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Net-Zero Emissions Energy Systems

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Providing energy services without net addition of carbon dioxide to the atmosphere

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Figs. 1-3
Table 1
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[Single-sentence summary]: Achieving an energy system that adds no CO_2 to the atmosphere will require focused innovation and cross-sector coordination. [*124 characters*]

[Abstract] Some energy services and industrial processes, such as long-distance freight transport, air travel, highly reliable electricity, and steel and cement manufacturing, are particularly difficult to provide without adding carbon dioxide (CO_2) to the atmosphere. Rapidly growing demand for these services, combined with long lead times for technology development and long lifetimes of energy infrastructure, make decarbonization of these services both essential and urgent. We examine barriers and opportunities associated with these difficult-to-decarbonize services and processes, including possible technological solutions and research and development priorities. A range of existing technologies could meet future demands for these services and processes without net addition of CO_2 to the atmosphere, but their use may depend on a combination of cost reductions via research and innovation, as well as coordinated deployment and integration of operations across currently discrete energy industries. [*129 words*]

People do not want energy itself, but rather the services that energy provides and the products that rely on these services. Even with substantial improvements in efficiency, global demand for energy is projected to increase markedly over this century (*1*). Meanwhile, net emissions of CO_2 from human activities—including not only energy and industrial production, but also land use and agriculture—must approach zero to stabilize global mean temperature (*2, 3*). Indeed, international climate targets such as avoiding more than 2 °C of mean warming are likely to require an energy system with net-zero (or net-negative) emissions later this century (Fig. 1) (*3*).

Energy services such as light duty transportation, heating, cooling, and lighting may be relatively straightforward to decarbonize by electrifying and generating electricity from variable renewable energy sources (such as wind and solar) and dispatchable (i.e. "on-demand") non-renewable sources (including nuclear energy and fossil fuels with carbon capture and storage). However, other energy services essential to modern civilization entail emissions that are likely to be more difficult to fully eliminate. These difficult-to-decarbonize energy services include aviation, long-distance transport, and shipping; production of carbon-intensive structural materials such as steel and cement; and provision of a reliable electricity supply that meets varying demand. To the extent carbon remains involved in these services in the future, net-zero emissions will also entail active management of carbon.

In 2014, difficult-to-eliminate emissions related to aviation, long-distance transportation and shipping; structural materials; and highly-reliable electricity totaled ~9.2 Gt CO_2, or 27% of global CO_2 emissions from all fossil fuel and industrial sources (Fig. 2). Yet, despite their importance, detailed representation of these services in integrated assessment models remains challenging (*4, 5, 6*).

Here, we review the special challenges associated with an energy system that does not add any CO_2 to the atmosphere (a net-zero emission energy system). We discuss prominent technological opportunities and barriers for eliminating and/or managing emissions related to the difficult-to-decarbonize services; pitfalls in which near-term actions may make it more difficult or costly to achieve the net-zero emissions goal; and critical areas for research, development, demonstration, and deployment. Our scope is not comprehensive; we focus on what now seem the most promising technologies and pathways. Our assertions regarding feasibility throughout are not the result of formal, quantitative economic modeling; rather, they are based on comparison of current and projected costs with stated assumptions about progress and policy.

A major conclusion is that it is vital to integrate currently discrete energy sectors and industrial processes. This integration may entail infrastructural and institutional transformations, as well as active management of carbon in the energy system.

Aviation, long-distance transport, and shipping

In 2014, medium- and heavy-duty trucks with mean trip distances of >160 km (>100 miles) accounted for ~270 Mt CO_2 emissions, or 0.8% of global CO_2 emissions from fossil fuel combustion and industry sources (estimated using *11, 12, 13*). Similarly long trips in light-duty vehicles accounted for an additional 40 Mt CO_2, and aviation and other shipping modes (such as trains and ships) emitted 830 and 1,060 Mt CO_2, respectively. Altogether, these sources were responsible for ~6% of global CO_2 emissions (Fig. 2). Meanwhile, both global energy demand for transportation and the ratio of heavy- to light-duty vehicles is expected to increase (*13*).

Light-duty vehicles can be electrified or run on hydrogen without drastic changes in performance except for range and/or refueling time. In contrast, general-use air transportation and long-distance transportation, especially by trucks or ships, have additional constraints of revenue cargo space and payload capacity that mandate energy sources with high volumetric and gravimetric density (*7*). Closed-cycle electrochemical batteries must contain all of their reactants and products. Hence, fuels that are oxidized with ambient air and then vent their exhaust to the atmosphere have a substantial chemical advantage in gravimetric energy density .

Battery- and hydrogen-powered trucks are now used in short-distance trucking (*8*), but at equal range, heavy-duty trucks powered by current lithium-ion batteries and electric motors can carry ~40% less goods than can trucks powered by diesel-fueled, internal combustion engines. The same physical constraints of gravimetric and volumetric energy density likely preclude battery- or hydrogen-powered aircraft for long-distance cargo or passenger service (*9*). Autonomous trucks and distributed manufacturing may fundamentally alter the energy demands of the freight industry, but if available, energy-dense liquid fuels are likely to remain the preferred energy source for long-distance transportation services (*10*).

Options for such energy-dense liquid fuels include the hydrocarbons we now use, as well as hydrogen, ammonia, and alcohols/ethers. In each case, there are options for producing carbon-neutral or low-carbon options that could be integrated to a net-zero emissions energy system (Fig. 1), and each can also be interconverted through existing thermochemical processes (Table 1).

Hydrogen and ammonia fuels

The low volumetric energy density of hydrogen favors transport and storage at low temperatures (-253°C for liquid hydrogen at atmospheric pressure) and/or high pressures (350 to 700 bar), thus requiring heavy and bulky storage containers (*14*). To contain the same total energy as a diesel fuel storage system, a liquid hydrogen storage system would weigh roughly six times more and be about eight times larger (Fig. 3A). However, hydrogen fuel cell or hybrid hydrogen-battery trucks can be more energy efficient than those with internal combustion diesel engines (*15*), requiring less onboard energy storage to achieve the same traveling range. Toyota has recently introduced a heavy duty (36,000 kg), 500 kW fuel cell/battery hybrid truck designed to travel 200 miles on liquid hydrogen and stored electricity, and Nikola has announced a similar battery/fuel cell heavy duty truck with a claimed range of 1300 to 1900 km, comparable to today's long-haul diesel trucks (*16*). If hydrogen can be produced affordably without CO_2 emissions, its use in the transport sector could ultimately be bolstered by the fuel's importance in providing other energy services.

Ammonia is another technologically viable alternative fuel that contains no carbon and may be directly combusted in an engine or may be cracked to produce hydrogen. Its combustion must be carefully controlled to minimize production of highly oxidized products such as NO_x (*17*). Furthermore, like hydrogen, ammonia's gravimetric energy density is considerably lower than that of hydrocarbons such as diesel (Fig. 3A).

Biofuels

Conversion of biomass currently provides the most cost-effective pathway to non-fossil, carbon-containing liquid fuels. Liquid biofuels at present represent about 4.2 EJ of the roughly 100 EJ of energy consumed by the transport sector worldwide. Currently, the main liquid biofuels are ethanol from grain and sugar cane and biodiesel and renewable diesel from oil seeds and waste oils. They are associated with substantial challenges related to their life-cycle carbon emissions, cost, and scalability (*18*).

Photosynthesis converts <5% of incident radiation to chemical energy, and only a fraction of that chemical energy remains in biomass (*19*). Conversion of biomass to fuel also requires energy for processing and transportation. Land used to produce biofuels must have water, nutrient, soil, and climate characteristics suitable for agriculture, thus putting biofuels in competition with other land uses. This has implications for food security, sustainable rural economies, and the protection of nature and ecosystem services (*20*). Potential land-use competition is heightened by increasing interest in bioenergy with carbon capture and storage (BECCS) as a source of negative emissions (that is, carbon dioxide removal), which biofuels can provide (*21*).

Advanced biofuel efforts include processes that seek to overcome the recalcitrance of cellulose to allow use of different feedstocks (e.g., woody crops, agricultural residues, and wastes), to achieve large-scale production of liquid transportation fuels at costs roughly competitive with gasoline (e.g., U.S. $19/GJ or U.S. $1.51/gallon of ethanol) (*22*). As technology matures and overall decarbonization efforts of the energy system proceed, biofuels may be able to largely avoid fossil fuel inputs such as those related to on-farm processes and transport, as well as emissions associated with induced land use change (*23, 24*). The extent to which biomass will supply liquid fuels in a future net-zero emissions energy system thus depends on advances in conversion technology, competing demands for bioenergy and land, the feasibility of other sources of carbon-neutral fuels, and integration of biomass production with other objectives (*25*).

Synthetic hydrocarbons

Liquid hydrocarbons can also be synthesized by industrial hydrogenation of feedstock carbon, such as the reaction of carbon monoxide and hydrogen by the Fischer-Tropsch process (*26*). If the carbon contained in the feedstock is taken from the atmosphere and no fossil energy is used for the production, processing, and transport of feedstocks and synthesized fuels, the resulting hydrocarbons would be carbon-neutral (Fig. 1). For example, emissions-free electricity could be used to produce H_2 by electrolysis of water, which would be reacted with CO_2 removed from the atmosphere either by direct air capture or photosynthesis (which in the latter case could include CO_2 captured from the exhaust of biomass or biogas combustion) (*27, 28*).

At present, the cost of electrolysis is a major barrier. This cost includes both the capital costs of electrolyzers and the cost of emissions-free electricity; 60 to 70% of current electrolytic hydrogen cost is electricity (Fig. 1C) (*28, 29*). The cheapest and most mature electrolysis technology available today uses alkaline electrolytes (such as KOH or NaOH) together with metal catalysts to produce hydrogen at an efficiency of 50 to 60% and a cost of ~ U.S. $5.50/kg H_2 (assuming industrial electricity costs of U.S. $0.07/kWh and 75% utilization rates) (*29, 30*). At this cost of hydrogen, the minimum price of synthesized hydrocarbons would be $1.70 to $1.50/liter of diesel equivalent (or $5.50 to $6.50/gallon and $42 to 50 per GJ; assuming carbon feedstock costs of $0-100 per ton of CO_2 and very low process costs of $0.05/liter or $1.50 per GJ (*28*)). For comparison, H_2 from steam reforming of fossil CH_4 into CO_2 and H_2 currently costs $1.30 to 1.50 per kg (red line in Fig. 3B) (*29, 31*). Thus, the feasibility of synthesizing hydrocarbons from electrolytic H_2 may depend upon demonstrating valuable cross-sector benefits, such as balancing variability of renewable electricity generation, or else a policy-imposed price of ~$400 per ton of CO_2 emitted (which would also raise fossil diesel prices by ~$1.00/liter or ~$4.00/gallon).

In the absence of policies or cross-sector coordination, hydrogen costs of $2.00/kg (i.e., approaching the cost of fossil-derived hydrogen and synthesized diesel of ~$0.79/liter or $3.00/gallon) could be achieved, for example, if electricity costs were $0.03/kWh and current electrolyzer costs were reduced by 60 to 80% (*29*) (Fig. 3B). Such reductions may be possible (*33*) but may require centralized electrolysis (*34*) using less mature but promising technologies, such as high-temperature solid oxide or molten carbonate fuel cells, or thermochemical water splitting (*30, 35*). Fuel markets are vastly more flexible than instantaneously balanced electricity markets due to the relative simplicity of large, long-term storage of chemical fuels. Hence, using emissions-free electricity to make fuels represents a critical opportunity for integrating electricity and transportation systems to supply a persistent demand for carbon-neutral fuels while boosting utilization rates of system assets.

Direct solar fuels

Photoelectrochemical cells or particulate/molecular photocatalysts directly split water using sunlight to produce fuel by artificial photosynthesis, without land-use constraints associated with biomass (*36*). Hydrogen production efficiencies can be high, but costs, capacity factors, and lifetimes need to be improved to obtain an integrated, cost-advantaged approach to carbon-neutral fuel production (*37*). Short-lived laboratory demonstrations have also produced liquid carbon-containing fuels using concentrated CO_2 streams (*38*) (Fig. 1H), in some cases using bacteria as catalysts.

Outlook

Large-scale production of carbon-neutral and energy-dense liquid fuels may be critical to achieving a net-zero emissions energy system. Such fuels could provide a highly advantageous bridge between the stationary and transportation energy production sectors, and may therefore deserve special priority in energy research and development efforts.

Structural materials

Economic development and industrialization are historically linked to the construction of infrastructure. Between 2000 and 2015, cement and steel use persistently averaged 50 and 21 tons per million dollars of global GDP, respectively (~1 kg per person per day in developed countries) (*4*). Globally, ~1,320 and 1,740 Mt CO_2 emissions emanated from chemical reactions involved with manufacture of cement and steel, respectively (Fig.2) (*12, 40, 41*); altogether, this equates to ~9% of global CO_2 emissions in 2014 (purple and blue in Fig. 1). Although materials intensity of construction could be substantially reduced (*42, 43*), steel demand is projected to grow by 3.3% per year to 2.4 billion tons in 2025 (*44*) and cement production is projected to grow by 0.8 to 1.2% per year to 3.7 to 4.4 billion tons in 2050 (*45, 46*), continuing historical patterns of infrastructure accumulation and materials use seen in regions such as China, India and Africa (*4*).

Decarbonizing the provision of cement and steel will require major changes in manufacturing processes, use of alternative materials that do not emit CO_2 during manufacture, or carbon capture and storage (CCS) technologies to minimize the release of process-related CO_2 to the atmosphere (*39*) (Fig. 1B).

Steel

During steel making, carbon (coke from coking coal) is used to reduce iron oxide ore in blast furnaces, producing 1.6 to 3.1 tons of process CO_2 per ton of crude steel produced (*41*).This is in addition to CO_2 emissions from fossil fuels burned to generate the necessary high temperatures (1100 to 1500 °C). Reductions in CO_2 emissions per ton crude steel are possible through use of electric arc furnace (EAF) “minimills” that operate using emissions-free electricity; efficiency improvements (such as top gas recovery); new process methods (such as “Ultra-low CO_2 Direct Reduction,” ULCORED); process heat fuel-switching; and decreased demand via better engineering. For example, a global switch to ultrahigh-strength steel for vehicles would avoid ~160 Mt CO_2 annually. The availability of scrap steel feedstocks currently constrains EAF production to ~30% of global demand *(47, 48)*, and the other improvements reduce—but do not eliminate—emissions.

Prominent alternative reductants include charcoal (biomass-derived carbon) and hydrogen. Charcoal was used until the 18th century, and the Brazilian steel sector has increasingly substituted charcoal for coal to reduce fossil CO_2 emissions (*49*). However, the ~0.6 tons of charcoal needed per ton of steel produced require 0.1 to 0.3 hectares of Brazilian eucalyptus plantation (*49, 50*). Hundreds of millions of hectares of highly productive land would thus be necessary to meet expected charcoal demands of the steel industry, and associated land use change emissions could outweigh avoided fossil fuel emissions, as has happened in Brazil (*49*). Hydrogen might also be used as a reductant, but quality could be compromised because carbon imparts strength and other desirable properties to steel (*51*).

Cost notwithstanding, capture and storage of process CO_2 emissions has been demonstrated and may be feasible, particularly in designs such as top gas recycling blast furnaces, where concentrations and partial pressures of CO and CO_2 are high (40 to 50% and 35% by volume, respectively; Figs. 1G and 1E) (*52, 53*).

Cement

About 40% of the CO_2 emissions during cement production are from fossil energy inputs, with the remaining CO_2 emissions arising from the calcination of $CaCO_3$ (typically limestone) (*54*). Eliminating the process emissions requires fundamental changes to the cement-making process and cement materials, and/or installation of carbon-capture technology (*45*) (Fig. 1G). CO_2 concentrations are typically ~30% by volume in cement plant flue gas (compared to ~10 to 15% in power plant flue gas; *55*), improving the viability of post-combustion carbon capture. Firing the kiln with oxygen and recycled CO_2 is another option (*56*) but it may be challenging to manage the composition of gases in existing cement kilns that are not gas-tight, operate at very high temperatures (~1500 °C), and rotate (*57*).

A substantial fraction of process CO_2 emissions from cement production is reabsorbed on a time scale of 50 years through natural carbonation of cement materials (*58*). Hence, capture of emissions associated with cement manufacture might result in overall net negative emissions as a result of the carbonation of produced cement. If complete carbonation is ensured, captured process emissions could provide an alternative feedstock for carbon-neutral synthetic liquid fuels.

Outlook

A future net-zero energy system must provide a way to supply structural materials such as steel and cement, or close substitutes, without adding CO_2 to the atmosphere. Although alternative processes might avoid liberation and use of carbon, the cement and steel industries are especially averse to the risk of compromising the mechanical properties of produced materials. Demonstration and testing of such alternatives at scale is therefore a priority. Unless and until such alternatives are adopted, eliminating emissions related to steel and cement will depend on CCS.

Highly reliable electricity

Modern economies demand highly reliable electricity; for example, demand must be met >99.9% of the time (Fig. 1A). This requires investment in energy generation or storage assets that will be used a small percentage of the time, when demand is high relative to variable or baseload generation.

As the share of renewable electricity has grown in the U.S., natural gas-fired generators have increasingly been used to provide generating flexibility because of their relatively low fixed costs (Fig. 3B), ability to ramp up and down quickly (*59*), and the affordability of natural gas (*60*). In other countries, other fossil fuel sources or hydroelectricity are used to provide flexibility. We estimate that CO_2 emissions from such "load-following" electricity were ~4,000 Mt CO_2 in 2014 (~12% of global fossil-fuel and industry emissions), based loosely on the proportion of electricity demand in excess of minimum demand (Fig. 2) (*61*).

The central challenge of a highly reliable zero-carbon electricity system is thus to achieve the flexibility, scalability, and low capital costs of electricity that can currently be provided by natural gas-fired generators—but without emitting CO_2. This might be accomplished by a mix of flexible generation, energy storage, and demand management.

Flexible generation

Even when spanning large geographical areas, a system in which variable energy from wind and solar are major sources of electricity will have occasional but substantial and long-term mismatches between supply and demand. For example, such gaps in the U.S. are commonly tens of petajoules (40 PJ=10.8 TWh=24 hours of mean U.S. electricity demand in 2015) and span multiple days- or even weeks (*62*). Thus, even with continental-scale or global electricity interconnections (*62-64*), highly reliable electricity in such a system will require either very substantial amounts of dispatchable electricity sources (e.g., generators or stored energy) that operate less than 20% of the time or corresponding amounts of demand management. Similar challenges apply if most electricity were produced by nuclear generators or coal-fired power plants equipped with carbon capture and storage, suggesting an important role for generators with higher variable cost, such as gas turbines using synthetic hydrocarbons or hydrogen as fuel (Fig. 1P; see, e.g., *65*).

Equipping dispatchable natural gas, biomass, or syngas generators with carbon capture and storage (CCS) could allow continued system reliability with drastically reduced CO_2 emissions. When fueled by syngas or biomass containing carbon captured from the atmosphere, such CCS offers an opportunity for negative emissions. However, the capital costs of CCS-equipped generators are currently considerably higher than for generators without CCS (Fig. 3B). Moreover, CCS technologies designed for generators that operate a large fraction of the time (with high "capacity factors"), such as coal-burning plants, may be less efficient and effective when generators operate at lower capacity factors (*66*). Use of CCS-equipped generators to flexibly produce back-up electricity and hydrogen for fuel synthesis could help alleviate temporal mismatches between electricity generation and demand.

Nuclear fission plants can operate flexibly to follow loads if adjustments are made to coolant flow rate and circulation, control and fuel rod positions, and/or dumping steam (*67, 68*). In the U.S., the design and high capital costs of nuclear plants have historically obligated their near-continuous "baseload" operation, often at capacity factors >90%. If capital costs could be reduced sufficiently, nuclear power might also become a cost-competitive source of load-following power, but costs may be increasing over time in some places (*69, 70, 71*). Similar to CCS-equipped gas generators, the economic feasibility of next-generation advanced nuclear plants may depend on flexibly producing multiple energy products such as electricity, high-temperature heat, and/or hydrogen.

Energy storage

Reliable electricity could also be achieved through energy storage technologies. The value of today's energy storage is currently greatest when frequent cycling is required, such as for minute-to-minute frequency regulation or price arbitrage (*72*). Cost-effectively storing and discharging much larger quantities of energy over consecutive days and less frequent cycling may favor a different set of innovative technologies, policies, and valuation (*72, 73*).

Chemical bonds. Chemical storage of energy in gas or liquid fuels is a key option for achieving an integrated net-zero emissions energy system (Table 1). Stored electrolytic hydrogen can be converted back to electricity either in fuel cells or by combustion in gas turbines (power-to-gas-to-power or P2G2P; Figs. 1F, 1P, red curve in 3D); commercial-scale P2G2P systems currently exhibit a round-trip efficiency (i.e. energy out divided by energy in) of >30% (*74*). Regenerative fuel cells, in which the same assets are used to interconvert electricity and hydrogen, could boost capacity factors, but would benefit from improvements in round-trip efficiency (now 40 to 50% in proton-exchange membrane designs) and chemical substitutes for expensive precious metal catalysts (*75, 76*).

Hydrogen can also either be combined with non-fossil CO_2 via methanation to create renewable methane or can be mixed in low concentrations (<10%) with natural gas or biogas for combustion in existing power plants. Existing natural gas pipelines, turbines, and end-use equipment could be retrofitted over time for use with pure hydrogen or richer hydrogen blends (*77, 78*), although there may be difficult trade-offs of cost and safety during such a transition.

Current mass-market rechargeable batteries serve high-value consumer markets that prize round-trip efficiency, energy density, and high charge/discharge rates. Although these batteries can provide valuable short-duration ancillary services (such as frequency regulation and backup power), their capital cost per energy capacity and power capacity makes them expensive for grid-scale applications that store large quantities of energy and cycle infrequently. For an example grid-scale use case with an electricity cost of $0.035/kWh (Fig. 3D), the estimated cost of discharged electricity using current lithium-ion batteries is roughly $0.14/kWh ($39/GJ) if cycled daily, but rises to $0.50/kWh ($139/GJ) for weekly cycling. Assuming that targets for halving the energy capacity costs of lithium-ion batteries are reached (e.g., ~$130/kWh of capacity) (*73, 79, 80*), the levelized cost of discharged electricity would fall to ~$0.29/kWh ($81/GJ) for weekly cycling. Cost estimates for current vanadium redox flow batteries are even higher than for current lithium-ion batteries, but lower cost flow chemistries are in development (*81*). Efficiency, physical size, charge/discharge rates, and operating costs could in principle be sacrificed to reduce the energy capacity costs of stationary batteries. Not shown in Fig. 3D, less-efficient (e.g., 70% round-trip) batteries based on abundant materials such as sulfur might reduce capital cost per unit energy capacity to $8/kWh (with a power capacity cost of $150/kW), leading to a levelized cost of discharged electricity for the grid-scale use case in the range of $0.06-0.09/kWh ($17-25 per GJ), assuming 20-100 cycles per year over 20 years (*81*).

Utilization rates might be increased if electric vehicle batteries were used to support the electrical grid (vehicle-to-grid, V2G), presuming that the disruption to vehicle owners from diminished battery charge would be less costly than an outage would be to electricity consumers (*82*). For example, if all of the ~150 million light duty vehicles in the U.S. were electrified, 10% of each battery's 100 kWh charge would provide 1.5 TWh, which is commensurate with ~3 hours of the country's average ~0.5 TW power demand. It is also not yet clear how owners would be compensated for the long-term impacts on their vehicles' battery cycle life, whether periods of high electricity demand would be coincident with periods of high transportation demand, whether the ubiquitous charging infrastructure entailed would be cost-effective, whether the scale and timing of the consent, control, and payment transactions would be manageable at grid-relevant scales (~30 million transactions per 15 minute period), or how emerging technologies and social norms (such as shared autonomous vehicles) might affect V2G feasibility.

Potential and kinetic energy. Water pumped into superposed reservoirs for later release through hydroelectric generators is a cost-effective and technologically mature option for storing large quantities of energy with high round-trip efficiency (>80%). Although capital costs of such pumped storage are substantial, long lifetimes of reservoirs result in competitive levelized costs of discharged electricity when cycled at least weekly (Fig. 3D). Major barriers are the availability of water and suitable reservoirs, social and environmental opposition, and constraints on the timing of water releases by non-energy considerations such as flood protection, recreation, and the storage and delivery of water for agriculture (*83*). Underground and undersea designs, as well as weight-based systems that do not use water, might expand the number of possible sites, avoid non-energy conflicts, and allay some social and environmental concerns (*84-86*).

Electricity may also be stored by compressing air in underground geologic formations, underwater containers, or above-ground pressure vessels. Electricity is then recovered with turbines when air is subsequently released to the atmosphere. Diabatic designs vent heat generated during compression and thus require an external (emissions-free) source of heat when the air is released, reducing round-trip efficiency to <50%. Adiabatic and isothermal designs achieve higher efficiencies (>75%) by storing both compressed air and heat, and similarly efficient underwater systems have been proposed (*84*).

Thermal energy. Thermal storage systems are based on sensible heat (e.g., in water tanks, building envelopes, molten salt, or solid materials such as bricks and gravel), latent heat (e.g., solid-solid or solid-liquid transformations of phase-change materials), or thermochemical reactions. Sensible heat storage systems are characterized by low energy densities (36-180 kJ/kg or 10-50 $\mathrm{Wh_{th}}$/kg) and high costs (*84, 87, 88*). Future cost targets are $<\$15/\mathrm{kWh_{th}}$; *89*). Thermal storage is well-suited to within-day shifting of heating and cooling loads, whereas low efficiency, heat losses, and physical size are key barriers to filling week-long, large-scale (e.g., 30% of daily demand) shortfalls in electricity generation.

Demand management

Technologies that allow electricity demand (Figs. 1A, 1B, and 1C) to be shifted in time (load-shifting or load-shaping) or curtailed to better correlate with supply would improve overall system reliability while reducing the need for underutilized, flexible back-up generators (*90, 91*). Smart charging of electric vehicles, shifted heating and cooling cycles, and scheduling of appliances could cost-effectively reduce peak loads in the U.S. by ~6% and thus avoid 77 GW of otherwise needed generating capacity (~7% of U.S. generating capacity in 2017) (*92*). Managing larger quantities of energy demand for longer times (e.g., tens of PJ over weeks) would involve idling large industrial uses of electricity—thus underutilizing other valuable capital—or effectively curtailing service. Exploring and developing new technologies that can manage weekly or seasonal gaps in electricity supply is an important area for further research (*93*).

Outlook

Non-emitting electricity sources, energy-storage technologies, and demand management options that are now available and capable of accommodating large, multi-day mismatches in electricity supply and demand are characterized by high capital costs compared to the current costs of some variable electricity sources or natural gas-fired generators. Achieving affordable, reliable and net-zero emissions power systems may thus depend on substantially reducing such capital costs via continued innovation and deployment, emphasizing systems that can be operated to provide multiple energy services.

Carbon management

Recycling and removal of carbon from the atmosphere (carbon management) is likely to be an important activity of any net-zero emissions energy system. For example, synthesized hydrocarbons that contain carbon captured from the atmosphere will not increase atmospheric CO_2 when oxidized. Integrated assessment models also increasingly require negative emissions to limit the increase in global mean temperatures to 2 °C (*94-97*), for example via afforestation/reforestation, enhanced mineral weathering, bioenergy with CCS, or direct capture of CO_2 from the air (*20*).

Capture and storage will be distinct carbon management services in a net-zero emissions energy system (e.g., Fig. 1J and 1E). Carbon captured from the ambient air could be used to synthesize carbon-neutral hydrocarbon fuels or sequestered to produce negative emissions. Carbon captured from combustion of biomass or synthesized hydrocarbons could be recycled to produce more fuels (*98*). Storage of captured CO_2 (e.g., underground) will be required to the extent that uses of fossil carbon persist and/or that negative emissions are needed (*20*).

For industrial CO_2 capture, research and development are needed to reduce the capital costs and costs related to energy for gas separation and compression (*99*). Future constraints on land, water, and food resources may limit biologically mediated capture (*20*). The main challenges to direct air capture include costs to manufacture sorbents and structures, energize the process, and handle and transport the captured CO_2 (*100, 101*). Despite multiple demonstrations at scale (~15 Mt CO_2/year are now being injected underground (*99*)), financing carbon storage projects with high perceived risks and long-term liability discharge remains a major challenge (*102*).

Discussion

We have estimated that difficult-to-eliminate emissions related to aviation, long-distance transportation and shipping; structural materials; and highly-reliable electricity represented more than a quarter of global fossil fuel and industry CO_2 emissions in 2014 (Fig. 2). But economic and human development goals; trends in international trade and travel; the rapidly growing share of variable energy sources (*103*); and the large-scale electrification of other sectors all suggest that demand for the energy services and processes associated with difficult-to-eliminate emissions will increase substantially in the future. For example, in some of the Shared Socioeconomic Pathways that were recently developed by the climate change research community to frame analysis of future climate impacts, global final energy demand more than doubles by 2100 (*104*); hence, the magnitude of these difficult-to-eliminate emissions could in the future be comparable to the level of total current emissions.

Combinations of known technologies could eliminate emissions related to all essential energy services and processes (Fig. 1), but substantial increases in costs are an immediate barrier to avoiding emissions in each category. In some cases, innovation and deployment can be expected to reduce costs and create new options (e.g., *33, 73, 105, 106*). More rapid changes may depend on coordinating operations across energy and industry sectors, which could help boost utilization rates of capital-intensive assets. In practice, this would entail systematizing and explicitly valuing many of the interconnections depicted in Fig. 1, which would also mean overcoming institutional and organizational challenges to create new markets and ensure cooperation among regulators and disparate, risk-averse businesses. We thus suggest two parallel broad streams of R&D effort: (1) research in technologies and processes that can provide these difficult to decarbonize energy services; and (2) research in systems integration that would allow for the provision of these services and products in a reliable and cost-effective way.

We have focused on provision of energy services without adding CO_2 to the atmosphere. However, many of the challenges discussed here could be reduced by moderating demand, such as by substantial improvements in energy and materials efficiency. Particularly crucial are the rate and intensity of economic growth in developing countries and the degree to which such growth can avoid fossil-fuel energy while prioritizing human development, environmental protection, sustainability, and social equity (*4, 107, 108*). Furthermore, many energy services rely on long-lived infrastructure and systems such that current investment decisions may lock in patterns of energy supply and demand (and thereby the cost of emissions reductions) for half a century to come (*112*). The collective and reinforcing inertia of existing technologies, policies, institutions, and behavioral norms may actively inhibit innovation of emissions-free technologies (*113*). Emissions of CO_2 and other radiatively active gases and aerosols (*109*), from land use and land-use change could also cause substantial warming (e.g., *110*).

Conclusion

We have herein enumerated energy services that must be served by any future net-zero emission energy system and have explored the technological and economic constraints of each. A successful transition to a future net-zero emission energy system is likely to depend on the availability of vast amounts of inexpensive, emissions-free electricity; mechanisms to quickly and cheaply balance large and uncertain time-varying differences between demand and electricity generation; electrified substitutes for most fuel-using devices; alternative materials and manufacturing processes for structural materials; and carbon-neutral fuels for the parts of the economy that are not easily electrified. The specific technologies that will be favored in future marketplaces are largely uncertain, but only a finite number of technology choices exist today for each functional role. To take appropriate actions in the near-term, it is imperative to clearly identify desired endpoints. If we want to achieve a robust, reliable, affordable, net-zero emission energy system later this century, we must be researching, developing, demonstrating and deploying those candidate technologies now.

[*5,412words excluding abstract*]

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Figures

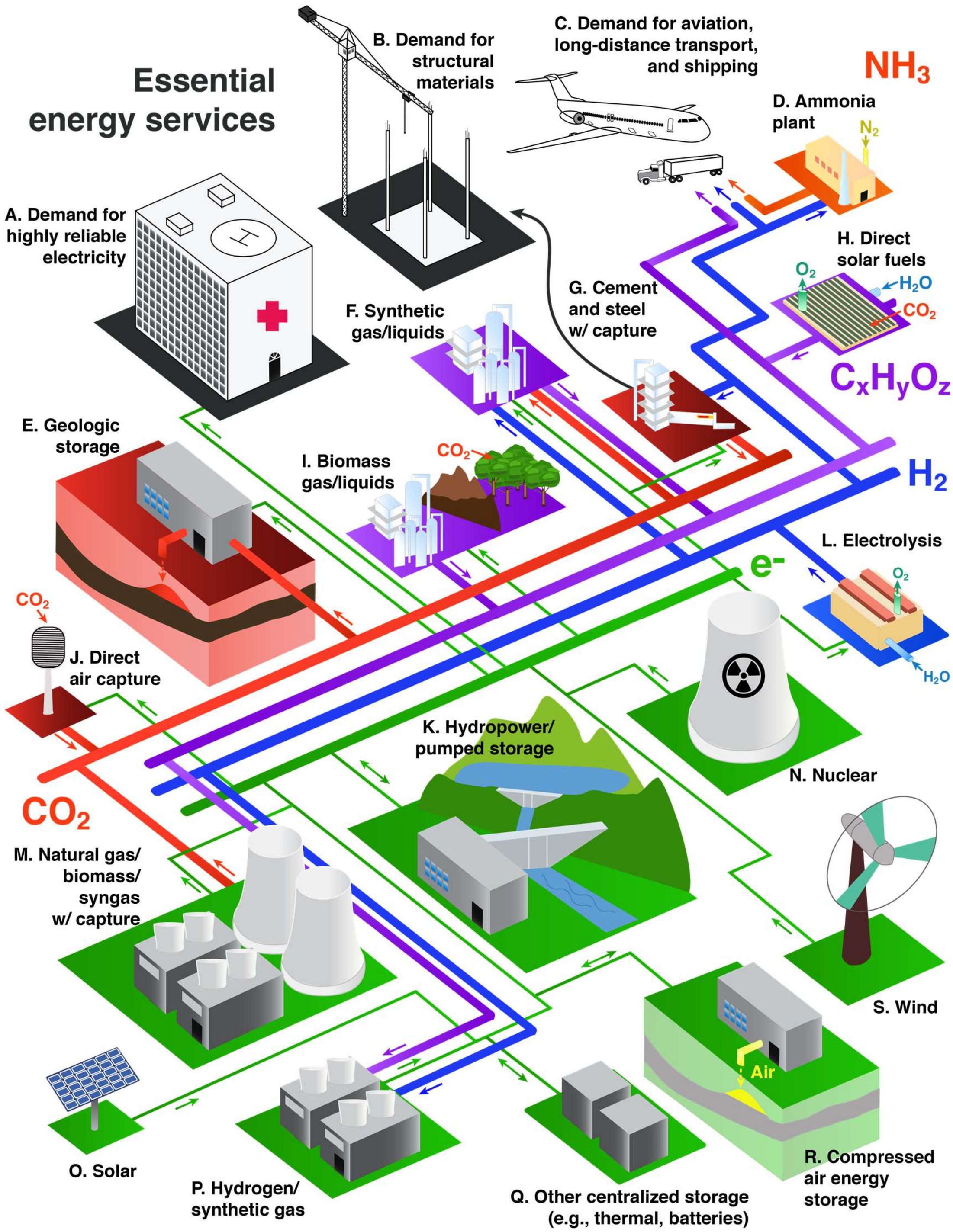

Fig. 1. Schematic of an integrated system that can provide essential energy services without adding any CO_2 to the atmosphere. Colors indicate the dominant role of specific technologies and processes: electricity generation and transmission in green, hydrogen production and transport in blue, hydrocarbon production and transport in purple, ammonia production and transport in orange, carbon management in red, and end uses of energy and materials in black.

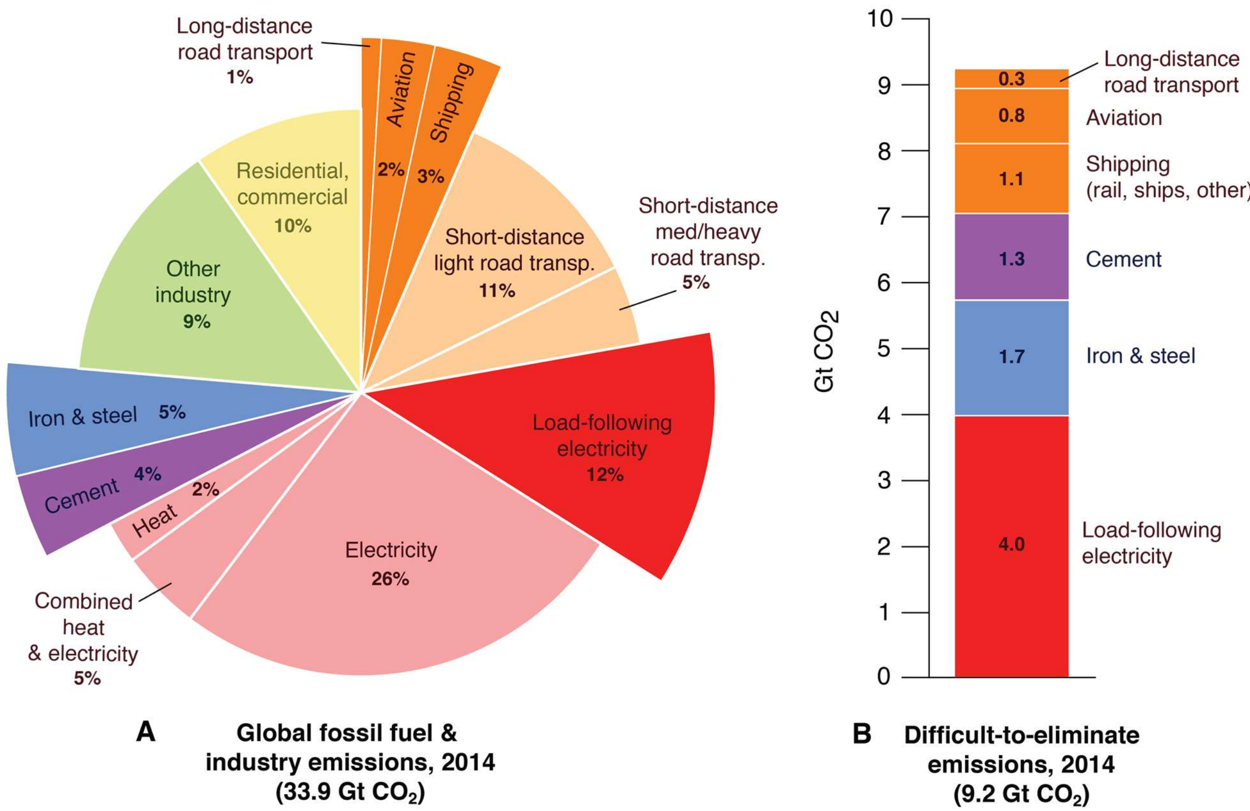

Fig. 2. Difficult-to-eliminate emissions in current context. Estimates of CO_2 emissions related to different energy services, highlighting (e.g., by longer pie pieces in **A**) those services that will be the most difficult to decarbonize, and the magnitude of 2014 emissions from those difficult-to-eliminate emissions. Note that the shares and emissions shown here reflect a global energy system that still relies primarily on fossil fuels and that serves many developing regions. Both the shares (**A**) and the level of emissions (**B)** related to these difficult-to-decarbonize services are likely to increase in the future. Totals and sectoral breakdowns shown are based primarily on data from IEA and EDGAR 4.3 databases. The highlighted iron & steel andcement emissions are those related to the dominant industrial processes only; fossil energy inputs to those sectors that are more easily decarbonized are included with direct emissions from other industry in the Other industry category. Residential and commercial emissions are those produced directly by businesses and households, and Electricity, CHP (combined heat and power), and Heat represent emissions from the energy sector. See *Supplementary Materials* for further details.

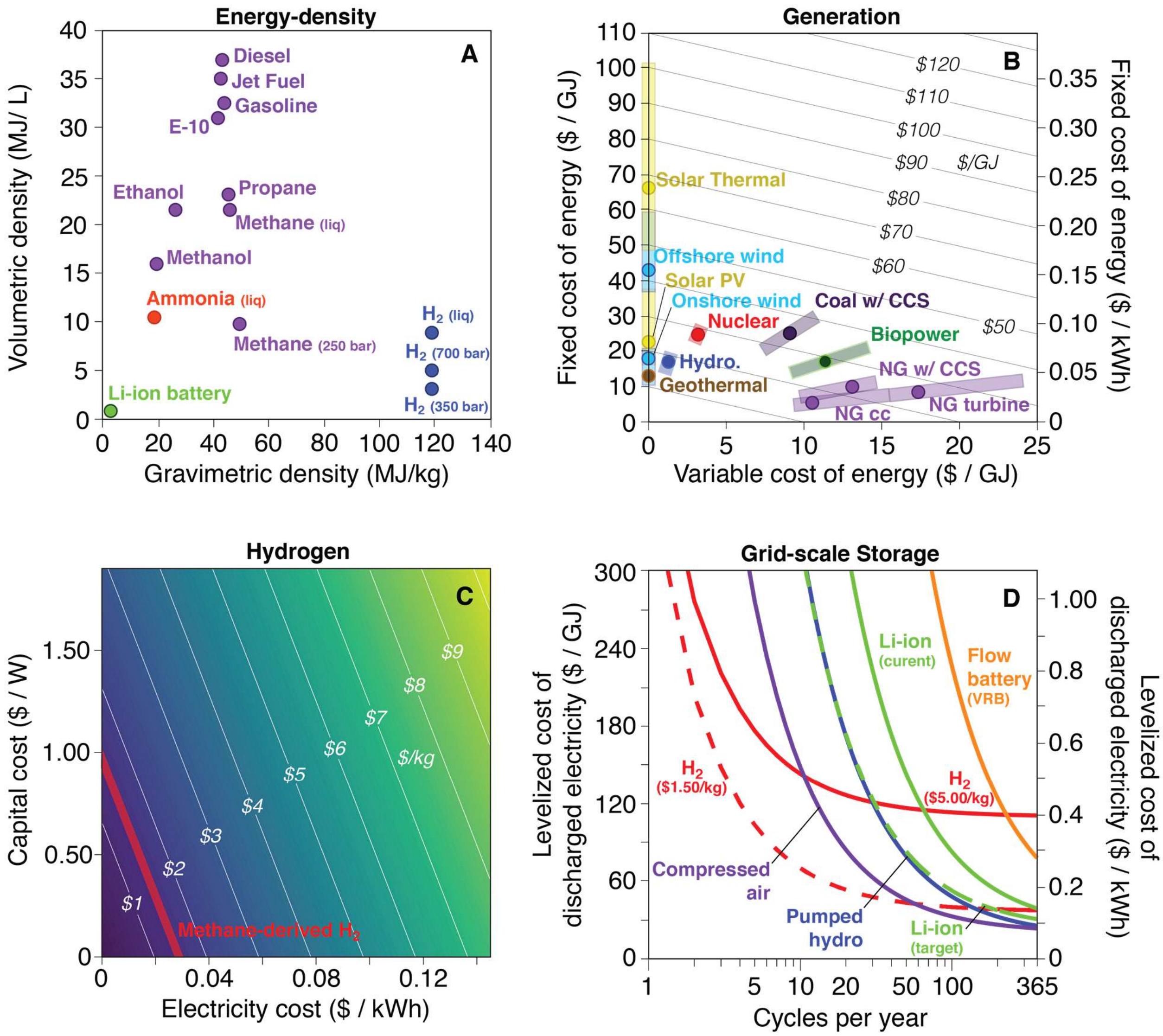

Fig. 3. Comparisons of energy sources and technologies. Panel **A** shows the energy density of energy sources for transportation, including hydrocarbons (purple), ammonia (orange), hydrogen (blue), and current lithium ion batteries (green). Panel **B** shows relationships between fixed capital versus variable operating costs of new generation resources in the U.S., with shaded ranges of regional and tax credit variation and contours of total levelized cost of electricity, assuming average capacity factors and equipment lifetimes. “NG cc” is natural gas combined cycle. (*113*). Panel **C** shows the relationship of capital cost (i.e. electrolyzer cost) and electricity price on the cost of produced hydrogen (i.e. the simplest possible electricity-to-fuel conversion) assuming a 25-year lifetime, 80% capacity factor, 65% operating efficiency, 2 year construction time, and straight-line depreciation over 10 years with $0 salvage value (*29*). For comparison, hydrogen is currently produced by steam methane reformation at costs of ~$1.50/kg H_2 (~$10/GJ; red line). Panel **D** compares the levelized costs of discharged electricity as a function of cycles per year, assuming constant power capacity, 20 year service life, and full discharge over 8 hours for daily cycling or 121 days for yearly cycling. Dashed lines for hydrogen and Li-ion reflect aspirational targets. See *Supplementary Materials* for further details.

Table 1. Key energy carriers and the processes for interconversion. Processes listed in each cell convert the row energy carrier to the column energy carrier. Further details about costs and efficiencies of these interconversions are available in the *Supplementary Materials*.

from \ to	e^-	H_2	$C_xO_yH_z$	NH_3
e^-		• Electrolysis (*29*) ($5-6/kg H_2)	• Electrolysis + methanation • Electrolysis + Fischer-Tropsch	• Electrolysis + Haber-Bosch
H_2	• Combustion • Oxidation via fuel cell(*115, 116*)		• Methanation ($0.07-0.57/m^3 CH_4) • Fischer-Tropsch ($4.40 to $15.00/gallon of gasoline-equivalent)	• Haber-Bosch ($0.50-0.60/kg NH_3)(*116*)
$C_xO_yH_z$	• Combustion	• Steam reforming ($1.29-1.50/kg H_2) • Biomass gasification ($4.80-5.40/kg H_2)		• Steam reforming + Haber-Bosch
NH_3	• Combustion	• Metal catalysts (*117*) (~$3/kg H_2) • Sodium amide (*118*)	• Metal catalysts + methanation/Fischer-Tropsch • Sodium amide + methanation/Fischer-Tropsch	