

Novel Nanomaterials for Hydrogen Production and Storage

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Using hydrogen energy as an alternative renewable source of fuel is no longer an unrealized dream, it now has real-world application. The influence of nanomaterials on various aspects of hydrogen energy, such as hydrogen production, storage, and safety, is considerable.

Keywords: hydrogen energy ; storage ; nanomaterials

1. Nanomaterials Used in Hydrogen Production

Hydrogen production involves four different methods: (1) photoelectrochemical (PEC) water splitting, (2) solid-state hydrogen storage, (3) photocatalytic hydrogen production, and (4) proton exchange membrane fuel cells (PEMFCs). Photocatalysis involves the following reaction: photogenerated electrons and holes at the conduction and valence bands lead to the redox reaction, resulting in hydrogen and oxygen production. Efficient photocatalysts are expected to possess: (1) suitable band gaps and structures to absorb sunlight/UV light, leading to hydrogen- and oxygen-evolution half-reactions; (2) good charge transfer ability for electrons and holes, with low recombination rates; and (3) high surface area for catalytic activity. Fujishima and Honda first reported the successful use of TiO₂ anode and Pt cathode for solar-driven water splitting for hydrogen production [1]. In 1979, Bard designed a water splitting system that operates photocatalytically, using particles/powders as semiconductor photocatalysts [2]. PEC water splitting is considered the primary approach and TiO₂ is the best choice of semiconductor for PEC water splitting [3][4]. TiO₂ band gap is 3.2 eV, hence it is difficult to absorb visible and infrared light for solar water splitting, this is why metal or non-metal ion doping has been involved in narrowing down the band gap of TiO₂, so that TiO₂ is also functional under visible light [3][5]. C-doped TiO₂ nanocrystalline films possess high water splitting performance with enhanced conversion efficiency (11%) and photoconversion efficiency (8.35%), besides these credentials, they were active under visible light, which was an added advantage [6]. Grimes et al. demonstrated TiO₂ nanotube arrays for PEC water splitting yielding a photoconversion efficiency of 16.5% under UV light. The nanotube system owing to its nanotubular architecture, achieves superior electron lifetime and enhanced charge separation [7][8][9][10][11][12][13]. TiO₂ and fluorine-doped tin dioxide (SnO₂:F, and FTO), which is commonly used for preparing transparent conductive oxides (TCO), has been reported for their contribution in PEC cells [14]. ZnO is yet another popular wide band gap semiconductor, predominantly used for PEC water splitting applications [15][16]. Ion doping [17][18][19] and visible light sensitization with narrow band gap semiconductors [20][21][22] have expanded the light absorption range and improved the performance of PEC. ZnO nanostructures were doped with shallow Al donor levels, with added Ni for improved optical absorption [23]. Oval core/shell α -Fe₂O₃ nanorod nanoarrays, modified with thin WO₃/TiO₂ overlayers, have been reported to result in enhanced photo efficacy [24]. Other authors controllably tuned the ZnIn₂S₄ microstructure for enhanced visible light-mediated hydrogen evolution [25][26][27]. In the past decades, innumerable reports have addressed the critical requirements of photocatalysts [5][28][29][30][31][32].

Nanomaterials such as CdS, SiC, CuInSe₂, and TiO₂ have been used for photocatalytic hydrogen production [4][33][34][35] and demonstrated for their enhanced photocatalytic properties. Currently, Nb₂O₅ [36], Ta₂O₅ [37], α -Fe₂O₃ [38][39], ZnO [15][16], TaON [40], BiVO₄ [41][42], and WO₃ nanomaterials have been explored [43]. In most of these, band gap limitation can lower H₂ production [44]. To resolve this issue, noble metal/ion doping, sensitization and metal ion implantation techniques have been attempted. In noble metal doping, Pt is the best; but it is extremely expensive, so Ag, Ru, Pd, Ni, Cu, and Ir have been explored in parallel [45][46][47][48][49][50][51][52][53]. Incorporation of co-catalysts with photocatalyst nanomaterials for photocatalytic hydrogen production has also been attempted.

Loading cocatalysts onto photocatalysts to lead to hydrogen or oxygen evolution sites has enhanced photocatalytic splitting of water. In the past, transition metals, metal oxides, metal sulfides and noble metals, such as Pt, Ru, Au, and metal oxides, such as NiO_x, Rh/Cr₂O₃, etc., were well utilized as water reduction cocatalysts by entrapping electrons [5]

[29]. IrO₂, RuO₂, Rh₂O₃, Co₃O₄, and Mn₃O₄ metal oxides have been able to function as effective oxidation cocatalysts by entrapping the holes [28]. Researchers have loaded noble metals and metal sulfides as dual cocatalysts (Pt–Ag₂S and Pt–CuS), which could result in efficient separation of photogenerated electrons and holes for enhanced hydrogen evolution [54][55].

2. Nanomaterials Used in Hydrogen Storage

Various hydrogen storage systems have been explored for hydrogen storage applications [56][57][58][59][60][61][62][63][64]. These include metal hydrides, complex hydrides, chemical hydrides, adsorbents and nanomaterials (nanotubes, nanofibers nanohorns, nanospheres, and nanoparticles), clathrate hydrates, polymer nanocomposites, metal organic frameworks, and others [56][57][58][59][60][61][64][65][66][67]. However, as mentioned earlier, none of the currently available materials meet all these requirements, and the hydrogen content, release temperature and reversibility requirements are especially hard to meet. The other major option is solid-state hydrogen storage in light metal hydrides [68][69][70][71][72][73][74] and complex hydrides such as alanates [75][76], amides [77][78], borohydrides [78][79][80] and their combinations [81][82]. An offset of light metal hydrides are the alkali/alkali earth metal hydrides NaH, LiH, and MgH₂. Interstitial, or metallic, hydrides such as PdH_x are formed by transition and rare earth elements. Covalently bound hydrides such as AlH₃ and NH₃BH₃ are also used, but have their own limitations. Recently, the focus has been more on boron hydrides such as LiBH₄, alanates such as NaAlH₄, and even systems containing multiple phases, such as LiBH₄+MgH₂. Yet, most of these store 5 wt.% hydrogen and face kinetics and reversibility issues because of its complex nature and the presence of multiple phases after dehydrogenation. This being the case, the other alternative method of increasing the hydrogen sorption kinetics is nanostructuring. Stable crystallites of 5–10 nm were reported in a MgH₂TiH₂ system [83], smaller particles with sizes less than 10 nm have also been used. In 2005, the breakthrough pioneering work on nanoconfined borane in mesoporous silica enabled major changes in their hydrogen desorption properties, paving the way for a new beginning [84]. Additional effects have been identified, such as better mechanical stability and thermal management during cycling via incorporating carbon materials [85][86][87].

Carbonaceous materials are an attractive option for hydrogen storage owing to its adsorption ability, high specific surface area, pore microstructure, and low mass density. Despite numerous reports on hydrogen uptake by carbon materials, the actual mechanism of storage remains a mystery. The interaction is possibly based on van der Waals attractive forces (physisorption) or by chemisorption. The physisorption of hydrogen, limits the hydrogen-to-carbon ratio restricted to less than one hydrogen atom per two carbon atoms (i.e., 4.2 mass %). In chemisorption, this is realized as in the case of polyethylene [88][89][90]. Dillon et al. presented the first report on hydrogen storage in carbon nanotubes [91], which activated ripples worldwide in carbonaceous materials research. Now, it is known that hydrogen can be physically adsorbed on activated carbon and be “packed” more densely on the surface and inside the structure of carbon, as if it is compressed. The best results using carbon nanotubes, are verified to correspond to a hydrogen storage density of about 10% of the nanotube weight [92].

Fullerenes are currently one of the most popular carbon allotropes with a close-caged molecular structure [93]. They are able to react with hydrogen via the hydrogenation of carbon–carbon double bonds and so have been used for hydrogen storage. A maximum number of nearly 60 hydrogen atoms can be attached inside (endohedrally) and outside (exothermally) the spherical fullerene surfaces. Thus, a stable C₆₀H₆₀ isomer is obtained with a theoretical hydrogen content of ~7.7 wt.%. It seems that the fullerene hydride reaction is reversible at high temperatures. The 100% conversion of C₆₀H₆₀ indicates that 30 moles of H₂ gas will be released from each mole of fullerene hydride compound, but this reaction requires high temperatures ranging from 823–873 K [94][95].

Hydrogen can also be stored in glass microspheres of approximately 50 μm diameter. These microspheres can be loaded with H₂ through heating these glass microspheres to increase their permeability to hydrogen. A pressure of approximately 25 MPa, resulting in a storage density of 14% mass fraction and 10 kg H₂/m³ is reported [94]. At 62 MPa, a bed of glass microspheres can store 20 kg H₂/m³. The release of hydrogen is through reheating the spheres, which increases the permeability of hydrogen. Carbon-based sorbents, synthesized from various organic precursors, can be structured into various carbon forms, such as carbon nanotubes [96][97][98], fibers [96][98], fullerenes [98][99], and activated carbons [100][101]. These structurally diverse forms can be tuned for hydrogen gas storage. Metal–organic frameworks (MOFs) are highly porous, crystalline solids consisting of a periodic array of metal clusters linked through multi-topic organic struts [102][103]. Other highly porous, crystalline materials include zeolitic imidazolate frameworks (ZIFs) [104] and covalent organic frameworks (COFs) [105], which have also been considered as an option for hydrogen storage.

Nanocomposites consist of a polyaniline matrix that can be functionalized by catalytic doping or incorporation of a nanovariant. It has been reported that polyaniline can store 6–8 wt.% of hydrogen [106] and a study revealed a successful

hydrogen uptake of 1.4–1.7 wt.% [107]. With all this in mind, there is still an urgent need for the development of new reversible materials. Clathrates are a new class of materials for hydrogen storage [58], which are primarily hydrogen-bonded H₂O frameworks, where hydrogen molecules can be incorporated, making it useful for off-board storage of hydrogen.

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