

Self-Assembled Nanocomposites and Nanostructures

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Self-assembled nanocomposites and nanostructures are a class of molecules that spontaneously assemble directly through specific interactions or indirectly through their environment without any human intervention. The benefits of self-assembled nanocomposites or nanostructures have been reported several times for their prominent applications.

nanocomposites

nanostructures

self-assembled

photocatalyst

energy materials

thermoelectric materials

1. Self-Assembled Nanostructured Membranes for Photocatalysis

1.1. Self-Assembled Nanostructured Membrane Photocatalysts for Wastewater Treatment

Liu et al. prepared titanium dioxide films via a layer-by-layer self-assembly technique to immobilize TiO_2 nanoparticles using polyurethane (PU) and increase the adsorption capacity of the photocatalyst ^[1]. In addition, the photocatalytic performance and reusability of the films were investigated by the decomposition of MB under UV irradiation, and it was shown that the catalytic efficiency of the prepared films was still as high as 94.56% after five cycles and they could be reused six times without affecting the photocatalytic activity. Synthetic membranes are therefore promising candidate materials for wastewater treatment applications.

However, single nano- TiO_2 photocatalysts have low specific surface area, poor adsorption capacity for pollutants, and are easily agglomerated and difficult to recover, resulting in low catalytic efficiency for the photocatalytic degradation of low concentrations of organic matter. The researchers found that composite nano- TiO_2 materials can significantly improve the defects of single nano- TiO_2 materials which are prone to agglomeration. To overcome the defects of nano- TiO_2 particles, research and application of composite materials are gaining increasing attention ^{[2][3][4]}.

Zhang et al. successfully coated TiO_2 nanofibers onto ceramic hollow fiber membranes using a simple dip-coating technique to form TiO_2 nanofiber membranes with reticular morphologies, and they evaluated the performances of the TiO_2 nanofiber membranes in treating hyaluronic acid by monitoring the change in the total organic carbon values in water ^[5]. After six cycles, there was no significant loss of activity of the TiO_2 nanofiber hollow membrane.

Therefore, the TiO₂ nanofiber hollow membranes proposed possessed high stability during the removal of hyaluronic acid.

Bai et al. successfully synthesized a novel multifunctional carbon nanotube/ZnO/TiO₂ nanocomposite ultrafiltration membrane by hydrothermal synthesis and ultrasonic-assisted acid treatment [6]. Chang et al. obtained self-assembled nanoporous Ti with a smooth surface and many folds by a hydrothermal method using metallic titanium foam as the raw material [7]. A self-assembled layer of a strongly adherent 3D Na₂Ti₃O₇ nanowire network was grown on the surfaces of Ti particles and channels after alkaline hydrothermal treatment in a NaOH solution. The self-assembled TiO₂ nanowire networks were uniform, with lengths of 2–3 μm [7]. The self-assembled nanowire network utilized the wastewater degradation device for two different types of dyes, RhB (20 mg/L) and MB (20 mg/L), both of which showed good photocatalytic properties after UV irradiation for 60 min only, reflecting its good degradation effects. The above results illustrate the high performance of self-assembled nanoporous Ti as photoelectrolytic electrode materials.

In addition, Fe₃O₄-based nanocomposites can be used as good Fenton-like catalysts for the degradation of organic pollutants in water [8]. The introduction of magnetic Fe₃O₄ nanoparticles provides another advantage for nanostructured composites, and the magnetic properties of the prepared composites facilitate fast and easy separation during catalyst recovery and reuse [9].

Wang et al. successfully prepared an Fe₃O₄/rGO/metal–organic framework (MOF) composite with a dispersed interlayer structure by a hydrothermal method and investigated the degradation performances of these composites on phenol [10]. They found that the degradation of phenol was mainly dependent on the pH, and the degradation efficiency of phenol reached 80% within 2 min at pH = 3. The degradation rate of phenol decreased sharply when the pH was further reduced [11]. The phenol was completely removed within 16 min for all pH conditions. By exploring the role of the catalyst components in Fenton-like reactions, they found that the excellent catalytic performances of the Fe₃O₄/rGO/MOF composites were mainly due to the synergistic effect of the porous MOF shell and the internal Fe₃O₄/rGO [12]. The reusability of Fe₃O₄/rGO/MOF was tested by recovering the catalyst at the end of the reaction and reusing it in the next run. As shown, the catalytic activity of Fe₃O₄/rGO/MOF was maintained at 96% after five reuses.

2. Self-Assembled Nanostructured Membrane Photocatalysts for Photocatalytic Hydrogen Production

Photocatalyst modification using doped noble metal nanoparticles is an effective method to improve the photocatalytic performance. Dal'Acqua et al. prepared a multilayer composite by combining gold (Au) and titanium dioxide (TiO₂) nanoparticles (NPs) into self-assembled photocatalytic films (SAPFs) [13], forming a composite with larger specific surface area compared to those of conventional nanostructured catalysts. This facilitated the maximization of the photocatalytic activity. In this structured photocatalyst, hydrogen is produced in the polymer/(TiO₂-Au) nanoparticle network and also in the body of the polymer/(TiO₂-Au) NP assembly. Hydrogen is readily produced in large quantities under radiation, and the amount of hydrogen produced by the structured

photocatalyst increased linearly with increasing UV irradiation time. The SAPFs have great potential for renewable energy development due to their simple preparation process and excellent photocatalytic activity.

He et al. prepared nanoporous CoFe_2O_4 loaded with platinum and silver by dealloying [14]. They showed that the hydrogen precipitation rate of the resulting sample was as high as 2.36 mmol/h/g under full-spectrum irradiation, which was 24 times that of CoFe_2O_4 without platinum or silver. The H_2 -releasing activity did not decrease significantly after 32 h of continuous irradiation, indicating the excellent stability of this photocatalyst. The analysis showed that the silver NPs had a strong surface plasmon resonance (SPR) effect in visible light, resulting in effective visible light absorption. This effect expanded the range of light absorption and effectively improved light utilization, while Pt could act as an effective electron trap for electron–hole pair separation, effectively inhibiting electron and hole complexation. In addition, the simultaneous loading of Pt and Ag on CoFe_2O_4 produced a synergistic effect that contributed to its photocatalytic performance.

The morphological modification of photocatalysts and the construction of heterojunctions are considered to be the main means of significantly improving the performances of photocatalytic hydrogen evolution [15][16].

Zhang et al. successfully synthesized ZnS nanocrystals with different morphologies using cysteine as the sulfur source at different heating temperatures using a template-free method and evaluated the photocatalytic performances of the samples [17]. After a series of catalytic experiments, it was found that the inherent self-absorption and photon recirculation of photoluminescence played a key role in the photocatalysis. The photocatalytic activity was investigated by the degradation of RhB solutions. It is well known that the morphology of a material has an important influence on its properties. As shown by the photocatalytic results and the morphological images, ZnS-200 exhibits a simpler surface structure and better catalytic activity than ZnS-100 and ZnS-150, suggesting the idea that defect-rich edge states are advantageous in providing reactive sites and broadening the absorption range.

Bhirud et al. prepared hierarchical nanostructures of cubic-spinel-structured CdIn_2S_4 selectively by a hydrothermal method [18]. The effects of surfactants on the morphology and microstructure of cadmium sulfide were investigated using polyvinylpyrrolidone (PVP) and cetyltrimethylammonium bromide as surfactants. The cadmium sulfide samples prepared with PVP as the surfactant exhibited excellent photocatalytic activities, with a maximum hydrogen production rate of up to 3238 $\mu\text{mol/h}$.

3. Self-Assembled Nanocomposites for Energy Storage

3.1. Self-Assembled Nanocomposites for Lithium-Ion Batteries

Deng et al. reported a simple wet chemistry route for the large-scale synthesis of nearly monodispersed self-assembled SnO_2 nanospheres by direct hydrogen peroxide oxidation of bulk tin (Sn) metal in deionized water (DIW) with the assistance of polyvinylpyrrolidone (PVP) and ethylenediamine (EDA) at room temperature, using PVP as a spatial stabilizer to limit the nanocrystal-to-nanocrystal contact, effectively preventing aggregation of

nanocrystals [19] and causing particle aggregation to form nanospheres by minimizing the energy. The amount of PVP in the reaction system could lead to the controlled growth and self-assembly of SnO_2 nanocrystals. It contributed to the large-scale synthesis of self-assembled functional oxide nanostructures. Man et al. prepared SnO_2 porous nanotubes (PNTs) by electrostatic spinning self-assembly [20]. The hollow structure and nano- SnO_2 particles effectively increased the contact area between the electrolyte and the active material, alleviated the defects caused by volume expansion, and improved their electrochemical properties. It was found through testing that the SnO_2 -PNTs had excellent rate performances. After charge/discharge tests, the coulombic efficiency exceeded 99% and could still provide a reversible capacity of 1045 mAh/g after 160 cycles. As shown in the SEM images, there were no cracks on the electrode surface after cycling, indicating its excellent structural stability.

Molybdenum disulfide (MoS_2) is a layered transition metal disulfide that has also attracted interest as an electrode material for lithium batteries due to its important mechanical, electrical, and optical properties. It has been found that the addition of carbon-based conductive additives to MoS_2 can significantly improve the recyclability and testability of a material [21][22]. Das et al. prepared MoS_2 -carbon hierarchical nanostructures with different carbon compositions by hydrothermal self-assembly and investigated the application of the composite as a high-energy electrode for lithium-ion secondary batteries [23]. When the material was used as an electrode for lithium batteries, the binding of carbon provided significantly improved cycling stability and the carbon skeleton effectively inhibited particle agglomeration.

Carbon coatings usually cover the surface of an active material tightly and do not effectively release the large strain from the volume expansion, which in turn leads to increased resistance for the lithium ions to reach the active material inside. Graphene is used as a nanostructured electrode material for energy applications due to its special structure, excellent electrical conductivity, large surface area, and chemical stability [24].

Sun et al. realized the large-scale preparation of MoO_2 /graphene nanocomposites by uniformly encapsulating MoO_2 nanocrystals in graphene sheets and testing them as the positive electrodes of lithium-ion batteries. They found that the electrode material synthesized from MoO_2 /graphene nanocomposites had a significantly higher electrochemical performance than the bare MoO_2 electrode material [25]. The electrochemical performance of the MoO_2 /graphene nanocomposite was found to be significantly higher than that of the bare MoO_2 electrode material, with high cyclability and increasing reversible capacity. The coulomb efficiency approached 100% at high current densities, and the capacity reached up to 407.7 mAh/g after 70 cycles and up to 848.6 mAh/g at low current densities. The morphology remained pristine, further demonstrating the high stability of the graded nanostructures, excellent cycling performances, and good rate capability.

MOFs or porous coordination polymers have received special attention as a new class of hybrid nanoporous materials because of their high surface areas and unique structure [26][27]. Zhu et al. prepared porous $\text{ZnO}/\text{Co}_3\text{O}_4$ nanocomposite clusters by self-assembly [28], with an initial discharge capacity of up to 2049 mAh/g, which showed high reversible capacity of 957 mAh/g after 100 cycles.

3.2. Self-Assembled Nanostructured Supercapacitor Materials

A composite of polycation-functionalized reduction of graphene oxide (FRGO-p) and MnO_2 nanosheets (FRGO-p- MnO_2) was synthesized by the electrostatic precipitation method [29]. Since the self-assembly of the MnO_2 sheet effectively prevented the aggregation of MnO_2 , this material showed a strong capacitive performance and retained more than 89% of its initial capacitance after 1000 cycles. The layered nanostructure was prepared by an electrostatic self-assembly method [30]. The capacitance of the graphene supercapacitor was increased by more than 70%, the high-power density was increased by 15%, and the cycle life was increased. A graphene/carbon nanotube hybrid film with an interconnected carbon structure network was prepared by layer-by-layer (LBL) self-assembly technology [31]. The pure carbon electrode based on amine-based functionalized multi-walled carbon nanotubes (MWCNT- NH_2) and rGO self-assembly prepared by Byon et al. had a specific capacitance of 120 F/g. The specific capacitance of the MWCNT/rGO electrode assembled by hydrazine steam treatment was about 1.5 times higher than that of rGO.

A high-density hybrid film was prepared by self-assembly technology. The prepared activated carbon film could be used as the binder-free electrode of a supercapacitor. The constant current charge—discharge curve of the electrode showed that it had ideal charge—discharge characteristics and good reversibility [33].

Through the self-assembly of in situ carbon-fiber-coated WO_{3-x} , a DNA-like double helix WO_{3-x} /ultrafine fiber structure (DNA-WC) was designed. As an advanced supercapacitor material, it had excellent electrochemical properties, and its stability was greater than 94% after more than 5000 continuous cycles [34]. Stability is one of the important factors in the application of supercapacitors. The above materials have been studied in detail. With energy storage becoming a popular research subject, the application potential of supercapacitors in harsh environments has been deeply explored.

4. Self-Assembled Thin-Film Thermoelectric Materials for Energy Harvesting

4.1. Self-Assembled Thin-Film Thermoelectric Materials for Power Generation Using Waste Heat

Du et al. studied Sb-doped $\text{Mg}_2\text{Si}_{0.4}\text{Sn}_{0.6}$ materials and reached a consistent conclusion, showing that the Seebeck coefficient increased by about 300 $\mu\text{V/K}$ [35][36]. The thermoelectric figure of merit for $\text{Mg}_{2.2}\text{Si}_{0.7}\text{Sn}_{0.3}\text{Sb}_{0.01}$ was 0.64 at 723 K. Further studies of Sb-doped Mg_2Si and Mg_2Sn by Liu et al. showed a gradual increase in the Seebeck coefficient, with the thermoelectric figure of merit reaching about 1.3 at 700 K [37].

Polymer-based thermoelectric materials have been widely noticed for their good flexibility and low-density properties. To improve the energy conversion efficiencies of thermoelectric materials, polymer-based composites have become a popular research topic. Liu et al. prepared a functional film self-assembled from Bi_2Se_3 nanopillars using a solvothermal method [38], and the power factor of thermoelectricity was increased from 1.1 $\mu\text{W/cm}\cdot\text{K}^2$ of the sheet nanoflowers to 1.7 $\mu\text{W/cm}\cdot\text{K}^2$. The structures can significantly improve the performances of organic thermoelectric materials. Cho et al. prepared double-walled nanotubes (DWNT)-polyethyleneimine (PEI)/graphene-

PVP multilayer nanocomposites using an LbL assembly method, which uniformly bound active conducting elements into layered 3D hybrid organic nanostructures, significantly improving the electrical conductivity as well as the Seebeck coefficient [39]. Due to the increased conductivity of the nanocomposites, the power factor was also substantially increased, with a power factor of $1.9 \mu\text{W}/\text{cm}\cdot\text{K}^2$. Te-based thermoelectric thin-film materials have attracted widespread interest due to their excellent thermoelectric properties. Zhou et al. prepared Te thin films by electrodeposition on stainless-steel substrates (SSS) with high flexibility. The room temperature power factor was $3.21 \mu\text{W}/\text{cm}\cdot\text{K}^2$ and the thermal conductivity was $4.4 \text{ W}/\text{K}\cdot\text{m}$ [40].

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