Journal of Sol-Gel Science and Technology (2023) 106:406–421
https://doi.org/10.1007/s10971-022-05750-y

ORIGINAL PAPER: INDUSTRIAL AND TECHNOLOGICAL APPLICATIONS OF SOL-GEL AND HYBRID MATERIALS

Sol–gel research in China: a brief history and recent research trends in synthesis of sol–gel derived materials and their applications

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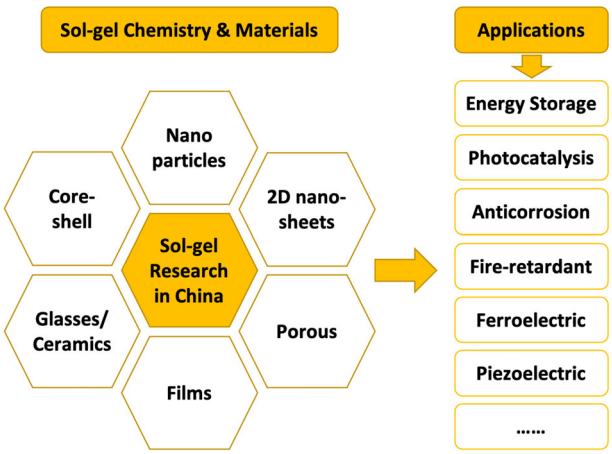
Received: 17 May 2021 / Accepted: 18 February 2022 / Published online: 11 March 2022

Abstract

The sol–gel process has become a rapidly growing research area in materials science. A variety of materials prepared via sol–gel routes have shown unique properties and characteristics that are difficult to achieve using conventional methods. In recent years, tremendous progress in sol–gel R&D has been made not only in the world but also in China. Here, this review provides a retrospective overview of the sol–gel history in China and summarizes recent progress and applications of sol–gel research in Chinese universities, institutes, and industries. It highlights some of the recent developments published by Chinese researchers in the last 5 years, ranging from the sol–gel synthesis of nanomaterials, bulk materials, and functional coatings, to their applications in the fields of energy conversion, energy storage, photocatalysis, etc. It is evident that sol–gel technology nowadays in China has evolved into a vibrant research area both in academia and industry.

Graphical abstract



Keywords Sol–gel · China · Historical notes · Nanomaterials · Functional coatings

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Highlights

- A brief overview of sol–gel history in China is given.
- Some of the recent developments of fundamental sol–gel research in China are highlighted.
- The commercialization of sol–gel technology in China is introduced.

1 A brief history of sol–gel research in China

Since the First International Workshop on Glasses and Glass Ceramics from Gels was held in Padova, Italy in 1981, sol–gel technology has become an emergent research focus for scientists and researchers from all over the world [1, 2]. Through reactions such as hydrolysis and polymerization of highly reactive precursors, multiple forms of materials, including bulk, powder, fiber, thin film, and monolith, can be produced combined with other processing procedures such as surface modification, self-assembly and phase separation [3]. As one of the most versatile methods for synthesis of a variety of materials, the sol–gel process enables precise control over their physical and chemical properties, creating materials with high purity, excellent homogeneity, controllable morphology, and unique microstructures [4–7].

The rapid development of sol–gel technology in the 1980s had attracted much attention from the scientific communities in China as well. Keeping pace with other parts of the world, many researchers from prestigious universities and research institutes in China began their research endeavor on developing new sol–gel-derived synthesis methods and materials. Research groups from organizations such as Zhejiang University, Shanghai Institute of Ceramics-Chinese Academy of Sciences (CAS), Institute of Optics and Electronics-CAS, and China Building Materials Academy made early contributions of sol–gel related research in China.

For the sol–gel research communities in China, the First National Sol–gel Science and Technology Conference held in Hangzhou, Zhejiang province in 1990 is perceived as the event that signified the beginning of China's sol–gel era. In the years following this conference, especially during the twenty-first century, considerable progress and developments both in fundamental research and practical applications of sol–gel technology has been achieved in China as in other major countries of the world [8]. From 2000 onwards, there has been a rapidly growing number of sol–gel-related scientific papers published in China, mainly by universities and research institutes. Figure 1 reveals that the scientific research papers containing "sol–gel" in the title/abstract/keyword (source: Web of Science, patents not included) in the last 10 years (2011–2020) has reached a staggering number of 73,216, out of which 27,252 were contributed by research organizations in China, accounting for 37.22% of the total production of sol–gel-related scientific papers (Fig. 3). Researchers in China also contributed 36.02% of the total citations of sol–gel-related papers from 2011–2020 (Figs. 2 and 3).

Table 1 summarizes the numbers of publications contributed by different research organizations in China, which shows that the CAS, University of Chinese Academy of Sciences, Tianjin University, Zhejiang University, and Jilin University were the most active research organizations in terms of the total number of sol–gel papers published in the last 10 years.

As mentioned above, the First Chinese National Sol–gel Science and Technology Conference was organized in Hangzhou in 1990. This conference attracted 30 participants

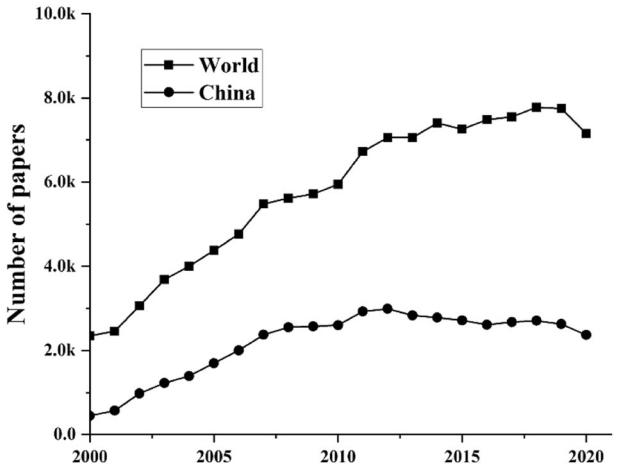

Fig. 1 Number of scientific papers containing "sol–gel" in title/abstract/keyword published per year in China and the world from 2000 to 2020

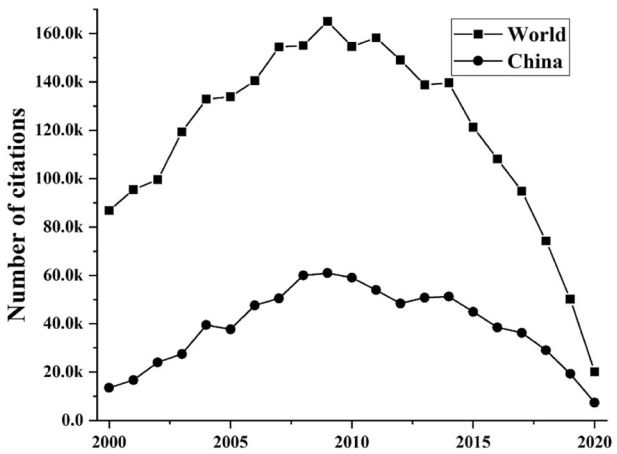

Fig. 2 Number of citations for papers in China and the world

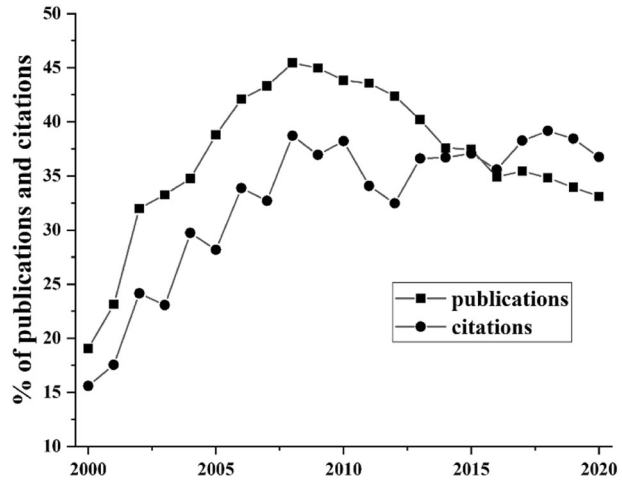

Fig. 3 Percentage of publications and citations in China over those in the world

Table 1 List of universities and research institutes in China that published the greatest number of publications from 2011 to 2020 (source: Web of Science)

Name of organization	Number of publications
Chinese Academy of Sciences	2434
University of Chinese Academy of Sciences	622
Tianjin University	519
Zhejiang University	506
Jilin University	475
Tsinghua University	462
Harbin Institute of Technology	440
Tongji University	434
South China University of Technology	394
Wuhan University of Technology	367

and over 20 papers were presented. Despite the limited number of attendees, this conference was the first national academic event focusing only on the topic of sol–gel technology. Years later, a second national conference was held in Dalian in 1997, and a third one was held again in Hangzhou in 2004. In 2006, this conference was hosted again by Zhejiang University in Hangzhou, and was renamed the Sol–Gel Symposium of China and International Forum. Since then, the Sol–Gel Symposium of China and International Forum started to be held regularly in different cities in China: Wenzhou (2006), Shenzhen (2008), Kunming (2014), Changsha (2016), and Xi'an (2018). Today, this biannual conference is widely recognized as the leading event in the sol–gel community in China. The latest conference held in Xi'an in October 2018 attracted more than 280 attendees from over 10 countries and 73 organizations. Four keynote speeches, 21 invited talks, 74 oral presentations, and 85 posters were presented at the conference on various topics including fundamentals of sol–gel chemistry, nanomaterials and nanostructures, nanostructured porous materials, functional new materials, etc. Despite the Shandong University-hosted conference originally scheduled for 2020 being postponed over the COVID-19 pandemic, it remains the most active sol–gel event in China, and constantly attracts increasing attention from both academia and industry in China.

To meet the continuously increasing needs for productive activities and social connections from the sol–gel community in China, the Chinese Ceramic Society initiated the establishment of the Sol–Gel Committee in 2008, and Professor Hui Yang has been acting as the Chair of the Committee since then. Promoted by the members of the Chinese Ceramic Society and Zhejiang University, the primary goal of this committee is to facilitate the Sol–Gel research and industrialization of sol–gel-based applications in China.

In August 2011, the 16th International Sol–gel Conference was successfully held in Hangzhou, China [9]. It was the first time that such an important and world-leading event in the field of sol–gel science and technology took place in China. With joint effort from the International Sol–gel Society and local organizing committee members from the Chinese Ceramic Society, Zhejiang University, and other organizations, this conference attracted 515 delegates from 39 countries. Overall, 3 keynote speeches, 3 award lectures, 16 invited talks, 116 oral lectures, and 302 posters covering 12 different topics were presented at the conference. This conference also underlined a shift of emphasis of sol–gel research from fundamentals to industrialization, indicating a change in research trends both in China and in the world.

2 Some highlights of recent sol–gel research in China

As discussed in previous section, there has been an explosion of publications in fundamental sol–gel research in recent years. The research interests extended from sol–gel synthesis and formation mechanism of functional materials to a wide spectrum of their applications. In this section, we provide a retrospective overview on some studies recently published in peer-reviewed scientific journals by Chinese researchers and scientists in the last 5 years. These publications were included in this review mostly because of the utilization and modulation of sol–gel processing for producing a variety of advanced materials with new functionalities or enhanced properties. It is also worth noting that this review only mentions a part of the substantial number of publications featuring sol–gel researchers from Chinese

organizations, and covers not only the most highly cited papers published in high-impact journals by Chinese researchers but different active areas of sol–gel research in China. Although this is not an exhaustive and detailed review that comments on every sol–gel research area and presents the totality of sol–gel research activities currently underway in China, it still gives a broad picture of the recent developments and progress made by sol–gel communities in China. This review will cover three major forms of sol–gel-derived materials, namely nanostructured materials, bulk materials, and functional coatings. It also discusses their applications in the fields of energy conversion, energy storage, photocatalyst, etc. By highlighting some of these outstanding contributions, we hope that this review will present an overview of the comprehensiveness and importance of China's sol–gel research in recent years.

2.1 Fabrication of nanostructured materials via sol–gel process

Materials with well-defined nanostructures have drawn great attention in the field of materials science owing to their unique physical and chemical properties that are distinctly different from those in conventional bulk forms. As an important synthesis method for nanostructured materials, solution-based processing techniques have been extensively explored, including co-precipitation, hydrothermal methods, solvothermal methods and sol–gel methods. Among these techniques, sol–gel-derived synthesis methods have proven to be particularly effective in the fabrication of nanostructured materials with uniform size distribution, specified morphology, and controllable chemical stoichiometry, thereby enabling the precise manipulation of their physical and chemical properties [8, 10, 11]. Thanks to these advantages, nanomaterials prepared through sol–gel approaches are widely explored in various areas such as electroceramics, microwave ceramics, electrical contact materials, energy conversion and storage materials, photocatalysts, etc. In this section, several studies on the sol–gel synthesis of nanomaterials in the forms of nanoparticles, 2D nanomaterials, and nanomaterials with core-shell structures are introduced, and their applications in energy conversion and storages as well as photocatalysts are further discussed.

2.1.1 Sol–gel-derived nanomaterials for energy conversion and storage applications

In recent years, the constantly growing demands for alternative and clean energy sources have triggered tremendous interest in energy conversion and storage research, promoting the developments of new electrochemical energy devices, such as lithium-/sodium-ion batteries, supercapacitors, and fuel cells. Seeking suitable electrode materials with superior electrochemical properties, high energy density, good cycling stability and high safety has always been an active topic of energy conversion and storage research [12, 13]. In this regard, many research groups in China have explored the use of sol–gel methods for preparation of alternative electrode materials [14–21]. The versatility of sol–gel processing allows precise designs of nanostructures with desired electrochemical properties for energy storage applications. For example, with great promise as the alternative to deliver better specific energy for lithium-ion batteries, lithium-sulfur batteries still suffer from many drawbacks such as poor ionic and electronic conductivity of S and Li_2S. To circumvent such challenges, Tao et al. from the Zhejiang University of Technology employed the one-step Pechini sol–gel method to prepare $Li_7La_3Zr_2O_{12}$ nanoparticle-decorated porous carbon foam (LLZO@C) as a filler in the poly(ethylene oxide) (PEO) electrolyte of a solid-state lithium-ion battery [18]. The LLZO@C sample exhibited a cross-linked hierarchical pore structure decorated with a single nano-crystalline garnet-structured LLZO (Fig. 4). The LLZO nanoparticle acts as an interfacial stabilizer to reduce the interfacial resistance between S and the ion/electron conductive matrix and improve the conductivity of PEO electrolyte. In another study, Luo et al. from Donghua University demonstrated silicon/mesoporous carbon/crystalline TiO_2 core-shell-shell nanoparticles via a two-step sol–gel coating process, which showed excellent lithium storage properties owing to the double-shell design that avoids direct contact of Si with electrolyte and provides better structural rigidity [19].

As an alternative substitute to Li-ion batteries, sodium-ion batteries have emerged due to their low-cost and rich Na abundance. However, finding suitable electrode materials for Na-ion batteries with good electrode kinetics and cycling stability remains a grand challenge. Guo et al. from Nanjing University of Science and Technology presented the sol–gel synthesis of cobalt sulfide (CoS_x) quantum dots embedded N/S-doped carbon nanosheets with two-dimensional heterostructure as electrode material for Na-ion batteries [20]. The ultra-small CoS_x quantum dots were embedded in situ within the ultrafine carbon nanosheets (Fig. 5), which made the heterogenous composites exhibit excellent rate capability and cycling stability as electrode material for high-performance sodium-ion batteries.

Sol–gel processing is also widely explored as a synthesis method to fabricate electrode nanomaterials with enhanced capacitance for supercapacitors. Through the one-step sol–gel process, Zhou et al. from Shanghai Institute of Ceramics-CAS prepared phosphorus/sulfur co-doped porous carbon using resorcinol and furaldehyde as precursors and phosphorus pentasulfide as doping sources. The as-prepared co-doped mesoporous carbon

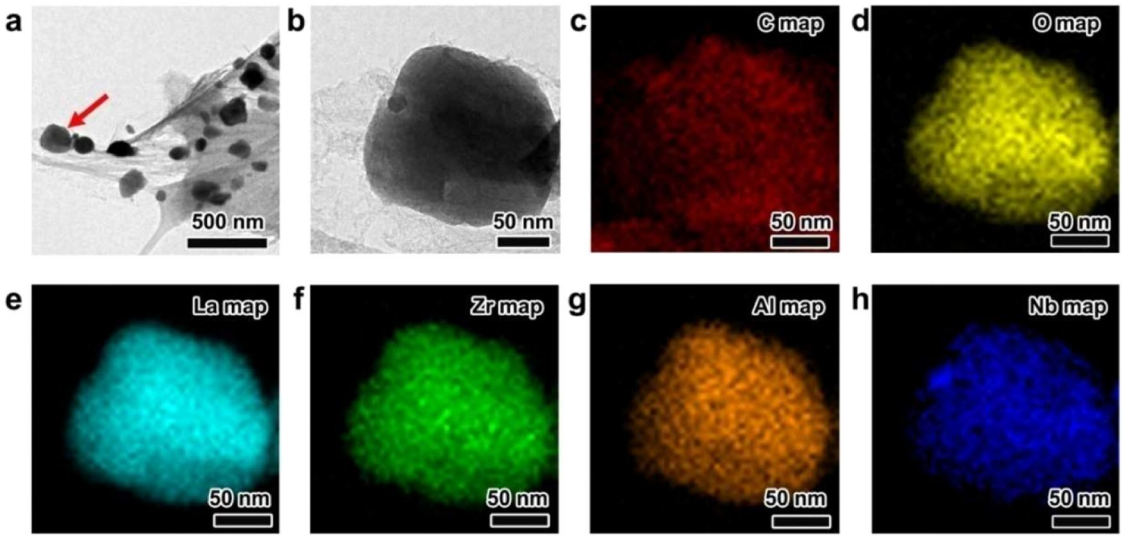

Fig. 4 TEM images and elemental mapping of the cross-linked porous structure and the LLZO nanoparticles. Reprinted with permission from ref. [18]. Copyright © 2017 American Chemical Society

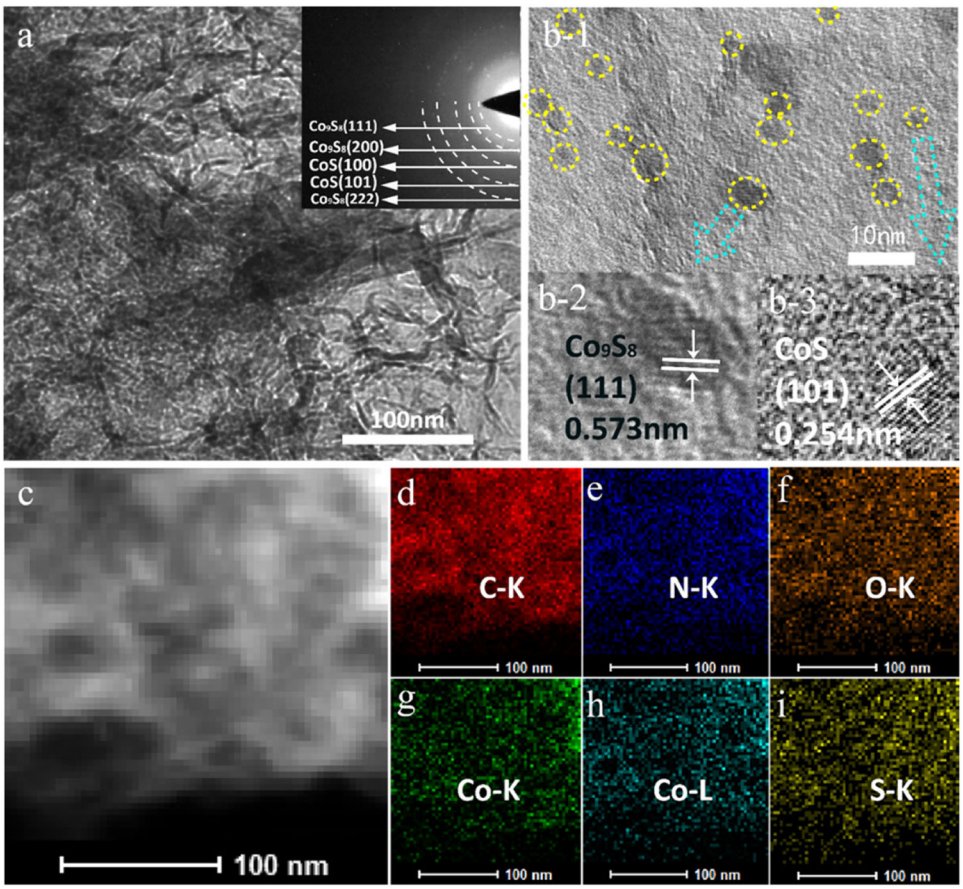

Fig. 5 TEM images (**a**, **b**) and STEM elemental mapping (**c**–**i**) of the CoS_x@NSC composite. Reprinted with permission from ref. [20]. Copyright © 2017 American Chemical Society

materials exhibited high surface area, enlarged pore size, and better graphitization because of the sulfur doping, resulting in the enhanced specific capacitance and catalytic activity for oxygen reduction reactions in supercapacitor applications [21].

2.1.2 Sol–gel derived nanomaterials for photocatalytic applications

Photocatalysis is another active research area of sol–gel-derived nanostructured materials [22–27]. Semiconductor-based

photocatalytic nanomaterials have been widely investigated for applications such as organic pollutant degradation, heavy metal removal, and hydrogen generation [25, 28, 29]. However, the photocatalytic performance of semiconductor photocatalysts may be significantly impaired by the fast recombination of photogenerated electron-hole pairs. One effective strategy to address this issue is the creation of electron- or hole-trapping sites within the semiconductor lattice by doping of transition metal or rare earth elements. It has been discovered that sol–gel processing is a particularly effective route for the synthesis of doped oxide semiconductors due to its refined control over their chemical stoichiometry [30, 31].

To date, the sol–gel process has been extensively adopted for facile synthesis of bismuth ferrite ($BiFeO_3$, or BFO) nanoparticles, a perovskite-type visible-light-driven oxide semiconductor photocatalyst with excellent photovoltaic properties and photocatalytic activity [22–25]. In recent years, the doping of Gd into the BFO lattice has been proven an effective way to enhance its photocatalytic activity owing to the increased ferroelectric domains in Gd-doped BFO [32]. For instance, Zhang et al. from China Jiliang University reported the sol–gel synthesis of Gd-doped $BiFeO_3$ nanoparticles with enhanced visible-light photocatalytic activity for Rhodamine B degradation, which was attributed to the increased optical absorption, enhanced separation and migration efficiency of photogenerated charge carriers, and the decreased electron-hole recombination [23]. In another study, Yang et al. from Xi'an University of Architecture and Technology investigated the photocatalytic activity of Gd^{3+}-doped perovskite BFO nanoparticles prepared via sol–gel processing for water splitting and hydrogen production. Their studies revealed that Gd^{3+} doping enhanced the photocatalytic activity of $BiFeO_3$ owing to the improved efficient generation, separation, and migration of electron-hole pairs, and that the magnetic property of Gd^{3+} doped BFO was enhanced to enable easier collection of the photocatalytic materials in solution [24].

Apart from the $BiFeO_3$-based nanomaterials, titania (TiO_2)-based photocatalytic nanomaterials are intensively studied for their high photocatalytic performance, low-cost and environmental friendliness [33]. To address the drawbacks associated with conventional TiO_2-based photocatalytic nanomaterials such as low quantum efficiency and low electronic properties, researchers have proposed many strategies, among which the creation of heterojunctions between TiO_2 nanoparticles and two-dimensional materials, such as graphene and MoS_2, is considered a promising strategy to enhance the photocatalytic performance of TiO_2 nanoparticles due to the inhibiting effect of electron-hole recombination, fast charge separation, and improved transfer of photogenic electrons. In this regard, Sheng et al. from Jiangsu University of Science and Technology demonstrated the sol–gel-synthesis of hexagonal boron nitride (H-BN) nanosheets/TiO_2 nanocomposites that exhibited excellent photocatalytic properties for the degradation of organic dyes [26]. The nanocomposites showed strong interaction between TiO_2 nanoparticles and h-BN nanosheets resulted from the formation of B-O-Ti bonds within the composites during the sol–gel process, which gave rise to improved charge transportation rate and reduced electron-hole recombination rate, thereby significantly enhancing the photocatalytic property of the nanocomposites compared to pure TiO_2 nanoparticles. In another work conducted by Li et. al from China University of Mining and Technology (Beijing), a mild sol–gel method combined with chemical stripping and self-assembly was employed for the synthesis of a 3D heterogeneous g-C_3N_4/TiO_2/kaolinite composite [27]. Due to the improved light harvesting and the increased separation and transfer efficiency that arose from the "sandwich" structure and chemically bonded interface in this ternary system, the composite exhibited enhanced visible-light absorption-photocatalytic degradation performance for ciprofloxacin removal and higher disinfection ability toward *S. aureus*. Similar to the previously mentioned study, the sol–gel process facilitated the formation of Ti-O-H bonds between TiO_2 nanoparticles and rich hydroxyl-containing kaolinite natural mineral. This strategy of utilizing sol–gel routes to synthesize heterogeneous composites provides new insights into the creation of photocatalysts with enhanced visible-light-driven photocatalytic properties.

2.1.3 Sol–gel synthesis of nanomaterials with core-shell structures

The sol–gel approach has proved to be an especially useful tool for the fabrication of nanoparticles with core-shell structures that exhibit unique properties in areas such as multiferroic materials, fluorescent and optical materials, and electrode materials for energy storage applications [34–36]. In 2017, Yang et al. from Donghua University introduced a facile low-temperature kinetics-controlled sol–gel approach for the preparation of Si nanoparticles encapsulated by an amorphous TiO_2 shell as anode material for Li-ion batteries [37]. To overcome the issues associated with conventional pure silicon-based anodes such as structural degradation and unstable solid electrolyte interphase growth, an outer layer of amorphous TiO_2 shell was coated on the surface of Si nanoparticles (as depicted in Fig. 6), leading to faster ionic mobility and electronic diffusion. The use of the sol–gel approach also presented several advantages such as low reaction temperature without the need for an annealing process and low-cost. As a result, the electrode incorporating core-shell nanoparticles exhibited excellent electrochemical performance and better safety performance owing

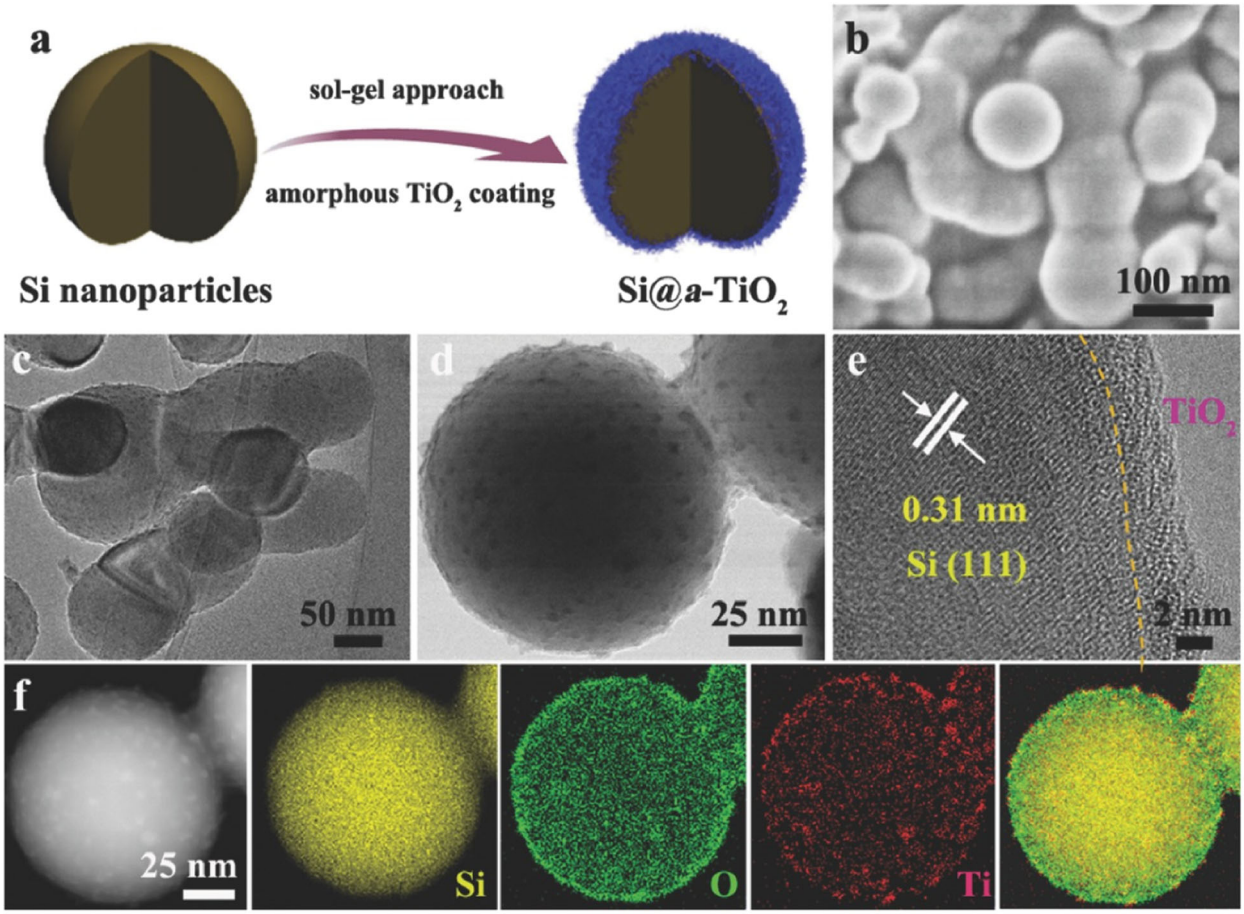

Fig. 6 Schematic illustration of synthetic route for the amorphous TiO_2 shell-coated Si core-shell nanoparticles (**a**), SEM (**b**), and TEM images (**c**–**e**) and STEM elemental mapping (**f**) of the Si@a-TiO_2 nanoparticles. Reprinted with permission from ref. [37]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

to the elastic behavior and higher lithium-ion diffusivity arising from the TiO_2 outer layer.

To solve the same issues with silicon anodes, Li, et al. from the University of Science and Technology Beijing took a similar scalable sol–gel approach to prepare SiO_x-TiO_2@C nanocomposite with watermelon-like structure, in which ultrafine anatase TiO_2 nanoparticles were uniformly distributed inside a carbon shell-coated silicon suboxide (SiO_x) core [38]. This unique structural design allowed faster transport of electrons and lithium ions because of the in situ formation of a Li_xTiO_2 phase from the TiO_2 nanocrystals. The outer carbon shell also facilitated the electronic conduction and the release of mechanical stress inside the Si component. By utilizing the benefits of this composite for lithium-ion battery anode materials, the electrode showed high specific capacity, excellent rate capability, and long-term cycling stability.

Wang et al. from Tsinghua University employed a sol–gel method to synthesize g-C_3N_4@TiO_2 core-shell structured photocatalysts for phenol degradation under visible-light irradiation. The photocatalysts with an increasing number of g-C_3N_4 layers showed higher photocatalytic activity due to the more effective charge separation and electron transfer promoted by the core-shell structure, and the sol–gel synthetic approach provided stronger bonding between the core and shell, resulting in reduced dissolution of the g-C_3N_4 material [39]. Liu et al. from Fudan University used a combined sol–gel/solvothermal/hydrothermal method to synthesize magnetic CoNi alloy-based core-shell microspheres for wideband microwave absorption applications [40]. As illustrated in the schematic of Fig. 7, both CoNi@SiO_2@TiO_2 core-shell-shell and CoNi@Air@TiO_2 yolk-shell microspheres were prepared. These unique structures gave rise to strong magnetic loss by the CoNi cores, excellent dielectric loss by the TiO_2 nanosheet shells, and the in-between air/SiO_2 intermediate layer as the impedance matching mediator, exhibiting significantly enhanced microwave absorption performance.

2.1.4 Sol–gel synthesis of 2D nanosheets

The sol–gel process is also a facile approach for the synthesis of nanostructured inorganic compounds with sheet-like shapes,

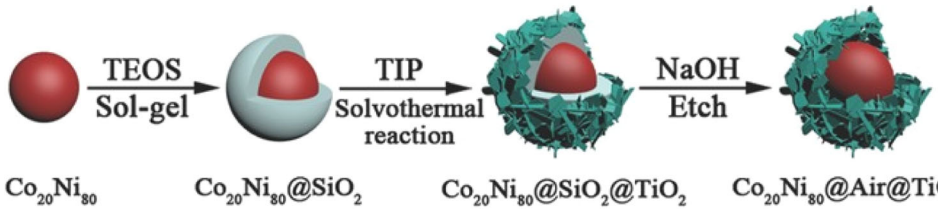

Fig. 7 Schematic illustration of synthetic route for the $CoNi@SiO_2@TiO_2$ and $CoNi@Air@TiO_2$ microspherical structures. Reprinted with permission from ref. [40]. Copyright © 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

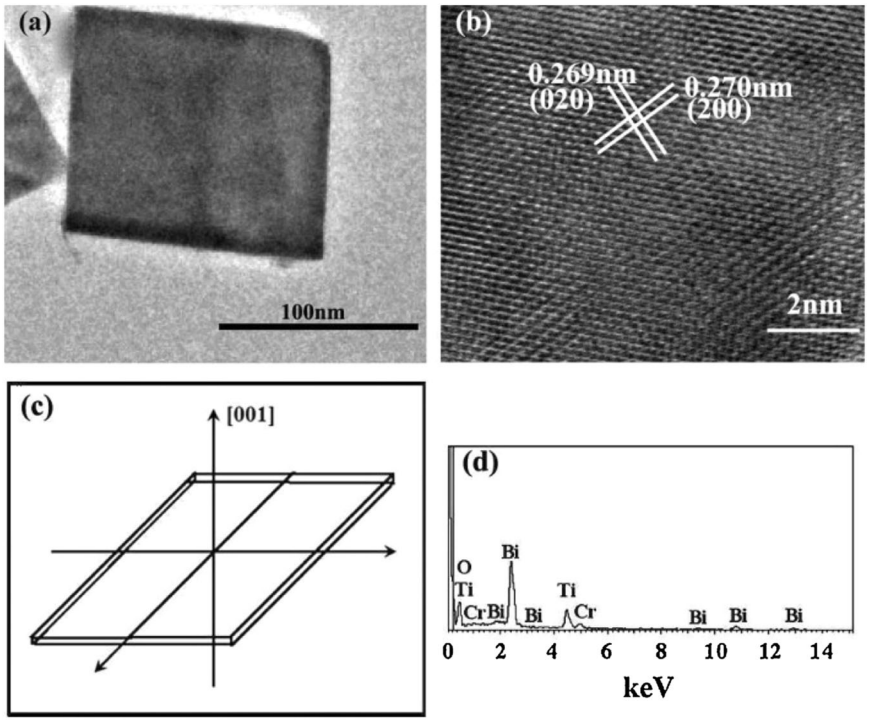

Fig. 8 TEM and HRTEM images, schematic illustration of the crystal orientation, and EDS pattern of the chromium-modified $Bi_4Te_3O_{12}$ nanosheets prepared via a sol–gel hydrothermal process [41]. Copyright © 2018 Elsevier Ltd

such as molybdenum disulfide [15], bismuth titanate [22, 41] and sodium manganese oxide [42]. The applications of sol–gel-derived 2D nanosheets range from photocatalysts and batteries to electromagnetic shielding materials. For example, Chen et al. from the South China University of Technology reported the synthesis of $Bi_4Te_3O_{12}$ nanosheets and chromium-modified $Bi_4Te_3O_{12}$ nanosheets via a sol–gel hydrothermal process for water pollutant degradation [22, 41]. The as-prepared [001] facet-exposed $Bi_4Te_3O_{12}$ nanosheets were rectangular in shape, ~100–150 nm in side length, and ~20 nm in thickness, and showed 79.2% of photodegradation efficiency of Rhodamine B under natural sunlight with good reusability. The $Bi_4Te_3O_{12}$ nanosheets with chromium modification revealed similar rectangular sheet-like nanostructure, and the addition of chromium could effectively improve their photocatalytic performance for hydrogen evolution and pollutant degradation due to the low recombination rate or the high separation efficiency for photogenerated electron-hole pairs (Fig. 8).

Zhang et al. from Shaanxi University of Science and Technology reported the preparation of a primary nanosheet $Na_{0.44}MnO_2{\cdot}Na_2Mn_3O_7$ heterojunction cathode via a sol–gel-assisted high-temperature sintering method [42]. The $Na_{0.44}MnO_2$ domain plays the role of both multidimensional channels to improve the rate performance and pillars for enhancing the cycling stability and Coulombic efficiency of $Na_2Mn_3O_7$. Li et al. from the Zhengzhou University of Light Industry reported the preparation of 2D ultrathin perovskite $LaNiO_3$ nanosheets with hierarchical porous structure via a facile sol–gel approach [43]. The $LaNiO_3$ nanosheets with a thickness of ~50 nm possessed large surface area and pore volume, as well as good rate capability and cycle stability, and excellent energy density and cycling stability performance when used in the corresponding asymmetric supercapacitor. Gu et al. from Nanjing University of Aeronautics and Astronautics reported the preparation of Fe_3N alloy/porous carbon nanosheet heterostructures prepared via sol–gel sacrificial template methods for microwave attenuation applications [44]. The microstructure and component content of the composite could be readily tuned by controlling the proportion of precursors, and the composite materials

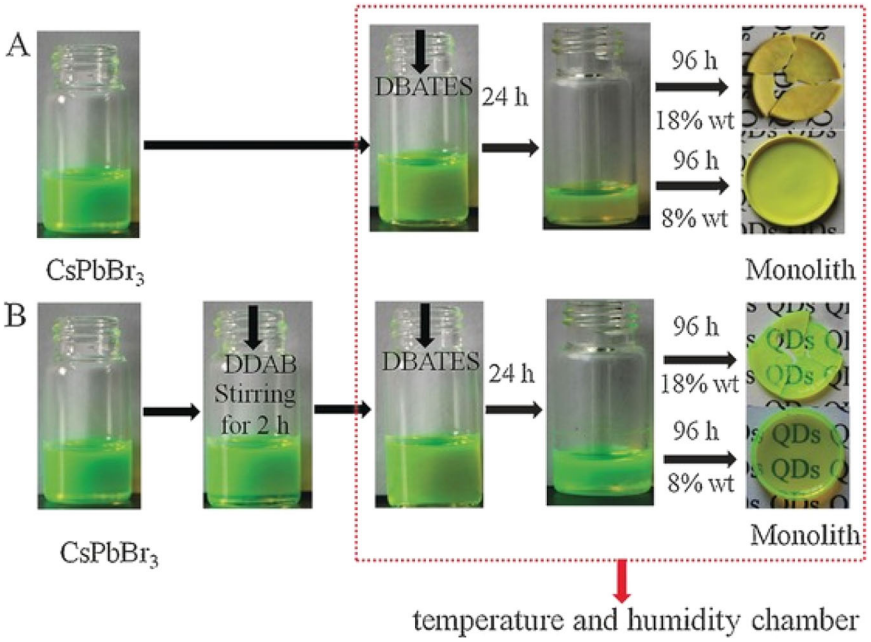

Fig. 9 Photographs of $CsPbBr_3$ QDs (**A**) and didodecyl dimethyl ammonium bromide (DDAB)-treated $CsPbBr_3$ QDs (**B**) solution after adding Di-sec-butoxyaluminoxytriethoxysilane (DBATES) into QDs solutions and their corresponding monolith. Reprinted with permission from ref. [50]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

exhibited excellent microwave absorbing performance as a result of sufficient magnetic loss capacity, strong conductivity, and polarization attenuation.

2.2 Fabrication of bulk materials via sol–gel process

2.2.1 Sol–gel synthesis of ceramics and glasses

As a liquid-phase process, the sol–gel process is also widely employed to obtain bulk materials in various forms such as glasses [45], ceramics, transparent glass ceramics [46], aerogels [47], and monoliths [48]. For instance, Zheng et al. from Zhejiang University developed a sol–gel derived method for the preparation of CeF_3-containing silicate glass-ceramic as UV absorber for biological protection [49]. By using $Ce(CH_3COO)_3$ and CF_3COOH as the Ce and F precursors, glass-ceramic materials could be created through in situ crystallization of CeF_3 at 400 °C and exhibited excellent UV-blocking performance.

Through a sol–gel process, Li et al. from Shanghai Jiaotong University [50] incorporated $CsPbBr_3$ perovskite quantum dots into a silica/alumina monolith, as illustrated in Fig. 9. The as-prepared transparent monoliths were highly luminescent with quantum yields up to 90% and stability under blue light illumination for 300 h, showing great potential as a color-converting layer for blue LEDs.

2.2.2 Sol–gel synthesis of metal oxide aerogels

As an important type of mesoporous material with a 3-D network of nanostructured skeletons, aerogels have attracted considerable attention in the field of sol–gel-derived bulk materials research [8, 47, 51–55]. In this review, we only briefly introduce some of the aerogel-related studies recently published in China. A more detailed review of aerogels and their high-temperature applications is presented in this special issue by Professor Jun Shen and his co-workers from Tongji University. In addition, another recently published review presented by Professor Feng et al. from the National University of Defense Technology comprehensively introduced the preparation of aerogels in combination with 3D printing and additive technology [55].

Aerogels have also begun to be adopted as pressure sensors owing to their good electrical conductivity, excellent mechanical properties, and ultralow density. Copper nanowire (CuNW) in the form of an aerogel presents great potential as a piezoresistive pressure sensor material owing to its excellent pressure-induced electrical resistance change, high electrical conductivity and natural abundancy. Through a one-step sol–gel synthesis method, Xu et al. from the Shanghai Institute of Ceramics-CAS prepared a CuNW-based aerogel for its application as a flexible pressure sensor [51]. In this synthesis process, the copper precursor was reduced and self-assembled into sol flocs simultaneously through a "bubble-controlled assembly" procedure, followed by freeze-drying treatment to form the CuNW aerogels. This synthetic approach enables tunable pore structures and densities of the nanowire aerogel, thus providing controllable sensitivities of the sensor via pore structure adjustment.

Due to their low densities, high surface areas, and fast mass transport, aerogels show great potential for heterogeneous applications in fields such as energy storage and conversion. Ou et al. from Fuzhou University developed a

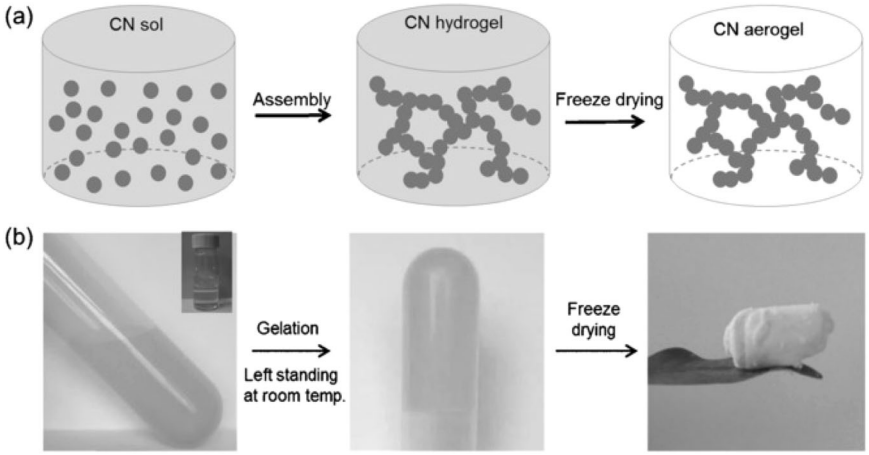

Fig. 10 Schematic illustration of the synthetic process for CN aerogels (**a**), and photos of as-prepared CN nanoparticle, hydrogels, and aerogels (**b**). Reprinted with permission from ref. [52]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

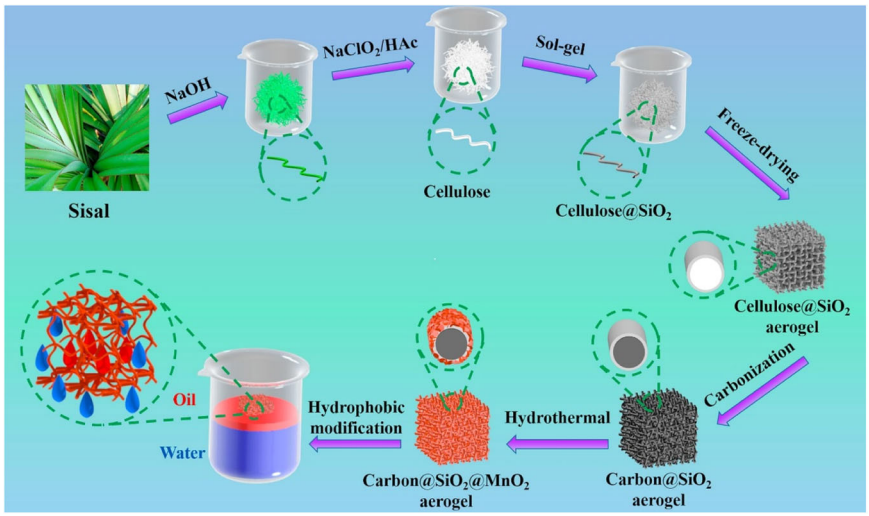

Fig. 11 Schematic illustration of the synthetic route for the carbon@SiO_2@MnO_2 aerogel. Reprinted with permission from ref. [56]. Copyright © 2018 Elsevier Ltd

facile sol–gel-based method to synthesize self-supported carbon nitride (CN) aerogels for photo-redox conversion of water under visible-light irradiation [52]. In this fabrication process, as illustrated in Fig. 10, self-supported CN aerogels were prepared using a self-assembly-based method. This unique method did not require the aid of strong acids and cross-linking agents as reported in previous studies, and the resulting materials exhibited excellent physical properties, significantly enhanced photocatalytic water splitting performance, as well as scalable and low-cost producibility.

Yuan et al. from Jiangsu University proposed the preparation of compressible, superhydrophobic, and multifunctional hierarchical biomass carbon@SiO_2@MnO_2 aerogel (HBCSM). In the synthetic process, the biomass carbon@SiO_2 (BCS) aerogel was firstly fabricated using the sol–gel process with additional freeze-drying and carbonization treatment (as illustrated in Fig. 11), and then MnO_2 nanosheets were in situ assembled on the surface of the BCS by hydrothermal treatment to form the HBCSM aerogel [56]. The resulting product exhibited excellent surface roughness, superhydrophobicity, absorption capacity for oils, and mechanical durability.

2.2.3 Sol–gel synthesis of hierarchically porous monoliths

Apart from aerogels, hierarchically porous monoliths are also widely studied sol–gel-derived bulk materials that exhibit multiple-level porosity integrated into a single architecture [57]. Guo et al. from Zhejiang University prepared hierarchically porous composites constructed by ZnAl-LDH/Al$(OH)_3$ nanosheets via a sol–gel route for adsorption of organic dyes [58]. The composites exhibited ultrafast adsorption rates and high adsorption capacities in the removal of the anionic dyes Methyl Orange and Congo Red due to ion exchange and hydrogen bonding on the surface of LDH and amorphous Al$(OH)_3$. In their subsequent work, hierarchically porous $ZnFe_2O_4$/C composites with interconnected macropores and co-continuous hollow skeletons were prepared by the sol–gel process with phase separation treatment and investigated as anode materials for Li-ion batteries [59]. The anode materials

exhibited some of the best Li-ion storage properties reported for $ZnFe_2O_4$-based materials.

2.3 Preparation of functional coatings through sol–gel process

2.3.1 Preparation of anticorrosion coatings on metallic surfaces via sol–gel process

The sol–gel process is widely recognized as a powerful method for producing a wide variety of inorganic or organic-inorganic hybrid coating materials [7, 60–63]. Unlike conventional preparation techniques for inorganic-based coatings such as vapor deposition (CVD/PVD) and electrochemical deposition, the sol–gel process does not require expensive equipment or high-temperature treatment, allowing the preparation of coating with high-performance and low-cost. Coatings with different functionalities and characteristics (anticorrosive, antireflective, anti-fogging, surface reinforcing, water-resistant, etc.) can be prepared via sol–gel routes on different substrate surfaces using deposition techniques such as dip-coating, spin-coating, or spray-coating.

Among these functional coating systems, anticorrosion coatings prepared via the sol–gel process are of great interest due to their outstanding anticorrosive properties, high surface hardness, versatile preparation method, low-temperature treatment, low toxicity and "green" synthesis route [60, 62, 63]. The sol–gel process involving the use of inorganic nanoparticles (silica, titania, zirconia, etc.) and organosilanes is a commonly used strategy for the preparation of organic-inorganic hybrid coatings with excellent anticorrosion performance as well as other characteristics such as heat resistance and wear resistance [64–66]. For instance, Li et al. from Zhejiang University developed a sol–gel derived method for the preparation of a two-layer anticorrosion coating incorporating silica and titania fillers in the bottom layer and a translucent colloidal silica sol–gel matrix in the topcoat (Fig. 12) [67]. This coating structure design allowed the deposition of thick and defect-free organic-inorganic hybrid protective films with excellent physical properties. On the other hand, the hydrophobic topcoat acted as a barrier that prevents water leakage into the inner structure.

Inspired by the natural "lotus leaf" phenomenon, many research efforts have been devoted to the development of artificial superhydrophobic coating surfaces that exhibit high water resistance, giving them great potential in scientific and industrial applications such as anticorrosion, self-cleaning, anti-fouling, anti-icing and oil-water separation [68, 69]. As one of the most suitable methods to fabricate anticorrosion films with high water resistance, the sol–gel process is a convenient approach to obtain superhydrophobic coatings that greatly delay water adsorption and prolong the service life of the undercoats or the underlying materials [64]. For example, Wu et al. from the Guilin University of Technology demonstrated the preparation of superhydrophobic and superoleophobic coatings using both low and high surface energy SiO_2 nanoparticles through a sol–gel process [70]. In this case, the coating surfaces obtained exhibited variable surface roughness and surface energy by adjusting the molar ratio of low and high surface energy nanoparticles, leading to tunable surface hydrophobicity. At a molar ratio of 2:4, the coating possessed the best superoleophobicity and mechanical properties, showing great potential in self-cleaning and anti-icing applications. Similarly, Wu et al. from Central South University of Forestry and Technology employed SiO_2 and TiO_2 nanoparticles combined with vinyltriethoxysilane to fabricate superhydrophobic surfaces by a sol–gel method on various substrates, such as wood, glass, and metal [71]. The superhydrophobic coating showed excellent mechanical durability arising from the hierarchical structure and strong interfacial adhesion of the coating and exhibited excellent durability under harsh environmental temperature and humidity.

Apart from fabricating the coating matrix itself, the sol–gel process also acts as a versatile method to synthesize functional additive components incorporated into anticorrosion coatings [72–74]. The introduction of additional nano- or micro-sized additives brings new functionalities to anticorrosion coatings, in particular, the "self-healing" function [75, 76]. In this case, microcapsules or

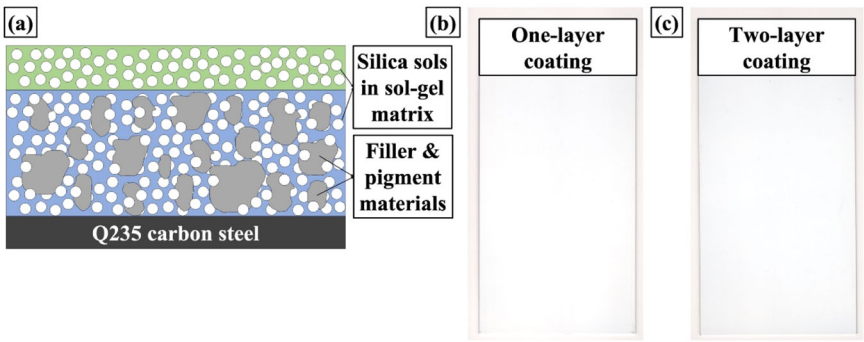

Fig. 12 Schematic illustration of the anticorrosion sol–gel coating structure and surface images of the as-prepared coatings. Reprinted with permission from ref. [67]. Copyright © 2019 MDPI

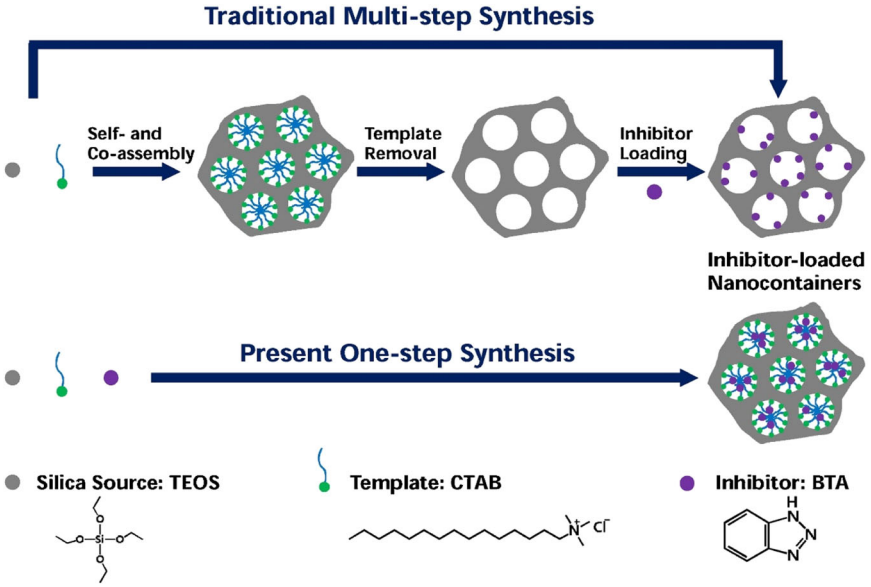

Fig. 13 Schematic illustration of the one-step synthetic route for inhibitor-loaded mesoporous SiO_2 nanocontainers. Reprinted with permission from ref. [77]. Copyright © 2018 Elsevier Ltd

nanocontainers encapsulating corrosion-inhibiting species or self-healing molecules can be activated upon exposure to external or internal stimuli (pH, heat, light, mechanical, etc.) to repair the coating damage or recover the barrier property. For example, Xu et al. from Zhejiang University proposed a one-step sol–gel method for the preparation of inhibitor-loaded mesoporous silica nanocontainers [77]. As illustrated in Fig. 13, the BTA inhibitor was first dissolved in CTAB surfactant micelles, followed by the formation of silica nanoparticle through hydrolysis and polycondensation of TEOS precursor. The self-healing nanocontainers obtained exhibited effective pH-responsive inhibitor-release behavior, and incorporation of the silica nanocontainers into the anticorrosion coating gave rise to the self-healing function and enhanced electrochemical properties of the coating for copper surface protection.

Ding et al. from Nanjing University of Science and Technology prepared corrosion inhibitor-loaded mesoporous silica nanoparticles modified with pseudorotaxanes as supramolecular nanovalves and incorporated them into a self-assembled nanophase particle barrier coating on a magnesium alloy surface [78]. The mesoporous nanoparticles exhibited active release of corrosion inhibitors upon exposure to alkali or Mg^{2+} stimuli, and the incorporation of the nanocontainers resulted in the successful construction of a feedback-active coating that possessed a superhydrophobic surface, effective self-healing property, and long-term anticorrosion performance.

2.3.2 Preparation of waterproof and fire-retardant coatings on fabrics via sol–gel process

Sol–gel-derived coatings can be utilized not only on metal surfaces but also with fabrics or textiles to offer better waterproof and fire-retardant functionalities [79–83]. Su et al. from the South China University of Technology, for example, proposed the fabrication of a highly durable superhydrophobic polydimethylsiloxane@silica coating on polyester fabric via a vapor-liquid sol–gel approach. [81]. The superhydrophobic textile possessed excellent durability in multiple solutions and solvents and exhibited high separation efficiency and good reusability in oil-water separation applications. In their following study, the fabrication of superhydrophobic and flame-retardant coatings on cotton fabrics via a one-pot sol–gel approach was demonstrated [79]. The fabrics showed excellent durability and self-cleaning properties as well as a strong physical barrier effect when exposed to fire. In addition, epoxy resin nanocomposites incorporated with H-BN can also be prepared via the sol–gel process, showing good thermal stability, flame retardancy, and smoke suppression [83].

2.3.3 Sol–gel preparation of functional films with ferroelectric properties

As one of the most commonly used chemical processes for the fabrication of thin/thick films, the sol–gel process is also the ideal method for the deposition of a variety of materials with ferroelectric and piezoelectric properties. In recent years, the field of ferroelectric thin films has been one of the main research focuses by research communities in China, especially in energy storage applications such as electrostatic capacitors and solid-state cooling devices [84–91]. For instance, Sun et al. from Inner Mongolia University of Science and Technology demonstrated the preparation of lead-free relaxor ferroelectric $(1-x)(Bi_{0.5}\ Na_{0.5})TiO_3$-$xBi(Ni_{0.5}Zr_{0.5})O_3$ (BNT-xBNZ, $x = 0$–0.5) film with various doping content of BNZ via a water-based sol–gel method [85].

With doping content $x = 0.4$, the film showed the best recoverable energy density and high energy storage efficiency, as well as strong fatigue endurance. In another case, Peng et al. from Guangxi University demonstrated the sol–gel preparation of $Pb_{0.97}La_{0.02}(Zr_{0.65}Sn_{0.3}T_{i0.05})O_3$ (PLZST) relaxor antiferroelectric (AFE) thin films that showed a large positive electrocaloric (EC) effect in a broad temperature range [86]. The PLZST film exhibited a large EC effect owing to the in-plane residual thermal tensile stress developed during the layer-by-layer annealing process and the high-quality film structure. The excellent dielectric relaxor dispersion around the dielectric peak due to the coexistence of multiple FE and AFE phases led to the broad EC temperature range.

$BiFeO_3$ (BFO) is another extensively studied ferroelectric thin film that can be readily prepared using the sol–gel method. Sol–gel-derived polycrystalline BFO thin film photoelectrodes exhibit small bandgap and good band positions for photoelectrochemical (PEC) water splitting. To this end, Liu et al. from Soochow University demonstrated the preparation of polycrystalline BFO films on Pt/Ti/SiO2/Si(100) substrates using the sol–gel method [87]. The BFO films were further decorated with Ag nanoparticles, which resulted in enhancement of the PEC properties of the films owing to the improved electron-hole pair separation efficiency and the localized surface plasmon resonance phenomenon of Ag nanoparticles.

3 Commercialization of sol–gel technology in China

With the rapid advancement of sol–gel technology, a wide range of novel materials with outstanding physical and chemical properties have been created for practical and industrial applications. In recent years, China has been contributing not only to the production of scientific papers but also to the application and industrialization of sol–gel technology. Looking at sol–gel-related patents (source: WIPO), there has been a rapid growth in the number of applied patents both in China and in the world since the beginning of the 21st century (Fig. 14). It can be seen that China's contribution to the sol–gel patent literature increased from 5.4% in 2000 to 64.9% in 2020 (Fig. 13 inset), and 54.1% of the 11,508 patents published in the world in the last 10 years from 2011 to 2020 were produced in China.

One of the most developed industrial applications of sol–gel technology in China relates to the production of colloidal oxide nanoparticles such as silica, alumina, and yttria. Usually dispersed in water or other solvents, these nano-sized oxides prepared via the sol–gel process possess high purity, good dispersibility, high stability, and high translucency along with many other benefits. Sol–gel oxides nanoparticles have been extensively used in polishing of

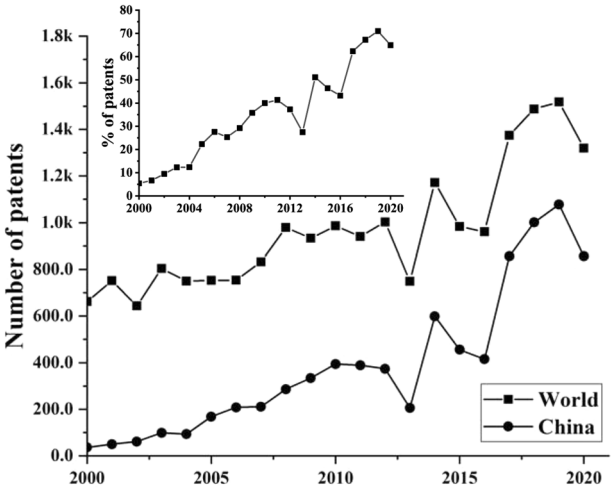

Fig. 14 Number of patents containing "sol–gel" in topic published per year in China and the world from 2000 to 2020. Inset is the percentage of patents published in China over patents in the world (source: WIPO)

optical components, fabrication of functional coatings, reinforcement of polymeric materials, loading structures of catalytic materials, and many other areas.

Sol–gel routes are also widely adopted to produce granules or nanopowders in industry. Using highly active compounds as precursors, granules, or nanopowders with high purity, well-defined size, controllable crystal shape, and structure can be synthesized by sol–gel derived methods. Unlike conventional "top-down" approaches such as ball-milling and laser ablation, the sol–gel process is a "bottom-up" approach that provides new alternatives to the large-scale production of inorganic oxide nanopowders with precise control over their physical and chemical properties. In light of these benefits, the sol–gel approach is extensively employed in the production of nanopowders used for electroceramics, microwave ceramics, electrical contact materials, nanocomposite granules, and so on.

Moreover, the sol–gel process has become an emerging technology for the fabrication of coatings or thin films with various functions, such as anticorrosive, antireflective, abrasion-resistant, superhydrophobic, self-cleaning, antifogging, antibacterial, and many others. The global market for sol–gel coatings has reached US$2.5 Billion in 2020 and is estimated to reach US$4.8 Billion by 2027 [92]. China's sol–gel coating market is forecast to reach US$833 Million by 2027 with a compound annual growth rate of 9.2%.

One aspect of the successful industrialization of sol–gel technology involves the application of sol–gel-derived coatings for the protection of metallic surfaces. For example, Jinfei Holding Group [93], an automobile parts manufacturer, has successfully implemented the use of sol–gel-derived organic-inorganic hybrid coatings for the surface treatment of aluminum alloy wheels, which provides better anticorrosive performance, mechanical properties, and

decorative characteristics for their products. Compared to conventional surface treatment techniques such as electroplating and anodic oxidation, the sol–gel processing technique produces less wastewater pollution, heavy metal contamination, volatile organic compound emission, etc.

4 Conclusion

Overall, this review presents a brief historical account of the sol–gel developments in China and discusses some of the recent advances of scientific research and development on sol–gel-related technology in China. Some examples of the applications of sol–gel-derived materials are demonstrated. Though this review only focuses on some aspects of technological achievements and does not depict the full scope of China's sol–gel technology developments, it can still be anticipated that more novel materials created using sol–gel technology can be discovered with the constant endeavors of researchers and scientists in China.

Acknowledgements This review was supported by the Zhejiang Provincial Natural Science Foundation of China under Grant No. LQ20E020008, by the Guangxi Department of Science and Technology-Zhejiang University Science, Technology and Innovation Cooperation Project (No. ZD20302002), by the Open Project of State Key Lab of Silicon Materials, Zhejiang University (SKL2020-11), and by the Fundamental Research Funds for the Central Universities (No. K20200042).

Compliance with ethical standards

Conflict of interest The authors declare no competing interests.

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