ARTICLE IN PRESS
JID: JPECS [m5G;November 8, 2017;16:41]
Progress in Energy and Combustion Science 000 (2017) 1–5
Contents lists available at ScienceDirect
Progress in Energy and Combustion Science
journal homepage: www.elsevier.com/locate/pecs

Perspective

Clean combustion: Chemistry and diagnostics for a systems approach in transportation and energy conversion

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This perspective article intends to show opportunities and provide food for discussion regarding some specific developments in the field of combustion. It reflects the author's views and makes no claim to be exhaustive. It is hoped that aspects and problems addressed in this article will stimulate intensified communication and collaboration toward more efficient processes and reduced combustion emissions. In the author's view, combustion science as a multidisciplinary field connecting chemistry, fluid dynamics, metrology, and high-performance computing with a variety of practical applications in energy, transportation, and industrial production is well positioned to help developing technologies for an integrated, sustainable future energy system.

1. Combustion's role? Some reflections on the status quo

Emissions from combustion processes have a large influence on air quality, environment, climate, and health [1,2]. They cause concerns about severe air pollution in cities and contributions to anthropogenic carbon dioxide [3,4]. Today, more than 80% of global primary energy consumption relies on fossil energy carriers [1,2,5]. High-energy-density liquid fuels are indispensable for transportation, with trends noted toward bringing in fuels of non-fossil origin. A fully renewable energy supply based on wind, water, and sun is suggested as technically viable [6] with the largest concern being temporal fluctuations. Also, research toward harvesting and storing renewable energy with chemistry is accelerating [7]. Nevertheless, global CO_2 emission has risen from about 22 Gt in 1990 to 36 Gt in 2015 [3]. With increasing global energy demand, anthropogenic fossil-fuel contributions to CO_2 emissions (of near 94% in 2015 [3]) are not expected to change rapidly [1,2,4]. Carbon dioxide, however, is only one pertinent issue regarding combustion emissions. Black carbon, for example, – particulate matter or soot from open burning and controlled combustion – is associated with significant climate impact [8] and reported to be among the most prominent global health hazard factors [9–12], contributing notably to risks such as diseases of the respiratory and cardiovascular systems.

It is too short a perspective, although quite common in public debate, to target only combustion emissions from ground transportation: Combustion is not synonymous with combustion engine. Introducing enough electric cars, so one rationale, would supersede combustion and eliminate its emissions. This perception, however, neglects not only that electric propulsion is not yet independent of fossil energy [13], but disregards also the multitude of combustion processes and devices in use, including aero-engines, stationary gas turbines and boilers, waste incinerators, household appliances, and industrial furnaces and processes for the production of glass, ceramics, steel, cement, and other major products. The CO_2 burden from cement making alone was approximately 2 Gt in 2015 or 5.6% of global CO_2 emissions [3]. Approximately half of this amount was generated in China [3], where further significant CO_2 contributions for some high-temperature industrial products (metals, plate glass, and chemicals) were recently evaluated [14]. A wide spectrum of such thermochemical energy conversion processes must thus be targeted with the aim of enhancing efficiency and reducing emissions while bringing in renewable energy sources, a large-scale transformation that will take enormous time and effort. Promising concepts and strategies will need interdisciplinary knowledge on fuels, energy conversion processes, systems, and infrastructure – knowledge in part available from combustion scientists and engineers.

- Combustion processes dominate today's energy and transportation systems to an extent that substantial changes will take time, especially with growing population and energy demands.
- Combustion science and technology can assist in the transformation of energy conversion systems toward efficient processes with low emissions.

2. Good fuels? Informed choices toward a future fuel portfolio

Future transportation fuels are discussed with today's liquid petroleum-based fuels as the benchmark. The question of tomorrow's best fuels, however, is not settled, and solutions will depend on many factors, including scientific understanding of their use and impact, energy density, toxicity, water solubility, emissions, availability, economic viability, adaptation to existing infrastructure, ease of end use, and a viable lifecycle analysis, with carbon neutrality and sustainability as key factors. Biomass-based fuels beyond first generation are often considered promising [15–18], but challenges include sufficiency of resources, large-scale energy-efficient, economic production, and the availability of "green" hydrogen for deoxygenation of feedstock with too high oxygen content.

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http://dx.doi.org/10.1016/j.pecs.2017.10.001

Please cite this article as: K. Kohse-Höinghaus, Clean combustion: Chemistry and diagnostics for a systems approach in transportation and energy conversion, Progress in Energy and Combustion Science (2017), http://dx.doi.org/10.1016/j.pecs.2017.10.001

Improvements in process technology and integration are reported [17,18], with research needs including, for example, higher efficiency for catalytic conversions. Biomass-based fuel additives offer chances to tailor fuel properties, but complications with such multi-component mixtures can arise for *e.g.*, mixture formation, ignition timing, and reaction chemistry. Fuel design from biomass should strive for suitable propulsion and emission performance of the fuel–engine system and then develop a viable and sustainable fuel production strategy [19].

Some fossil fuel options may continue to be relevant both in the short-term as well as in the more distant future, where their use will depend on the potential of re-activating CO_2. Currently, a cleaner fuel of choice for a range of applications might be natural gas (compressed or liquefied to simplify transportation). The fuel portfolio can also include more unconventional fossil sources such as shale gas and, potentially, methane hydrate. However, leakage of the potent greenhouse gas methane could lead to adverse effects that should be considered. With a future perspective of carbon capture [20] and recycling, *i.e.* using the waste product CO_2 as a renewable C1 building block in organic synthesis [21], liquid compounds can be accessed such as methanol, usable directly or upgradable to further useful chemicals such as dimethyl ether (DME), ethene, and fuels with a longer carbon chain [22]. As further synthetic fuel candidates, oxymethylene ethers (OMEs) receive attention [23,24] that are accessible from methanol and formaldehyde [24,25], itself a C1 energy carrier. Also, ammonia is being discussed as an energy vector [26]. Solar and wind power will be important for producing electricity renewably, but owed to their intermittent nature, energy storage issues will need attention: The current storage capacity, almost exclusively from pump-storage hydroelectricity, is estimated to be about 1% of the energy consumed worldwide [13], not suggesting, however, that long-term buffering could be achieved with this capacity. It could be extended beneficially by using liquid hydrogen-rich compounds – fuels.

It should not be overlooked that aviation fuels must adapt to specific criteria, including foremost high energy density. Also, combustion for stationary purposes, from household heating to gas-fired turbines for power generation, might consume similar energy as in the transportation sector. Here the challenge for new fuels lies in the hundreds of millions of consumers, with a vast diversity of combustion equipment that is adapted to a specific range of fuels, *i.e.* natural gases. Drastic changes in fuel properties, for example by introducing hydrogen, imply large-scale changes in equipment. Fuel-flexible devices would thus be a useful option.

Which fuel, for which purpose is a complex issue, and predictions are not easy. Ultimately, fuels should have a neutral carbon footprint and be derived from a source in infinite supply. To reduce particle formation, options may include compounds with only C1 chains such as OMEs, especially when they are liquid, not toxic, and can be sustainably produced. Process integration providing multiple conversion options between heat, electric power, chemical storage, and transportation fuels will be advisable [27,28]. Thermochemical energy conversion processes are valuable building blocks for future energy systems, where combustion specifically provides links between high-energy-density liquid carriers from different sources and thermal energy at the needed high temperature and pressure levels. Inherent complexity and yet unclear challenges regarding suitable interfaces of future energy conversion systems demand an interdisciplinary scientific approach. Revealing the physics and chemistry controlling these processes for real fuels and pertinent thermodynamic conditions can rely prominently on expertise gained from combustion.

- Whilst future's "best" fuel is not yet decided, high-energy-density liquid compounds are always useful for energy conversion and storage.
- Since change of fuels can entail large changes in infrastructure for production, delivery, handling, and conversion, such processes are best optimized jointly, considering a viable lifecycle analysis.

3. Cleaning up? Research needs for emission reduction and aftertreatment

Combustion emissions are composed of a diverse spectrum of chemical species of which only some are regulated so far [29]. These include gaseous species such as oxides of carbon, nitrogen, and sulfur, for which measurement procedures are available to control compliance with regulations. The chemical composition of emissions depends on both, fuel and combustion process. Understanding and controlling particulate matter or soot emission from combustion counts among the most pressing issues [9,11,30]. In terms of disability-adjusted life years (DALYs), ambient particulate matter (PM) pollution is estimated for 2015 to exhibit up to 16% influence on tracheal, bronchial, and lung cancer in almost all regions of Africa and Southeast Asia, and household air pollution from solid fuel burning a contribution of 4–8% on all health risks in most of the same regions [10]. A crucially important target is therefore to accurately assess black carbon impacts on the global atmospheric radiative energy balance, on human health, and air quality [30]. Further research to systematically understand the relationships between fuel, combustion process, soot formation mechanism, resulting particle properties, as well as soot emission and oxidation characteristics is highly needed [31,32]. Relevant details are lacking in the understanding of influences on soot mass and number concentration, size distribution, morphology, optical properties, chemical composition, active surface area, and reactivity [33]. Such properties must be reliably determined, especially for nanometer-size particles, requiring advances in measurement techniques. Furthermore, secondary organic aerosol (SOA), organic matter that forms in the atmosphere by oxidation from gas-phase precursors including aromatics and larger alkanes, may be a potentially underestimated combustion-related emission [29]. Research focusing on such aspects is the more acute since particulates from combustion are one of the best targets to alleviate climate forcing impacts and human health risks simultaneously.

Achieving lower emission levels, for existing and novel boundary conditions regarding fuel choice and combustion process, to meet tighter regulations or to reduce yet unregulated or "new" pollutants, will require a dedicated interdisciplinary approach. Developing lower-emission combustion systems demands improved scientific understanding of fuel- and condition-specific pollutant formation mechanisms, including the complex pathways of soot formation and oxidation. Development of new combustion concepts could be favorably coupled with research on suitable aftertreatment systems, involving the catalysis and materials science communities. Scientific evidence to inform policymakers, and affordable aftertreatment systems potentially suitable for retrofits might assist in speedy introduction of abatement measures in regions where this may be most needed. Similarly, efficient and affordable, potentially portable sensors must be available for local emission control. Reducing combustion emissions must focus on many applications, including ground and air transportation [29,34] but should not overlook the impact of low-tech devices such as rural cookstoves [35]. A wide research field thus continues to demand attention.

- Cleaner combustion concepts will profit from fundamental understanding and joint design of fuels, combustion and aftertreatment systems.
- Affordable technology, potentially for retrofits, is the key for fast improvements.

Please cite this article as: K. Kohse-Höinghaus, Clean combustion: Chemistry and diagnostics for a systems approach in transportation and energy conversion, Progress in Energy and Combustion Science (2017), http://dx.doi.org/10.1016/j.pecs.2017.10.001