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Catalysis in the development of clean energy technologies

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Abstract

Among the most enduring challenges for scientists and engineers interested in catalysis is the development of clean energy conversion processes. Much of the research reported in the first 100 volumes of *Catalysis Today* has been devoted to this subject; e.g., synthesis of clean fuels, catalytic combustion, natural gas utilization, and control of emissions from stationary and mobile sources, for example. There is a clear relationship between energy use and our standard of living, creating an ever increasing demand for affordable energy. At the same time, environmental considerations require that this energy be generated and used with minimal impact on our surroundings.

There are essentially three approaches to the use of fossil fuels to meet this challenge:

(1) Lower emission technologies: more usable Btu/lb emission;
(2) Higher efficiency: more usable Btu/Btu fuel;
(3) "Renewable" energy: usable Btu without consuming non-renewable fossil fuel.

Catalysis can and will play a central role in each of these approaches.

Keywords: Clean energy; Catalysis; Environmental catalysis

1. Introduction

There are a number of factors that drive the need to produce more energy with less economic and environmental impact.

1.1. Importance of energy

There is a clear correlation between energy use and standard of living. As an example, Fig. 1 shows data that supports this [1]: energy consumption per capita is directly and strongly correlated with per capita GNP. The result is an increasing demand for energy, especially in developing nations where energy efficiency is not as high as in the developed world. For example, Fig. 2 shows that per capita energy use (Btu per capita) as a function of per capita GNP is much higher in developing countries than in industrialized ones [2].

1.2. Uncertainty in energy prices

Although energy price fluctuations are inevitable, the resulting uncertainty appears to be greater in recent times. For example, Fig. 3 shows historic trends in natural gas prices [1], with the recent spikes being obvious. Corresponding trends in the price of oil add to the uncertainty. The result is added incentive to produce more usable energy at a lower cost.

These price changes can produce significant economic impacts. Table 1 shows the effect of a permanent increase of \$10/bbl in the price of oil [3]. Even this rather modest increase in oil price produces severe effects-job loss and economic disruption.

1.3. Environmental impact

The conversion of fuels to energy also has significant environmental impact. Because most of the conversion of fossil fuels to energy involves air-blown combustion processes, the trends in NO_x emissions are an indicator of

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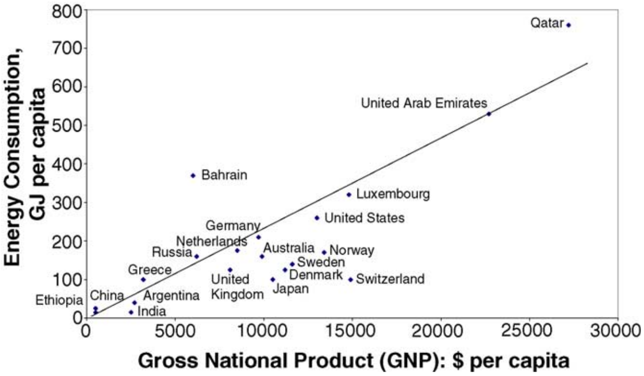


Fig. 1. Per capita energy consumption vs. per capita GNP (data from International Energy Agency).

this impact. Fig. 4 shows the historic trend in NO_x emissions [4], with the increase being obvious.

Collectively, these factors provide a clear incentive to develop energy conversion processes that are efficient and have minimal environmental impact.

There are three basic approaches to the use of fossil fuels to meet this challenge:

(1) Lower emissions: more usable Btu/lb emission;
(2) Higher efficiency: more usable Btu/Btu fuel;
(3) ''Renewable'' energy: usable Btu without consuming non-renewable fuel.

Examples of the challenges for catalysis research for each of these approaches are provided below.

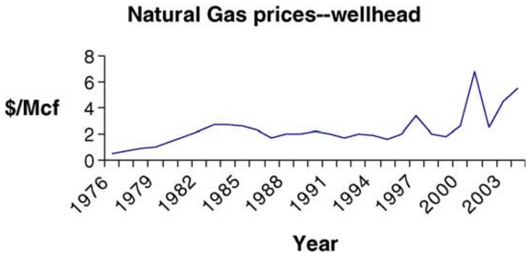


Fig. 3. Natural gas prices 1975–2003 (Ref. [1]).

1.4. Approach I: lower emission technologies – more usable Btu/lb emission

Catalysts have been used in a wide range of processes aimed at minimizing emissions from energy conversion processes. Among the more important applications are:

1. *Hydrogen-based energy conversion processes*, which eliminate the formation of NO_x that is inevitable in air-blown non-catalytic combustion. [If the hydrogen is free of hydrocarbons and CO, emissions resulting from these compounds are also eliminated in hydrogen-based combustion processes.] These hydrogen-based processes have perhaps received more recent attention than any of the other lower-emissions technologies. Hydrogen can be used for electric power generation, as a transportation fuel, in industrial boilers, and even for commercial and residential heating. Hydrogen is a clean-burning fuel that can be produced from a number of sources, including biomass [5–7], coal [8], and even solar energy [9]. Although many challenges remain before hydrogen can be widely used [10,11], there are clear environmental and

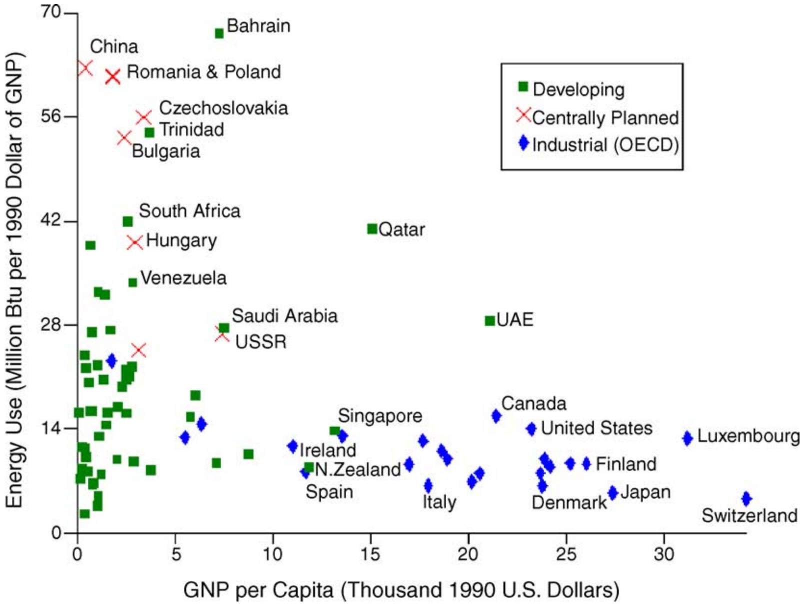


Fig. 2. Energy use per dollar GNP vs. per capita GNP (Ref. [2]).

Table 1
Macroeconomic impact of a permanent $10/bbl increase in the price of oil, change from baseline (Ref. [3])

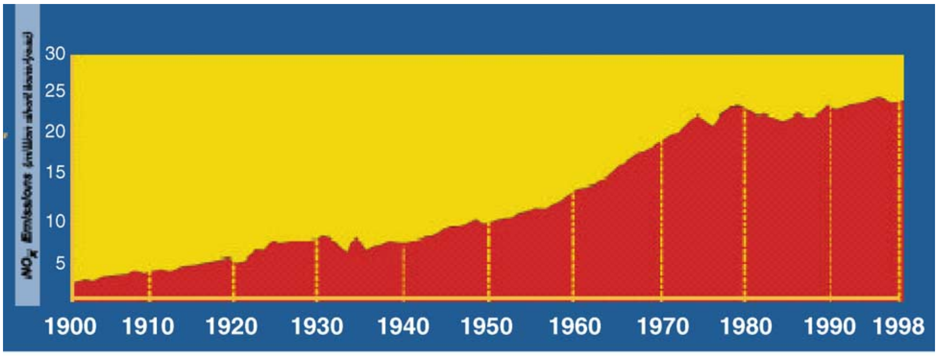
	Year 1	Year 2	Year 3
Real GDP growth (%)	−0.1	−0.4	−0.8
Real GDP (10^9 $$)	−5.6	−40.2	−121.4
Nominal GDP (10^9 $$)	−5.0	−21.1	−30.0
After-tax profits (10^9 $$)	11.2	9.7	7.0
Unemployment rate (%)	0.0	0.1	0.3

efficiency benefits [12,13]. For these reasons, the US DOE has a stated goal of replacing 2–4 quads of US energy use with hydrogen by 2010 [14]. Current US production of hydrogen is about 18 billion lb/year (corresponding to about 1.1 quads), almost all for chemical synthesis and processing [15]. If used solely to produce energy, this could fuel 20–30 million cars or power 5–8 million homes [15].

Catalysts will play perhaps the most critical role in the ultimate development of these processes [16,17]. It is outside the scope of this paper to describe in detail all the opportunities for catalysis in this area, but there are a number of challenges that must be addressed, from the production of hydrogen, through its distribution and storage, to its use in stationary and mobile fuel cells [18] and combustion. Some examples include the following (the use of hydrogen in fuel cells is discussed later in connection with fuel processors, which provide hydrogen to the fuel cell):

- *production*: today, steam reforming of methane accounts for 95% of hydrogen produced in the US [15]. Assuming this will continue to be the most important near-term source of bulk quantities of hydrogen [19], there are significant improvements needed in these catalysts to lower production costs [20]. These include minimizing deactivation [21] and improved reactor design (e.g., catalytic membranes [22,23]).
- *storage*: unless it is produced on-board (vide infra), hydrogen must be stored for use in mobile applications. Catalysts can be used to produce hydrogen storage media such as carbon nanotubes, which are synthesized by the decomposition of methane [24]. Catalysts are also important in catalyzing reversible chemical reactions that can be used to store hydrogen; e.g., methanol decomposition [25,26] or the formate–bicarbonate reaction [27].

2. *Automotive catalytic converters*, which have undergone significant study and change since their introduction in the 1970's. However, significant challenges remain [28,29], e.g., limiting deactivation [30] and developing catalyst formulations with minimal noble metal loadings [31]. An especially important challenge is the control of gaseous and particulate emissions from the more efficient lean-burn (diesel) engines [32,33], discussed more fully below.
3. *Catalytic combustors for gas turbines*, which are designed to meet even the most stringent NO_x emission regulations. These catalysts, typically based on Pd, combust natural gas at temperatures near 1300 °C, which is sufficiently low to limit NO_x emissions to 3–5 ppm [34]. Other catalysts such as perovskites [35] and hexaaluminates [36–38] are also being developed as a way to minimize costs while retaining sufficient high temperature activity. Challenges include reactor design (e.g., limiting the temperature to which the catalyst is exposed), deactivation [39], dynamics of the Pd–PdO transformation, low temperature activity (lightoff near 350 °C), kinetic modeling, and use of fuels other than methane [34,40,41].
4. *Post-combustion $SO_x$and/or NO_xremoval processes*, which are applied both to stationary and mobile sources. For example, the combustion of coal for electrical power requires removal of both SO_x and NO_x, typically using a combination of scrubbers to remove SO_x followed by selective catalytic reduction of NO_x using ammonia [42]. Low temperature SO_x/NO_x processes based on carbon-supported catalysts, designed to operate at more accessible places in the process, have also been studied [43]. For other combustion processes, a wide range of reducing agents for NO_x have been investigated in laboratory studies, but the most commercially applicable are urea [44], ammonia [45–47], and CO/H_2 produced



Source: U.S. Environmental Protection Agency, "National Air Pollution Emission Trends Report, 1900 - 1998," March 2000.

Fig. 4. US NO_x emissions, 1900–1998 (Ref. [4]).

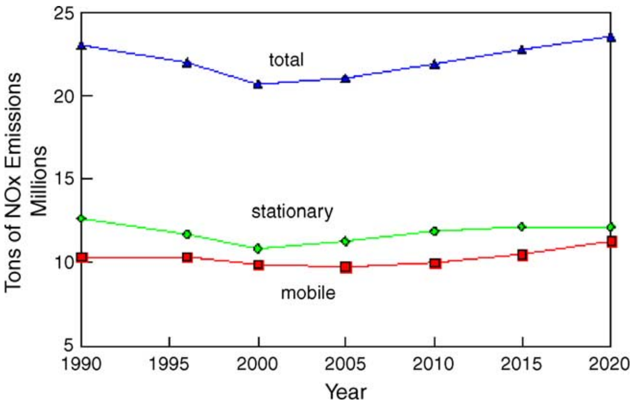


Fig. 5. US NO_x emissions by source, 1990–2020 (Ref. [55]).

from intentional operation of the combustion process at rich conditions [48,49]. Catalysis research is currently focused on deactivation [45,50–52] and selectivity of the reduction reaction in the presence of excess oxygen [53,54].

1.4.1. Example – NO_x control from mobile sources

Using NO_x emissions as an indicator of gross energy usage, data show that roughly half of these emissions in the US are from mobile sources (Fig. 5) [55]. Because of the higher efficiency of diesel engines, there is an incentive to increase the proportion of vehicles using this technology. However, without emission controls, these engines produce unacceptably high levels of NO_x and particulate matter, which will be severely restricted in the future (Fig. 6) [56]. Catalysts are being developed to remove NO_x from the exhaust gases using a number of approaches [57,58], including:

- NO_x traps, in which the NO_x (primarily NO) is oxidized to NO_2, which reacts with a base metal and is periodically regenerated (Fig. 7).
- Selective catalytic reduction using added urea or CO + H_2 generated on-board [59].

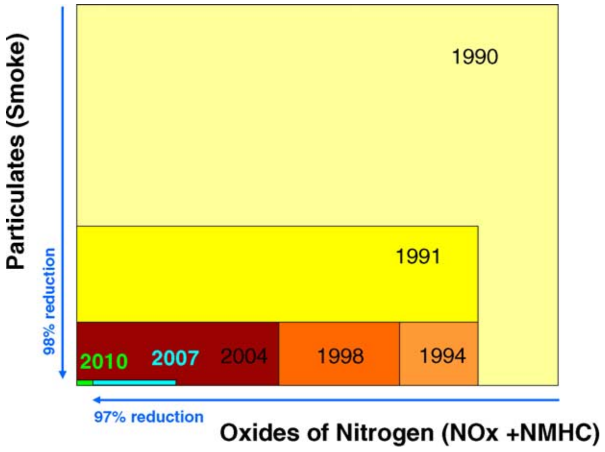


Fig. 6. Increasing emission restrictions on diesel engines (courtesy of Caterpillar, Inc.).

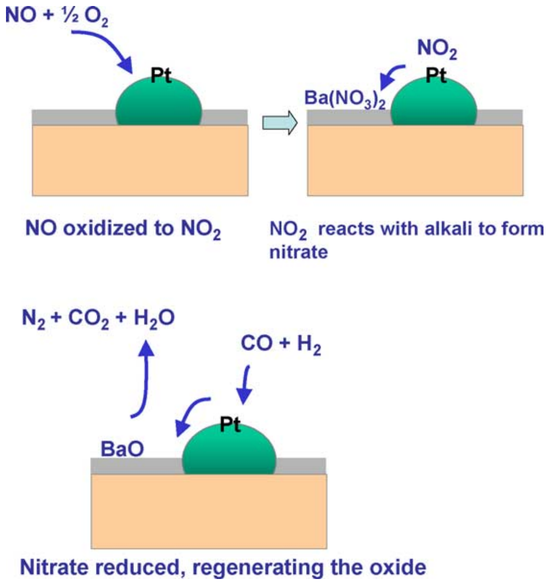


Fig. 7. Catalytic NO_x trap, showing both the oxidation of NO to NO_2 on Pt (followed by its reaction with a base, BaO in this case, to form a nitrate), and subsequent regeneration using CO and H_2.

There are significant challenges to both approaches because of the presence of particulate matter, and contaminants such as sulfur.

Another approach is the direct catalytic decomposition of NO. This reaction is thermodynamically favorable at diesel exhaust conditions (typically 300–400 °C):

$$2NO \leftrightarrow O_2 + N_2$$

$$\Delta G_{350\,^{\circ}C} = -39.5\,\text{kcal/mol}$$

[Despite the thermodynamic feasibility of the reaction at these conditions, Goralski and Schneider have recently pointed out that there are equilibrium limitations to achieving the extremely high fractional conversions of NO required to meet the stringent requirements of upcoming emission standards [60].] A number of metals are active for this reaction [61–66], with Pt and Pd showing the highest activity. However, these metals tend to deactivate due to oxygen poisoning of the surface [8,67]. This is illustrated in Fig. 8, which shows data from a study by Burch and Watling on a Pt/alumina catalyst [68]. The initial activity for the reaction diminishes rapidly as oxygen atoms accumulate on the surface after a few turnovers. However, metal particle size and support can affect the reaction. Wang et al. recently studied the effects of support and particle size for supported Pt for the direct NO decomposition and found that although molecular oxygen is produced at temperatures around 350 °C, N_2O is also produced at these temperatures [69], suggesting oxygen accumulation on the surface. At higher temperatures, N_2O production decreases, and steady state NO decomposition to N_2 and O_2 is found at 600 °C, without apparent self-poisoning for a period of 1 h. Possible sintering was also examined. On a silica support, larger Pt clusters

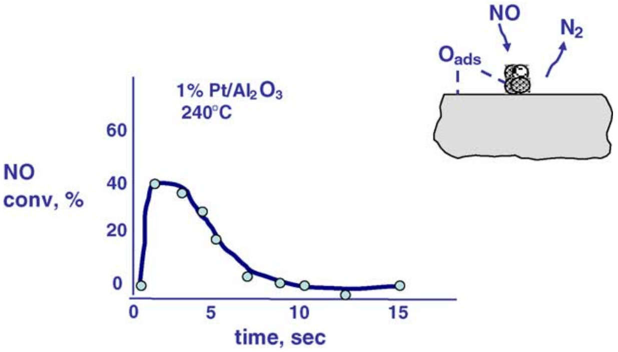


Fig. 8. Self-poisoning of a Pt/alumina catalyst in the direct decomposition of NO by accumulation of O-atoms on the surface (adapted from data in Ref. [68]).

resisted sintering, while nanometer-sized particles sintered during the reaction.

1.5. Approach II: higher efficiencies – more usable Btu/Btu fuel

Recent comprehensive studies of the energy flow for the US show that out of a total consumption of about 100 exajoules (1 exajoule = 10^{18} joules), about 60% is "rejected" (Fig. 9) [70]. Though much of the rejected energy is thermodynamically inevitable, this clearly suggests there are opportunities to improve efficiency in energy conversion processes. Closer examination of this energy flow shows that utilities/power and transportation are both the largest energy-consuming sectors, and also those which offer the greatest potential benefit from higher efficiencies (Table 2).

Table 2
Energy efficiency of various US energy-consuming sectors

Sector	Percent of total energy consumed[a]	Rejected energy/total energy consumed for that sector
Utilities/power	36	0.67
Transportation	26	0.80
Industrial	22	0.20
Residential/commercial	18	0.25

[a] Based on data in Ref. [70].

1.5.1. Fuel cells

One technology that is being carefully evaluated for use in stationary power and transportation applications is the fuel cell [71]. These systems offer significant potential to improve energy conversion efficiencies (Fig. 10) [72,73]. Among the various types of fuel cells, proton exchange membrane (PEM) technology is being pursued by a number of companies because its low operating temperature, response to transients [74], and compact size make it ideal for a number of residential [75], commercial, and military [76] applications.

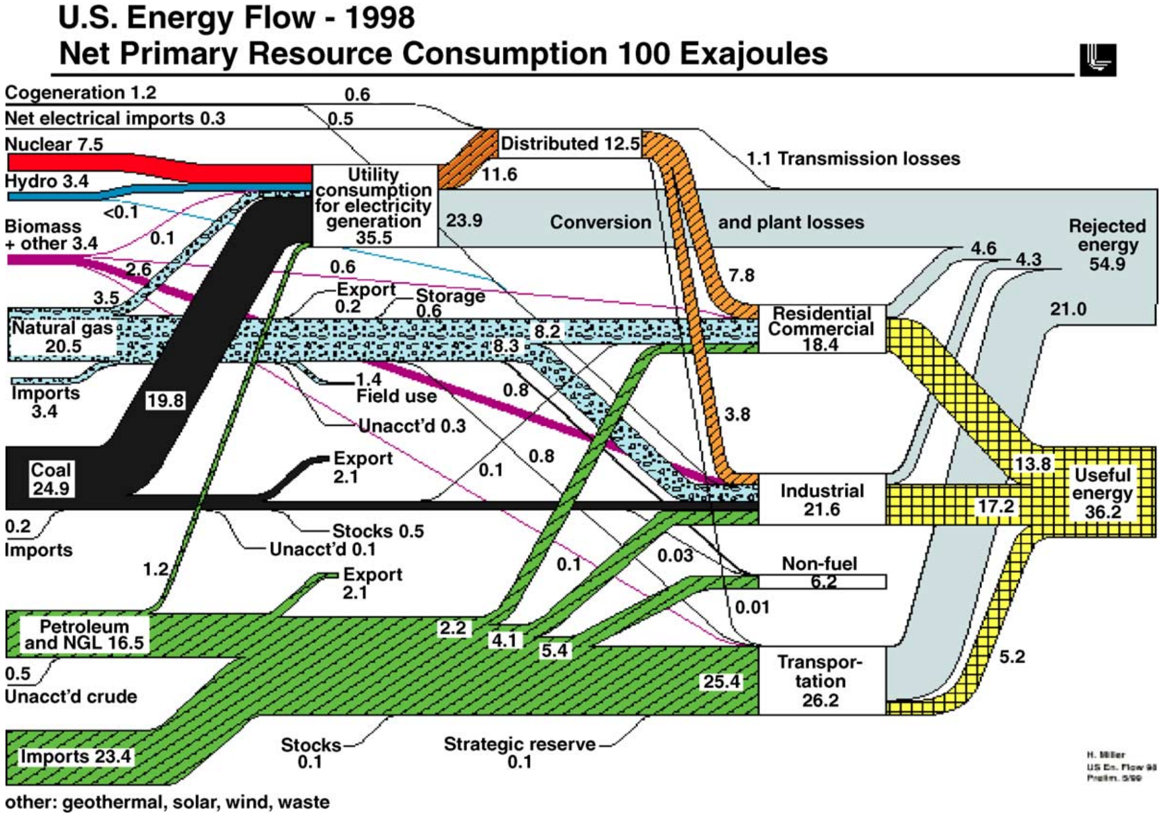


Fig. 9. Energy flow in the US, 1998 (Ref. [70]).

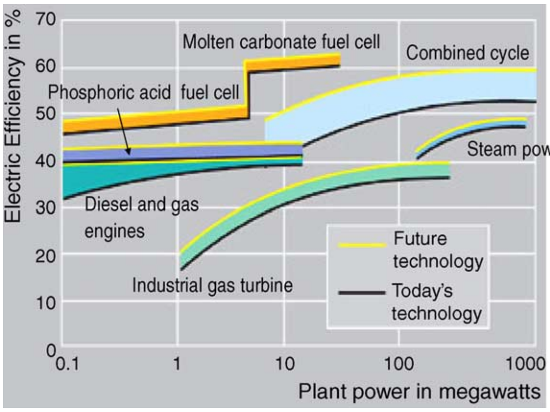


Fig. 10. Energy efficiency of various processes as a function of size (Ref. [72]).

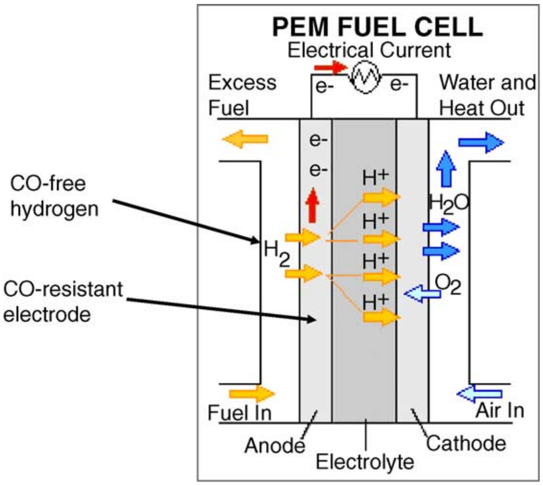


Fig. 11. Schematic of a PEM fuel cell (Ref. [77]).

1.5.2. Example – fuel processors for fuel cells

1.5.2.1. Catalysts for preferential oxidation. Fig. 11 shows a schematic of a PEM fuel cell [77], which uses catalysts both in the fuel cell itself and in the fuel processor, which supplies the CO-free hydrogen to the Pt electrode. For liquid fuels (e.g., methanol, diesel, or gasoline), this processor consists of several steps, as shown in Fig. 12. Farrauto et al. have recently reviewed the key catalyst development needs for these PEM fuel cells, and show that the preferential CO oxidation (PROX) step, the last before the fuel cell, is critical [78]. In this step, CO must be reduced from about 1% to less than 10–20 ppm, in the presence of ∼35% hydrogen, to avoid poisoning the Pt electrode. A number of different metals have been investigated for this reaction, including CuO [79,80], Pt [81–83], Au [84–86], and Rh [87], among others. Dumesic and co-workers have recently applied computational methods to explain differences among Au, Pt, and Cu catalysts [88]. Roberts et al. have also shown that the reverse water gas shift reaction can limit the apparent CO conversion, and that external transport limitations can be significant [89].

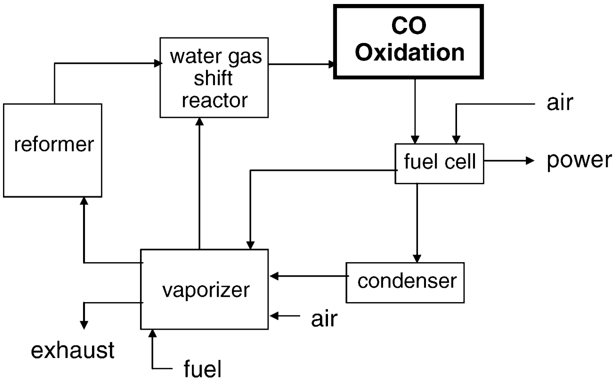


Fig. 12. Process flow diagram of a fuel processor for liquid fuels.

Promoters have been found to significantly improve selectivity for CO oxidation versus oxidation of hydrogen. For example, the Roberts group [90], Goodwin and colleagues [91], and Farrauto and co-workers [92] have shown that Fe-promoted Pt catalysts have substantially higher activity and selectivity than Pt alone. Similar results have been shown for Fe promotion of Au [93].

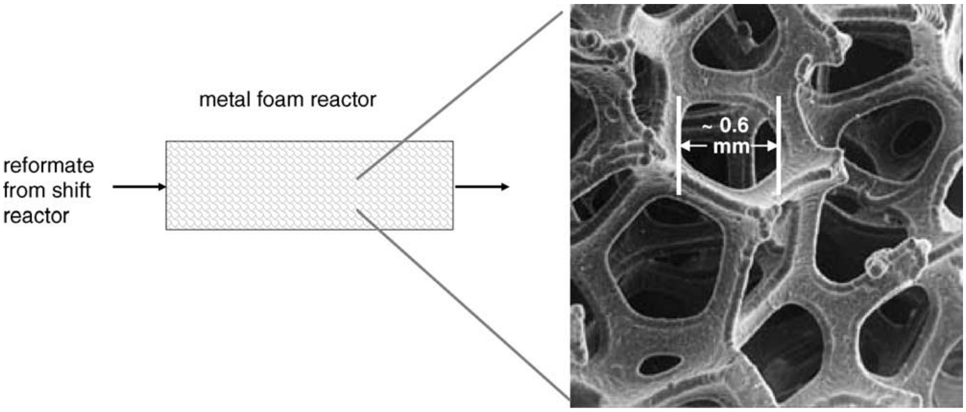


Fig. 13. Morphology of metal foams; these materials have been used for CO oxidation in fuel processors (Ref. [90]).

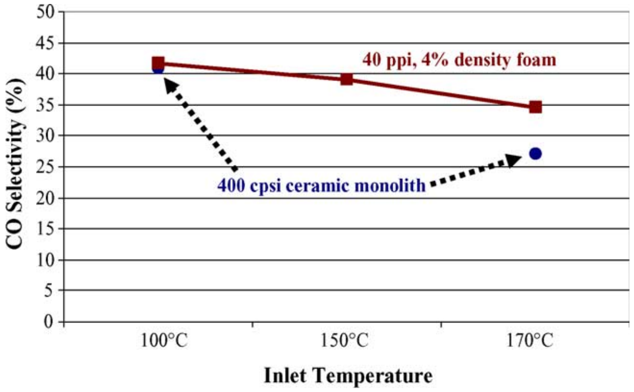


Fig. 14. CO selectivity as a function of reactor inlet temperature, for catalytic metal foams and a directly comparable conventional straight-channel ceramic monolith (Ref. [90]).

1.5.2.2. Metal foams. Metal foams are an example of a novel reactor design being studied as alternatives to conventional straight-channel monoliths for the PROX reaction [90]. These foams offer the potential advantages of radial heat and mass transfer (which is not possible in straight-channel monoliths), and integrated cooling by metallurgical bonding of the foam to cooling tubes. Fig. 13 shows the morphology of these foams, which typically have void volumes of 85–95%, and 20–40 pores per inch (ppi). Fig. 14 shows that these materials compare favorably with straight-channel monoliths at nominally identical catalyst loadings, especially at higher inlet temperatures where thermal gradients may adversely affect selectivity [90,94].

1.6. Approach III: "Renewable" energy: usable Btu without consuming non-renewable fossil fuel

Worldwide, renewable energy sources (including wood as well as other materials), account for about 19% of total energy usage [73]. In the US, the corresponding value is roughly 11%, with biomass/solid waste and hydro power accounting for most of this total (Fig. 15) [95]. Despite the potential of biomass gasification as a source of synthesis gas for production of clean fuels, 98% of all energy from biomass is currently produced by direct combustion [96]. There is a clear incentive, and potential, for the use of gasification and other thermal methods (e.g., pyrolysis) to convert biomass to synthesis gas and finally to clean fuels. Large scale plants, ranging from 15 to 70 $\mathrm{MW_{th}}$ are being developed in Europe [97]. Catalysts will play an important role in the development of these processes. Some examples of the challenges facing catalysis are provided below for gasification-based processes.

1.6.1. Example – biomass conversion to clean fuels and energy

1.6.1.1. Catalytic gasification of biomass. In addition to traditional non-catalytic gasification processes, catalysts can be used in the gasifier itself to lower the operating temperature, making the process more economically feasible. Typically, temperatures above roughly 750 °C at a minimum are required for non-catalytic gasification [98,99]. Although these temperatures increase the overall reaction rate and reduce tar formation [100], the high energy consumption needed to operate at these temperatures adversely affects the economics. Gasification catalysts can lower the operating temperature to around 600 °C [101], resulting in substantial savings. A common gasification catalyst that has been widely investigated is dolomite [102,103], an inexpensive carbonate [$\mathrm{CaMg(CO_3)_2}$]. However, rapid deactivation by tar formation at the lower temperatures of catalytic gasification [104] has led to the search for more tar-resistant, but still active, gasification catalysts. Ni-based catalysts are more active than dolomite and have been widely studied [105,106]. However, these catalysts are usually based on steam reforming formulations [107], and have not been optimized for the catalytic gasification reactions. They also suffer from deactivation by tars [104,108], $\mathrm{NiAl_2O_4}$ spinel seems to be less susceptible to deactivation, while retaining reasonable activity [109]. Among new materials that are being tested, Rh-ceria has recently shown promising results [101,107,110], but these materials are likely to be too expensive for large-scale use.

Thus, one of the challenges for catalysis in the conversion of biomass to clean fuels is the development of cost-effective gasification catalysts that are resistant to deactivation, and which will help improve the overall economics.

1.6.1.2. Gas cleanup. Although low in sulfur compared to coal, biomass-derived syngas may contain up to several hundred ppm sulfur, depending on the source [111,112]. Typically, sulfur must be removed to levels approaching 1 ppm upstream of catalysts used for fuel synthesis [113,114]. In addition, removal of tars, alkali, chlorine, ammonia, and particulate are required for virtually all downstream conversion processes [115–119]. One recent study suggests that the direct adaptation of current gas cleaning systems to the cleanup of biomass-derived syngas will not be sufficient to remove contaminants to desired levels [120]. In this study, catalytic tar removal was found to be the critical step.

As an example of the research needs in this area, catalysts for the decomposition of tars downstream of the gasifier (as opposed to in-bed tar removal, as discussed above in connection with catalytic gasification) are typically based on Ni catalysts, which operate at 700–900 °C [118]. These catalysts are subject to deactivation, but there are relatively few systematic studies of the deactivation process, or how to regenerate the catalyst, particularly in-situ. There are significant differences in reactivity among the various classes of compounds that are collectively classified as tars [121,122], suggesting that multi-component catalysts may be needed to maximize tar decomposition rates while minimizing deactivation. Perhaps periodic regeneration using steam [121] or hydrogen could be used lengthen

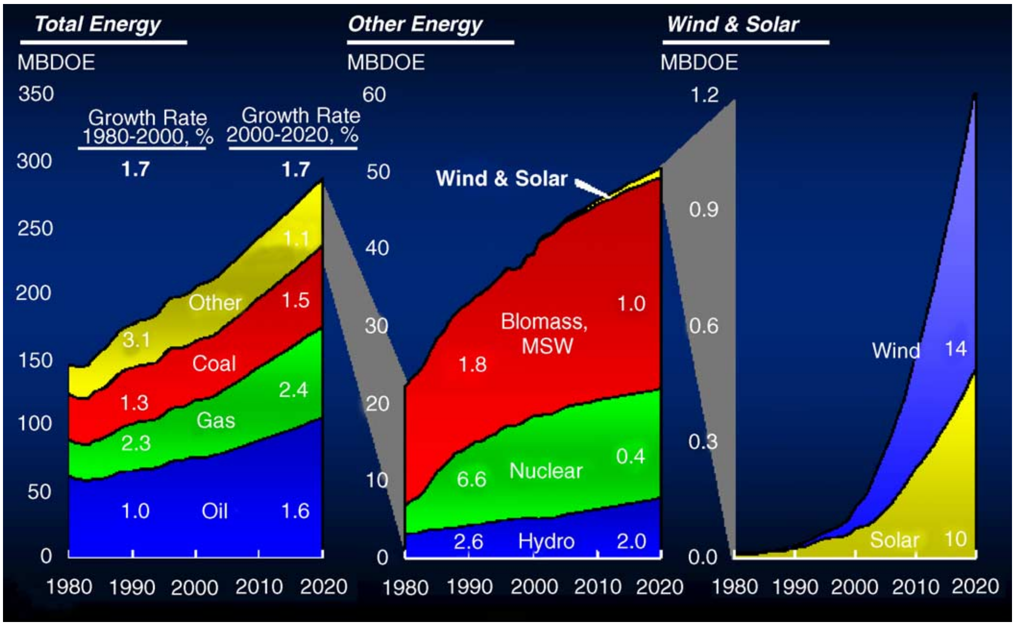


Fig. 15. Total energy use in the US by source, 1980–2020 (Ref. [95]).

the catalyst life. Some novel approaches have been reported, such as the use of pulse corona discharge [123], but there is still substantial room for innovation.

One objective of catalysis research in gas cleanup is to intensify the processes/materials required to remove all the contaminants, simplifying them into as few continuous operations as possible. Examples include combining alkali and particulate removal [116], and catalytic tar and ammonia decomposition on a particulate-resistant support [124,125]. The end products of the overall gas cleanup process should be useful products, if possible. For example, Harrison et al. have shown that partially reduced ceria can be used at 500–700 °C for removal of H_2S to levels below 20 ppm, and regenerated using SO_2 to produce elemental sulfur, the most useful end product of such a process [126,127].

1.6.1.3. Catalytic conversion of biomass-derived syngas to clean fuels. In principle, the cleaned syngas from biomass gasification can be converted into clean fuels using well-established technologies, such as Fischer–Tropsch synthesis [128–131] higher alcohol synthesis, or other syngas conversion processes. It can also be used as a high-Btu gaseous fuel [132], e.g., for catalytic combustion in gas turbines [133] or other combustion processes. Challenges that remain for these technologies include novel catalytic reactor designs tailored to the typically smaller scale of biomass conversion processes [134], improved attrition resistance for Fischer–Tropsch catalysts [135,136], catalysts for downstream adjustment of the H_2/CO ratio for specific end products [137], catalytic reforming of biomass-derived liquids (e.g., from pyrolysis) [138], and even production of hydrogen-rich gases for subsequent hydrogen-powered energy devices [138].

2. Summary

Catalysis will play a key role in the continuing development of clean energy conversion processes, which is essential to our standard of living. Challenges include minimizing emissions from these processes, improving energy conversion efficiencies, and developing viable means of converting biomass and other renewable resources into clean energy. There is substantial room for innovation in these areas: process intensification to couple unit operations into more efficient steps, understanding and minimizing catalyst deactivation, limiting emissions, producing hydrogen and ultra-clean fuels for transportation and using all of this knowledge to develop economically viable, sustainable energy conversion processes.

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