



Catalytic nanoarchitectonics for environmentally compatible energy generation

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Environmentally compatible energy management is one of the biggest challenges of the 21st century. Low-temperature conversion of chemical to electrical energy is of particular importance to minimize the impact to the environment while sustaining the consumptive economy. In this review, we shed light on one of the most versatile energy-conversion technologies: heterogeneous catalysts. We establish the integrity of structural tailoring in heterogeneous catalysts at different scales in the context of an emerging paradigm in materials science: *catalytic nanoarchitectonics*. Fundamental backgrounds of energy-conversion catalysis are first provided together with a perspective through state-of-the-art energy-conversion catalysis including catalytic exhaust remediation, fuel-cell electrocatalysis and photosynthesis of solar fuels. Finally, the future evolution of catalytic nanoarchitectonics is overviewed: possible combinations of heterogeneous catalysts, organic molecules and even enzymes to realize reaction-selective, highly efficient and long-life energy conversion technologies which will meet the challenge we face.

Introduction

Since the industrial revolution in the 18th century, we have been pursuing technologies to produce food and energy for daily life. Looking back through the 20th century, there emerged two groundbreaking technologies which have enabled the vast growth of human activity in terms of population and economy: the mass-production of fertilizer from nitrogen in air (the Haber-Bosch (HB) process) and energy-generation by combustion systems. Currently, the world's annual demand for fertilizer is industrially manufactured through the HB process [1]. Most transportation, such as aircrafts, cars and ships, are powered by combustion systems including gasoline and/or diesel engines, and moreover, 70% of the world's electricity demand is supplied by fossil-fuel power plants [2].

These traditional energy-conversion technologies are strongly related to an elemental technology controlling chemical reactions on demand: *catalysis*. The rate-determining reaction of the HB process, $N_2 + 3H_2 \rightarrow 2NH_3$, is an uphill reaction which goes forward only in the presence of iron-oxide catalysts [1]. Combustion systems

are always equipped with exhaust-remediation catalysts comprising precious-group metals (PGMs) which remediate the toxic exhaust at much lower temperatures than the combustion temperature of fuels [3,4].

To maximize the energy efficiency and minimize the materials cost, huge effort has been expended to tailor the structure of catalysts. In particular, exhaust-remediation catalysts (Fig. 1(a) and (b)) have experienced generations of development to meet the harsh catalytic conditions in exhaust: extremely large space-to-volume ratio ($SV > 10,000 \text{ s}^{-1}$) of the reactant gas and high temperatures often exceeding $1000 \text{ }^\circ\text{C}$ [3–5].

The development of exhaust-remediation catalysts has been based on structure tailoring of the material at different scales. First, the exhaust catalysts are shaped as a co-axial filter with narrow channels to minimize the turbulent flows which strongly inhibit mass transport (Fig. 1(c)) [4]. The usual channel size is of the order of 1000 to $100 \mu\text{m}$ where the Reynolds number is smaller than the turbulent-flow limit, 2200 [6]. In some cases, the channels are designed such that the exhaust repeatedly circulates through the channel to increase the retention time and promote sluggish remediation of burn-resistant particulate matter (PM25) [5].

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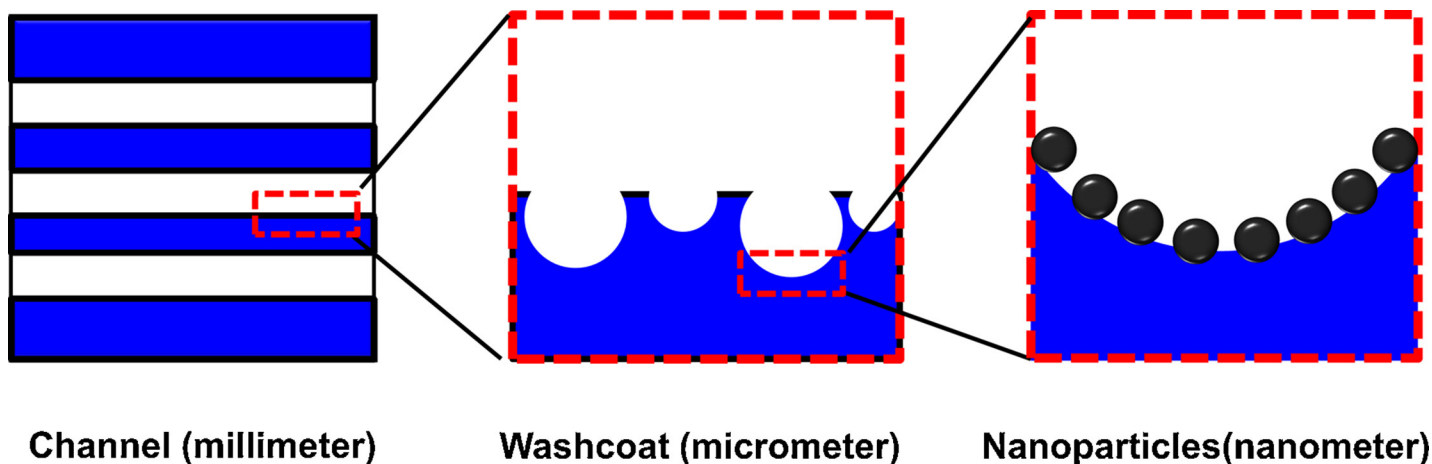


FIGURE 1

Architecture of exhaust catalysts, ranging from milli, micro and nanometers.

The channel surface of catalysts is further coated with porous layers (usually called washcoats) to increase the surface area up to $0.1\text{--}10\text{ m}^2\text{ g}^{-1}$ (Fig. 1(d)) [4]. The pore size is adjusted to the mean free path of the molecular diffusion of the reactant gas; 10 to $0.1\text{ }\mu\text{m}$ to improve the mass transfer [6]. Finally, nanometer-sized catalytic centers are dispersed and immobilized on the surface of the pores to maximize the catalytic activity (Fig. 1(d)). The catalytic centers usually consist of 1000 through 10,000 atoms [3,4]. Here it is worth noting that such structural control of *individual* components often provides incremental improvements in the overall performance. In order to realize efficient exhaust remediation, the components must not only be carefully structure-tailored, but also rationally combined in an *integrated* way from millimeter through nanometer levels, just like the construction of elaborate architectures.

Several years ago, a new concept emerged in the field of materials science called *nanoarchitectonics*. This novel concept of nanoarchitectonics was originally proposed by Masakazu Aono as a new paradigm to integrally construct required material architectures by arranging at the nanoscale [7,8]. Although nanotechnology mainly utilizes individual techniques at the nanoscale, nanoarchitectonics harmonizes mutual interactions and components to build up hyperfine structures, as architectures with nanoscale building blocks. Atomic and molecular-level manipulations, chemical functionalization, control of self-organization and physical field-induced orientational control should be mutually combined to create new materials with the desired nanoarrangement. This concept has recently been applied in many research categories including materials preparation [9–11], physicochemical fabrication [12–14], and biomedical applications [15–17]. For catalytic nanoarchitectonics, we particularly focus on tailoring the molecular composition, surface-atomic arrangement, and microstructures for innovative catalysts.

Backgrounds: problems and requirements

Challenges for heat-energy conversion

Combustion engines extract available energy from fuels including hydrogen, fossil fuels and/or biomass fuels, through thermo-mechanical converters operating at temperatures higher than the ignition points of fuels in air ($>400\text{ }^\circ\text{C}$). The combustion is always

accompanied by exhaust containing carbon monoxide and toxic nitrogen oxides, NO_x [3]. The exhaust causes severe impacts on living conditions especially in emerging countries where ground transportation is rapidly increasing [18,19].

In addition, the efficiency of such thermo-mechanical converters is always limited by the operating temperature. When the converters work at temperatures lower than the usual heatproof limit ($<1000\text{ }^\circ\text{C}$), the efficiency is lower than 40% [20]. The unused chemical energy, which was stored in the fuels, is irreversibly dissipated to the environment as heat loss. This heat loss not only degrades the fuel-to-energy efficiency and promotes the depletion of fossil fuels, but also contributes to global warming through a synergy effect with green-house gases contained in the exhaust, such as methane (CH_4) and nitrous oxide (N_2O) [11].

Different alternatives have been developed, including nuclear power plants and solar power plants, yet none of these power sources has replaced combustion engines, primarily because of the scalability [21]. It is worth noting that even nuclear power plants are not free from heat loss because they still rely on thermo-mechanical converters to extract energy from fission fuels.

Catalytic energy conversion at low temperatures

There is a possible way to reduce this unfavorable heat loss and exhaust: energy-extraction at atmospheric temperature. NO_x is generated through the high-temperature reaction of O_2 and N_2 in the oxidant [3]. Heat loss occurs only at the interface of two objects with different temperatures. When the energy-extraction is performed at atmospheric temperature, both the NO_x generation and heat loss will be substantially suppressed. However, such atmospheric-temperature energy-extraction is insufficient to supply the energy demands of practical machinery such as cars or electronic devices because of sluggish fuel-oxidation at low temperatures.

To accelerate low-temperature energy-extraction from fuels, the most versatile way is to activate the fuel molecules by promoting the formation of chemical bonds with another catalyst molecule which is *not* directly involved in the reaction. When the fuel molecule forms chemical bonds with the catalyst molecule, the electrons used for the chemical bonds of the fuel molecule partially move to the newly formed bonds, resulting in a weakening of

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