Net-zero emissions energy systems

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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 (e.g., long-distance freight transport, air travel, highly reliable electricity, 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 the challenge to decarbonize these services both essential and urgent. We examine barriers and opportunities associated with these most difficult-to-decarbonize services, including possible technological solutions and research and development priorities. A range of existing technologies could meet future demands for these services without net addition of CO_2 to the atmosphere, but their use may ultimately depend upon a combination of cost-reductions via research and innovation, and coordinated deployment and integration of operations across now-discrete energy industries. [*129 words*]

People do not demand energy *per se*, they demand services that energy provides and 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 by 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 2 °C of mean warming are likely to require a net-zero (or net-negative) emission energy system later this century (*3*).

Some energy services may be relatively straightforward to decarbonize by electrifying and generating electricity from some combination of variable renewable energy sources (e.g., wind and solar) and dispatchable non-renewable sources (e.g., nuclear and fossil fuels with carbon capture and storage). However, other energy services that are essential to modern civilization entail emissions that are likely to be more difficult to fully eliminate, including: (1) aviation, long-distance transport, and shipping; (2) carbon-intensive structural materials such as steel and cement, and (3) a highly reliable electricity supply that meets varying demand. Detailed representation of these difficult-to-decarbonize services in integrated assessment models remains challenging (e.g., *4, 5, 6*).

Here, we review the special challenges associated with an energy system that does not add any CO_2 to the atmosphere (what we will refer to as a "net-zero emission energy system"; Fig 1). 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 ultimate 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.

A major conclusion is that it is vital to integrate currently discrete energy sectors and industrial processes—which may entail both infrastructural and institutional transformations as well as active management of carbon in the energy system. Our assertions regarding feasibility throughout are not the result of formal, quantitative economic modeling, but rather compare current and projected costs with stated assumptions about progress and policy.

Aviation, long-distance transport, and shipping

Light-duty vehicles can be electrified or run on hydrogen without drastic changes in performance, but requirements of range, refueling time, revenue cargo space, and payload capacity for general-use air transportation and long-distance transportation of freight by trucks or ships mandate energy sources with high volumetric and gravimetric density (*7*). Fuels combusted with ambient air with exhaust vented to the atmosphere have a substantial chemical advantage in gravimetric energy density compared to closed-cycle electrochemical batteries which must carry all their reactants as well as their products.

Although battery- and hydrogen-powered trucks are now used in short-distance trucking (e.g., *8*), at equal range the payload capacity of heavy-duty trucks powered by current lithium-ion batteries and electric motors is ~40% less than trucks powered by diesel-fueled, internal combustion engines. Similarly, physical constraints likely preclude battery- or hydrogen-powered aircraft for long-distance cargo or passenger service (*9*). Autonomous trucks and distributed manufacturing may fundamentally alter 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*).

Medium- and heavy-duty trucks with mean trip distances of >160 km (>100 miles) accounted for ~270 Mt CO_2 emissions in 2014, 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 cars and trucks accounted for an additional 40 Mt CO_2, and aviation and other shipping modes (e.g., trains and ships) emitted 830 and 1,060 Mt CO_2, respectively (altogether, ~6% of global CO_2 emissions; dark orange in Fig. 2). Meanwhile, both global energy demand for transportation and the ratio of heavy- to light-duty vehicles is expected to increase (*13*).

Hydrogen and ammonia

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 (e.g., 350-700 bar), involving heavy and bulky storage containers (*14*). To contain the same total energy, a liquid hydrogen storage system would weigh roughly six times more than a diesel fuel storage system and be about eight times larger (Fig. 3A). But hydrogen fuel cell or hybrid hydrogen-battery trucks can be more energy efficient than 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 battery-powered class 8 truck with a fuel cell system range extender claiming a 800-1200 mile range, similar to today's long haul diesel trucks (*16*). If affordably produced without CO_2 emissions, such uses of hydrogen in the transport sector could ultimately be bolstered by the fuel's importance in providing other energy services (see below).

Ammonia is another technologically viable alternative fuel that lacks carbon and may be directly combusted in an engine (Fig. 1D) or cracked to produce hydrogen. Its combustion must be carefully controlled to minimize production of highly oxidized products such as NO_x (*17*), and—like hydrogen—its gravimetric energy density is considerably lower than hydrocarbons such as diesel (Fig. 3A).

Biofuels

Conversion of biomass currently provides the most cost-effective pathway to non-fossil, carbon-containing liquid fuels (Fig. 1I). Liquid biofuels at present represent about 4.2 EJ of the roughly 100 EJ of energy consumed by the transport sector worldwide. Currently available liquid biofuels, primarily ethanol from grain and sugar cane and biodiesel and renewable diesel from oil seeds and waste oils, face substantial challenges related to their effective carbon footprint, cost, and scalability (*18*).

Photosynthesis converts <5% of incident radiation to chemical energy, and only a fraction of that remains in biomass (*19*). Conversion of biomass to fuel also requires energy for processing and transportation. Land producing biofuels must have water, nutrients, soil, and climate characteristics suitable for agriculture, putting biofuels in competition with other land uses with implications for food security, sustainable rural economies, and 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 (i.e. carbon dioxide removal), which biofuels can provide (*21*).

Advanced biofuel processes seek to overcome the recalcitrance of cellulose to achieve large-scale production of liquid transportation fuels at costs roughly competitive with gasoline (e.g., $19/GJ or $1.51/gallon of ethanol) (*22*). As technology matures and system-wide decarbonization efforts proceed, biofuels may be able to largely avoid fossil fuel inputs and emission associated with induced land use change (*23, 24*). The extent to which biomass will supply such liquid fuels in a future net-zero emissions energy system thus depends upon 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 (e.g., reaction of carbon monoxide and hydrogen by the Fischer-Tropsch process; Fig. 1F and Table 1) (*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" (synthetic gas/liquids, Fig. 1F). For example, emissions-free electricity (e.g., Fig. 1O, 1S, 1K, 1N, etc.) could be used to produce H_2 by electrolysis of water (Fig. 1L), which would be reacted with carbon removed from the atmosphere either by direct air capture (Fig. 1J) or photosynthesis (perhaps captured after combustion biomass or biogas; Fig. 1M/1I) (*27, 28*).

At present, the cost of electrolysis is a major barrier, including both the capital costs of electrolyzers and emissions-free electricity (60-70% of current electrolytic hydrogen cost is electricity) (*28, 29*). The cheapest and most mature electrolysis technology available today uses alkaline electrolytes (e.g., KOH or NaOH) with metal catalysts to produce hydrogen, at an efficiency of 50-60%, and at a cost of ~$5.50/kg H_2 (assuming industrial electricity costs of $0.07/kWh and 75% utilization rates) (*29, 30*). At this cost of hydrogen, the minimum price of synthesized hydrocarbons would be \$5.50-\$6.50 per gallon of diesel equivalent (or \$42-50 per GJ; assuming carbon feedstock costs of \$0-100 per ton of CO_2 and very low process costs of \$0.20 per gallon 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 \$1.30-1.50 per kg (red line in Fig. 3B) (*29, 31*). Equalizing current costs of H_2 produced by electrolysis and steam methane reformation would thus entail valuing of a cross-sector benefit, such as renewable electricity balancing, or a policy-imposed price of ~\$400 per ton of CO_2 emitted (which would also raise fossil diesel prices by ~\$4.00 per gallon). Natural gas can also be converted directly to liquid hydrocarbons at costs of \$0.99-\$1.21 per gallon of diesel equivalent (or ~\$8-\$9 per GJ; assuming a natural gas price of \$3.50-4.50 per thousand cubic feet) (*32*).

Absent policies or cross-sector coordination, hydrogen costs of \$2.00/kg (i.e., approaching the cost of fossil-derived hydrogen and synthesized diesel of ~\$3.00 per gallon) could be achieved, for example, if electricity costs were \$0.03/kWh and current electrolyzer costs were reduced by 60-80% (*29*) (Fig. 3B). Such reductions may be possible (*33*), and 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*). Efficiencies for production of hydrogen 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 (unstable) bacteria as catalysts.

Structural materials

Economic development and industrialization are historically linked to construction of infrastructure. Decarbonizing the provision of cement and steel will require either major changes in manufacturing processes, use of alternative materials whose manufacture does not produce CO_2, or carbon capture and storage (CCS) technologies to minimize the release of process-related CO_2 to the atmosphere (*39*) (Fig. 1B).

Between 2000-2015, cement and steel use consistently averaged 50 and 21 tons per million dollars of global GDP, respectively (1σ=2.6 and 1.6, respectively; ~1 kg per person per day in developed countries) (*4*). Figure 2 shows that ~1,320 and 1,740 Mt CO_2 emissions (4% and 5%) emanated from chemical reactions involved with manufacture of cement and steel, respectively (*12, 40, 41*) (altogether, ~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 0.8-1.2% to 3.7-4.4 billion tons in 2050 (*45, 46*), reflecting historical patterns of infrastructure accumulation and materials use in regions such as China, India and Africa (*4*).

Steel

Carbon (coke from coking coal) is used to reduce iron oxide ore during steel-making in blast furnaces, producing 1.6-3.1 tons of "process" CO_2 per ton of crude steel produced (*41*)—in addition to CO_2 emissions from fossil fuels burned to generate the necessary high-temperatures (1100-1500 °C). Reductions in CO_2 emissions per ton crude steel are possible through: electric arc furnace (EAF) "minimills" that operate using emissions-free electricity; efficiency improvements (e.g., top gas recovery); new process methods (e.g., ULCORED); process heat fuel-switching; and decreased demand via better engineering (e.g., a global switch to ultrahigh-strength steel for vehicles would avoid ~160 Mt CO_2 annually). The availability of scrap steel 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 biomass-derived charcoal 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*). ~0.6 tons of charcoal are required per ton of steel produced, requiring 0.1-0.3 hectares of Brazilian eucalyptus plantation (*49, 50*). Hundreds of millions of hectares of highly productive land would therefore be necessary to meet expected demands of the steel industry using charcoal, and associated land use change emissions could outweigh fossil emissions avoided, as has happened in Brazil (*49*). Hydrogen might also be used as a reductant, but carbon imparts strength and other desirable properties to steel, so the iron oxides would need to remain in hot plasma without negatively altering the characteristics of produced iron (*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 (40-50% and 35% by volume, respectively; Figs. 1G and 1E) (*52, 53*).

Cement

Approximately 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 cementitious materials, and/or installation of carbon-capture technology (*45*) (Fig. 1G). CO_2 concentrations are typically ~30% by volume in cement plant flue gas (cf. ~10-15% 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 is complicated because existing cement kilns operate at very high-temperatures (~1500 °C), are not gas-tight, and rotate (*57*). The cement and steel industries are also understandably risk-averse to alternative processes that could compromise the mechanical properties of produced materials.

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

Highly reliable electricity

Modern economies demand highly reliable electricity (e.g., demand met >99.9% of the time; Fig. 1A), requiring investment in 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 due to their relatively low fixed costs (Fig. 3B), operational ability to ramp quickly to follow large swings in electricity supply and demand (*59*), and the affordability of natural gas (*60*). We estimate that CO_2 emissions from such "load-following" electricity were ~ 3,978 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 (red in Fig. 2) (*61*).

The central challenge of a highly reliable electricity system is thus to replicate 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 (Figs. 1O and 1S) are major sources of electricity will have occasional but substantial (tens of petajoules; 40 PJ=10.8 TWh=24 hours of mean U.S. electricity demand in 2015) and long-term (e.g., days- or weeks-long) mismatches between supply and demand (*62*). Thus, even with continental-scale or global electricity interconnections (*62-64*), highly reliable electricity in such a system will require very substantial amounts of dispatchable electricity sources (e.g., generators or stored energy) or demand modifiers that operate less than 20% of the time. Similar logic applies if most electricity were produced by nuclear generators or coal-fired power plants equipped with carbon capture and storage (Figs. 1N and 1M), 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; Figs. 1M and 1E) 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 (more below). However, capital costs of CCS-equipped generators are currently considerably higher than generators without CCS (Fig. 3B). Moreover, CCS technologies designed for high capacity factor generators (e.g., 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 (e.g., by electrolysis or thermochemistry; Fig. 1L) for synthesis of fuels (Fig. 1F) could thus beneficially shift mismatches between either variable energy sources or dispatchable but inflexible generators and time-varying electricity demand out of the electricity sector.

Nuclear fission (Fig. 1N) can operate flexibly to follow loads by making adjustments 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 cf. *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 (e.g., Fig. 1L).

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 with fewer annual cycles 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 (Fig. 1L) can be converted back to electricity either in fuel cells or by combustion in gas turbines (i.e. power-to-gas-to-power or "P2G2P"; Figs. 1F, 1P, red curve in 3D), with commercial-scale P2G2P systems currently exhibiting a round-trip efficiency 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 40-50% in proton-exchange membrane designs) and chemical substitutes for more costly 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*), recognizing important trade-offs of cost and safety during such a transition.

Mature closed-cycle battery chemistries 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 (e.g., frequency regulation, 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 (Figs. 2Q and 3D). For example, assuming a grid-scale use case, the estimated cost of energy stored (i.e. electricity discharged) using current lithium-ion batteries (solid green curve in Fig. 3D) 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., ~36/MJ or \$130/kWh of capacity) (*73, 79, 80*), the levelized cost of stored energy would fall to ~\$0.29/kWh (\$81/GJ) for weekly cycling (dashed green curve in Fig. 3D). Cost estimates for current vanadium redox flow batteries (orange curve in Fig. 3D) 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 \$3-8/MJ (\$10-30/kWh), leading to a levelized cost of stored energy for the grid-scale use case in the range of \$2-40 per GJ (\$0.01-0.15/kWh, assuming 10-100 cycles per year) (*81*).

Utilization might be increased if tens of millions of electric vehicle batteries (1 PJ = 10% of 28 million 100 kWh charges) were used to support the electrical grid (vehicle-to-grid, V2G), presuming that the disruption to vehicle owners (i.e. diminished battery charge) would be less costly than an outage would be to electricity consumers (*82*). It is 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 (e.g., 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%) (Fig. 1K). Although capital costs of such pumped storage are substantial, long lifetimes of reservoirs result in competitive levelized costs of stored energy when cycled at least weekly (blue curve in Fig. 3D). Major barriers are the availability of water and suitable reservoirs, compounded by social and environmental opposition, as well as flexibility constraints of 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*).

Compressed air systems in underground geologic formations, underwater, or above-ground pressure vessels use electricity to compress the air recovered by turbines when air is subsequently released to the atmosphere (Figs. 1R and purple curve in 3D). 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 (Fig. 1Q) 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 Wh_{th}/kg); they remain expensive (*84, 87, 88*), but in some cases the ultimate cost target is <\$15/kWh_{th} (*89*). Thermal storage is well-suited to within-day shifting of heating and cooling loads, whereas 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 (i.e. load-shifting or -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*).

Carbon management

Integrated assessment models 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*). Carbon capture and storage will be separable services in a net-zero emissions energy system (e.g., Fig. 1J and 1E). Carbon captured from the ambient air (Fig. 1J) could be used to synthesize carbon-neutral hydrocarbon fuels (Fig. 1F) or sequestered to produce negative emissions (Fig. 1E). Carbon captured from combustion of biomass or synthesized hydrocarbons (Fig. 1M) could be recycled to produce more fuels (Fig. 1F) (*98*). Storage (e.g., geological; Fig. 1E) will be required to the extent that fossil fuels supply carbon and/or that negative emissions are needed (*20*).

For industrial 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/yr now being injected underground (*99*)), financing carbon storage projects with high perceived risks and long-term liability discharge remains a major challenge (*102*).

Discussion and Conclusions

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). 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, global final energy demand in some of the recently developed Shared Socioeconomic Pathways more than doubles by 2100 (*1*), 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, 104, 105*). More rapid changes may depend on coordinating operations across energy and industry sectors, which could help boost utilization rates of capital-intensive assets (i.e. systematizing and explicitly valuing the interconnections depicted in Fig. 1), but will require 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.

Factors other than technological and economic challenges will undoubtedly influence the technological makeup of future energy systems and how quickly those systems would be deployed. For example, 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 "leapfrog" fossil-fuel energy while prioritizing human development, environmental protection, sustainability, and social equity (*4, 106, 107*).

CO_2 emissions from land use and land-use change, as well as emissions of CH_4, N_2O, and other radiatively active gases and aerosols (e.g., methane and black carbon; *108*) could also cause substantial warming, and may in part be determined by different energy pathways (e.g., *109*). As a prominent example, bioenergy may avoid fossil-fuel emissions but spur emissions from land-use change (*110*).

Many of these 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 (*111*). The collective and reinforcing inertia of existing technologies, policies, institutions, and behavioral norms ("carbon lock-in") may actively inhibit innovation of emissions-free technologies (*112*).

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 (1) vast amounts of inexpensive, emissions-free electricity, (2) mechanisms to quickly and cheaply balance large and uncertain time-varying differences between demand and electricity generation, (3) electrified substitutes for most fuel-using devices,(4) alternative materials and manufacturing processes for structural materials, and (5) 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.

[*4994 words excluding abstract*]

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Figures

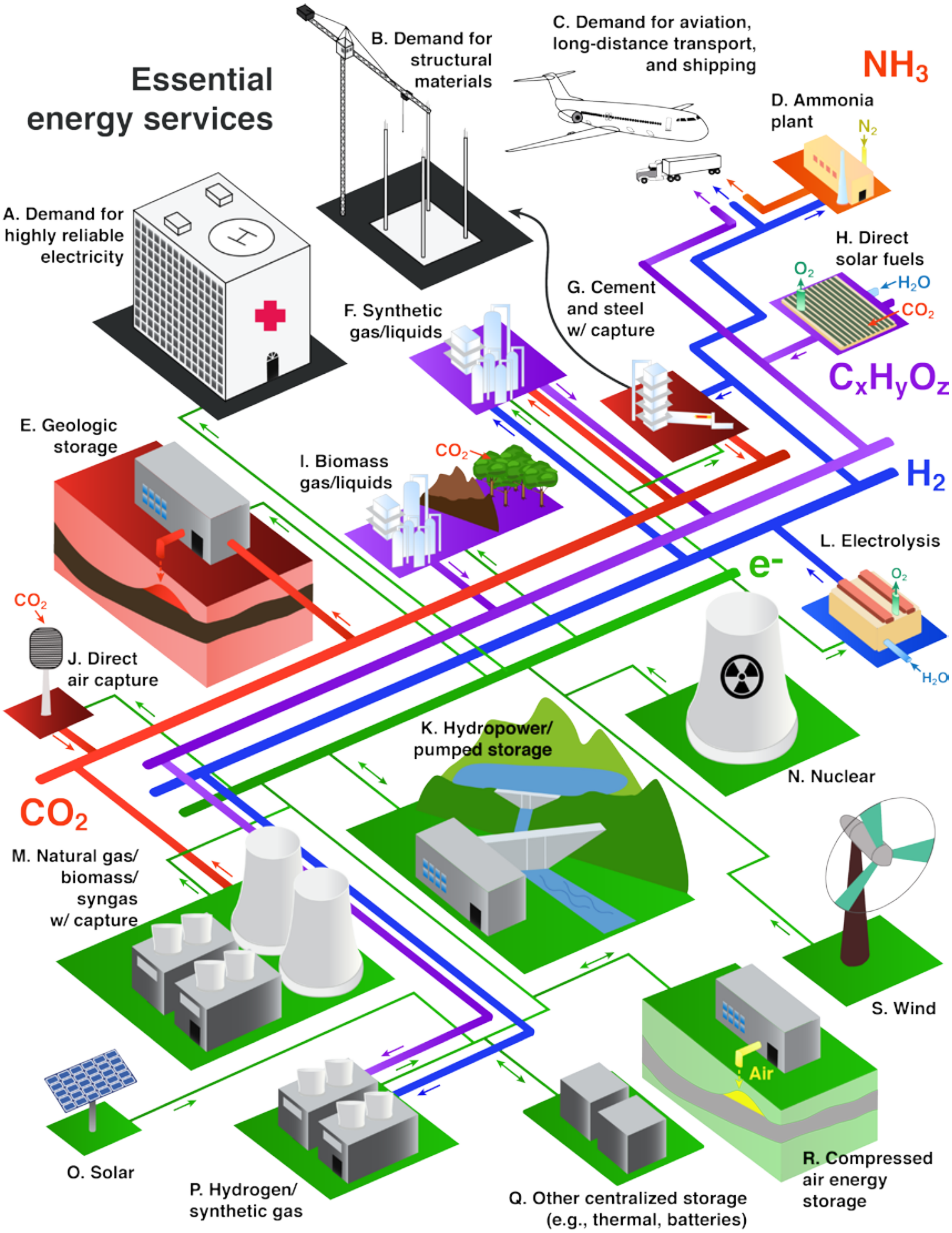

Fig. 1. Schematic of an integrated system that can provide essential energy services without adding any CO_2 to the atmosphere. Colors indicate the dominant role of specific technologies and processes: electricity generation and transmission in green, hydrogen production and transport in blue, hydrocarbon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

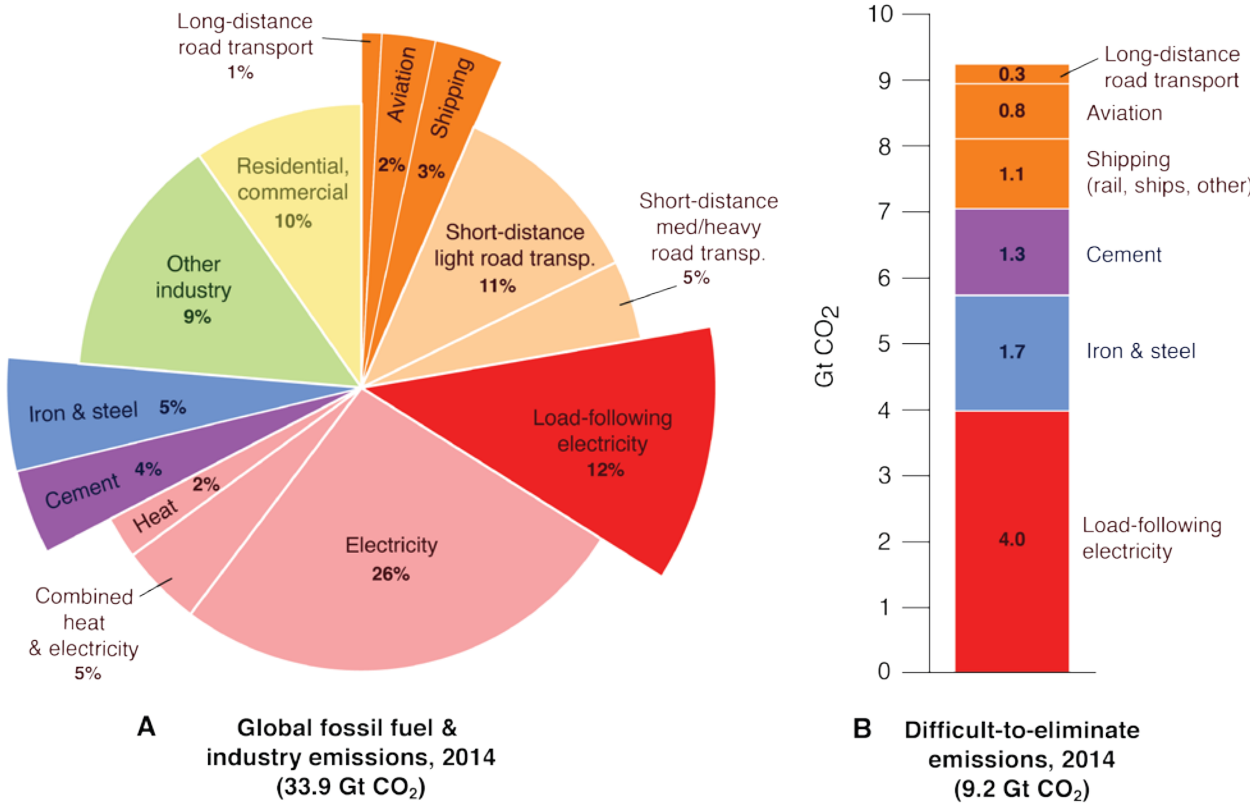

Fig. 2. Difficult-to-eliminate emissions in current context. Estimates of CO_2 emissions related to different energy services, highlighting (e.g., by longer pie pieces in **A**) those services that will be the most difficult to decarbonize, and the magnitude of 2014 emissions from those difficult-to-eliminate emissions. Note that the shares and emissions shown here reflect a global energy system still relying primarily on fossil fuels and with many still-developing regions. Both the shares (**A**) and the level of emissions (**B**) related to these difficult 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. “Iron and steel,” “Cement,” emissions highlighted 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.

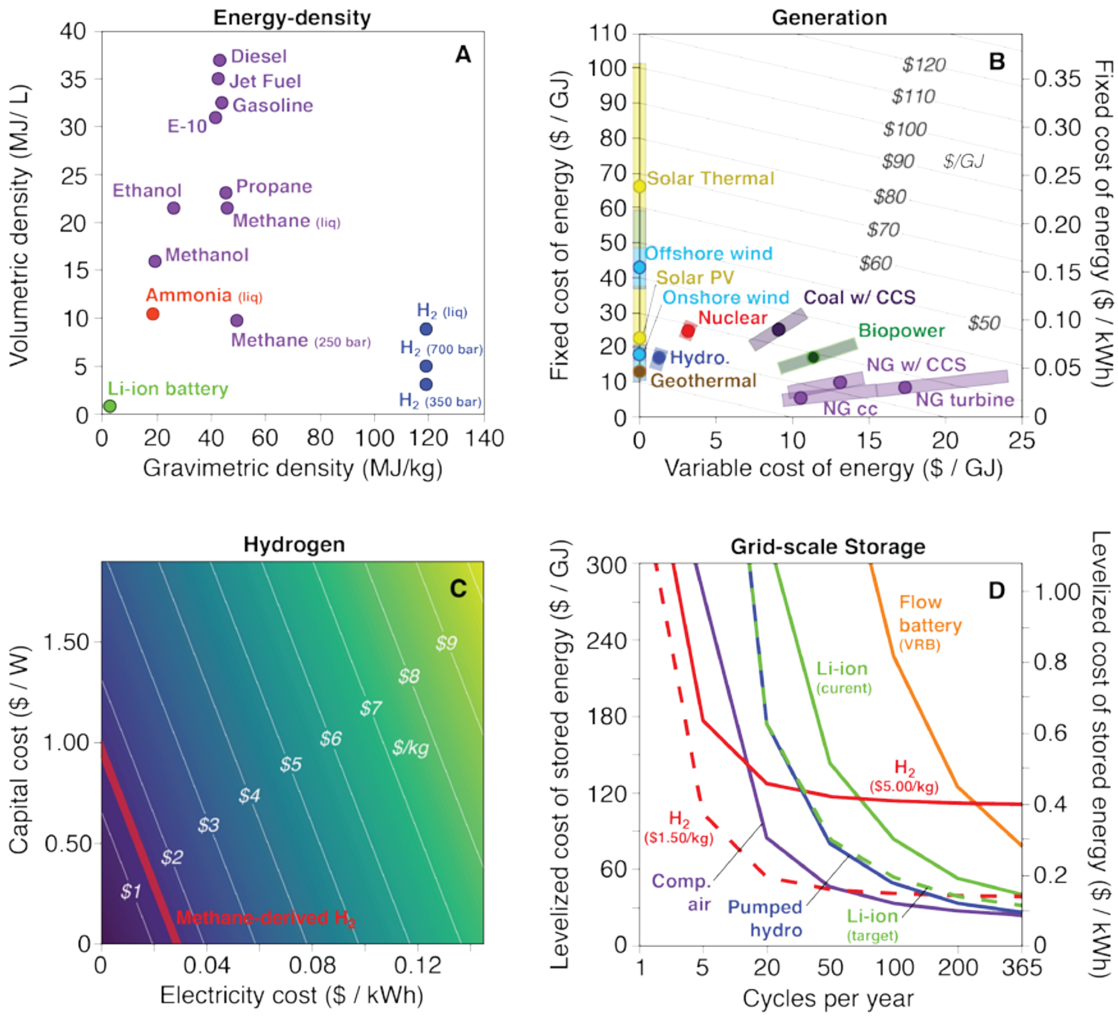

Fig. 3. Comparisons of energy sources and technologies. Panel **A** shows the energy density of energy sources for transportation, including hydrocarbons (purple), ammonia (orange), hydrogen (blue), and current lithium ion batteries (green). Panel **B** shows relationships between fixed capital versus 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 stored energy (i.e. 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*.

	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(*114, 115*)		• Methanation ($0.07-0.57/m^3 CH_4) • Fischer-Tropsch ($4.40 to $15.00 per gallon of gasoline-equivalent)	• Haber-Bosch ($0.50-0.60/kg NH_3)(*115*)
$C_xO_yH_z$	• 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 (*116*) (~$3/kg H_2) • Sodium amide (*117*)	• Metal catalysts + methanation/Fischer-Tropsch • Sodium amide + methanation/Fischer-Tropsch	