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The role of combustion science and technology in low and zero impact energy transformation processes

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ABSTRACT

Predictions made by climate researchers are highly worrisome and demand rapid action to avoid the threat of climate catastrophe. Global energy systems must be transformed as quickly as possible by minimising or avoiding net greenhouse gas emissions. There is broad agreement on this goal, demonstrated by international treaties such as the Sustainable Development Goals of the United Nations and the European Green Deal presented in 2019.

However, the practical measures required for the transition are the subject of heated discussion. Consensus on the goal, dissent on the pathway is how the situation can be summarized.

This opinion article aims to bring engineering sciences into the centre of the discussion. We are concerned that technological options that are important for our society from an ecological and economic point of view are being neglected. We plead for competition between all technological solutions to reach the goals in the best possible way and to consider feasibility, ease of transition, and economical and societal aspects.

We are convinced that the thermochemical utilisation of chemical energy carriers is an important component of future energy systems and is key to enabling climate neutrality. Biogenic and synthetic carbonaceous and carbon-free chemical energy carriers will be indispensable for reliable power generation and energy supply for mobility, industry, and buildings.

This opinion article is the result of intensive discussions between a group of more than fifty internationally renowned researchers who are scientifically engaged in thermofluids and energy process engineering. With this article we express our plea: Let us consider all options and explore new ideas that will move us towards a climate-neutral energy system!

Introduction

A key challenge of the 21st century is transforming the energy industry into a climate-neutral circular economy [1]. To meet this global challenge, energy must come from renewable energy sources and emission of greenhouse gases must be avoided. Governments mostly agree about this goal but not about how to achieve it. To meet climate targets, a technology-agnostic approach is indispensable, since the energy mix of the future will be more diverse and will include technologies that have not yet been well researched. Gradual, manageable transformation of energy systems towards climate neutrality will be made possible by including chemical energy carriers and processes for thermochemical and electrochemical energy conversion. These topics will be discussed using Germany as a case study as most German examples are similar to other parts of Europe and the world despite differences in energy mix, resources, and policy.

Realizing a climate-neutral circular economy requires electrification of the mobility, industry, and building sectors[1], as well as energy carriers from non-fossil fuel sources for energy storage and transport, which will largely depend on renewable electricity with limited energy contribution from biomass sources. Consequently, the worldwide demand for electrical energy will increase sharply in the coming years. However,

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[1] In 2018, total CO_2 emissions in Germany were 866 million tons. This came mainly from the electricity (35.9 %), industry (22.6 %), mobility (18.7 %) and building (13.5 %) sectors (total emissions excluding land use, land use change and forestry) [2].

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meeting this demand is not the only challenge faced by the energy sector.

In Germany, for example, around 65% of power plants will have to shut down in the next few years due to the phase-out of using nuclear energy and coal. These base-load capable power plants (which made a significant contribution to the electricity supply, with net electricity generation of 515 terawatt hours in 2019 [3]) now need to be replaced with renewable energy sources.

The most important sources of renewable energy[2] are wind and solar-powered photovoltaic plants. However, in Germany, their contribution is limited by geography and weather. Wind and sun-rich areas close to Germany that combine high potential for expansion with economic efficiency are mainly located in the coastal regions, southern Europe, and North Africa. To jointly benefit, reliable international partnerships must be established and technical challenges overcome. One of the challenges is the distance between production and consumption of the renewable power, which must be transported over long distances and stored with as little loss as possible. Time lag between availability and demand is also a challenge: Due to weather conditions, electricity generated using wind power and photovoltaic plants has massive production fluctuations [4].

Energy storage

Depending on requirements, energy can be stored electrically, electrochemically, mechanically, thermally, or chemically. These methods differ in their storage capacity and discharge duration. As Fig. 1 shows, chemical energy carriers combine a high storage capacity with the longest discharge durations[3].

Chemical energy carriers

Chemical energy carriers are distinguished by their origin. Significant fossil fuel chemical energy carriers are natural gas, crude oil, and coal, which were formed from organic material in geological prehistoric times. Synthetic chemical energy carriers are produced using renewable energy. To produce so-called e-fuels, electrical energy can be converted into hydrogen by electrolysis, then used and stored directly, or converted into liquid or gaseous synthetic fuels (by catalysis), which is more easily stored and higher in volumetric energy density. Synthetic chemical energy carriers include biofuels, such as ethanol.

Synthetic chemical energy carriers are carbonaceous or carbon-free. Carbonaceous energy carriers include synthetic natural gas and synthetic fuels such as e-fuels or biofuels. When consuming these energy carriers, the greenhouse gas carbon dioxide (CO_2) is released. However, if the carbon in the fuel originates from biomass, CO_2 that was removed from the atmosphere[4], or power and industry plant exhaust, there are zero carbon dioxide net emissions.

Using carbon-free chemical energy carriers avoids any CO_2 release. Examples include molecular hydrogen (H_2), ammonia (NH_3) [7], metals [8], and some metalloids and non-metals [9]. The reaction products escape into the air when H_2 and NH_3 are used, but they are usually harmless substances, such as water or molecular nitrogen. With metals such as iron, metalloids such as silicon, and non-metals such as sulphur, the reaction product has to be collected and returned to the production cycle. As discussed below, these different chemical energy carriers most probably will become available and usable at different times.

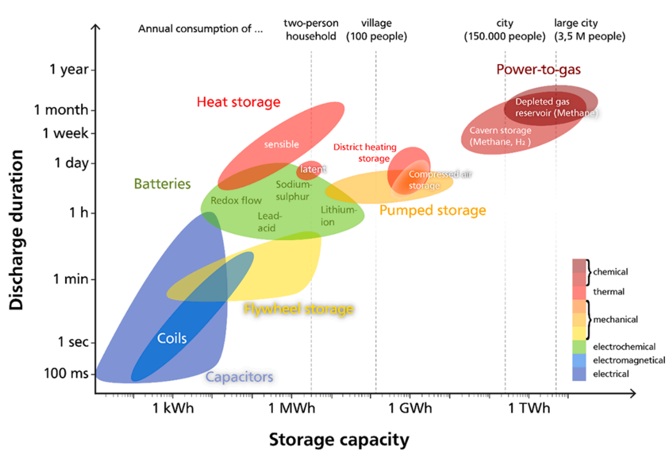

Fig. 1. Overview of storage capacity and discharge duration of storage technologies. Reproduced from [5].

Thermochemical energy conversion

The energy stored in chemical energy carriers from renewable sources can be used by employing electrochemical and thermochemical[5] processes. Electrochemical processes, such as fuel cells and redox flow batteries, are slowly becoming established on the market despite enormous development efforts, whereas thermochemical processes are widespread and particularly reliable. Thermochemical energy conversion of carbonaceous chemical energy carriers from renewable sources is of great importance, particularly during a transitional phase, which can last for decades. There is no time limit for the thermochemical energy conversion of carbon-free chemical energy sources. In many sectors, this approach will be important to energy supply in the long-term.

The phases

Development of a climate-neutral energy system needs time and resources. Worldwide, infrastructure must be rebuilt, new plants built, and innovative technologies conceived and developed.

To keep technological and economic risks manageable, we argue for a transformation that is as continuous as possible. In our opinion it makes sense to proceed rapidly, steadily, and with plannable steps that we classify in three higher-ranking phases:

- Short-term: Focus on drop-in technologies that use existing infrastructure and enable smooth transition. For example, natural gas can increasingly be replaced by synthetic natural gas. In this way, time is gained to prepare for parallel technology changes.
- Medium-term: Focus on technologies in which infrastructure is supplemented or newly built. By this time, there will be power plants that can be operated with 100 % carbon-neutral energy carriers, such as green hydrogen[6] as a substitute for natural gas (whereas only

[2] The term Renewable Energy is commonly used. It is therefore also used here, although it is physically not correct. It refers to technically usable forms of energy that are obtained from the conversion of renewable sources, such as wind and solar energy.

[3] The discharge duration indicates how long a storage unit can supply energy. It is calculated from the ratio of energy that can be stored and the withdrawal power.

[4] The cost for recovering CO_2 from the atmosphere is estimated to be between 92 and 232 USD per ton of CO_2 [6]. Assuming a liquid density of 737 kg/m^3 and a carbon mass fraction of 84.47% for RON95 E5 (EN228) [40], one liter of fuel will form 2.28 kg CO_2. Recovery from the atmosphere is hence equivalent to approximately 0.21 to 0.53 USD per litre of petrol.

[5] The technical term thermochemistry covers all high-temperature oxidation processes, regardless of whether the chemical energy sources are of fossil or synthetic origin.

[6] Green hydrogen is hydrogen produced by electrolysis with electricity from renewable energies or biomass.

admixtures of up to a maximum of approximately 20 % are possible in the short-term). Other technologies will be under development.
- Long-term: Focus on future technologies whose potential is apparent now but which needs to be explored in greater depth.

There is a great need for research and development (R&D) in all three phases. Research and development of drop-in technologies is naturally more applied, while future technologies require more fundamental investigations. Research on future technologies should pursue many different angles so as to identify the best solutions over time. Among the variety of technological options, thermochemical energy conversion based on chemical energy carriers can make a significant contribution at all three transformation stages of the energy system.

The costs

Currently, the use of electricity, fuels, and other types of energy is relatively inexpensive: In many countries, it costs little to nothing to release climate-damaging gases into the earth's atmosphere. A global CO_2-neutral and largely CO_2-free economy and lifestyle entails significantly higher costs, as long as the damage caused by CO_2 does not have a quantifiable economic value. These costs arise from the recovery and storage of the greenhouse gas from the atmosphere or its recovery from waste gas streams. Renewable energy is also not available for free: For example, there are considerable infrastructure costs for photovoltaics and wind power plants, as well as transport and conversion into electricity or e-fuels. The real cost of energy conversion processes must be charged within the framework of international agreements. The current CO_2 tax in Germany is EUR 25 per ton of CO_2, rising to EUR 55 per ton in 2025, which slowly becomes comparable to the price of CO_2 direct air capture of between USD 92 and 232 [6].

In our opinion, a reliable estimation of the costs, risks, and opportunities of the transformation process is hardly possible, which is in agreement with Ref. [10]. The appropriate estimation tool would be a mathematical-thermodynamic-economic model that describes the entire lifecycle of technologies in individual sectors as well as their multitudes of coupling in various scenarios. However, important data and boundary conditions required to create truly predictive models are still lacking.

When models are subject to considerable uncertainties, different technology paths should be assessed in parallel, and different energy carriers and energy conversion technologies should be considered. This approach is described as technology-agnostic. The decision for or against a particular technology path needs first its availability at a high technology readiness level resulting from systematic research and development. The use of a technology is then decided in the interplay between socioeconomic and political considerations. The long-term forecast which technologies are best suited and selected by different countries is therefore a challenge. What is already clear now, however, are the criteria that technologies must meet: Environmental compatibility throughout the entire lifecycle, security of supply, economic efficiency (CO_2 saving per Euro), and societal acceptance.

Thermochemical energy conversion technologies have the advantage that their reliability is proven and that distribution networks, power plants, heat generators, propulsion technologies, and other infrastructure are available. In the drop-in phase, which can be realized in the short-term, additional costs will only be incurred during the synthesis of chemical energy carriers. These costs arise mainly from the increase in renewable energy plants, such as wind power and photovoltaic plants, and their conversion into chemical energy carriers. Costs for the development of new energy conversion infrastructure specially adapted to future energy carriers will be added in the medium and long-term.

Energy sectors

A. Electrical power

In Germany, nearly all recent science-based studies conclude that CO_2-free technologies must be based primarily on renewable power generation. There is political consensus in Germany to end mining and use of coal as a fossil fuel by 2038. This is in agreement with the goals of the European Green Deal [11] and the Sustainable Development Goals of the United Nations [12].

The generation of electricity from hydropower and biogenic fuels is already contributing significantly to avoiding CO_2 emissions. However, both of these resources have limited availability as opposed to wind and solar energy, which will therefore play an even more important role in the future [13].

In Germany with a total installed peak power generation capacity of 225 GW, domestic wind power and photovoltaic plants contribute with an installed capacity of around 118 GW (in 2018). With an increased share of wind and solar and an increased transition to use electricity in mobility, industry, and domestic heating, it is foreseeable that the total installed power generation capacity must increase significantly. To compensate for the fluctuations in wind and solar described in the next paragraph, additional power capacity is required. With moderate restrictions in the reduction of the greenhouse-gas release, this substitution can be covered by, e.g., stand-by gas-powered plants. In case greenhouse gas emissions are to be reduced by ~90%, substitution with gas-powered plants is not permittable. Using substitution with wind and solar alone, the additional requirement installed power is predicted in the range of 250–600 GW [14]. The large variation in these studies result from differences in assumptions about energy imports, the intensity of sector coupling (e.g., spread of heat pumps and e-mobility), increases in energy efficiency, and technology developments (e.g., full-load hours of wind turbines). CO_2-neutral alternatives that prevent the installation of the mentioned overcapacities in wind and solar would be highly desirable.

Fluctuating renewable electricity

A central challenge with using renewable energy sources comes from the large fluctuations in generated power. Figs. 2 and 3 illustrate this for an annual cycle and a monthly cycle, respectively. Fig. 2 shows the maximum and minimum power output from the fluctuating wind and sun as hourly averages in 2019 for Germany. The values fluctuate between almost zero and less than half of the installed capacity of renewable energies. The fluctuations of the hourly mean power output are particularly pronounced in the winter months. At this time, nearly no solar energy is available, so it cannot compensate for fluctuations in wind energy.

The primary energy sources

Fig. 3 shows the contribution of primary energy sources to electricity demand in Germany. The upper limit of approximately 80 GW corresponds to the maximum power demand, which is currently met by hydro power, biogenic sources, nuclear energy, coal, gas, wind, and solar. The figure shows a strong dependence on the time of day and pronounced fluctuation due to weather conditions [16].

The fluctuations in renewable energy sources need to be compensated. Short-term fluctuations (in the range of minutes to half hours) are in the order of 10 to 50 GWh and can be compensated by pumped storage, compressed air storage, and other technologies, some of which are already available (Fig. 1). Demand control and other innovations will contribute to even more comprehensive solutions in the future. Batteries and thermal storage, such as in Carnot batteries, will probably be able to compensate for fluctuations over a few hours, up to an energy quantity of several hundred GWh. However, only chemical energy

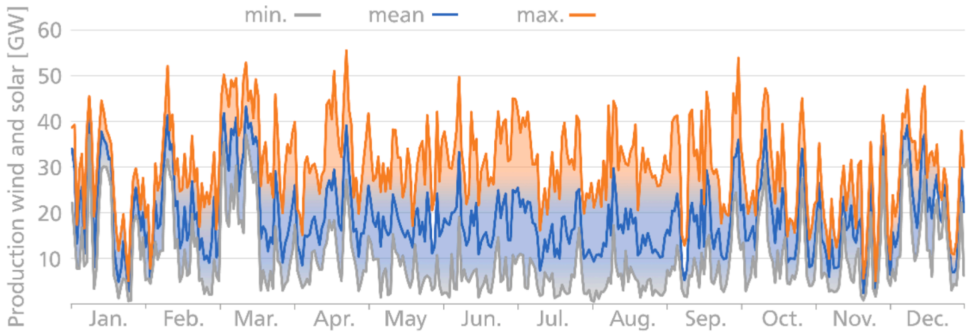

Fig. 2. Hourly averaged electricity generation from wind power and photovoltaics in Germany in 2019. Highlighted are mean, maximum, and minimum power output. Data from [15]. Fluctuations are particularly pronounced in the winter months.

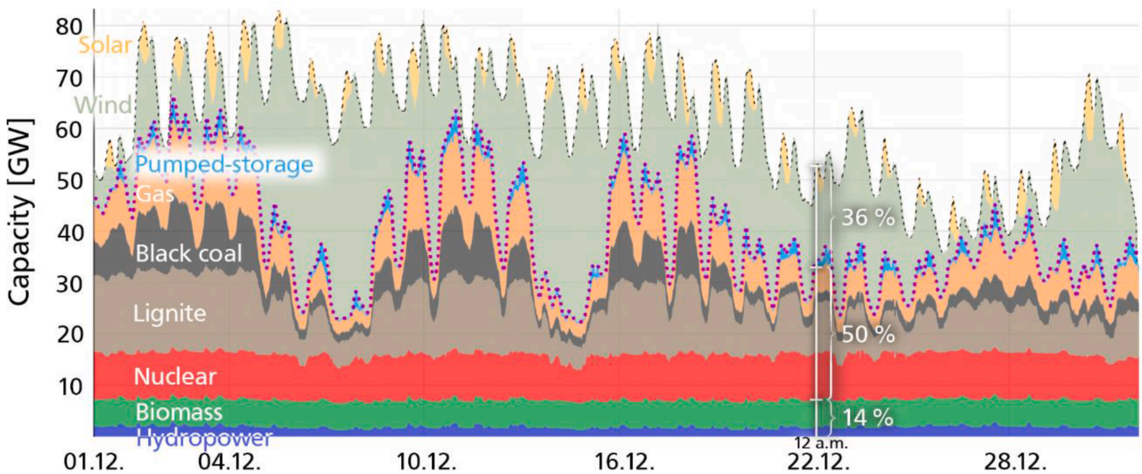

Fig. 3. Power generation from different sources and temporally fluctuating maximum power demand (upper, grey dotted line) in December 2019 in Germany; reproduced from [16]. The contribution from oil is negligible and hence not shown. The lower dotted line (magenta) marks the contributions from hydro power, biomass, nuclear energy, hard coal, lignite, gas, and pumped storage. The difference to the total output, which fluctuates over time, is the share of wind and solar energy. The example for 22 December 2019 at midday shows that wind and solar energy contributed around 36 % of the demand.

carriers can be used to compensate for fluctuations caused by periods of low wind and sunshine for days, the 'cold dark doldrums' [10], and supply electricity for gaps of several thousand GWh. The chemical energy carriers of the future must be generated from renewable energy sources, whose conversion back to electricity requires controllable power plants.

The locations for producing chemical energy carriers from solar and wind energy must be selected in a way that guarantees economic efficiency and security of supply. In Germany, domestic sites for wind power and photovoltaics have low public acceptance and, because of unfavourable weather conditions, wind-power plants have low capacity factors, which reduces their economic efficiency.

International cooperation

The lack of sufficient renewable energy sources in many countries makes international cooperation with countries in Southern Europe and North Africa a sensible option. The German Federal Government's National Hydrogen Strategy [17] points to this. The vision is that, in southern regions, electricity from renewable sources could be stored in suitable chemical energy carriers (power-to-X) [18] and imported to metropolitan areas to be used in classic thermal power plants. Gas and steam combined-cycle power plants are particularly suitable for this. How a high degree of efficiency can be achieved over the entire process chain[7] is the subject of current discussions.

The scenario outlined here could reduce pressure on domestic electricity production from renewable sources. In Germany, this approach would contribute to the base load requirement and control capacities of up to 65 GW that will be lost due to the phase-out of nuclear and coal power. It will also help to meet the predicted high demand for renewable energy [10]. In forecasts, the demand for purely domestic power generation would be up to 600 GW of installed capacity, which is five times the current levels. Such a diversified energy system can only be realised if all technical, economic, social, and political influencing factors are considered.

Power plants

To compensate for long phases without wind and sunshine that occur in combination with the low temperatures that occur in Germany for

[7] When green hydrogen is used, the efficiency of the entire process chain, including transport, is approximately 28 %. In accordance with [19], 95 % efficiency was assumed for short- and long-distance transport, 70 % for electrolysis, 70 % for power-to-liquid, and 63 % for the combined-cycle process [20].

several hundred hours almost every year[8], a weather-independent power plant capacity of at least 80 GW is necessary. According to different scenarios from three studies [10,19,21], Fig. 4 shows a demand for controllable power plant capacity between 80 and 140 GW, which is in the order of the current power plant capacity of about 100 GW. These power plants could potentially be operated with chemical energy carriers produced from renewable sources.

In Germany, only gas turbine power plants, gas and steam combined-cycle power plants with high efficiencies of up to 63 % [20], and gas engines are able to meet this challenge after nuclear and coal power ceases. The main reasons for this are:

- These plants are load flexible and can react within minutes to fluctuations in power generation from renewable sources.
- Even a short-term sharp increase in demand can be reliably covered using natural gas. In an initial phase, fossil natural gas could mainly be used, as it enables electricity generation with CO_2 emissions that are around 60 % [22] below those of lignite-fired power plants. However, the use of natural gas requires a significant reduction in methane slip[9] because of the high greenhouse gas potential of methane.
- Fossil natural gas can be replaced by synthetic natural gas from renewable sources in a continuous transformation process. In view of the existing, efficient gas infrastructure, this is a considerable strategic advantage.
- Fossil natural gas can be enriched in the short-term with green hydrogen from renewable sources. This is important for the continuous transformation process.
- After further development of existing gas turbine technology [24], fossil natural gas can be completely replaced by green hydrogen in the long-term.

Perspective

The phases of the transformation process described above can be realized reliably and with manageable risk using gas turbine technology. Continuous change of technology can be achieved even with proportions

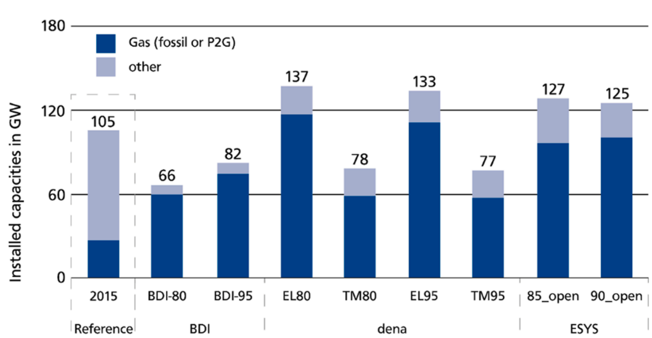

Fig. 4. Scenarios for the demand for controllable power plant capacities in Germany in 2050, reproduced from [14], where the scenarios are explained in detail.

of about 20 % by volume of hydrogen in natural gas [24, 25]. With the higher hydrogen content in synthetic natural gas, today's gas turbine technology must be further developed to accommodate the specific physical and chemical properties of hydrogen.

In the medium-term, consideration should be given to the possibility of either removing CO_2 from the cycle through safe long-term storage (Carbon Capture and Storage, CCS[10]) or its further use (Carbon Capture and Utilization, CCU, which is a particularly sensible option for a future circular economy). The retained greenhouse gas can, for example, be used to produce synthetic fuels containing carbon. In combination with the use of biomass or synthetic hydrocarbons produced with atmospheric CO_2, even negative CO_2 emissions can be achieved [1,27,28]. According to the IPCC [29], this is necessary to achieve the 1.5°C target.

In the long-term, chemical energy carriers from renewable sources can enable a completely carbon-free cycle while avoiding CO_2 emissions. If this is done successfully, CO_2 no longer needs to be recovered from the atmosphere or the exhaust gas stream.

In addition to the renewably produced green hydrogen discussed above, other chemical energy carriers can be considered, including hydrogen-ammonia mixtures [30-32], ammonia [33,7,34], metals such as iron [8], metalloids such as silicon [35], and non-metals such as sulphur [36]. These energy carriers are available in the liquid or the solid phase and have higher volumetric energy densities than gaseous hydrogen or synthetic natural gas. This facilitates easier transport over long distances, for example, from regions with higher sun and wind levels than Germany.

The potential of metals, metalloids, and non-metals as chemical energy carriers for large-scale industrial applications has not yet been studied in detail. Currently, the use of these solids, which are based on elements relatively common in the earth's crust, is associated with higher technological risks than green hydrogen. This raises worthwhile questions for future research and development.

B. Mobility

Mobility takes up a large fraction of total energy used, with a share of about 30 % in Germany and a contribution of CO_2 emissions of 16–19 %. When discussing transition of the energy system, it is important to distinguish between contributors, which all have different energy needs and requirements. Based on energy demand, the main applications here are road-based passenger and heavy-goods transport, aviation, and ships, of which personal passenger transport is by far the highest percentage, and aviation the lowest. An important consideration for all areas is the high potential to improve energy conversion efficiencies. The World Energy Outlook 2018 states, "Energy efficiency is the key mechanism that curbs oil consumption in cars." Of course, we need to replace fossil fuels by CO_2-neutral synthetic or biomass-derived fuels. Energy efficiency remains as task of utmost importance, simply to limit fuel costs as far as possible. Another commonality across all combustion-based energy conversion technologies in the mobility sector is the need for further reduction of pollutant emissions. Both aspects might be realised using new synthetic fuels with beneficial properties, such as Fischer-Tropsch fuels, methanol, and oxymethylene ethers (OME_x).

Electrification

Electrified propulsion is often used in rail-bound transport, both of people and goods. This is an excellent option due to the low friction and the use of overhead lines. It is also widely agreed that for the short distances involved in personal passenger transport and delivery traffic, battery-electric vehicles (BEV) are a good solution for several reasons. One is that BEV produce no harmful emissions during operation, which

[8] For example, in the period from January 16 to 25, 2017, there was a foggy calm almost everywhere in Germany. Wind turbines and solar power plants with a combined installed capacity of 91 GW fed only about 4.6 GW into the power grid during this time but electricity demand was about 63.1 GW (Scientific Services of the German Bundestag, reference number WD 5-3000-167/18 (2019)).

[9] Methane slip refers to the proportion of methane emitted into the atmosphere during production, transport, and use. In 2013, the global mean fugitive emission rate, according to [23], was approximately 2.3 %.

[10] The CCS process is listed by the US National Academy of Engineering as one of the 14 Grand Challenges for Engineering [26].

is very important for reducing air pollution, especially in large cities. The well-to-wheel efficiency of about 70 % for delivering propulsion energy from renewable electricity [19] is high, though it does not consider energy demand for battery production, which is known to be a substantial part of the energy and CO_2 balance.

While promising for many application scenarios, electric vehicle technology faces many challenges when meeting the full range of transportation needs. Besides the well-known limitations of specific energy and energy density, and the current high cost, the benefits become most obvious when considering tank-to-wheel, which is often the basis for political discussions and European legislation. It is, however, important to include the actual CO_2 footprint of electricity production and other harmful emissions that are often ignored [37]. Transition to electrified personal passenger transport requires extensive charging infrastructure, which is difficult to provide, especially in cities. Another important aspect is that for many of the industrialized and densely populated parts of the world, renewable electricity might not be produced locally, but is imported, requiring long-distance transport. This is best achieved with chemical energy storage. BEV would therefore need an extra energy conversion step, from chemical to electric energy, at a conversion efficiency of about 60 %, which reduces overall efficiency substantially.

Biofuels

Biomass is a precious and versatile energy carrier that can be used for various applications, such as power generation, heat generation (especially high-temperature industrial processes), and as fuel for propulsion in gas turbines and internal combustion engines [38]. Consequently, there is competition for biomass in the energy market due to its limited availability. In the mobility sector, liquid fuels made from biomass have been proposed and might be useful for applications that are difficult to electrify, such as medium and long-distance road-based transport, ships, and aviation, where chemical energy carriers with high energy density are needed. Especially in road-based transport, drop-in fuels successively added in larger quantities to conventional fuels allows for the use of existing distribution systems and a gradual and non-disruptive transition to CO_2-neutral operation.

Much research has been done on biofuels. Many possible candidates have been found and some are already in use: sugar and starch-based ethanol for spark-ignition engines and fatty acid methyl esters (FAME) for compression-ignition engines. However, first-generation biofuels are made from the edible part of plants, which are part of the food chain; ethical issues are obvious. As an alternative, second-generation biofuels made from the ligno-cellulosic part of the biomass have great potential and different molecular structures, such as alcohols [39] and furanic species [40], have been discussed. Because of the limited amount of available biomass, biofuels can only make a partial contribution to the overall need for mobility, though it is an important contribution.

Carbon-based synthetic fuels

Synthetic fuels made from renewable resources span a wide range of production technologies and molecule structures. They could, for instance, come from gasification of biomass to syngas with a subsequent synthesis leading to hydrocarbon or oxygenated hydrocarbon fuels. Fischer-Tropsch synthesis, which leads to linear and branched hydrocarbon fuels, has already been in use in aviation with the benefits of reduced particulate emissions and that the synthesised fuels closely resemble the properties of fossil aviation fuels. This is important for worldwide compatibility. Other examples considered for internal combustion engines are methanol, dimethyl ether, and OME_x, where properties beneficial to knock resistance and reduction of emissions were found. However, if the carbon/hydrogen sources are from biomass, the same restrictions regarding their potential contribution to reducing CO_2 apply as those mentioned above.

Carbon-based e-fuels are made from available CO_2 streams that might result from non-carbon-neutral processes, or from direct air capture (DAC) of CO_2. The latter is particularly interesting, since, while DAC is thermodynamically challenging and hence energy-intensive and often said to be too costly, the extra cost of DAC per litre of petrol is only about USD 0.21 to 0.53 (see footnote 4), which seems well within typical fluctuations of the price for petrol. The current CO_2-tax in Germany is EUR 25 per ton of CO_2, which translates to about 6 cents per litre of petrol.

Two examples of carbon-based e-fuels are methanol and OME_x, which are used for spark-ignition and compression-ignition engines, respectively. Bio-hybrid fuels are an attempt to address the volatility of CO_2, biomass, and renewable electricity by combining these resources in a flexible way [41]. This leads to a large variety in the molecular structure of potential fuels, which means that they can be optimised for the desired fuel properties.

Carbon-free synthetic fuels

Examples of carbon-free e-fuels are ammonia, which can be made from molecular nitrogen using the Haber-Bosch process [42], and hydrogen, which can be made from water electrolysis. Both have received a lot of attention as potential chemical energy carriers for use in the mobility sector, because renewable electricity needs to be stored over periods of time, may not be produced locally and needs to be transported over long distances. Ammonia is unlikely to become an option for individual passenger transport because of its toxicity. However, it has been considered as a potential fuel for ship engines. Hydrogen is presently considered for many applications. In the mobility sector, hydrogen is often discussed in the context of fuel cells, which have great potential if the significant additional costs of manufacturing and operation (fuel cells require a high degree of purity of hydrogen during operation) can be reduced, and lifetime and operational experience in field tests [43–45] can be improved. Because of high costs and a comparably short service-life (due to unsolved materials problems in electrodes and membranes), fuel cells are only slowly gaining acceptance in the market despite enormous development efforts.

Alternatively, hydrogen could be used in thermo-chemical energy conversion processes in internal combustion engines. This has its challenges because, despite its simple molecule structure, hydrogen has very complex combustion properties that are substantially different to those of hydrocarbon fuels. They include very high burning velocities [46], low ignition energies and wide flammability limits [47], low volumetric energy density, and the tendency to form thermo-diffusive instabilities [48], which can drastically increase burning rates. All of these issues need to be considered for safety and stable operation of combustion devices. However, despite these challenges, hydrogen also has opportunities, especially for internal combustion engines, where the combustion properties mentioned could lead to particularly high thermal efficiencies.

Perspective

Two of the most-discussed options for the individual mobility sector are battery-electric and hydrogen-powered fuel-cell vehicles. Infrastructure and politics pose challenges to the development of distribution networks for electrical energy and hydrogen. Without charging and filling stations, the new technologies will not find buyers, which is a chicken-and-egg situation: A well-developed distribution network is needed to make the new options attractive to potential users – but the expensive network can only be operated economically if many customers use it. The transformation process comes with considerable risks.

Gas turbines for air traffic, internal combustion engines for ships, and hybridized drivetrains with internal combustion engines will be important in the future and provide opportunities for a non-disruptive energy transition. This is because the use of renewable and carbon-neutral carbonaceous or carbon-free chemical energy carriers has several benefits:

In the short-term, bio or synthetic fuels that have properties very similar to conventional fuels can be used as drop-in fuels until successively increasing quantities (up to neat) of renewable fuels are available. The Brazilian market has demonstrated the feasibility of using sugarcane-based ethanol for internal combustion engines; South Africa has demonstrated the use of Fischer-Tropsch fuels for aviation. While they are not perfect examples (as the Brazilian ethanol is a first-generation biofuel with the problems discussed above and the South African synthetic fuels were based on coal-derived syngas), they demonstrate the potential for seamless and non-disruptive transition. Similarly, synthetic natural gas and hydrogen can be used in mobile gas engines, effectively reducing CO_2 emissions, as discussed in the previous section for stationary gas turbines for power generation. The existing natural gas grid could be used for distribution if added hydrogen concentrations remain low.

For internal combustion engines in the long-term, fuels can be optimised jointly along with engines to provide clean energy conversion with ultra-high efficiencies. Hydrogen is one option for this, though carbon-based bio or synthetic fuels are equally possible and have advantages in ease of handling and safety. The joint optimization of fuel and engine is currently difficult for aviation because of the long life of aircraft engines and the need for world-wide compatibility. The use of ammonia for efficient hydrogen storage is interesting for mobile applications due to its high volumetric energy density and is promising for application in well-controlled environments [34]. This potential has been realised in the development of the first ammonia-fuelled ship engines [49]. Fundamental questions regarding process control and pollutant formation remain unanswered.

Like chemical energy carriers with high specific energy and energy density, liquid bio and synthetic fuels could facilitate the establishment of a world-wide connected market for renewable energy.

It is not possible to predict which technologies will prevail in the long-term and which will dominate the market. There are too many uncertainties regarding infrastructure, technology development, and political framework conditions. However, to meet the climate targets in a cost-efficient manner, several mobility technologies should be developed to high technology readiness levels (TRL) in parallel.

C. Industry

For all industrial sectors, the cost-effectiveness of climate protection measures is particularly important due to enormous global competition. Recent scenarios [50,19] assume disproportionately lower CO_2 reduction in the industrial sector. The imbalance should be offset by higher reductions in the mobility and building sectors.

Industry is a major factor in the energy statistics of many countries. In Germany, industrial processes account for 29 % of primary energy consumption. The main contributors are the metal industry (22.3 %), refineries and coking plants (10.1 %), glass, ceramic, cement, and lime (7.4 %), pulp and paper (6.4 %), and food and feed stuff (5.6 %) [51]. In other industrialized countries, of course, the distribution of industries is different. On a world-wide scale, approximately 29.4 % of greenhouse gas emissions are produced by industry, either through end-energy use in industry (24.2 %) or by direct product-related CO_2 emissions (5.4 %, mainly through calcination) [52]. The IPCC report AR5 [52,53] summarises the major efforts needed to reduce the carbon footprint of industry. The energy intensity in industry could be reduced by up to 25 % through wide-scale replacement and deployment of best available technologies and by a further 20 % through innovation in energy efficiency technologies. Real changes can be achieved by a shift to carbon-neutral fuels, product innovation (e.g., alternatives to cement), CCS and CCU, and electrification.

However, electrification has its limitations, as many industrial processes need high temperatures, which makes the replacement of fuels difficult. The required process temperatures for various industrial sectors and the relative amount of energy put into the individual temperature ranges is shown for German industry in Fig. 5. Most of the processes need temperatures above 1000°C, consuming a substantial fraction of the required energy.

Each CO_2 reduction measure in each industry sector mentioned needs to account for these temperature requirements and their different process characteristics and reactor concepts. This includes reformers (chemical industry), blast furnaces (steel industry), glass melting furnaces, rotary kilns (cement industry), black liquor boilers (pulp and paper), and lime shaft kilns (sugar production). This heterogeneity of processes prevents a simple analysis of which type of burner works with which future fuel for each application, since the combustion system layout may be very different and require an in-depth analysis of the specific situation.

In addition, for many processes, the design of a combustor in industry is different to other energy applications, as their job is not just to release heat from the fuel with low emissions. More importantly, the strong interdependence of the spatial and temporal heat release and the quality of the product to be produced, such as foamed clay (porosity), lime (reactivity), carbon black (specific surface area), iron ore sintering (density), metal annealing (ductility, hardness) must be considered. Therefore, flame–product interaction must be analysed carefully before switching fuels, for example, from natural gas to hydrogen. Many industrial applications rely on luminous non-premixed flames to transfer radiative heat to a product for processing. In hydrogen flames, radiation is of minor importance as soot is absent. The flame–product interaction analysis must also include the fact that a fuel often acts as an additional reaction partner to the product, such as the reduction of metals by hydrogen or carbon.

The specific challenges in CO_2-lean industry processes depend on the details of the process to be considered. A complete overview cannot be given here. However, a few examples can illustrate the diversity and opportunities in the process/product tasks to be tackled:

- Most glass melting furnaces are operated by natural gas, using highly preheated air (up to 1400°C) as oxidizer. Oxyfuel-fired furnaces[11] have been on the market for many years [54] to make products like high-temperature borosilicate glass. A switch to hydrogen and all-electric furnaces is under discussion; all-electric furnaces already

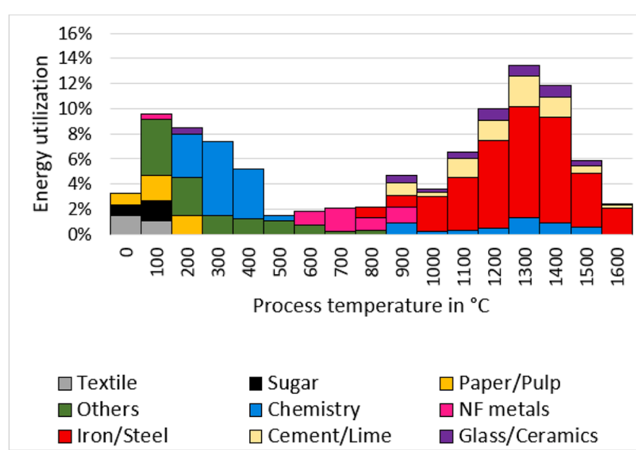

Fig. 5. Process temperature requirements in various industrial sectors and energy utilization shares of temperature intervals for Germany, adapted from [54].

[11] The term "oxy-fuel" describes an atmosphere mainly composed of CO_2, H_2O and O_2 in the combustion zone.

exist for smaller production rates and specialty glasses. Whether hydrogen combustion (or mixed hydrogen/natural gas combustion) influences the glass properties is being examined. As the optical properties of glass depend on the mineral composition, the use of solid biomass is not feasible, as the biomass ash may harm glass quality. Therefore, discussion about biomass in the glass industry is about biogas and syngas produced by gasification.

- Cement clinker production, in contrast, is robust when it comes to the integration of ash minerals. In recent years, coal has been replaced, often by waste-based fuels (up to 70 % in Germany [55]). As these fuels contain biomass, this leads to a reduction in the CO_2 footprint and production costs because of the low or even negative fuel price. As cement production is associated with product-related CO_2 emissions due to the calcination of $CaCO_3$, the pressure to reduce its CO_2 footprint is strong. Thus, the cement industry in Europe is currently the main driver for CCSU technologies [56], although detailed knowledge on oxyfuel combustion under cement conditions is scarce. Further research [57] is going into alternative production processes for cements with low Ca/Si ratio.
- If waste incineration cannot be combined with an industrial production process, because, for example, the heating value is too low, other options are needed. Worldwide municipal waste will double to approx. 8.6 billion tons per day before 2050 [58]. Today, more than 50 % of waste ends up in open-dump or controlled landfills. Only 13 % of this is recycled, which is the preferred option for waste treatment [59]. However, thermochemical treatment of waste in combustion (or gasification) units (either as fluidised beds or grate-firing systems, which are the most prominent reactor principles for waste incineration) will remain important, especially when legislation forces the shutdown of landfills. Waste is a potentially harmful and difficult fuel of high physical and chemical heterogeneity. Cheap, safe, and low-cost incineration solutions are required. Reliable, detailed combustion models for highly heterogeneous waste do not exist [60].
- Various methods to reduce the CO_2 footprint of steel-making have been examined [61]. The preferred solution currently under discussion is to replace the pulverized coal (as reducing agent) with hydrogen in the classical blast-furnace process [62]. Transition to direct reduction of iron ore, with hydrogen in shaft kilns or fluidised beds, in combination with electric arc furnaces [63], is anticipated but requires research and large-scale investment. As measurements inside reactors densely packed with particles are almost impossible, models are needed to account for the detailed particle-based processes of the solid-liquid phase change of the iron inside the reactor.

Many industrial processes are extremely sensitive to small changes, which is why modifications in technology are made very hesitantly. Long service life-times of the plants and high investment costs also make rapid technology changes difficult.

Perspective

High-temperature industrial processes can be very diverse, which leads to the need for individually adapted solutions. However, some of the major trends towards CO_2-lean industrial production processes can be summarised as follows:

- In industry, a large proportion of energy is required at temperatures above 1000°C. This means that thermochemical conversion/combustion will remain important, as it is difficult to replace, particularly in energy-intensive, high-temperature processes [64, 65], such as steel, glass and ceramics, stone and earth, and acetylene.
- Coal and oil will be replaced by natural gas and hydrogen, which will increasingly come from renewable sources. Shorter flame lengths and reduced radiation intensity must be carefully considered when switching to hydrogen as a fuel to ensure that product quality is not influenced [66].
- There will be an increased use of recycling strategies and hydrogen as a fuel/reduction agent [67], and the integration of renewably-generated electricity: This development is predicted by current studies, especially for iron and steel, aluminium, and copper.
- The use of CCS and CCU processes will increase, especially in industries that generate fuel-related CO_2 and product-related CO_2 emissions, such as the cement and lime industry. Radically new concepts, such as super-stoichiometric oxyfuel[12] combustion, are under discussion [68].
- The use of thermally recoverable waste and biomass will increase. The use of biomass for industrial processes to reduce CO_2 emissions is expected to focus on residual biomass for cost reasons; it has to be kept in mind that the total availability of biomass is limited [69].
- The development of novel products that fulfil quality criteria but can be produced with lower energy intensity and with a reduced carbon footprint will increase.

Because of the large variety of processes, reactor types, fuels, and products to be treated or generated, scientific understanding is not as advanced as for more standardised combustion systems, like in gas turbines. The industrial furnace business is characterised by small to medium enterprises, which typically have limited R&D capacities. Therefore, we plead for significant research efforts in the coming years to improve efficiency of industrial processes and reduce their CO_2 footprint.

D. *Buildings*

In terms of climate friendliness, the building sector has a lot of catching up to do, though there is great potential. The strongest motivation is increased energy efficiency[13] and the increased use of renewable energies. For Germany, the dena study [50] assumes that an integrated approach, in which the building envelope and technology are optimised as a unit, is the most economical. Packages of measures are proposed that consist of these components:

- Improved insulation of existing buildings (as a result of a significant increase in the annual renovation rate).
- Improved efficiency of existing building services (as a result of a significant increase in the annual replacement rate of old systems).
- Increased use of heat pumps operated using renewable electricity.
- Use of synthetic fuels, especially in hybrid systems, such as electricity-gas hybrid heating systems and gas-solar thermal hybrid heating systems.
- Use of climate-friendly building materials [71,72].

In the form of synthetic fuels chemical energy carriers will play an important role in the building sector of the future, especially in scenarios with low total cost and in combination with other measures. This is particularly relevant for existing buildings, for which retrofitting with heat pumps is not feasible [73]. For these older buildings, further reduction of CO_2 emissions will be made technically possible by successively adding synthetic natural gas and green hydrogen to the natural

[12] To avoid CO_2 recirculation for temperature control in oxyfuel combustion processes, the cement industry examines super-stoichiometric supply of oxygen to the rotary kiln, i.e. the super-stoichiometric oxygen acts as diluent to reduce the mean combustion temperature. The surplus of oxygen is consumed in the downstream calciner, where the highly endothermic calcination reaction moderates temperature.

[13] In Germany, new buildings must be constructed in accordance with the Energy Saving Ordinance [70]. Greenhouse gas emissions can also be significantly reduced by refurbishing existing buildings.

gas network and by using synthetically produced heating oil as a substitute for fossil fuel. This enables a continuous transition, since the existing distribution system, storage, and heating technology can be used as before.

Fuel flexibility of systems needs to increase where the energy carrier properties may strongly fluctuate (such as various amounts of hydrogen mixed to the natural gas) and further reduce the emission of pollutants, including nitrogen oxides and carbon monoxide, while maintaining the same high efficiency levels.

Implications for future research

Synthetic chemical energy carriers are indispensable for storing renewable energies, with one of the main reasons being that they enable a non-disruptive transition. For fast and feasible transformation into a sustainable energy circular economy, we plead for a technology-agnostic research approach. It should include the production of chemical energy carriers based on renewable energies, as well as electrochemical and thermochemical energy conversion processes.

As thermochemical energy conversion processes are the focus here, the following discussion concentrates on them. To support and develop promising approaches, increased efforts in basic and applied research are necessary. From our point of view, the following topics are particularly important:

- Processes for the utilisation of carbon-free and carbonaceous but carbon-neutral chemical energy carriers produced with the use of renewably-generated electricity.
- CO_2 recovery technologies.
- Methods for increasing process efficiencies.
- Reduction of pollutant formation during thermochemical utilisation of synthetic chemical energy carriers, especially under varying operating conditions and when using new fuel compositions.
- Complementary theoretical and experimental investigation of single and coupled reaction-transport processes, which significantly influence the efficiency and environmental impact of thermochemical energy conversion processes.
- Further development of numerical simulation methods using physically consistent mathematical models that enable stringent integration into computer-aided development processes. This requires benchmarking experiments for model development and validation.

CO_2 is released during the thermochemical use of carbonaceous chemical energy carriers, including synthetic natural gas and liquid synthetic fuels. The carbon footprint of these energy sources produced from renewable energies can be made neutral. CCS and CCU technologies are particularly suitable for stationary applications, as they can be used to directly remove the greenhouse gas emitted by power plants or industrial processes from the exhaust gas stream. CCS and CCU technologies are divided into pre-combustion capture, post-combustion capture, oxyfuel (see footnote 11), and chemical-looping processes [74-76].

For the example of the oxyfuel process, its future application requires a significant improvement in understanding [77]. It is still not sufficiently well understood how the CO_2- and oxygen-rich atmosphere in a combustion chamber changes the chemical reaction and physical transport processes in comparison to combustion with air. Basic scientific and technical investigations should be carried out with the aim of realising systems with significantly shorter development times.

There is also a need for research on gas turbines, particularly topics related to thermoacoustic phenomena [78-80], flame stability near the lean extinction limit [79,81], the degree of burn-out required to reduce methane slip, the tolerable addition of hydrogen to natural gas, and the conversion to operation with 100 % hydrogen [80,82]. In addition, conventional combustion processes in heavy industry and other industrial sectors should be examined for possible changes so that they can tolerate the highest possible and potentially variable hydrogen admixtures in the future.

The net CO_2 emissions in the transport sector can be avoided using carbon-neutral biofuels or synthetic fuels. For carbon-based renewable fuels, the CO_2 must be removed from the atmosphere to achieve zero climate impact. Synthetically produced kerosene for use in aircraft gas turbines must have strictly defined physical and chemical properties, as demanded by international air traffic regulations. The research questions concern conflicts of objectives between energy conversion efficiency, flame stability, and pollutant formation.

Synthetic chemical energy carriers for future low-emission, clean, and efficient internal combustion engines (which are synthesized using renewable electricity potentially combined with CO_2 and lignocellulosic biomass) include hydrogen, synthetic natural gas, heavier Fischer-Tropsch fuels, and the class of bio-hybrid fuels that are typically oxygenated hydrocarbon compounds. Compared to conventional Diesel and petrol, fuels such as methanol, oxymethylene ether, and dimethyl carbonate have a lower energy density. Due to their specific physical-chemical properties, however, their potential is very high. Increases in efficiency through higher knock resistance [83,84] or the joint reduction of soot and nitrogen oxides [85,86] are conceivable. Fundamental knowledge of chemical kinetics, liquid spray behaviour, combustion, and pollutant formation is needed for many species in this wide class of molecules for the joint design of fuels and engines.

If carbon-free chemical energy carriers are used, CO_2 separation from point sources and CO_2 recovery from the earth's atmosphere become increasingly redundant. However, if the energy carrier does not consist of atmospheric elements such as hydrogen or nitrogen, the reaction product must be recycled [8].

Of the carbon-free energy carriers, the first to be introduced will be molecular hydrogen for energy technology and automotive applications, as indicated by The National Hydrogen Strategy of the German Federal Government [17]. There is a need for research, especially on the molecular-dynamic and reaction-specific properties of hydrogen. Flame stability, flame dynamics, thermal nitrogen oxide formation, and the safety-relevant flammability and ignition limits all require detailed investigation.

Besides the thermochemical use of pure hydrogen, a mixture with ammonia is also conceivable [33,7,34]. Ammonia can be synthesized from renewable energy sources and has a much higher volumetric energy density than hydrogen. This facilitates longer-term storage and transport over long distances. There is a need for research regarding the tendency of ammonia to form nitrogen oxides, its high toxicity, and reaction-specific properties, such as low flame velocity [87,7]. A mix with hydrogen has great potential. New study results show that hydrogen-ammonia mixtures will allow higher degrees of turbulence without the risk of flame extinction [32]. This could enable higher power densities to be achieved while reducing residence time in the combustion chamber, which also has a positive effect on the development of nitrogen oxides following the thermal production route.

The thermochemical use of metals and metal oxides lies even further in the future. The use of elementary iron as an energy carrier was recently proposed; it would enable the storage and long-distance transport of renewably produced energy in a reduction-oxidation cycle [8]. The oxidation of iron for the release of thermal energy is conceivable in dust firing or fluidised-bed processes. Use in existing power plants could be possible, which is a major advantage when implementing new technologies. The chemical reduction of the iron oxides that result from the thermochemical use can be carried out in several ways. One possibility is the use of green hydrogen from renewable energy sources, as there are synergies with recent projects in the steel industry [88]. Overall, the oxidation–reduction cycle has great potential as an energy carrier for renewable energies. The research questions arising are of an extremely fundamental nature. They concern combustion characteristics, thermodynamic properties of the overall process, and economic efficiency. Regarding recycling of the reaction product, this option seems to be particularly suitable for stationary use.

Whether carbonaceous or carbon-free energy carriers are used, their thermochemical energy conversion leads to the release of pollutants, including classic air pollutants such as nitrogen oxides (NO_x), sulphur dioxide (SO_2), carbon monoxide (CO), and particulate matter. Unburned hydrocarbons, which can result from incomplete conversion of the fuel, and formaldehyde, as a reaction by-product of oxygenated fuels, can also be produced. Further reduction in pollutant formation is essential to be able to operate thermochemical plants within the framework of future statutory emission limits. This includes primary measures, such as lean premixed combustion [89–91] and exhaust gas recirculation [92–94], and secondary measures using catalytic converters [95–97].

The energy system should be transformed quickly. But the development of new technologies takes time. Many processes can be accelerated with numerical simulation tools. Over recent decades, numerical simulation of technical systems using high-performance computers has become established as an independent scientific discipline that interlocks theory and experiment, particularly in combustion science and technology. This development was made possible by steadily increasing computing power at reduced costs as well as progress in increasingly more accurate mathematical models, as targeted in internationally organized workshops [98-102]. Numerical simulation methods enable shorter development times and improve quality of individual components and entire combustion systems. In addition, numerical simulations allow complete digitalisation of process chains, starting with the first steps of development, up to the finished product, and continuing across the entire lifecycle, including disposal or recycling.

Simulation methods should be continuously extended to include the physical and chemical sub-processes relevant to new scenarios, such as those discussed above, for instance combustion of pulverized fuels (biomass, metals), combustion with high levels of CO_2, ultra-lean combustion including ignition and flame-stabilisation, plasma-assisted combustion, combustion at high pressure, soot formation from oxygenated fuels, fuels such as ammonia, iron, or sulphur, and flame instabilities and their interaction with turbulence. Simulation methods are needed for industrial processes that describe flame-product interaction to then be able to reliably predict and design product quality. These topics all require fundamental research, including experiments and direct numerical simulations, as well as development of models and their integration into a multi-physics simulation framework. With higher complexity of the physical processes and increasing simulation fidelity, simulation time and cost increases. The most accurate and expensive models are not always desirable for industrial use. Hierarchies of models should be developed that include uncertainty estimates to make the trade-offs between simulation cost, turnaround time, and accuracy flexible.

With higher fidelity in both experiments and simulations, the amount of available data increases: data analysis has become a very complex yet promising topic [103]. Machine learning techniques should be explored for their use in data analysis [104] to improve understanding and knowledge and in model development. This new field needs lots of research that goes all the way to operation, monitoring, and control of combustion devices.

Summary

For a reliable and feasible transformation of the energy system we plead for a technology-agnostic research approach. Thermochemical energy conversion processes enable continuous transition towards a renewable and carbon-neutral future that does not require the disruptive change associated with infrastructural barriers and high technological risk.

For thermochemical energy conversion, from our point of view the most important research questions are:

- How can energy conversion efficiencies of thermochemical energy conversion processes be improved to substantially higher values?
- How can we operate thermochemical energy conversion systems with synthetic chemical energy carriers in a way that is not only sustainable but also reliable, cost effective, and at the highest possible efficiency?
- How can we optimally coordinate thermochemical energy conversion systems and synthetic chemical energy carriers?
- How can the fluctuations in electricity from renewable sources be compensated for with thermochemical energy conversion systems in a way that is safe, clean, sustainable, and cost-effective?
- How can we accurately map the relevant physical-chemical processes using mathematical models and reliably predict them with numerical simulations?
- How can we measure the coupling of chemical reactions and physical transport processes precisely and accurately?
- How can we use the methods of artificial intelligence to predictively simulate thermochemical energy conversion processes, analyse physical-chemical mechanisms, and develop new, precisely controllable thermal energy converters?

There are already many options for transforming the energy system and further exciting innovations are on the horizon. We should openly explore, test, and use them as they bring us closer to the main goal: A climate-friendly future.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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