; normalized for the number of carbons) of a series of common platform chemicals made from the indicated source feedstock[425,426], in which a value below 0 indicates a thermodynamically favourable process, and a value above 0 indicates a thermodynamically unfavourable process that requires energy input for the conversion. Zero kJ mol^{-1} is indicated by a bold line. The larger orange circles in the background represent the energy available (based on energy to burn in kJ mol^{-1}, normalized for the number of carbons) in each feedstock. In assessing the conversion of these feedstocks to other chemicals, data indicate that the transformation of CH_4 to formic acid, ethanol, methanol and dimethyl ether is energetically downhill whereas, in the case of CO_2, only its conversion to formic acid is energetically favourable. CH_4 and polymers require the lowest energy to be converted to other chemicals and have the most energy to burn. Food waste and biomass have a medium level of energy to burn and require a relatively high energy to convert to many chemicals. CO_2 needs the most energy to be converted, and it has no energy to burn. Comparing the enthalpies of formation and energies of combustion allowed us to be reaction-agnostic, a simplification enabling this qualitative comparison on the potential of each feedstock. To fully assess the suitability of any of these feedstocks in a given process, a full energetic analysis would be needed, in addition to a life cycle analysis assessing all energy inputs, including the transportation of feedstocks.

free energy change. Alternatively, CO_2 captured in an aqueous medium as bicarbonate can readily be reduced by hydrogenation to formate. The bicarbonate–formate cycle affords the ability to operate near equilibrium, with moderate changes in temperature and pressure. Furthermore, the economic and safety benefits of aqueous formate and bicarbonate salts may offer a practical approach for seasonal energy storage for small communities[167]. In addition to serving as a hydrogen-based energy carrier, carbon-based LOHCs reuse carbon and keep it in play for further use in circular economies. The feedstocks described in the next section could provide the source of carbon for the large-scale development of LOHCs.

Areas most likely to benefit from hydrogen as a carbon alternatives

Here, we have focused on some of the challenges specific to the transportation of H_2, but it is notable that H_2 and H_2 carriers can provide storage for excess electricity from otherwise curtailed generation (when generation is reduced below capacity because of demand or transmission constraints)[173] and for large-scale, long-term energy storage[174,175]. Current analyses[176,177] have focused on the storage of H_2 in salt caverns; however, storing H_2 in the chemical bonds of LOHCs will provide opportunities for geographically agnostic long-duration storage. Furthermore, H_2 can replace carbon fuels in large-scale industrial processes. For example, the utilization of clean H_2 as a reductant in the production of iron has received substantial attention[25,178–180]. However, there remains a need for fundamental scientific research to position industry to develop cost-effective ways to store and use H_2[135,181–188].

Keeping carbon in play

We define keeping carbon in play as a carbon cycle in which every carbon atom within products and waste streams is reused, ideally multiple times. Here we identify multiple feedstocks that could be used, including CO_2, biomass, food wastes, plastics/polymers and biogenic CH_4. In the illustration of feedstock energies in Fig. 2, CH_4 and plastics have the highest enthalpy of combustion, as can be determined from the size of the orange circles. The conversion of these two feedstocks into many major platform chemicals is also thermodynamically favourable, given that most of the products, shown as small dark circles, have a negative enthalpy of formation, as illustrated by the fact that they lie below 0 kJ mol^{-1} and within the green band. Food waste and biomass are about equivalent, having somewhat less energy to burn (smaller orange circles) than CH_4 or plastic, and their conversion to major platform chemicals is generally less energetically downhill, or even uphill, as illustrated by the majority of the small dark product circles lying above 0 kJ mol^{-1}. The most difficult to convert and with the least energy to burn (0 kJ mol^{-1}) is CO_2. This property is unfortunate when considering CO_2's abundance (Table 1). Therefore, converting non-CO_2 sources of carbon to new materials makes more energetic sense, followed by filling any remaining carbon needs with CO_2.

Transportation of these wastes over long distances is not practical from an economic or carbon footprint point of view. A possible solution is the development of small modular reactors at or close to the source of the feedstock, which would likely involve new chemical processes. This will require access to renewable energy at the point of generation to avoid increased GHG emissions and complete life cycle analyses of the overall process would be needed.

The described feedstocks will be quite complex, with many types of carbons as well as a variety of other constituents. Due to the complexity of these feedstocks, energy-efficient separations are an additional need. Currently, separations processes consume 10–15% of US energy. Therefore, the development of energy-efficient separations will be needed to keep CO_2 generation low.

Feedstocks

This section focuses on the availability and the pros and cons of using major non-fossil carbon sources, including CO_2, biomass, food waste, plastics waste and CH_4, as carbon feedstocks. This information is summarized in Table 1 and discussed below. The technical advances

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Table 1 | The attributes of renewable carbon feedstocks including availability, quality, impurities, energy content, and energy input needed for conversion to desired products are summarized here

Feedstock	Amount	Pros	Cons	Fundamental science needs	Impurities
Carbon dioxide	Global: 455 billion tons[195] USA: 48.5 million tons from biogenic point sources*	Abundant; could fulfil all carbon needs Will contribute to both decarbonization and net-negative emissions	Is dilute in the air or ocean and needs to be separated and concentrated Impurities cause problems with catalysts and separations materials Reactive impurities, such as those found in flue gases, will need additional solutions Very stable; requires substantial energy input for reuse Impurities have stronger binding (sulfur dioxide and nitrogen oxides)****	Efficient ways to separate/concentrate or selective catalysts for dilute streams Reactive separations Catalysts robust to flue gas impurities	Major impurities: Air: nitrogen (up to 78%), oxygen (up to 21%) and water (up to 18%) Seawater[405–409]: sodium, potassium, magnesium, calcium and sulfate Flue gas[410]: water, sulfur oxides and nitrogen oxides[411–413] Minor impurities: Air: carbon monoxide, nitrogen oxides, hydrogen sulfide, hydrogen chloride, ammonia, hydrogen, sulfur dioxide, mercury and cadmium Seawater: boron, bromine and chlorine
Biomass	Global[414]: Agricultural: 3.0 billion tons (dry weight)** Forestry: 1.8 billion tons*** USA:**** Agricultural: 149 million dry tons Forestry: 287 million dry tons[242] Manures: 29 million dry tons	Potential to replace off-road fuel applications and some chemicals***** Derived from photosynthesis, currently the most cost-effective mechanism to remove large amounts of carbon dioxide from the atmosphere Net energy balance to convert a given feedstock to a product suggests a roadmap for viable feedstocks Provides for highly distributed uses	Processing can be difficult for current industrial processes Lignin is a challenge for some conversion technologies (e.g., fermentation), requiring significant energy input Attention required to avoid land use change concerns Substantial water content and water requirements for cultivation Local processing needed Fertilizer used to support growth is energy-intensive as currently produced and has negative environmental impacts, such as greenhouse gas emissions of nitrous oxide, which is 300x as potent as carbon dioxide, and water quality degradation	Efficient separations Distributed processing and the related fundamental science for modular systems Improved quantification of the flow of carbon and nutrients among agricultural and industrial systems	Polycyclic aromatic hydrocarbons Volatile organic compounds Heavy metals[39,232] Dioxins[415]
Food Waste	Global: 1.4 billion tons[416] USA: biogenic portion of municipal solid waste, landfill gas, food and human waste, fats, oils and greases, 190 million dry tons[242] Food waste only, 14 million dry tons[417]	Substantial energy available; much less energy input needed for conversion to useful products, and landfilling leads to methane emissions Some infrastructure exists for separations Currently, it costs energy and money to dispose of; switching to a feedstock adds value	High water content can increase the energy needed to convert Complex feedstock (currently combined with other wastes, and may contain plastics, paper products, metal, etc.) creates challenges with current catalysts Need to prioritize redistributing waste food to humans and animals, with only spoiled food chemically converted, so availability of feedstock is unknown Local processing needed	Efficient separations Catalysts that can stably and selectively operate in a complex feedstock Distributed processing and the related fundamental science for modular systems	Many and varied, from heavy metals to plastics, to drugs, to water
Plastics	Global: 460 million tons USA: 40 million tons[418]	Substantial volume Pure plastics have substantial energy available for recycling and upcycling Chemical recycling allows conversion of multilayer packaging films	Mixed plastics characterized by impurities, additives and different process parameters that can make them difficult to reuse/recycle[419] Multi-layered films and integrated polymers pose a challenge for mechanical recycling Requires significant energy input Local processing needed Pyrolysis produces 10–20% light gases and 5–10% char, which requires disposal or recycling[420]	Catalysts with appropriate selectivity and that can handle mixed plastics Catalysts stable to chlorides Distributed processing and the related science for modular systems	Anti-oxidants and ultraviolet stabilizers like butylated hydroxytoluene, tris (2,4-di-tert-butylphenyl) phosphite and bis(2,2,6,6-tetramethylpiperidin-4-yl) decanedioate Plasticizers and lubricants like di-(2-ethylhexyl) adipate, chlorinated paraffins (for PVC), calcium stearate, esterified Montan wax Flame retardants like tetrabromobisphenol A and decabromodiphenyl oxide Fillers and reinforcements like calcium carbonate Colourants: white=titania; black=carbon black

Table 1 (continued) | The attributes of renewable carbon feedstocks including availability, quality, impurities, energy content, and energy input needed for conversion to desired products are summarized here

Feedstock	Amount	Pros	Cons	Fundamental science needs	Impurities
Methane	Global: 167,000 billion tons[295] 10–22 billion tons (biogas and biomethane only[421,422])****** USA: 8,900 billion tons	Abundant Will contribute to both decarbonization and net-negative emissions Contains a large amount of energy Use removes a greenhouse gas 27–30x as potent as carbon dioxide	Impurities cause problems with catalysts and separation materials Non-renewable source Higher global warming potential than carbon dioxide (27–30x)[423] Stranded gas is uneconomic to transport Selective activation of C–H is difficult Potential for leaks	Capture of biogenic methane Catalysts with appropriate selectivity Efficient processing of stranded methane	Hydrogen sulfide[424], water, carbon dioxide, hydrocarbons, mercury, nitrogen and helium

Multiple substantial carbon feedstocks exist without the need to extract new fossil fuels. Each feedstock has different availability, quality, impurities, energy content and energy input needed for conversion to desired products. Based on these features, each stream may be suited to specific ideal products. Those attributes are summarized here. *Calculated based on emissions from the corn ethanol, wine and beer industries. **Considers the major global crops (wheat, maize, rice, soybean, barley, rapeseed, sugarcane and sugar beet) in the selected countries/regions with large biomass potential (Europe, USA, Canada, Brazil, Argentina, China and India) and assumes an average crop residue moisture content of 8%, based on values in PHYLLIS database (https://phyllis.nl/Browse/Standard/ECN-Phyllis). An alternate source suggests that the amount of biomass could be highly variable, ranging from 5 to 95 billion tons dry weight, with variability due to assumptions on crop yield trends, land use, population, diet changes, and so on[245]. ***1.3 Gt per year goes to fuel purposes and 0.9 Gt per year is primary residues for a total of 2.2 Gt per year[414], or 1.8 Gt per year dry weight, assuming reported weight was fresh and using a 16.67% average moisture content. ****Excludes energy crops (239 million dry tons). *****We want to avoid the 30% of the gasoline market that represents the light-duty and medium-duty vehicles, which will be electrified. ******Biogas is projected to be available in quantities sufficient to meet a noticeable fraction of future fueling needs, but only at costs that are five to ten times the current price of fossil fuels.

required for the efficient processing of these feedstocks, each of which have unique challenges for conversion[189–194], are discussed. Here, we focus on the common challenges of feedstock processing, which include the need for new catalysts, separations, and low-temperature and low-pressure processes that are tolerant to impurities.

CO_2. Around 455 billion tons of CO_2 have been released into the atmosphere from fossil sources since 1850 (ref. [195]), making CO_2 an abundant source of carbon. The IPCC states[196,197] that just stopping the generation of CO_2 will not solve the problem – we have enough CO_2 in the atmosphere that we must also remove CO_2 from either the air or the ocean to avoid a significant climate impact; therefore, using it as raw material would be ideal. Although Fig. 2 shows that the conversion of CO_2 to the majority of precursor materials is not energetically favourable, the use of CO_2 as a C_1 precursor of fuels and feedstocks is likely to be needed at some level for closing the carbon cycle, in cases in which there is not adequate carbon from other source streams.

CO_2 exists in the air, in the ocean and as a by-product of industrial processes. The CO_2 in the air or ocean water is very dilute, with many different constituents from which it will need to be separated (Table 1). Surmounting the challenge of capturing CO_2 directly from the air, direct air capture (DAC), or ocean, direct ocean capture (DOC), will be required to benefit the climate. Most industrial sources of CO_2, such as flue gases, contain more concentrated streams of CO_2 and so are considered more practical carbon feedstock sources. However, flue gases have added challenges, as they also tend to include reactive contaminants, such as H_2O, SO_x and NO_x. If not adequately removed, these contaminants can degrade the sorbents used in the capture and purification process, or poison CO_2 utilization catalysts (Table 1). Therefore, separation processes must be tolerant of these impurities for the large-scale separation and purification of CO_2. Reviews of the fundamental challenges of CO_2 capture and use are available for the interested reader[64,198].

The current state of the art represents the many advances in collecting and utilizing CO_2. The separation of CO_2 from multicomponent mixtures, such as flue gas, is achieved using membranes, cryogenic distillation, solid adsorbents or solvent-based absorption[199–205]. Monoethanolamine is the current industry choice for amine scrubbing at concentrations up to 30 wt%. However, this process is characterized by high energy and freshwater consumption for the regeneration process, as well as the thermal degradation of the solvent and the generation of toxic waste and corrosive fumes[203]. New alternatives include ionic liquids, for which the energy consumption is 40% lower, phase change adsorbents (blends of amine/water/alcohol) and, more recently, electrochemically mediated separations[206–210]. For DAC, technologies include alkaline solutions such as KOH, solid sorbents (such as zeolites, supported amines or porous carbon)[211], alkali carbonates, and porous supports, such as MOFs/covalent organic frameworks[212–214]. For flue gas, water-lean solvents have been reported to exhibit the highest efficiency for solvents[215] and the cheapest costs at US$39 per tonne of CO_2[216] in the published literature. For both flue gas and DAC, there is a need for the development of sorbents and membranes, as well as engineering design for the integration of the capture[217–219]. Moreover, regeneration is extremely important for process intensification[201,220–222]. Technologies for direct ocean capture are potentially efficient and inexpensive, as the ocean naturally captures the CO_2, and only technologies to release CO_2 are required; the current state of the art relies on membranes or an electrochemical approach, that is, electrodeionization, electrodialysis, or electrochemical-pH swing[223–227], to remove CO_2 from the surface of the water and then either utilize it for fuels and chemicals or store under the sea floor. Another example of CO_2 utilization is the co-production of glycols and methanol from CO_2, epoxide and H_2. This process has a 100% theoretical atom efficiency, as waste H_2O is consumed in situ to produce a second product (glycol) and generate the two valuable products with no waste[228]. Finally, a route to create sustainable aviation fuel from CO and CO_2 waste from steel mills has recently been demonstrated[229–231].

Agricultural and forestry byproducts and other forms of biomass (biomass). A recent assessment from the DOE concluded that the USA has the potential to produce at least 1 billion dry tons of biomass resources annually without adversely affecting the environment[39,232–235]. Specifically, this biomass will be composed of carbonaceous materials gleaned from agriculture, forestry, organic wastes, purpose-grown crops and algal products. This amount of biomass could produce enough biofuel, bioenergy and bioproducts to displace fossil-based

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aviation fuel (at the 50% blend currently required by the aviation industry)[236–241] with additional material remaining for biobased chemicals. This can be achieved with minimal impacts on the production of food or other agricultural products[242]. Globally, there are enough feedstocks from sustainable sources to meet the needs of the aviation industry in 2050, as part of the US DOE sustainable aviation fuel challenge[243] (up to 540 million tons of sustainable aviation fuel)[244,245].

Photosynthesis by green plants, algae and some prokaryotic organisms requires CO_2 and occurs naturally the world over. It is thus one of the most effective ways to remove CO_2 from the atmosphere[246]. The agriculture and forestry sectors already manage photosynthesis to meet human goals and have a substantial footprint, covering 36.5% (agriculture) and 31.2% (forestry) of the Earth's land surface[247,248]. These sectors also already provide food, fibre, bioenergy and environmental co-benefits for society and are expanding their scopes to include long-term CO_2 removal[249,250].

The benefit of converting organic waste to biofuel, bioenergy and bioproducts is twofold – it generates something useful while also reducing GHG emissions. Purpose-grown biomass crops, or those that are grown specifically for energy rather than food, have a more complicated picture, however. Their deployment must be balanced with food, feed and fibre production; soil, water and biodiversity protection; and cultural/historical land uses[251–254]. Biomass contains nitrogen, sulfur, chlorine, alkali and alkaline earth metals, which can complicate their processing, including poisoning catalysts and contaminating products, if their presence is not properly accounted for (Table 1). Improper thermal treatments of biomass feedstocks can release environmental pollutants, including volatile organic compounds, polyaromatic hydrocarbons and dioxins[40]. Demolition lumber treated with older wood preservatives and biomass grown on soils contaminated with heavy metals requires special consideration in processing or should be avoided altogether to prevent the release of heavy metals and/or pentachlorophenol into the environment[39,40,232].

The current state of the art in using biomass has led to a number of advancements. Biomass and algae have important chemical functionalities, including nitrogen and oxygen groups, within their backbone structures, which can aid in the replacement of fuels and chemicals provided by fossil fuels. Their use as biomass feedstocks can reduce carbon and water intensity as well as sequester carbon in soils as soil organic matter or carbonized biomass (biochar). Microalgae contain a substantial amount of triglycerides, which can be extracted and converted to biodiesel[255–257] and bioethanol[40], and they have been explored for cultivation from open and closed growing systems[258,259]; their potential for production based on composition has also been investigated[260].

Biomass has long been studied for renewable chemical and fuel production, and it heavily relies on the thermal, chemical, biochemical or catalytic transformation to products[261–265]. Most of the chemicals derived from the pyrolysis of biomass have biochar as a by-product, which can be used as a soil amendment[266]. Modelling studies suggest biochars are able to durably store carbon with negative emissions and reduce impact on land, energy and water use compared with fossil fuels[267]. Many industries are expanding to produce cosmetics, biofuels and lubricants from cultivated biomass (for example, ~200 biorefineries in the USA, including ExxonMobil, RTI International, Absolute Energy LLC, Ace Ethanol LLC and POET) and algae (for example, Algenol, Sapphire, GreenFuel and Solazyme).

Food waste. Some 1.4 billion tons of food is wasted per year globally, roughly one-third of the global food production, amounting to ~US$1 trillion and 26 exajoules (EJ) of energy (a quarter of the US energy consumption) wasted[268–270]. Although reducing or redistributing excess food to avoid waste should be a primary goal, this represents an important carbon source, given that the food waste from all parts of the supply chain that cannot be used by humans and animals is unlikely to go to zero.

There are a number of areas that have developed the current state of the art to use food waste as feedstocks. Valorization of these food wastes could help enable a circular economy. Extraction of value-added compounds[271], such as phenolic acids, terephthalic acid, p-cymene and limonene, can be used to produce antioxidants, polyethylene terephthalate resin and polyester films, cosmetics, and pharmaceuticals[268]. Although technology exists to valorize the remainder of the waste via gasification, liquefaction, hydrothermal treatments and pyrolysis into a variety of products[272–274], energy efficiency and selectivity need to be improved, while reducing CO_2 production and recovering and reusing any resulting CO_2. Reliable routes to convert wet and dry feedstocks of variable quality at different scales must be developed. Aside from the energy demand of high-temperature processing, the main products, bio-oils, hydrothermal liquefaction-oils and biochars, are of relatively low-value. However, these products can be upgraded to higher-value products, such as electrodes, carbon nanotubes, graphene, fuels and chemicals, and/or they can be used as media to store carbon[275].

Polymers. If plastic demands follow current projections, global plastic waste volumes will increase from 380 million tons per year in 2016 to 460 million tons per year by 2030 (ref. 276). This demand will elevate the already monumental environmental problem of plastic waste management to a new level. In 2021, US consumers recycled only 5–6% of the 40–50 million tons of their plastic wastes[277,278]. Polyethylene (high density and low density) and polypropylene account for over 50% of discarded waste plastics[279]. Therefore, this waste represents a resource opportunity for producing chemicals and materials. In addition, more than 10 million tons of polyester fibres and textiles are manufactured every year, with less than 1% efficiently recycled[280]. Factors that affect yield of liquid transportation fuels are the presence of additives and contaminants[281], including mixed plastics[282], in the waste as feedstocks. Additionally, many current catalytic and non-catalytic processes for polymer decomposition or upcycling are not sufficiently selective, leading to additional separations. For example, the deconstruction of polyvinylchloride produces HCl, which creates added complexity in the process. Developing the science to convert fossil-derived polymer waste is crucial to recover carbon from this waste stream. Ultimately, as we use biogenic sources to create polymers, the science can be directly translated to similar polymers towards the reuse of newly derived polymers.

There are several advances that establish the current state of the art in using polymers for feedstocks. They include competing technologies that are being developed for utilizing polyolefin wastes as feedstocks for liquid transportation fuels, including thermal pyrolysis[283,284], and gasification to syngas followed by Fischer–Tropsch synthesis[285,286]. In addition, there has been a limited number of studies reported in the literature for converting plastic wastes by catalytic hydrogenolysis[287,288]. Note that catalytic pyrolysis in the presence of H_2 (20–50 atm; 370–450 °C) has been reported to avoid char formation and rapid catalyst deactivation[289,290]; however, excessive C–C bond scission[291,292] is a major challenge, because it reduces the overall liquid yield and leads to the formation of undesirable light gas compounds. For polyester materials, such as polyethylene terephthalate, multiple

catalytic and non-catalytic depolymerization approaches have been developed, including aerobic oxidation, hydrolysis[83], alcoholysis, glycolysis, aminolysis, hydrogenolysis, enzymatic depolymerization and pyrolysis, which allow the reutilization of the monomers for repolymerization to virgin polymers, avoiding the need for producing these monomers from fossil oil[84,85]. To ensure that the CO_2 released in the majority of these processes does not increase greenhouse gases, capture for reuse or sequestration would be required.

Methane. CH_4 is one of the Earth's most abundant carbon-containing molecules. It is the major component of shale gas and is found as methane hydrates and coalbed methane, which is natural gas found in coal deposits[293,294]. The amount of CH_4 in proven reserves is estimated to be 167,000 billion tons globally[295]. CH_4 is also a major component of biogas that results from the decomposition of organic matter. Given that CH_4 has ~27–30 times the global warming impact of CO_2, this is a substantial contributor to global warming, making an additional case for the importance of finding ways to utilize it[46,296]. Estimates show that landfills account for 17% of total US CH_4 emissions[270].

Today there are over 330 operational biomethane projects in the USA focused on biomass and/or food wastes as well as over 2,200 operational biogas plants that displace petroleum-based hydrocarbons[39,242,297,298]. The fossil-derived CH_4 used as feedstock in many industrial processes could be substituted by non-fossil-produced CH_4, if it can be economically transported to the processing site and purified with a net-zero emissions[35] footprint[295]. Typical impurities associated with CH_4 include CO_2, hydrogen sulfide, NH_3, siloxanes, water vapour, oil, nitrogen, hydrates and C2–C5 hydrocarbons. Ultimately, we see biogenic CH_4 and/or stranded CH_4, rather than fossil-derived CH_4, as the non-fossil or waste sources of carbon.

Several developments have resulted in the current state of the art for using CH_4 as a feedstock. CH_4 can be used to produce value-added products. CH_4 steam reforming and the water–gas shift reaction are currently used to commercially produce H_2, although these processes generate CO_2 as a by-product. There are several potentially promising reactions that do not produce CO_2, including dry reforming to produce syngas ($CO + H_2$), which can be used as a feedstock to produce chemicals and fuels[299,300], as well as CH_4 pyrolysis or thermocatalytic decomposition to produce H_2 and solid carbon[301–304]. However, the high dissociation energy of the first C–H bond (440 kJ mol^{-1}) necessitates high reaction temperatures that can result in rapid catalyst deactivation[299].

Comparison of Feedstocks. Above, we have discussed the current state of the art for the individual feedstocks. Of course, not every initially intriguing effort towards decarbonization turns out to be practical. We note the industrial abandonment of algae as a feedstock[305] and the continuing asperities that preclude the industrialization of cellulose-derived ethanol[306]. Technology that seems to be advancing well include the fermentative conversion of flue gas by LanzaTech[307,308], the electrochemical reduction of CO_2 and its subsequent conversion being scaled up by Twelve[309,310], and the use of municipal solid waste being pursued by Fulcrum BioEnergy[311,312]. All of those technologies target sustainable aviation fuel[305–308,310–312].

As we consider the various feedstocks' ability to be converted into platform chemicals, the comparison in Fig. 2 shows that CH_4 and plastics have the highest enthalpy of combustion, and the formation of many major platform chemicals from them is thermodynamically favourable. The routes to platform chemicals using CH_4 include C1 and C2 chemistry of synthesis gas (CO plus H_2) made by steam reforming[313]. The routes starting from plastics will depend on the type and purity of the feedstock. If it is a mixed feedstock then either steam gasification or pyrolysis could be effective[314], the former yielding syngas, the latter yielding mixed hydrocarbons[315] that would need to be fractionated, much like petroleum into streams (olefins, aromatics and oxygenates) that could be converted further. Similar routes extend from biomass and food waste. Processing of waste plastic would be needed to avoid downstream contamination by halogens (polyvinylchloride and additives). Speciating the plastics into easily depolymerized fractions, such as polystyrene and poly(ethylene terephthalate), can allow the production of repolymerizable monomers (styrene[316], terephthalic acid and ethylene glycol[317]). Alternately, carbonization of either CH_4[301] or waste plastic[318] can produce valuable carbon products and H_2. Each process would benefit from research on the catalysts and reactors. In summary, all the major platform chemicals[319,320] made from petroleum are accessible starting with waste or non-fossil carbon sources.

Catalysis, catalyst degradation and new processes for non-fossil feedstocks

Reusing carbon from complex feedstocks (CO_2, biomass, food waste, polymers and CH_4) requires processes and systems to be tolerant to feedstock variations and impurities. Ultimately, both catalysts and the modules in which they reside must be durable and resistant to degradation. The primary mechanism of catalyst deactivation is a result of the deposition of reaction products or impurities (Table 1) that encumber active sites. Chemical degradation leads to phase and/or composition changes in the catalyst that may be irreversible or require high temperatures and reactive environments to regenerate the original material[321]. Fundamental research in catalyst degradation and deactivation, as well as robust regeneration methods, requires operando methods to analyse the catalyst evolution on the requisite time scales and with increasingly complex streams[322]. As the quantity of impurities will be substantially higher in the heterogeneous feedstocks present in reusing carbon streams, an understanding of the fundamental principles governing the reactivity, selectivity and stability of catalytic materials is vital[189].

The variability in makeup and impurities in future feedstocks (see Table 1) will likely reduce the efficiency of current catalytic processes. Therefore, developing accelerated testing methods to allow understanding and responding to temporal changes will be useful to enable predictive models of deactivation[323]. Improved sensors and detectors will enable this response, as well as automation to respond in real time to allow agile catalytic processes. Providing well-defined test feedstocks will be an important approach. As an example, a detailed research hierarchy for separation systems was proposed, which could be applied to catalyst discovery for efficient carbon conversion and utilization[324]. In this study, instead of using idealized mixtures, it was proposed that a series of well-defined exemplar mixtures be developed to bridge the gap between fundamental studies and practical applications. This hierarchy maximizes the impact of early-stage research to identify promising catalysts that will be resilient to varying carbon quality. In a related example, in the 1980s, the Argonne Premium Coal Sample Bank was established to provide the research community with well-characterized coal samples so experimental results could be compared, allowing differences to be attributed to experimental technique rather than sample variation. The carbon feedstocks of the future will be complex and heterogeneous, underscoring the need to standardize feedstock studies and prioritize feedstocks for their most suitable application.
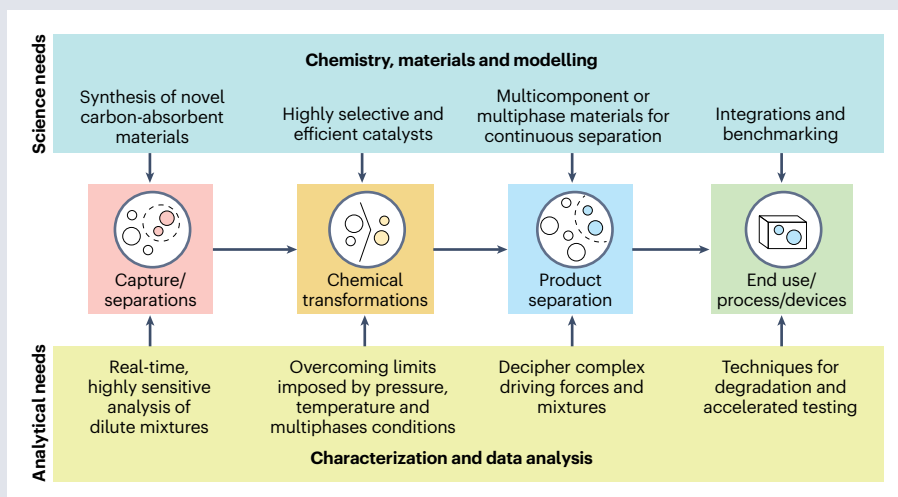
Replacing fossil carbon with non-carbon alternatives or non-fossil carbon at an industrial scale poses substantial scientific challenges. To address them, the scientific community will need to develop experimental, computational and data science tools to better understand chemical transformations, catalyst design and performance, and separations (refer to the figure in Box 1).

The complex carbon feedstocks we discuss in this Roadmap are often beyond the limits of current analytical instrumentation approaches. As shown in the bottom of the figure, the observation of real-time processes of multiple elements (for instance observing the metals in catalysts and the carbon in the feedstock at the same time), potentially concurrently in all phases (gas, liquid and solid; see the figure, right-middle), and considering scales ranging from the single atom to mesoscale and macroscale will be a critical advance. The new approach of co-designing catalysts and separations materials will need to be coupled to rapid characterization and data modelling for the development of the most efficient and effective materials. This will require carbon-focused operando tools that use advanced imaging, scattering, spectroscopy and correlative characterization. The X-ray, neutron and nanoscience user facilities as well as laboratory-scale instrumentation can provide benchmarks and mechanistic information (see the figure, left).

The design of improved catalytic systems will continue to benefit from the partnership between experiment and computation (see the figure, left-middle)[427–433]. Data science and machine learning, although relatively new in their application to catalysis and separations problems, offer substantial potential for further advancement. The use of digital twins in which experiments and high-fidelity models are run in parallel and learn from each other, will accelerate research and minimize experimental trial-and-error and cost (see the figure, right)[434,435]. Extracting design principles from enzymes, nature's catalysts that are highly selective under mild conditions with Earth-abundant metals, has the potential to enable the functional implementation of those principles into synthetic catalysts. Recent breakthroughs in foundational models, particularly in generative artificial intelligence, create new opportunities to analyse and learn from decades of multimodal data on chemical conversions. These advancements can guide the development of innovative materials that enable the selective steering of chemical reactions, favouring desired products over competing pathways[436].

Historically, data from the catalysis community has not been stored in accordance with FAIR (findable, accessible, interoperable and reusable[437]) data principles, making data science use challenging. Advances in machine learning will enable the extraction of chemical information from literature[438], creating a new generation of artificial intelligence tools[439] capable of complex reasoning tasks to support chemical discoveries. However, consistently curated data, for both good and failed catalysts, is needed throughout the catalysis community, and the need to create a community database has been recognized[440–450]. Such a broad and deep database of chemical reactions, chemical reaction conditions, and catalyst and separation materials would enable the implementation of techniques to mine new features from deep data by enabling comparisons across complex reaction schemes difficult to do unaided by data science. As increasingly more data are available in these databases, standardized feedstocks would also help by providing data that can be directly compared.

It is challenging and laborious to study the degradation and deactivation of catalysts, especially during early-stage research. However, methods like those used to study automotive exhaust catalysts or fluid catalytic cracking catalysts should inspire new approaches to understand catalyst aging and deactivation in carbon conversion research[325]. These new methods must manage the added complexity of varying feedstocks, impurities and reaction conditions. To provide the chemical insight necessary to develop new materials and predictive models of catalyst lifetime and performance (Box 1), in situ and operando analytical methods coupled with accelerated testing protocols need to be developed[326]. Machine learning methods have accurately forecasted aging processes in industrial reactors, demonstrating that data-driven models have the potential to outperform mechanistic/physics-based models[327]. Further application of machine learning could be an important contributor to accelerating our scientific discoveries by learning the nonlinear relationships between feedstock variability, process conditions and chemical degradation from experimental and computational data[327,328] (Box 1).

As stated, the development of small modular reactors at or close to sources of non-fossil feedstocks is needed, which will require catalysts

and separations materials that can be easily integrated (see Fig. 3). As depicted in Fig. 3, the distributed, heterogeneous feedstocks of the future may necessitate modular and flexible conversion systems inclusive of low-temperature processes (Fig. 3, upper left); alternative energy inputs for activation, such as microwave or plasma (Fig. 3, upper middle); integration of reactions and separations (Fig. 3, upper right); and robust, interchangeable units that are plug-and-play for catalysts and feed trains (Fig. 3, bottom)[329]. The scale of these systems will depend upon multiple factors. The first is the availability, local sourcing and handling of feedstocks. Additional considerations are the scaling factor of reactors and the use profile, market size and distribution mode of the product(s). Finally, the availability of primary (and ideally renewable) energy inputs will be a critical consideration. Therefore, this modular approach could include farm-scale units (~tons of feedstock per day) that fit onto a truck bed or into a shipping container[330–333], as well as larger-scale flexible modules (~hundreds of tons of feedstock per day) that can be combined to match feedstock availability[334]. This future requires understanding the scaling factors for the new novel reactor designs that leverage alternative energy inputs to drive low-temperature, low-pressure[49,335,336] and associated handling/processing challenges, as well as business models for distributed chemical processes.

Lower temperature and pressure processes

Most industrial processes require high temperatures and/or pressures to make reactions favourable or to overcome sluggish kinetics. To be less energy-intensive and operate in distributed chemical processes, catalysts that operate at lower temperatures and pressures are needed. Biological catalysts, such as metalloenzymes, which operate at atmospheric temperatures and pressures, can act as inspiration[337–341].

To drive chemical reactions at lower temperatures, there is an effort to move away from thermal heating methods. The most advanced of these technologies uses electrochemical potential as the primary driving force[342]. Electrochemical methods are used to induce many oxidation and reduction reactions[343], offering the possibility to reduce energy input compared with thermal processes, as long as there are appropriate low-overpotential catalysts. Additionally, paired electrolyses produce valuable redox products from the same energy input. Although our current understanding of thermal reactivity and kinetics far exceeds our knowledge of electrochemical reactions, recent studies have uncovered parallels between these two types of energy inputs[344]. For example, a study analysed the rate for the same Ni, N-doped carbon catalyst generating CO from CO_2 via electrochemical reduction or through thermal reaction conditions. The kinetic data were plotted with respect to temperature and potential, and the authors developed a generalized reaction driving force that suggests that the catalyst used for the studies could facilitate faster kinetics electrochemically than thermally. The authors see an exciting opportunity for further comparisons, at the same time cautioning that there are additional factors in catalytic activity that make correlation between the two driving forces challenging[345]. Although recent research efforts in this area have advanced our understanding, the transition

Catalyst

Approach: low-severity (temperature, pressure) catalysis

Relevance: enables decentralized processing by reducing safety hazards, facilitating automation and expanding the operator pool

Modular feed trains

Approach: robust, plug-n-play, modular feed trains
Relevance: enables feedstock diversity

Energy input

Approach: alternative energy input for electrochemical and chemical activation (e.g., microwave and non-thermal plasma)

Relevance: Enables renewable energy utilization and downscaling

State of current chemical industry

Catalytic processing | Separations

Top product

Bottom product

Reactive separations

Approach: integration of reactions and separations (e.g., reactive membranes)

Relevance: enables process intensification (capital expense savings) and downscaling

Modular catalytic units

Approach: interchangeable, plug-n-play, modular catalytic units (analogous to filter cartridges)

Relevance: enables product diversity through quick unit changeover in response to new market demands
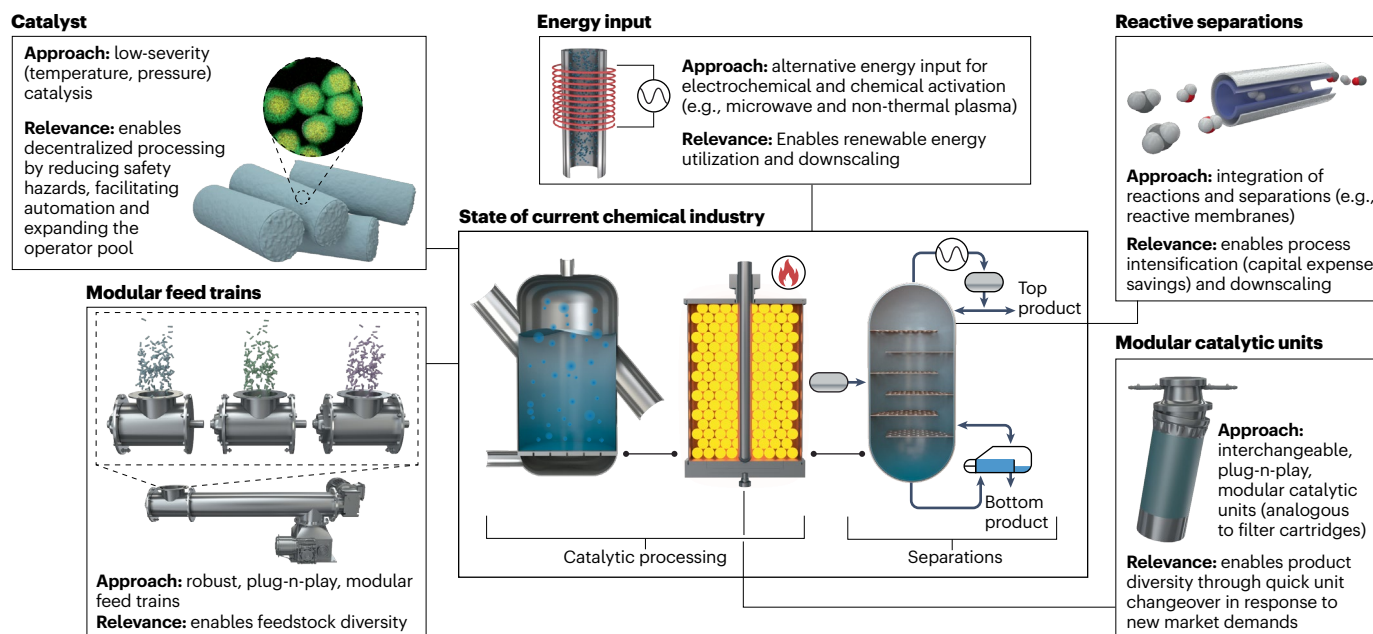
Fig. 3 | Modularity is the future of the chemical industry. Modularity and flexibility in the future chemical industry to process complex carbon source streams relies upon distributed feedstocks. The central image exemplifies the state of the current chemical industry, which focuses on unit operations that scale favourably with volume due to centralized large-volume feed streams. These unit operations are typically rigid, unable to respond to changes in feedstock type, composition, or product demand. A modular chemical industry implies flexibility, the ability to respond to changes in the quality of the distributed feedstock or product demand in real time. Robust catalysts operating at lower temperatures and pressures, stored in exchangeable canisters, could enable a modular chemical industry. Feedstocks will change based on location or season, so feed trains must seamlessly respond to this diversity. Thermal energy from Joule or induction heating or heat pumps sourced from renewable energy inputs rather than typical direct or indirect heating by combustion reduces energy requirements and mitigates the environmental impact. Intensification of unit operations into a single unit reduces the footprint of the chemical plant of the future.

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towards electrochemical operations faces challenges in comparison with the prevailing thermal methods, including the risk associated with the relatively limited knowledge base, the high cost of electrocatalysts, the lower selectivity of reactions at higher yields and the inherent capital costs associated with developing the new infrastructure. Much can be learned by drawing analogy with the decades of work that took H_2 electrolysers for water splitting from concept to commercialization[346]. Similarly, some commercial organic products are already produced by electrochemical means, including nylon[347,348]. These operational processes can provide critical data for scaling these new electrochemical processes for converting complex carbon feedstocks.

Alternative processes to reduce the reaction temperature and energy demand, while overcoming activation barriers, include non-thermal plasma, that is, utilization of electrically energized gas[349]; Joule heating, that is, use of electric current flowed through resistance to generate heat[350]; induction heating, that is, heating by internally induced electric current[351]; and electromagnetic heating, that is, taking advantage of electrically conductive and inductor materials to utilize electromagnetic field-induced heating[352]. There are advantages and challenges to using these alternative processes. For example, non-thermal plasma can be turned on and off quickly, making it compatible with the intermittent nature of renewable energy[353] in the absence of widespread long-term energy storage options. However, its non-selective nature requires new catalysts for improved selectivity and an understanding of plasma–catalyst interactions. In addition to using these processes by themselves, there is promise to enable operations at lower temperatures by combining them into tandem processes, for instance combined photo–thermal[354], electrochemical–thermal[355] or plasma–thermal[356,357] stimuli. Albeit a bench-scale example, the use of tandem plasma–thermochemical stimuli showed that activation of CO_2 and CH_4 can be achieved[358]. Electrochemical–thermochemical tandem reactions enable the efficient conversion of CO_2 to value-added products that cannot be achieved using either process alone, and if renewable energy is used as the excitation source, the CO_2 footprint can be further reduced[355]. In all cases, we need design rules and concepts to integrate these non-thermal methodologies with new catalysts and separations materials and processes. It is also critical to evaluate the full coupled processes with TEA and LCA to ensure that CO_2 emissions are decreasing and that we understand the process energy efficiency.

Biological systems, such as micro-organisms, powered by electrochemically or photochemically generated energetic intermediates (electrons, H_2, reduced carbon and reduced nitrogen)[359–363] may provide the delocalized and centralized production of fuels and chemicals from renewable energy and atmospheric gases. Because these are living systems, scale-up will require additional considerations for practical use. In particular, additional research is required in the biological engineering of the micro-organisms and ecosystems to achieve the desired products, their coupling with electrode-generated species for energy or product conversion, and the surrounding technology required to maintain purity in an energy-rich biological broth that is continuously exposed to air.

Separation needs for complex feedstocks
To effectively use complex carbon feedstocks, improved separation processes will likely be needed. Separation steps could be implemented before a chemical transformation to reduce the complexity of the feedstock and/or after a chemical transformation

to separate the desired product. In the USA, chemical separations (distillation, cyrogenic, solvent extraction, and evaporation) are extremely energy-intensive and currently use 10–15% of US energy[70]. Advances in energy and separation efficiency are thus essential[364–366].

There are many needs for new separation materials and processes to keep carbon in play, many of which are similar to those needed for catalysts. For instance, separation materials will need to be stable over long periods of time and insensitive to impurities. They will also need to operate in streams that vary spatially and temporally, as well as functioning at low temperatures[324,367]. For gas separations, current challenges include poisoning by contaminants, penetrant-induced plasticization, physical aging, and balancing permeability and selectivity. By contrast, liquid separations have difficulties when it comes to designing solute–solute selectivity beyond what has been achieved for water-solute separations. Beyond that, understanding and preventing fouling of adsorbents and membranes, and managing concentrates, such as brines, from ion exchange and membranes are essential advances.

Here, we explain the needs for separating complex feedstocks at low temperatures and coupling separations with catalytic reactions (that is, reactive separations). To accelerate scientific progress, we need both fundamental and applied investigations of separation materials and catalytic processes across a range of feedstocks. This should vary from ideal systems to systems under realistically complex conditions, using standardized feedstock studies[324] and testing conditions to maximize their generalizability and comparability in separations.

Reactive separations
To achieve the most economical and energy-efficient chemical conversions and separations, the co-development of both aspects should be considered at the development stage[368,369]. Reactive separations are a process intensification, that is, the improvements in a process achieved by combining unit operations[368,369]. For reactive separations, chemical reactions would be combined with separations processes, such as distillation, extraction, absorption or crystallization. Using reactive separations can increase product yield and selectivity, with decreased energy requirements, given that they can surpass equilibrium limitations. This also means they need lower capital investment and operating costs than typical catalytic processes. For example, reactive distillation has been used to decrease energy costs and increase the yield of fatty acid alkyl esters from vegetable oil, animal fat and waste cooking oil as compared with conventional biodiesel processes[370]. In other examples of reactive separations, reactive membranes have been used to maximize yield and reduce energy consumption in H_2 production from hydrocarbons[371], and reactive absorption and crystallization have been used to increase yields and energy efficiency compared with when CO_2 capture and utilization are performed separately[372]. Although these integrated processes offer new and exciting opportunities for energy-efficient, cost-efficient and atom-efficient separations, there are still many scientific and engineering challenges that need to be addressed based on the separation process.

Reactive separations are expected to play a major role in carbon dioxide capture and utilization, especially in DAC[35]. Conventional carbon dioxide capture and storage uses amines, ionic liquids, solid sorbents, membranes and porous materials to capture CO_2 for liquefaction and storage[373,374]. These processes typically rely on thermal swing sorption or capturing CO_2 at one temperature and raising the temperature to release it, followed by compression and transport for subsequent storage. Collectively, this makes CO_2 capture and storage highly energy-intensive.

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Reactive adsorption is the process by which CO_2 can be captured and converted through a chemical transformation. Examples include conversion to CH_4 (methanation), or to CO and water in the presence of H_2 (reverse water–gas shift, or dry reforming of CH_4)[375]. Another reactive separation approach is to directly mineralize CO_2 from the water–gas shift reaction. For example, $CaSiO_3$ or Mg_2SiO_4 can react with CO and H_2O to mineralize CO_2 as $CaCO_3$ or $MgCO_3$, respectively, for long-term storage while shifting the reaction equilibrium to form more H_2[376]. In addition to conventional thermal pathways, combining CO_2 capture with nonthermal CO_2 utilization pathways, such as electrochemical CO_2 reduction[377], plasma catalysis[358,378,379] or microwave catalysis[379] could increase the overall energy efficiency while reducing the infrastructure requirements. We need improved understanding of the kinetics and mechanisms of these nonthermal processes to aid in the design of new catalysts and energy-efficient processes for CO_2 capture and utilization. LCA and TEA will also play a critical role in ensuring reactive separations are compliant with the circular carbon economy and will help prioritize further investigations.

Although energy considerations are key to optimizing reactive separations, atom efficiency is just as important. Most separations require the selective concentration and isolation of a target molecule from its waste, which is energy intensive. However, a circular economy offers chemical processes designed to not just minimize waste, but to repurpose or eliminate it entirely. As we strive to close the carbon cycle, we hope to emulate strategies in which high atom and energy efficiency are built into the future design of chemical processes.

Areas most likely to benefit from keeping carbon in play. The fuel, chemical and polymer industries are those most likely to effectively reduce our fossil carbon needs. In particular, sustainable aviation fuel has received substantial attention as this sector is unlikely to move from carbon-based fuels any time soon, if ever. Many chemicals and polymers that form the foundation of other industries (plastics for lightweighting, clothing and manufacturing) are built upon carbon-based platform chemicals. Developing efficient and cost-effective mechanisms to provide reused carbon rather than fossil carbon, in a way that can be used in the existing infrastructure, would be ideal.

Designing an integrated future

In this Roadmap, we have discussed the science and technology needed to achieve carbon neutrality for difficult-to-electrify segments of the economy, focusing on alternatives to carbon, and keeping carbon in play. However, how do we meet the USA's and the world's aggressive carbon neutrality targets by 2050 — which is only 26 years away? The typical timeline from discovering to deploying technologies takes 15–20 years, meaning there is only time for one round of such development to meet this target. Therefore, we must make significant changes in how we do research so that we can provide solutions more rapidly[25]. An integrated approach is needed to develop efficient processes and materials that accelerate technologies at all TRLs. The processes need to be economically viable and meet regulatory and manufacturing needs, while at the same time reducing emissions and maintaining cognizance of social and environmental impacts.

In *Cradle to Cradle*[380], William McDonough and Michael Braungart asked readers to perform a thought experiment, imagining that they had been given the assignment of designing the industrial revolution. Looking at the consequences, they articulate that the system design would have included a variety of negative features such as putting billions of pounds of toxic material into the air, water and soil every year, and producing materials so dangerous that they will require constant vigilance by future generations. The point of course is that these consequences were not anticipated, and, most importantly, there was no design considered or implemented. Unlike the industrial revolution and its undesirable outcomes, we now have the opportunity to plan and design a more sustainable, energy-efficient and environmentally responsible economy. We aspire to replace the current linear economy with a circular economy, in which carbon is used and reused over decades in more than one application[381].

Processes of the future

The transition from fundamental discovery to optimization of systems with real feedstock streams, to deployment, needs to happen at an accelerated pace, if we are to meet the US target dates. Figure 4 shows an aggressive timeline from fundamental research to fully deployed technology (Fig. 4, top) that could be accelerated by embracing the key, use-driven fundamental challenges discussed here (Fig. 4, bottom),
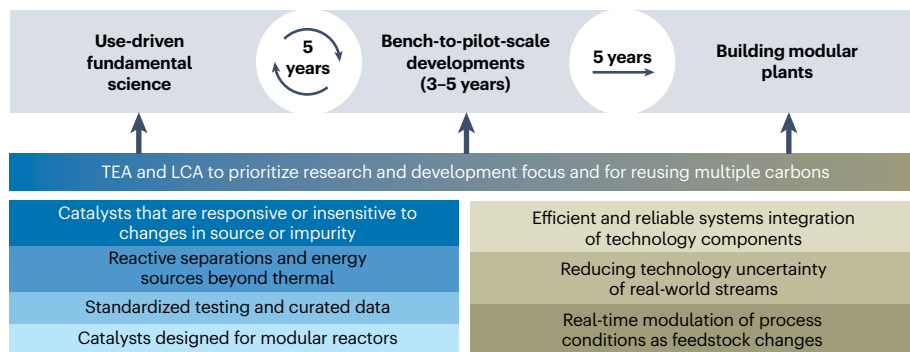

Fig. 4 | An aggressive timeline to close the carbon cycle. An example of an aggressive timeline to translate fundamental science to modular industrial plants to enable defossilization of the difficult-to-electrify segments of our society. Across the top (light blue) are shown the parallel and closely interconnected developments of fundamental science and pilot-scale activities. Implementing techno-economic analysis (TEA) and life cycle analysis (LCA) at all stages of technology development should result in an acceleration of the timeline. A continuous feedback loop between use-driven fundamental science and bench-to-pilot-scale developments will accelerate the transition to technology deployment, potentially reducing the time for building flexible plants from 20 years to 10 years. On the bottom, specific developments needed in catalyst and technology discovery are highlighted. Early-stage research is shown on the left, developments in more advanced stages are shown on the right. Although all these challenges are substantial, with focused research at a variety of technology scales, they should be achievable.

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with direction interaction and connection between the fundamental science and the ultimate end use (Fig. 4, upper left). TEA and LCA, shown in the middle portion of Fig. 4, should offer both acceleration in the timeline, because they are being utilized earlier in the cycle, and early feedback on promising catalysts and separations. If a variety of complex carbon-based feedstocks are to be implemented, a modular chemistry industry for distributed feedstocks and modular reactors will likely be needed to allow for differences in feedstocks. For instance, temporal variation in a given location, such as temperature or humidity, may result in different catalysts or separation materials. Similarly, a modular approach will also allow for feedstock variation in different geographical locations (see Fig. 3).

Reactive separations will likely play an increasing role in future chemical manufacturing, given their process intensification, energy-efficiency and modularity advantages. Broadly, catalysis and separations require integrated design to achieve the highest efficiency. Reactants must be separated from complex waste feedstocks, followed by catalytic conversion at low temperatures and pressures into products with atom efficiency using catalysts comprising Earth-abundant metals[382]. Finally, they need to be separated from reaction mixtures as high-purity, value-added chemicals. Integrated design is a central element for success going forward, and it will benefit from fundamental science developments. Integrating processes, such as reactive separations, offers some enticing benefits, but it will also present substantial challenges to overcome. In integrated reactive separations, there is potential for mismatches between the rates of capture and conversion under given reaction conditions. This would lead to a build-up of reactive intermediates and induce unwanted reactions or impact the reaction equilibrium. Therefore, we need catalysts and adsorbents that are designed to work together (co-designed), which can improve reaction synergy, matching the kinetics and thermodynamics of both processes[377].

We suggest coupling computational methods and data science with experiments to accelerate the pace of discovery and translation to technology. This approach will benefit from standardized feedstocks and testing conditions as well as transparent, complete and well-curated datasets from both separations and catalysis researchers (see Fig. 4 and Box 1). We anticipate that, as these databases grow and algorithms that can accurately extrapolate from such datasets are developed, data science will play a larger role in advancing our knowledge. A closed mass balance of the products analysed will fulfil researchers' needs for system separations and catalysis. Understanding the post-separation products will identify co-contaminants in the presence of which reactions can be performed or pre-treatment separation processes that must be designed.

Data science informed by theory and integrated experiments can assist in the design of modular systems that will require accurate and granular process models. This is particularly true in electrifying chemical processes for which optimal configurations are currently unknown and we still need to identify design principles to build the modules and processes. The incorporation of such models in TEA and LCA can be enabled by surrogate models developed with artificial intelligence (see Box 1).

Circularity can be evaluated at the level of an individual process, a suite of processes and a full system, which in this case could consider all of the processes needed to close the carbon cycle as shown in Fig. 1b, or could be defined as a smaller set of integrated processes. To achieve the highest efficiency, molecular insights must be combined with process-level and systems-level understanding. Process modelling, LCA and TEA will play critical roles in integrating the design of separations and reactions (see Box 2). Conventionally, TEA, LCA, and other process and systems designs are considered only after the science has reached a higher level of maturity. However, we propose integrating these analyses in an iterative, prospective way, rather than retrospectively, to enhance the speed of multiscale system insights. Furthermore, the coordination of multiple processes at a systems level may help future processes to maximize production, while minimizing wasted heat and energy, as well as reagent circulation. Designing materials and processes in an integrated fashion presents an opportunity to further accelerate the translation of materials that can perform in realistic environments (Fig. 4). More broadly, the rapid demand for implementation will require input from diverse disciplines, even beyond the domain sciences, including environmental, economic and equity evaluations.

TEA and LCA provide quantitative metrics as to the cost and environmental footprint of a new product or process. Since an intensified and/or decarbonized process affects other parts of an industrial plant, it is important that TEA includes the entire process. Similarly, LCA and sustainability indicators should be holistic over the entire supply chain, including analysis of CO_2 emissions, solvent toxicity and other waste streams[383]. However, the difficulties in performing both TEA and LCA are not only the broad distribution of evolving technologies, but also the lack of consistent models and unified guidelines[384]. The National Energy Technology Laboratory has developed LCA and TEA toolkits, with consistent updates as changes occur[385–393]. TEA and LCA are crucial even in fundamental and low-TRL efforts, as they can be used to understand economic feasibility, environmental impact and energy-consuming units (reactors, separations and pumps) that should be prioritized for defossilization, and they quantify the supply chain in terms of resources and emissions. However, new technologies are often without comprehensive information and can vary substantially, making it difficult to assess technologies systematically and consistently. Although further discussion is beyond the scope of this Roadmap, we need favourable regulatory and legislative foundations to accelerate the development of new technologies. Given the uncertainty and lack of guidance from policies for selecting emerging products and processes, scenarios analysis and comparison of various processes should be conducted to help guide and focus further research and development[394].

Energy and environmental justice

A roadmap for decarbonization and de-fossilization would not be complete without considering the people and living systems affected, and the environmental impact. As new processes are developed and deployed at scale to address hard-to-electrify sectors, decision-makers should recognize past cumulative inequities; work with communities to make decisions about new facilities or retrofits; analyse the distribution of risks and benefits among demographic groups, such as populations disadvantaged by race or income; and mitigate inequities. Disadvantaged populations suffer from a combination of economic, health, and environmental burdens, which may include high poverty and unemployment rates or exposure to pollution[395]. Justice concerns can apply to any step of the full life cycle of an energy or manufacturing technology, including extraction, processing, transportation, consumption and waste disposal[396]. It is important to ask who is on the frontlines of the energy or manufacturing transition[397] and the ultimate impact on the natural world.

Many pathways towards net-zero emissions described above have justice implications. For example, redesigning polymeric materials can contribute to the dismantling of polluting chemical

Box 2

Accelerated translation through use-driven fundamental science

Use-driven fundamental science, or fundamental science being targeted at a specific application, coupled with techno-economic analysis and life cycle analysis, may substantially accelerate technology deployment. Most typically, fundamental discovery is conducted in a way that points to an application, but there is often little understanding of the real limitations or needs if one wanted to deploy the fundamental discoveries at some later stage. This lack of understanding can lead to technologies that appear to have promise at the bench scale but don't address key limitations of current processes. To switch to a use-directed approach, conversations would begin with industry about what the limitations in their current or proposed processes are and key challenges to overcome. This might include cost, energy input, product purity, catalyst degradation or any number of other challenges. Working with industry to understand these challenges upfront and performing techno-economic and life cycle analyses even at the early stages of catalyst or separation materials development could avoid materials that won't be able to overcome the current challenges and accelerate discovery based on the identified challenges. To be clear, there is value in fundamental discovery that is not closely connected to an application, as this provides new insights for challenges even further down the road. However, in a case such as closing the carbon cycle, in which there are clear fundamental challenges and clear industrial challenges to overcome, a use-directed approach becomes critical to move forward rapidly.

This strategy has been used to improve separations of CO_2. Although CO_2 separations will need to be performed at massive scales, molecule-level interactions govern the macroscale phenomena. As an example, recent advances in solvent-based, post-combustion carbon capture using water-lean solvents were achieved by tightly weaving fundamental experimental and theoretical studies with process engineering, and they were guided by industry partners' years of experience in operating commercial units. Here, a research and development cyclic logic sequence was established with organic and computational chemists, chemical engineers, and industry in which the sequence begins with molecular design, followed by property prediction. Subsequently, synthesis and characterization of target derivatives, coupled with experimental measurements, are conducted, which feed model validation, and the cycle begins anew for the next parameter of need. Continued dialogue through this loop provides insights from each scientific discipline at each stage, allowing the team to learn quickly and pivot, if needed. This tightly integrated approach allows a multidisciplinary team to gain a predictive understanding of critical properties from libraries of thousands of prospective molecules more quickly than compartmentalized make–measure–model efforts.

In one representative example, industry indicated that viscosity was a limiting problem for mass and heat transfer in carbon capture,
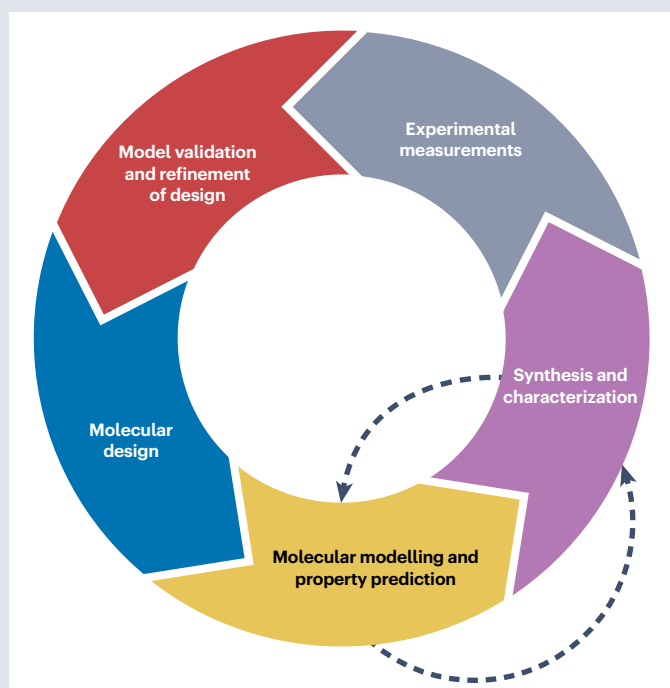
and process engineering suggested that solvents with different molecular structures would help solve it. These fundamental studies discovered unique environments in water-lean solvents in which complex chemical equilibria exist between dissolved CO_2 and chemically fixated CO_2 that result in heterogeneous nano-structuring of the ionic liquid domains, and these regions directly influence macroscale properties such as viscosity and CO_2 transport. The interplay between the chemical equilibria and the nanostructures, although complex, was found to be controllable. Continued commitment to this concept of molecule-level refinement ultimately led to the design of a leading solvent optimized to have the lowest regeneration enthalpy (2.0 GJ t^{-1} CO_2[215]) and total costs of capture (US\$39 t^{-1} CO_2[216]), both of which are below the DOE targets of 2.3 GJ t^{-1} and US\$46 t^{-1}, respectively. By tuning the solvent structure, a tenfold increase in mass transfer[451], a 20% reduction in regeneration energy and a 10^3 reduction in viscosity as compared with previous derivatives were achieved. The foundational knowledge of how the molecule-level interactions impact macroscale properties has enabled the molecule-level redesign of multiple leading solvents for CO_2 capture, accelerating their development by 3–5 years, and it has provided a deep fundamental understanding of molecule-level structures and interactions for acid gas separations.

industry infrastructure currently concentrated in disadvantaged communities[398]. Distributed energy systems may enable more equitable outcomes in community access and self-sufficiency, reliability, and resilience, compared with more centralized systems[399].

Others argue that a new social science research agenda is needed for H_2 to better understand the level and distribution of social effects[400]. DAC technologies, including communities around plants, pipeline infrastructure and storage, will require social acceptance[401].

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Glossary

Carbon dioxide capture and storage

A process in which a relatively pure stream of carbon dioxide (CO_2) from industrial and energy-related sources is separated (captured), conditioned, compressed and transported to a storage location for long-term isolation from the atmosphere — sometimes referred to as carbon capture and storage. This could result in long-term storage (hundreds to thousands of years).

Carbon dioxide capture and utilization

A process in which CO_2 is captured and then used to produce a new product. If the CO_2 is stored in a product for a climate-relevant time horizon, this is referred to as carbon dioxide capture, utilization and storage. Only then, and only combined with CO_2 recently removed from the atmosphere, can carbon dioxide capture, utilization and storage lead to CO_2 removal. Carbon dioxide capture and utilization is sometimes referred to as carbon dioxide capture and use.

Circular economy

Materials, products and services are designed for reuse for the same application and are kept in circulation for as long as possible, seeking to extract maximum value and create minimum waste from all parts of the process.

Clean hydrogen

Molecular hydrogen (H_2) produced with zero or next-to-zero carbon emissions.

Decarbonization

Typically refers to a reduction of the carbon emissions associated with electricity production, industry and transport.

Defossilization

Reduction of the use of fossil-derived chemicals, fuels and materials.

Direct air capture

(DAC). CO_2 capture from the air.

Direct ocean capture

CO_2 capture from the ocean.

Distributed chemical processes

Chemical processes operated at scales that are 2–3 orders of magnitude smaller than the centralized refineries we have today, typically handling on the order of tons to thousands of tons of feedstock per day. These distributed processes may operate at lower capital equipment utilization and/or efficiency than typical refineries to take advantage of lower operating costs.

Keeping carbon in play

A circular carbon cycle in which every carbon atom within products and waste streams is reused, ideally multiple times.

Large-scale energy storage

Storage greater than 1 GWh.

Linear economy

Raw materials are collected and transformed into products that consumers use and discard after a single use.

Long-duration energy storage

Storage systems to provide energy for time scales greater than 100 h.

Net-zero CO_2 emissions

Net-zero CO_2 emissions are achieved when anthropogenic CO_2 emissions are balanced globally by anthropogenic CO_2 removals over a specified period. Net-zero CO_2 emissions are also referred to as carbon neutrality. See also net-zero emissions.

Net-zero emissions

Net-zero emissions are achieved when anthropogenic emissions of greenhouse gases to the atmosphere are balanced by anthropogenic removals over a specified period. When multiple greenhouse gases are involved, the quantification of net-zero emissions depends on the climate metric chosen to compare the emissions of different gases (such as global warming potential, global temperature change potential and others, as well as the chosen time horizon).

Defining the baseline conditions of our current fossil fuel usage is important for evaluating justice metrics, so that proposed technologies can be compared with past or existing technologies. Replacing coal-fired power plants with renewable sources has been projected to result in the avoidance of premature deaths and reductions in hospitalizations because of reduced particulate exposures, especially for disadvantaged communities[402]. Adverse effects may be experienced by workers in declining industries, such as mines, petroleum refineries and manufacturing plants for combustion engine automobiles[397].

Social acceptance of new technologies can only be achieved if social factors, such as health, environmental and economic injustices, are acknowledged and investigated[403]. The circular economy must be just by design[404].

Conclusion and outlook

In this Roadmap, we imagine a future in which there is no waste — in which every item that is created and used can be reused productively at the end of its life, even if for a different purpose. We hope that products will be designed to be carbon-neutral and not with the landfill in mind. Crucially, we should not have to compromise our lifestyle in our efforts to close the carbon cycle and meet the net-zero CO_2 emission goals. Although electrifying with carbon-free or low-carbon energy sources will be a critical component of decarbonization, the segments of our economy that are not easily electrified will continue to need carbon to support the Earth's population. In these areas, defossilization is a critical part of achieving net-zero CO_2 emissions.

The need to replace fossil carbon with non-fossil carbon or H_2 creates a wealth of opportunities in both basic and applied research. With the current research approach, it is highly unlikely that the challenging goals of carbon neutrality in 2030 will be met. We must rethink how discovery science can be more rapidly deployed into higher TRLs. This will entail the implementation of TEA and LCA early in the discovery process, working closely with integrated research teams encompassing complementary disciplines, so that the accelerated advancements needed to achieve the bold US goals can be realized. An additional key element of advancement is the implementation of data science in the discovery and implementation phase, to advance discovery and deal with the heterogeneous nature of the feedstocks. We predict that implementing these developments into modular reactors that can adapt to fluctuations in feedstocks, are co-designed for conversion and separation, and can be placed at the point of feedstock generation will result in chemical processes that meet the targets stated in this Roadmap and ultimately are cognizant of social and environmental impacts. We also posit that these stretch goals can better be achieved if research is conducted in a more collaborative manner, thereby opening opportunities for equitable research.

Therefore, to meet our net-zero CO_2 emissions targets in 2050, we need to change our scientific approach, with support from

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intergovernmental legislation and policies. Achieving defossilization in the hard-to-electrify segments of our economy, by providing alternatives to carbon and keeping carbon in play, will be a momentous step towards this goal. If we can bring this vision to fruition at the urgent pace needed, we can reduce the levels of atmospheric CO_2 and its consequent impact on the environment while moving towards improving energy security and equity.

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