

Advanced Carbon-Metal Oxide Nanocomposites for Sustainable Solar Energy Conversion and Environmental Remediation

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Abstract

Sunlight is abundant and free, and a photocatalyst that uses it to make fuel or to break down a pollutant addresses two problems at once: the need for clean energy and the need for clean water and air. Metal oxide semiconductors such as TiO_2 , ZnO and Fe_2O_3 are the workhorses of photocatalysis because they are cheap, stable and non-toxic, but three faults limit them. Most absorb only ultraviolet light, which is a small slice of the solar spectrum; the electron-hole pairs they generate recombine before they can react; and fine powders are hard to recover and reuse. Coupling the oxide to a carbon phase addresses each fault. Graphene, reduced graphene oxide, carbon nanotubes, carbon quantum dots and graphitic carbon nitride act as electron acceptors and conductive highways that pull photogenerated electrons away from the oxide and suppress recombination, they adsorb pollutants and concentrate them at the reaction site, and several of them absorb visible light or sensitise the oxide to it. This review collects the current understanding of these carbon-metal oxide nanocomposites for solar energy conversion and environmental remediation. We set out the photocatalytic fundamentals and the specific jobs the carbon phase does, the heterojunction schemes (type-II, Z-scheme, S-scheme and Schottky) that govern charge separation, and the synthesis routes used to build the composites. We then survey performance across solar hydrogen production, CO_2 photoreduction to fuels, and photovoltaic and photoelectrochemical devices, followed by environmental applications in dye and organic-pollutant degradation, water disinfection, and magnetically recoverable systems. We close with the problems that still limit deployment: visible-light efficiency, long-term stability and reuse, product selectivity in CO_2 reduction, and the gap between milligram-scale tests and real treatment volumes.

Keywords: photocatalysis; carbon-metal oxide nanocomposite; solar energy conversion; hydrogen evolution; CO_2 reduction; environmental remediation; dye degradation; heterojunction

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1. Introduction

Two of the century's hardest problems, securing clean energy and cleaning up polluted water and air, share a possible answer in a single technology. A semiconductor photocatalyst absorbs light, generates mobile charges, and uses them to drive chemical reactions: splitting water into hydrogen, reducing carbon dioxide to fuels, or oxidizing an organic pollutant to harmless products. When the light is sunlight, the process runs on a free and renewable input and produces no secondary waste of its own, which is why photocatalysis is treated as a green route to both energy and remediation [1,4]. Metal oxide semiconductors have carried this field for decades. TiO_2 , ZnO , Fe_2O_3 , WO_3 and related oxides are inexpensive, chemically robust, non-toxic and easy to make, and TiO_2 in particular remains the reference material. Their limitations are equally well known. Most wide-gap oxides absorb only ultraviolet light, which is about 5% of the solar spectrum, so they waste most of the sunlight that reaches them. The electron-hole pairs they generate recombine within nanoseconds, wasting the absorbed energy as heat before it can do chemistry. And as fine suspended powders they are difficult to separate from treated water and to reuse [1,3].

Combining the oxide with a carbon phase attacks all three faults at once, which is what has made carbon-metal oxide nanocomposites one of the busiest areas in photocatalysis [2,3]. A conductive carbon such as graphene accepts the photogenerated electron from the excited oxide and carries it away, so the electron and hole are separated in space and recombine much less; this single effect often doubles or better the measured activity [1,4]. Carbon phases also adsorb organic pollutants onto their large surface, concentrating the target where the reactive species are made, and several of them, carbon quantum dots and graphitic carbon nitride in particular, absorb visible light themselves and extend the composite's response across more of the solar spectrum [5,7]. This review is organized around those ideas. Section 2 sets out the photocatalytic fundamentals and what each type of carbon contributes. Section 3 covers synthesis. Section 4 surveys solar energy conversion, and Section 5 environmental remediation. Section 6 is a candid account of the open problems, and Section 7 concludes.

2. Photocatalytic fundamentals and the role of the carbon phase

2.1 How a semiconductor photocatalyst works

A photocatalyst works because a photon with energy

above the semiconductor's band gap promotes an electron from the valence band to the conduction band, leaving a positively charged hole behind. The excited electron is a strong reductant and the hole a strong oxidant, and if they reach the particle surface before recombining, they drive redox reactions there. In water and air the most important products are reactive oxygen species: holes oxidise water or hydroxide to hydroxyl radicals, and electrons reduce dissolved oxygen to superoxide, and these radicals are what actually break down most pollutants [4,5]. The same charges, directed differently, reduce protons to hydrogen or carbon dioxide to fuels. The band gap sets which wavelengths the material can use, and the band positions set which reactions the electrons and holes are energetic enough to drive.

2.2 Why bare metal oxides fall short

Three properties limit a bare oxide. The band gap of the common oxides is wide, around 3.0 to 3.2 eV for TiO_2 and ZnO , so they respond only to ultraviolet light and ignore the visible bulk of sunlight.

Recombination is fast, so only a fraction of the generated charges ever reach the surface to react. And recovery is awkward, because a nanopowder dispersed in treated water is expensive to filter out and easy to lose. Doping, facet engineering and coupling to other semiconductors have all been used to address these faults, and pairing with a carbon phase has become one of the most effective and general of these strategies [1,3].

2.3 What each carbon phase contributes

The carbon component is chosen for what it adds. Graphene and its derivatives, graphene oxide and reduced graphene oxide, are the most used: their high conductivity and large surface make them excellent electron sinks and transport layers, so they draw electrons off the excited oxide and suppress recombination, and their surface adsorbs organic pollutants and brings them to the reaction site [2,3]. Carbon nanotubes play a similar conductive role in one dimension. Carbon quantum dots add two functions bare graphene lacks: they absorb visible light and can sensitise the oxide to it, and they can act as up-conversion or electron-shuttle centres, so decorating an oxide with carbon dots raises both light harvesting and charge separation [6,7,10]. Graphitic carbon nitride is different again, a visible-light semiconductor in its own right with a band gap near 2.7 eV, so pairing it with an oxide builds a genuine two-semiconductor junction rather than a semiconductor-plus-conductor contact [4,5]. Activated carbon, cheap and highly porous, is used mainly as an adsorbent scaffold that also allows magnetic or oxide components to be anchored [19].

2.4 Heterojunction schemes and charge transfer

How the two phases are joined at the interface decides how well charge separates, and several

junction types are used. In a conventional type-II heterojunction, electrons and holes move in opposite directions to the two components, separating the charges but at the cost of some redox power. The Z-scheme and the newer S-scheme (step-scheme) were developed to keep the strong reductant and strong oxidant while still separating charge: they recombine the less useful electron and hole across the interface and retain the most energetic ones, which is why S-scheme oxide-carbon-nitride composites reach high activity in both water splitting and CO_2 reduction [4,11]. Where a metal is present, a Schottky junction at the metal-semiconductor contacts traps electrons, and plasmonic metal nanoparticles such as silver or gold add visible-light absorption through localised surface plasmon resonance, as used in silver-decorated TiO_2 -graphene photoanodes [13]. The graphene or carbon phase frequently serves as the conductive mediator that makes these schemes work, shuttling charge between the two semiconductors [2,4].

3. Synthesis and design of the nanocomposites

The synthesis route fixes how intimately the oxide and carbon are joined, which in turn fixes how well charge crosses between them, so the interface is the real target of these methods. Hydrothermal and solvothermal routes are the most common: a metal precursor and a carbon source, often graphene oxide, are heated in a sealed vessel so the oxide nucleates and grows directly on the carbon sheets, and the same step frequently reduces graphene oxide to reduced graphene oxide, giving a well-bonded interface in one operation. This is how the rGO- TiO_2 composites used for CO_2 reduction and the defect-rich rGO/ TiO_2 -x used for hydrogen evolution were made, the latter in a single one-pot hydrothermal step that reduced the graphene oxide and introduced oxygen vacancies into the TiO_2 at the same time [8,9]. Thermal and pyrolytic routes build the composite by decomposing a precursor: graphitic carbon nitride is made by heating melamine, and metal-organic frameworks are pyrolysed into oxide-in-carbon composites, as in the MOF-derived $\text{TiO}_2/\text{g-C}_3\text{N}_4$ heterojunction and the ZIF-8-derived ZnO -carbon on reduced graphene oxide [15,20].

Sol-gel, co-precipitation and electrospinning are used where particle size or fibre morphology must be controlled. Green synthesis is a growing branch, using plant extracts as reducing and capping agents to make oxide and carbon composites without harsh chemicals, and agricultural wastes as the carbon source, as in the leaf-extract route to reduced graphene oxide- ZnO and the coconut-shell activated carbon used in magnetic TiO_2 catalysts [17,19].

4. Solar energy conversion

4.1 Photocatalytic hydrogen production and water

splitting

Splitting water with sunlight to make hydrogen is the headline goal, because it stores solar energy in a clean fuel. Carbon-metal oxide composites raise the hydrogen rate mainly by suppressing recombination and extending light absorption. A defect-rich reduced graphene oxide-TiO₂-x composite produced hydrogen at

13.6 mmol per hour per gram under solar illumination and 947 micromol per hour per gram under visible light alone, with the graphene suppressing recombination and the oxygen vacancies extending absorption into the visible [9]. Carbon quantum dots have a large effect at low loading: decorating iron oxide nanowires with carbon dots raised the photocurrent for water oxidation many times over, and nitrogen- and sulfur-doped carbon dots on a TiO₂ photoanode lifted the photoelectrochemical water-oxidation current to about

2.1 mA per square centimetre, roughly 1.3 times bare TiO₂, by improving charge injection [7,10]. Two-semiconductor designs push further: an S-scheme heterojunction of cadmium selenide quantum dots on polymeric carbon nitride reached a hydrogen evolution rate above 20 mmol per hour per gram while keeping strong redox power [11].

4.2 Carbon dioxide photoreduction to solar fuels

Reducing carbon dioxide with sunlight does double duty, storing energy in a fuel and consuming a greenhouse gas, but it is harder than water splitting because the reaction is slow and gives a mixture of products. Carbon-metal oxide composites help by separating charge and, in some cases, by adsorbing carbon dioxide on the carbon surface near the active sites. A reduced graphene oxide-TiO₂ composite reduced carbon dioxide to methane with higher activity than either graphite oxide or bare anatase, the graphene improving both charge separation and visible response [8]. ZnO quantum dots on potassium niobate nanosheets raised the methanol production rate over the bare nanosheets by promoting charge separation [12]. The same S-scheme cadmium selenide-carbon nitride composite that produced hydrogen also reduced carbon dioxide to carbon monoxide, showing that one well-designed junction can serve both solar-fuel reactions [11]. Selectivity toward a single useful product remains the central difficulty.

4.3 Photovoltaic and photoelectrochemical devices

Beyond powder photocatalysis, carbon-metal oxide composites improve devices that convert light to electricity or to a driven electrochemical reaction. In dye-sensitised solar cells the photoanode is usually mesoporous TiO₂, and adding reduced graphene oxide improves electron transport while a plasmonic metal extends absorption: a silver-decorated TiO₂-reduced graphene oxide photoanode reached a

power conversion efficiency of about 7.3%, close to 1.8 times the plain TiO₂ cell, and the same electrode also degraded pollutants, linking energy harvesting and remediation in one material [13]. In photoelectrochemical cells the carbon-dot-modified TiO₂ photoanodes discussed above raise photocurrent and charge-injection efficiency, which is the same physics applied to a wired electrode rather than a suspended powder [10]. Table 1 collects representative solar-conversion results.

Composite	Application	Reported performance	Ref.
rGO/TiO ₂ -x (defect-rich)	H ₂ evolution	13.6 mmol h ⁻¹ g ⁻¹ (solar); 947 μmol h ⁻¹ g ⁻¹ (vis)	[9]
NS-carbon dots / TiO ₂	PEC water oxidation	~2.1 mA cm ⁻² at 1.23 V (1.3x bare TiO ₂)	[10]
CdSe QDs / carbon nitride (S-scheme)	Water splitting + CO ₂	H ₂ >20 mmol h ⁻¹ g ⁻¹ ; CO production	[11]
rGO-TiO ₂	CO ₂ reduction	CH ₄ above graphite oxide and anatase	[8]
ZnO QDs / KNb ₃ O ₈	CO ₂ to methanol	Methanol rate raised over bare nanosheets	[12]
TiO ₂ / rGO / Ag (plasmonic)	Dye-sensitised solar cell	PCE ~7.3% (~1.8x bare TiO ₂)	[13]

Table 1. Representative solar-energy-conversion results for carbon-metal oxide nanocomposites. Values are from the cited studies under each study's own conditions and are not directly comparable across rows. PEC = photoelectrochemical; PCE = power conversion efficiency; QD = quantum dot.

5. Environmental remediation

5.1 Degradation of dyes and organic pollutants

The largest body of work is on breaking down organic pollutants, especially the colored dyes that textile and other industries discharge into water. The mechanism is the reactive-oxygen chemistry described earlier: hydroxyl radicals and holes attack the dye and mineralise it, ideally to carbon dioxide and water. Carbon-metal oxide composites consistently beat their bare-oxide parents because the carbon suppresses recombination and adsorbs the dye. A NaTiO₃-graphitic carbon nitride heterojunction completely degraded methylene blue within 25 minutes under visible light [14]. An eco-friendly reduced graphene oxide-ZnO composite reached about 99% methylene blue removal under sunlight, above the 96.5% of bare ZnO and far above

the 66% of reduced graphene oxide alone [17]. A functionalized graphene-TiO₂-strontium hexaferrite catalyst degraded methylene blue and rhodamine B under visible light through the synergy of the ternary junction [16], and a defect-rich TiO₂-graphitic carbon nitride heterojunction handled the harder case of phenolic wastewater with good reusability [15]. Reviews of graphitic carbon nitride and its heterojunctions, and of newer MXene and metal-sulfide couplings, report the same pattern across many dye systems [5,23,24].

5.2 Adsorption and combined adsorption-photocatalysis

Carbon phases contribute a second remediation mechanism that runs alongside photocatalysis: adsorption. Their large surface area pulls pollutants out of solution and holds them close to the catalytic sites, so removal is faster than photocatalysis alone would give, and the two effects reinforce each other. Depositing graphene oxide-ZnO on a porous polyurethane foam, for example, combined the foam's high adsorption capacity with the photocatalyst, so a large fraction of the dye was captured on the surface before being degraded [21]. In practice the adsorbed pollutant is then mineralised under light, which regenerates the surface, so a good carbon-oxide composite is both a sorbent and a catalyst [2].

5.3 Water disinfection

The same reactive oxygen species that break down dyes also kill microorganisms, so carbon-metal oxide photocatalysts double as disinfectants. Composites of iron oxide, ZnO and carbon dots have inactivated bacteria such as *Escherichia coli* under light, with the carbon dots improving light trapping and charge transport and so the yield of the radicals that damage cell membranes [22]. This gives a single material that can both detoxify chemical pollutants and disinfect water, which is attractive for point-of-use treatment.

5.4 Magnetic separation and reuse

A photocatalyst is only practical if it can be recovered and reused, and this is where a magnetic oxide earns its place in the composite. Adding a magnetic phase such as Fe₂O₃ or a magnetic ferrite lets the catalyst be pulled out of treated water with a magnet rather than filtered, then redispersed for the next batch. A reduced graphene oxide-zinc copper ferrite catalyst removed about 95% of methylene blue and was recovered magnetically across repeated cycles, though its activity fell to about 55% by the fifth cycle, which is typical and points to the durability problem [18]. A magnetic Fe₃O₄-activated carbon-TiO₂ catalyst degraded about 98% of methylene blue and survived seven cycles with little loss, the activated carbon adding adsorption and the magnetite adding recoverability [19].

Reusability data of this kind, rather than a single high first-pass number, are what indicate practical value. Table 2 collects representative remediation results.

Composite	Target / mode	Reported performance	Ref.
NaTiO ₃ / g-C ₃ N ₄	Methylene blue (visible)	Complete degradation in 25 min	[14]
rGO@ZnO (green synthesis)	Methylene blue (sunlight)	~99% (vs 96.5% ZnO, 66% rGO)	[17]
TiO ₂ -graphene / Sr-hexaferrite	MB and rhodamine B (visible)	Enhanced degradation via ternary junction	[16]
TiO ₂ / g-C ₃ N ₄ (defect-rich)	Phenolic wastewater	High degradation; reusable and stable	[15]
rGO / ZnCuFe ₂ O ₄ (magnetic)	Methylene blue; magnetic reuse	~95% removal; ~55% after 5 cycles	[18]
Fe ₃ O ₄ / AC / TiO ₂ (magnetic)	Methylene blue; magnetic reuse	~98% removal; stable over 7 cycles	[19]

Table 2. Representative environmental-remediation results for carbon-metal oxide nanocomposites. Conditions differ between studies; values indicate reported performance, not a controlled comparison. MB = methylene blue; AC = activated carbon.

6. Open problems and future directions

Several specific problems separate these encouraging results from large-scale use, and naming them precisely is more useful than a general call for further study.

Visible-light efficiency. Much of the solar spectrum is still wasted, because the workhorse oxides absorb only ultraviolet and the carbon phase alone cannot fully fix that. The strongest route is a genuine visible-light heterojunction, an S-scheme oxide-carbon-nitride or a carbon-dot-sensitised oxide, rather than a semiconductor-plus-conductor contact, and reporting apparent quantum yield at a stated wavelength rather than a bare degradation percentage would let these designs be compared honestly [4,11].

Stability and reuse. Activity that falls by half over five cycles, as several magnetic catalysts show, is not good enough for a treatment plant. Photocorrosion of the oxide, fouling of the carbon surface and loss of catalyst on each recovery all need long-run testing, and durable recovery, ideally magnetic, should be reported over many cycles rather than a few [18,19].

Selectivity in CO₂ reduction. Carbon dioxide photoreduction gives a mixture of carbon monoxide, methane, methanol and hydrogen, and steering it toward one useful product remains the central obstacle to solar fuels. Interface and single-site design, guided by operando spectroscopy, is the route to selectivity rather than to raw rate alone [11,12].

From the flask to real volumes. Most results come from milligram catalyst loads degrading millilitres of a single model dye under a lamp. Real effluent is a mixture at variable pH with competing species, and treating it at volume needs immobilised or monolithic catalyst forms, realistic reactor design, and testing against mixed pollutants under actual sunlight [2,3].

Understanding and predicting the interface. The charge-transfer path in these composites is often inferred rather than measured, and the active site is not always clear. Operando characterization and computational screening of oxide-carbon pairings would move the field from trial-and-error toward designed interfaces [1,4].

7. Conclusions

Carbon-metal oxide nanocomposites work because the carbon fixes the three chronic faults of metal oxide photocatalysts. A conductive carbon such as graphene draws off photogenerated electrons and suppresses recombination, carbon phases adsorb pollutants and concentrate them at the reaction sites, and visible-active carbons such as carbon dots and graphitic carbon nitride extend the response across more of the solar spectrum, while a magnetic oxide makes the catalyst recoverable. Across solar hydrogen production, CO₂ photoreduction, photovoltaic and photoelectrochemical devices, and the degradation, disinfection and magnetic-recovery tasks of environmental remediation, these composites consistently outperform their bare-oxide parents.

What stands between these results and deployment is specific: better use of visible light through genuine heterojunctions rather than conductor contacts; durability and recovery proven over many cycles; product selectivity in CO₂ reduction; and the step from model dyes in a flask to real effluent at volume. Progress on those points, rather than incremental gains in a single degradation number, is what will turn these materials into working solar and environmental technologies.

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Declarations

Conflicts of interest: The authors declare no competing interests.