Applied Catalysis B: Environmental 221 (2018) 530–555

Contents lists available at ScienceDirect

Applied Catalysis B: Environmental

journal homepage: www.elsevier.com/locate/apcatb

Review

Current status, opportunities and challenges in catalytic and photocatalytic applications of aerogels: Environmental protection aspects

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ARTICLE INFO

Article history:
Received 15 May 2017
Received in revised form 22 July 2017
Accepted 2 August 2017
Available online 25 August 2017

Keywords:
Aerogel catalyst
Aerogel photocatalyst
Environmental remediation
Air cleaning
Water cleaning

ABSTRACT

Aerogels are an exceptional class of materials which are of interest for several high-performance applications thanks to their extraordinary physical properties such as extremely high porosity, high specific surface area, and extremely low density combined with very versatile synthesis approaches. Since their invention by S. Kistler, various aerogels have been explored for catalytic and photocatalytic applications, though in the recent decades, several breakthroughs regarding different aspects ranging from more efficient catalysis in organic synthesis, energy-related processes, and catalytic environmental depollution by aerogels have been made. For both, catalytic and photocatalytic performances, aerogel monoliths prepared from sol–gel approaches accompanied with an appropriate drying technique, in particular supercritical point drying, have become striking alternative catalysts or catalyst supports compared to the presently used ones prepared from traditional wet synthesis approaches. In this article, aerogels in environmental remediation processes as heterogeneous catalyst and photocatalyst for depollution of air and aqueous media are fully addressed, and recent achievement in this context is thoroughly reviewed.

Contents

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http://dx.doi.org/10.1016/j.apcatb.2017.08.012
0926-3373/

1. Introduction

Aerogels are sol–gel derived porous materials with extraordinary properties, such as a high porosity, extremely low density, enormous active surface area and very low thermal conductivity [1]. The unique properties of aerogels originate from the combination of the specific properties of nanomaterials magnified by their macroscale assembly making aerogels attractive materials for several applications ranging from thermal and acoustic insulation, and catalysis to biomedical and pharmaceutical applications [2–4]. Among high-performance applications, *e.g.* as thermal insulators [4,5], aerogels have been recognized as catalysts or catalyst supports in various reactions [6,7].

Historically, the first aerogels were introduced by Kistler in the 1930s [8] by applying the supercritical drying method for the first time. However, in the following years, due to the progress in precursor chemistry and drying methods, various aerogels having organic as well as inorganic or even hybrid building blocks emerged [9]. Basically, irrespective of the building blocks and components, aerogels refer to solid materials having a three-dimensional porous network with interconnected micro/mesopores obtained by drying technologies that are able to conserve the initial wet gel structure [1].

Following the major interest of chemical industry in designing innovative materials as a solid catalyst, in recent years, major interest has been given to tailor-made aerogels as heterogeneous catalysts or catalyst supports for various reactions. In fact, Kistler [10] was a pioneer to recognize the catalytic applications of aerogels which were later pursued by several scientists, mainly Teichner, Baiker, Pajonk, and other groups [11–16]. In the 1990s, a series of reviews addressing the different gas and liquid phase catalytic aspects of aerogels particularly with the prospect of environmental protections were published by the same scientists [6,7]. So far, aerogels and their composites have been employed in both liquid–solid, and gas–solid catalyzed reactions in several domains, such as environmental protection, organic synthesis including partial oxidation, epoxidation, nitroxidation, hydrogenation [16,17] as well as energy-related applications [14,15,18–20]. Aerogels were also considered as solid biocatalysts for specific biochemical syntheses or as an active component of biosensors [21–24].

Since the support is playing a pivotal role in catalysts design, the use of materials with deliberately tailored properties is highly desirable. The suitability of aerogels as a catalyst support is related to their fascinating textural and structural properties in combination with the versatility of the synthesis route, *e.g.* the sol–gel technique, which gives flexibility of controlling the texture, composition, homogeneity and structural features of solids from a molecular level [12,16,20]. Additionally, the sol–gel technique opens up new possibilities to tailor the materials properties through the addition of different components, *i.e.* dispersion of the various oxides or metals in an aerogel matrix, to design and tailor the chemistry of aerogel toward the particular catalytic reactions [20].

Catalysts are essential components for the treatment of air and water pollutants on the way to a sustainable and clean environment [25]. Aerogels from various molecular precursors are recognized as active heterogeneous catalysts for several catalytic and photocatalytic environmental remediation purposes [6,26,27].

To preserve the air quality, control of the emission of hazardous volatile organic compounds (VOCs), nitrogen oxides (NOx; NO, NO_2, N_2O, *etc.*), carbon monoxide (CO), greenhouse gases *i.e.* methane (CH_4) and carbon dioxide (CO_2), *etc.* being released from transportation, municipal and industrial sectors and combustion is necessary.

Nitrogen oxides, NOx gases are generated from the reaction of nitrogen, oxygen, and hydrocarbons at high temperatures. The primary sources of NOx emission are traffic, industry, refineries and power plants [28,29]. Catalytic conversion of NOx into less harmful gases, such as N_2 and H_2O, is quite a matured field, and several review papers exist covering this topic [30–32]. The selective catalytic reduction (SCR) of NOx with various reductants such as NH_3, hydrocarbons, CO and soot particles over various catalysts and catalyst supports are reported for NOx abatement, so far [33]. However, ammonia or aqueous solutions of ammonia have been the main reductants used in the selective reduction of NOx, especially in the gas turbines, power plants, and waste incinerators [33]. In this contribution, we specifically focus on the SCR of NOx using NH_3 as a reductant by exploiting vanadium supported various metal oxides or carbon aerogel catalysts.

Aerogels are also recognized as suitable candidates for the adsorption/absorption as well as catalytic combustion of VOC vapors from effluents due to their tunable textural *e.g.* high specific surface area and porosity and structural properties *e.g.* density and monolithic structures as well as tunable surface chemistry [34]. In this paper, combustion catalysis of various VOCs using noble metal catalysts such as Pt, Pd, *etc.* supported carbon aerogels and transition metal oxides are studied [26].

Aerogels are also attractive for CO removal from an enriched hydrogen stream which is produced on board vehicles through different reforming and fuel oxidation processes [127] – hydrogen being an ideal fuel for proton-exchange membrane fuel cells (PEMFC). The PEMFC has drawn significant attention due to its low operation temperature, high efficiency, high power density and its environmentally benign exhaust gases [126]. However, during hydrogen production, a significant amount of CO is produced that causes a serious poisoning of the anode catalyst (*e.g.* platinum or palladium) of the PEMFC. Therefore, it is necessary to remove CO from the hydrogen stream with a minimum loss of hydrogen [128]. So far, several strategies for CO removal have been proposed [122], among them the preferential oxidation of CO (CO-PROX) as the most effective catalytic process [129,130]. In this regard, inorganic aerogels based on noble metals as well as various bimetallic compounds supported on inorganic aerogels have shown better promises for CO-PROX compared to the conventional catalysts.

Aerogel contribution for methane reforming with CO_2 is also discussed from the perspective of environmental protection aspects, even though this process is more known for the production of clean H_2 energy and synthesis gases (CO/H_2) as a feedstock for Fischer–Tropsch synthesis of hydrocarbons. Noble metals, transition metals, and their bi-metallic doped metal oxide aerogels have been exploited to catalyze the methane reforming process. Beside catalyst preparation methods, structural parameters, coke resistivity and operational conditions like the type of the reactor are extremely influential in the efficiency of the process that would be elaborated in this review paper.

Photocatalyst technology using semiconductors, mainly TiO_2 or TiO_2 based composites, is another effective way to deal with the problems concerning pollution abatement in gaseous environments [35]. The ease of implementation in commercial settings as well as low energy requirement that uses the sun or artificial light as energy sources and the applicability to a wide range of indoor and outdoor pollutants (*i.e.* NOx, VOC, bacteria, airborne particles, *etc.*) make this process an attractive choice in the degradation of contaminants [36,37]. However, traditional methods of sample preparation for photocatalytic reactions using powder form samples are not efficient enough with respect to catalyst regeneration, and consequently, from an economic and recycling standpoint [38]. Studies have shown that the preparation of photocatalysts in a certain geometry with the high active surface area for instance in two – (*e.g.* films) or three-dimensional structures (*e.g.* aerogels) for loading of the large amount of photoactive phases without any loss in catalyst during the process is becoming a compelling and critical