Annual Methodological Archive Research Review
http://amresearchreview.com/index.php/Journal/about
Volume 4, Issue 3 (2026)

Online ISSN 3007-3197
Print ISSN 3007-3189

http://amresearchreview.com/index.php/Journal/about

Bio-Inspired Photocatalysts for Sustainable Organic Synthesis: Design Principles, Photophysical Mechanisms, Light-Matter Interactions, and Green Catalysis

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ABSTRACT

Photocatalysis provides access to radical, radical ion, and electronically excited intermediates without relying on high temperatures or stoichiometric redox reagents. However, visible light operation alone does not guarantee environmental sustainability, highlighting the need for a structure-photon-function-sustainability framework to evaluate bio-inspired photocatalysts. Productive performance relies on the coordinated management of photon absorption, excitation energy migration, charge separation, interfacial charge transfer, catalytic turnover, and reactor-scale photon transport. Natural photosynthesis supplies the functional design for this coordination through antenna networks, directional electron transfer chains, spatially separated redox sites, proton-coupled processes, compartmentalization, and self-repair. These biological principles are closely connected to molecular and supramolecular photocatalysts, porphyrin-, flavin- and quinone-inspired systems, peptide and protein assemblies, bio-

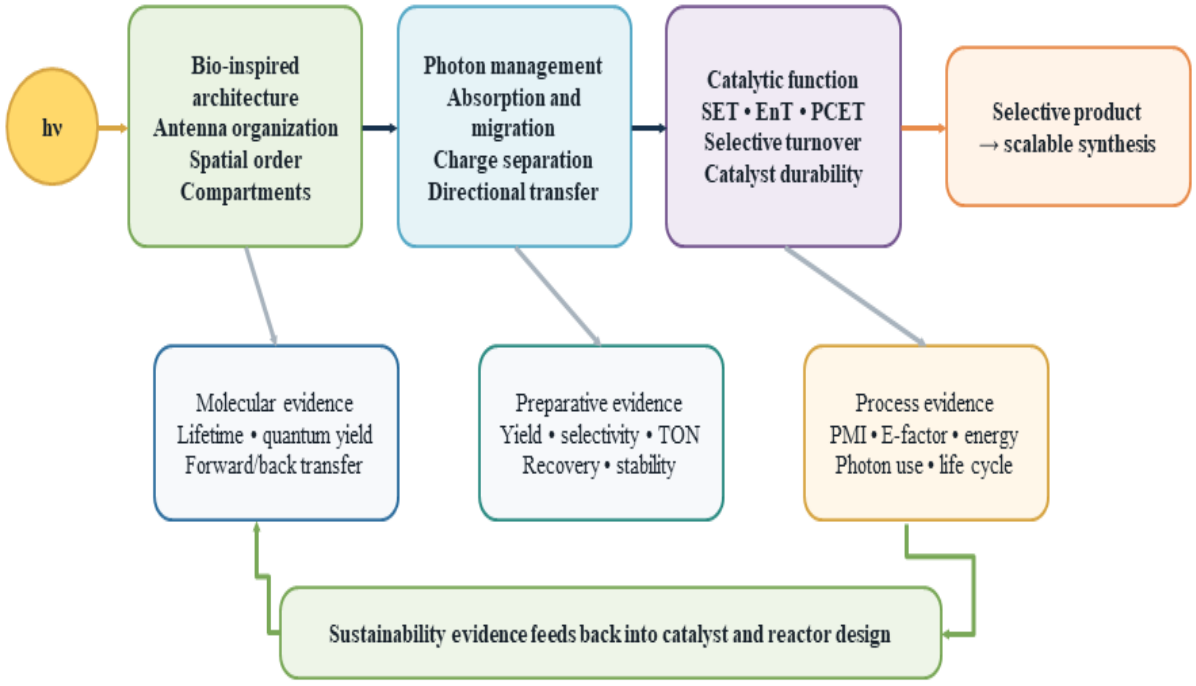
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Online ISSN 3007-3197
Print ISSN 3007-3189
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derived carbons, metal-organic and covalent organic frameworks, and biohybrid platforms. Mechanistic pathways are correlated with synthetic applications, including selective oxidation and reduction, C-C and C-heteroatom bond formation, biomass valorization, CO_2 incorporation, and cascade reactions. These are further evaluated against critical sustainability metrics catalyst abundance, solvent selection, photon and electrical efficiency, recyclability, process mass intensity, E-factor, and life-cycle boundaries. The central conclusion is that the principal obstruction is not a shortage of photoactive materials, but rather a lack of integration among mechanistic evidence, catalyst durability, photon accounting, reactor engineering, and sustainability assessment. Finally, design rules and reporting priorities are proposed to translate biological inspiration into scalable, environmentally credible organic synthesis.

Keywords: Bio-Inspired Photocatalysis; Visible-Light Photoredox Catalysis; Artificial Photosynthesis; Charge Separation; Sustainable Organic Synthesis; Green Metrics

Graphical abstract



Bio-inspired photocatalyst design linking synthetic efficiency with environmental sustainability

Introduction

The synthesis of pharmaceuticals, agrochemicals, functional materials and commodity chemicals are never apart from modern society, but conventional methods still depend on fossil-derived feedstocks, rare metals, hazardous solvents, multistep operations and high temperature. Yield is therefore an incomplete measure of process quality [1]. A

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route that gives nearly quantitative conversion can still generate a large environmental burden through protecting groups, excess reagents, dilute operation, solvent-intensive work-up or energy demanding purification, green chemistry instead requires a whole process view including atom economy, step economy, toxicity, catalyst production, energy consumption, solvent recovery, waste treatment and end of life management [1, 2]

In photocatalysis, photon energy is used to partially replace the thermal or chemical activation of a reaction. A catalyst's redox and energy transfer characteristics are temporarily altered by absorption, providing selective access to radical ions, neutral radicals, triplets, and electron-hole pairs [3]. As a result, under very mild conditions, visible light photoredox catalysis has increased synthetic space through oxidation, reduction, cross coupling, C-H functionalization, cycloaddition, and asymmetric transformations [3, 4]. Additionally, it naturally interacts with organocatalysis, electrocatalysis, biocatalysis, and transition metal catalysis [4].

Mild irradiation alone does not make an operation sustainable. Photons are formally traceless, but a photon production and delivery system are needed and for artificial light, electric power is needed. A large number of reactions are low concentration, with donors/acceptors in stoichiometric amounts, require precious metal photosensitizers, and end with chromatographic purification. The environmental benefits of a photocatalytic approach are not just the reduction of synthesis time, but also the elimination of toxic chemicals, the increase in selectivity, or the ability to use a large amount of feedstock that is not used in other synthesis pathways [5, 6].

The natural photosynthesis process is more challenging to model. It distributes light capture, excitation migration, charge separation, redox accumulation and catalysis among spatially organized components [7]. Protein scaffolds and membranes are used to regulate distances, orientations, polarity and proton pathways and repair and regulation maintain function under varying sunlight. The term “bio-inspired” is used for systems that emulate such functionality, but not necessarily of biological origin [8]. Thus, biomimetic chromophores, supramolecular donor-acceptor arrays, compartmentalized catalysts, peptide assemblies, bio-derived carbons, porous frameworks and semiconductor enzyme hybrids are all included in the category [7, 8].

In the existing reviews, biological inspiration, excited state mechanism, photocatalytic materials and organic synthesis and green metrics are usually considered as independent topics. This separation makes the causal relationship between architecture and charge dynamics, charge dynamics and selectivity, and selectivity and process sustainability difficult to see [7]. An integrated structure-photon-function-sustainability framework is the novelty of the present review. It reveals the practical design rules, namely the antenna organization, directional transfer, confinement, coupled proton and electron motion and adaptive structures, and it reveals the interchanges that are not apparent when dealing with qualitative green claims [9]. In addition to the photon transport, catalyst lifetime, material intensity, and hazard and scale-up, molecular performance is evaluated with bio-inspired activity to be compared with an environmentally credible synthetic function [1].

The review is structured in a causal sequence instead of a list of different classes of catalysts. First, the rules of biological organization are translated into molecular and

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mesoscale design rules [9]. These rules are then applied to the excited state mechanism and the interaction with light and matter, and their consequences for preparative organic synthesis. The green metrics and process boundaries are applied once the chemical function is defined, so that one can make a fair comparison at the same product quality [10]. This sequence is followed by challenges and future directions that are generated from failures. This design allows the distinction of an architectural feature that is truly biomimetic in terms of controlling photon or charge flow from one that is only biomimetic in appearance [9].

The critical assessment is based on three questions. Is there a measurable mechanistic benefit from biological inspiration, for example faster forward transfer, reduced recombination or better substrate orientation [9]? Does this benefit hold true for preparative turnover and scale-up, where optical gradients and impurities and catalyst change are significant? Does the overall process remove material, hazard or energy burdens compared to a viable alternative [1]? A positive answer to the first question alone delivers interesting photophysics, not sustainable synthesis. The novelty of the review is that it asks for the three types of evidence and it specifies the experiments required to link them together [9].

Natural Blueprint and Bio-Inspired Design Rules

Natural photosynthesis accomplishes three goals harvesting a large solar spectrum, building an adequate charge separated state with a long lifetime and steering the resulting redox equivalents to catalysis. Antenna pigments enhance the effective absorption cross section of a smaller reaction center, and controlled spacing of the pigments in the antenna enables the migration of excitation energy without all the absorbers need to engage in charge separation [11]. Excitation is converted to vectorial electron transfer by reaction centers, and oxidation and reduction chemistry separated by membranes. Kinetic coordination is the transferable lesson; absorption is only useful when energy migration, charge separation, and catalytic turnover happen more rapidly than relaxation and recombination [7, 11].

Both composition and spatial organization are important. A good forward transfer occurs when the donor-acceptor distance is not very large while back electron transfer can happen in a similar way or the excited state can be quenched if the coupling is too strong. Hydrogen bonding, π stacking, electrostatics, host guest recognition or metal coordination provide means for supramolecular assembly that control distance and orientation and maintain dynamics required to enter substrates and remove products. Coordinating cages and other coordinating hosts are also able to pre concentrate substrates and stabilize high energy intermediates. However, trapping is counterproductive if the pores exclude the reagents, the products bind too tightly, or if the chromophores are aggregated as a result of the dense packing in the pores [12].

Chromophores that are bio-inspired replicate functionalities rather than precise pigment structures. Strong visible absorption is provided by porphyrins and chlorins, while peripheral substitution modular metal coordination adjusts solubility, aggregation, and redox potential [13]. Flavins are promising for aerobic oxidation and metallaphotoredox chemistry, including aqueous systems, because they combine accessible excited state redox chemistry with hydrogen bonding and proton linked

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reactivity [13, 14].

Quinones act as electron-proton relay systems and have the ability to channel sensitized oxidation away from singlet oxygen pathways and into electron transfer processes [14]. Their drawback arises from the same property that makes them useful: the reactive redox states involved can undergo side reactions or irreversible degradation [13].

Programmable self-assembly, chiral environments and defined binding pockets are afforded by peptides and proteins. Short chromopeptides can form fibres or sheets, and proteins can almost molecularly precisely position sensitizers and catalytic cofactors [15]. Biohybrid platforms combine an abiotic absorber with an enzyme or whole cell to achieve efficient light capture and biological chemo, regio and stereoselectivity. Delivering electrons is still challenging when an enzyme active site is buried or not well coupled to the photosensitizer mediators increase accessibility, but cause diffusion losses and separation problems [16]. Biocatalysts can also be short lived due to oxidative stress and to photothermal heating [15, 16].

Bio-based carbon dots, graphene-like materials and biomass derived porous carbons provide tunable surface chemistry and aqueous compatibility and may have less critical element dependency. Their heterogeneity can help with adsorption and interfacial transfer, but makes mechanistic assignment difficult due to the different emissive states, defects, and active sites present depending on the precursor and synthesis [17]. The following are requirements for reporting in order to achieve reproducibility feedstock origin, thermal history, heteroatom content, particle size distribution and purification. When assessing the sustainability of a material, it is not enough if the source of the material is renewable; the energy used in the carbonization process, the low yield, and the poorly controlled release of nanoparticles must also be taken into the assessment boundary [17].

Light harvesting, charge transport and catalysis are modularly separated in metal–organic frameworks and covalent organic frameworks. Pores reorganize substrates close to the reactive sites and ordered donor–acceptor linkages can increase the absorption [18]. Immobilization can enhance recovery and establish oxidation and reduction domains. Despite this, the performance of the system is still dependent on crystallinity, defect density, linker stability, electronic coupling between nodes and linkers [19] etc. Apparent framework activity can come from leached complexes, photodeposited nanoparticles, or transformed surfaces [18, 19, 20] and therefore, operando characterization and solution-phase controls are essential.

In these classes, there are five structural levels that need to be co-optimized the electronic structure of the chromophore, the immediate coordination environment, donor-acceptor geometry, mesoscale morphology and organization at the level of the compartments. To conclude we can see from natural systems that a single “ideal” absorber is not enough [7]. Broad absorption must be followed by directional energy migration; the separated charges must reach selective catalytic sites; and the assembly must tolerate substrate and product exchange, impurities, and repeated excitation. The most widely applicable bio-inspired principle is functional division with controlled communication the biochemical components are specialised, but the distances, the redox potentials and the timescales are matched.

The absorption breadth is not the only criterion for antenna design. When the excitation

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energy has to be transferred in a useful assembly, it is necessary that the energy does not fall into low energy defects or excimer states [7]. Natural antennae have moderate electronic coupling and multiple partially redundant transfer paths to achieve robust transfer artificial arrays typically employ fewer chromophores, which results in greater sensitivity to a non-optimally oriented donor-acceptor pair [21]. Time resolved anisotropy, site selective spectroscopy and structure dependent kinetics can help to determine whether the added chromophores really expand the productive absorption cross section or only the optical density. The antenna effect proves to be useful only if the extra absorbed photons possess a certain "chemical potential" when they arrive at the reaction center [7, 21].

Compartmentalization is a means of managing mutually incompatible reactions. Communication may be regulated by membranes, pores, or redox mediators and oxidation/reduction reactions may take place in different microenvironments [8]. This separation may inhibit charge recombination and prevent the oxidation of enzymes or reduced products by an oxidative intermediate. It can also lead to new restrictions transport of mediators across an interface can become turnover limiting, local pH differences can alter the speciation of the catalysts, and the membrane resistance can decrease current [12]. Local concentrations and fluxes should therefore be measured, not compartment function, when designing. The appropriate comparison is the otherwise similarity system, with the same optical density and catalyst loading, that is uncompartmentalized [8, 12].

Adaptive behavior is also added by taking inspiration from the biological world. During operation, protein scaffolds reorganize, protonation states change and damaged parts are replaced [22]. Artificial catalysts tend to be optimized for the static state and are often only active in the dynamic state. Substrate driven reorganization may be possible by reversible coordination and non-covalent assembly, but too much lability results in component loss [23]. The sweet spot for design is between rigidity (to maintain electronic pathways) and controlled dynamics (to allow for turnover and recovery). Operando methods are needed to determine if an adaptive change restores the desired architecture or creates a chemically different catalyst [22, 23, 24]. These bio-inspired chromophore and material classes are summarized in **Table 2.1**.

Table 2.1. Representative bio-inspired chromophore and material classes for photocatalysis.

Class	Advantages	Limitations
Chlorins & porphyrins	Modular metal coordination, strong visible absorption, peripheral substitution tunes solubility, aggregation and redox potential	Aggregation and redox behavior must be adjusted carefully through substitution

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Online ISSN 3007-3197
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Class	Advantages	Limitations
Flavins	Accessible excited state redox chemistry with hydrogen bonding and proton coupled reactivity, effective for aerobic oxidation, metallaphotoredox and aqueous systems	Reactivity is sensitive to hydrogen bonding environment and proton availability
Quinones	Electron-proton relays can redirect sensitized oxidation from singlet oxygen toward electron transfer chemistry	The same reactive redox states can undergo side reactions or irreversible degradation
Peptides, proteins & biohybrids	Programmable self-assembly, chiral environment, defined binding pockets, biohybrids combine efficient light capture with biological chemo, regio, and stereoselectivity	Electron delivery is difficult when the active site is buried or poorly coupled, mediators add diffusion losses, oxidation stress and photothermal heating shorten biocatalyst life
Bio-derived carbons	Tunable surface chemistry, aqueous compatibility, lower dependence on critical elements	Heterogeneity complicates mechanistic assignments; reproducibility requires detailed precursor and synthesis reporting
Metal-organic & covalent organic framework	Modular separation of light harvesting, charge transport and catalysis, pores pre-organize substrates, immobilization can improve recovery	Performance depends on crystallinity, defects and linker stability, apparent activity may originate from leached species, requiring operando controls

Photophysical and Photochemical Mechanisms

Upon photoexcitation, a molecule is promoted vertically to a Franck-Condon state, which subsequently undergoes vibrational relaxation, internal conversion, fluorescence, intersystem crossing, electron transfer, or chemical reaction. Ensuring productive catalysis will depend on a reactive state whose lifetime must be sufficiently long to react with a substrate or to transfer energy in a pre-organized assembly [25]. Absorption coefficient is not enough to predict performance because a strongly absorbing state can have a long lifetime and not react to chemistry. However, a weakly absorbing triplet may dominate the reactivity if its lifetime and redox properties are such that its bimolecular quenching is favorable [25, 26]. **Fig. 3.1** shows photophysical

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Volume 4, Issue 3 (2026)

Online ISSN 3007-3197
Print ISSN 3007-3189
http://amresearchreview.com/index.php/Journal/about

pathways from photoexcitation to catalytic turnover.

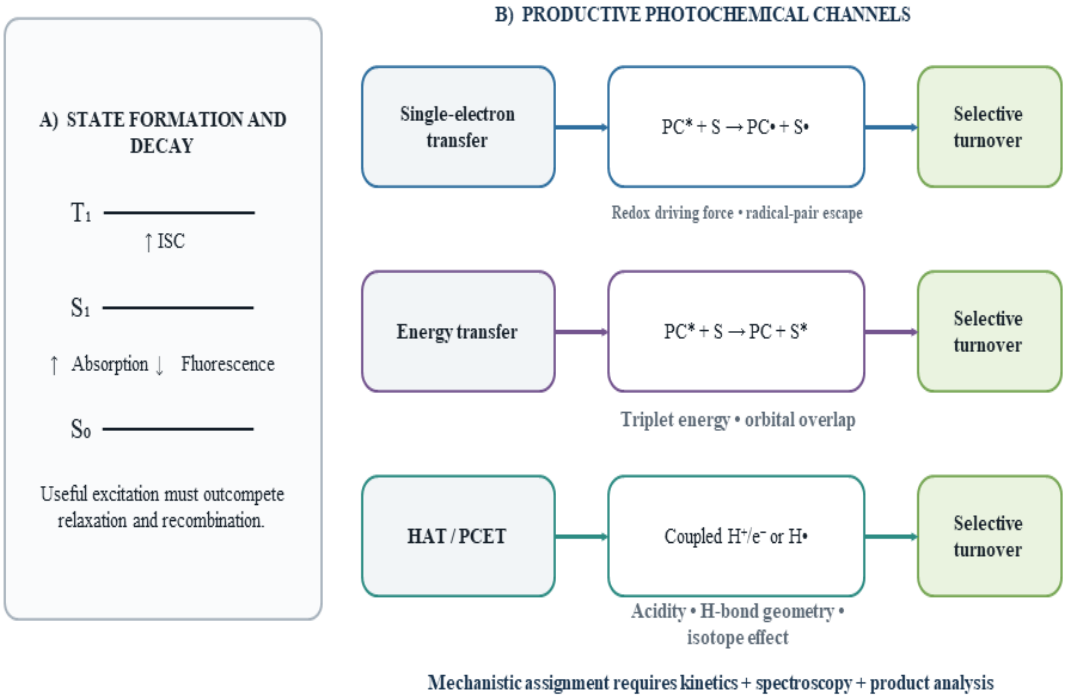


Figure 3.1. Photophysical pathways from photoexcitation to catalytic turnover.

A triplet state is populated by intersystem crossing by spin conversion. The advantages of ruthenium and iridium complexes make up for the cost and scarcity of the heavy atoms required for the spin-orbit coupling. Heavy atom-free organic photosensitizers can reach the triplet state via donor-acceptor charge transfer, symmetry breaking, conformational motion or a small singlet-triplet energy gap. Additional features of framework organization include limitation of nonradiative motion and tuning of electronic coupling. The key design challenge for acceleration of useful triplet formation is to avoid low-energy states that decrease the driving force for electron or energy transfer [27].

Single electron transfer proceeds through oxidative or reductive quenching of the excited photocatalyst. Although excited state redox potentials are frequently used to determine thermodynamic feasibility, reorganization energy, diffusion, electronic coupling, and the fate of the radical pair also affect productive flux. Initial efficient quenching event may be lost through back electron transfer. Fragmentation, deprotonation, bond formation or capture by the catalyst, must then compete on the appropriate timescale. Rather, a substrate is sensitized in a net electron transfer free manner, which allows for cycloadditions, isomerizations and bond activation that are not accessible by ground state pathways [28].

Hydrogen atom transfer (HAT) is a single elementary step in which an electron and a proton are transferred together from the same donor bond. Proton-coupled electron transfer (PCET) is the broader description: the electron and the proton may move in one concerted step or in two sequential steps, and in both cases their motions remain

Online ISSN 3007-3197
Print ISSN 3007-3189

kinetically coupled. The dearth of high energy charged intermediates and the ability to selectively activate O-H, N-H and C-H bonds make PCET a useful reaction for the pharmaceutical industry [29]. Its behavior is influenced by the redox potential and acidity, hydrogen bonding and proton acceptor geometry. Bio-inspired secondary coordination spheres can reorganize bases and substrates in the way that enzyme active sites do, but the assignment of a concerted PCET pathway cannot rest on a kinetic isotope effect alone and must be supported by thermodynamic, kinetic and spectroscopic evidence [29, 30].

Oxygen photosensitization is one of the competing mechanistic manifestations. Type I pathways are those in which electrons or hydrogens are transferred, resulting in the formation of superoxide, hydroperoxyl species or substrate radicals; and Type II pathways are those in which energy is transferred to ground state oxygen to yield singlet oxygen [31]. Both can coexist and the balance varies with the oxygen concentration, solvent, sensitizer triplet energy and substrate. Product distribution, isotope effects, EPR detection and direct near infrared detection are not always strictly selective, and are therefore best used together prior to assigning a pathway [31, 32].

Mechanistic characterization needs to be linked to preparative turnover in the ultrafast photophysics. Time correlated fluorescence, transient absorption, electrochemistry and accessible states (steady state absorption and emission) determine formation and decay EPR identifies trapped charges and radicals and operando vibrational or X-ray methods determine speciation of the catalyst [33]. Wherever possible, measurements should be made at the photon fluxes and catalyst concentrations that are actually used for catalysis. Even though states irrelevant under LED irradiation can be populated by high laser fluence, recombination and trapping cannot be uniquely identified by empirical multiexponential fits [34]. Use spectroscopy, electrochemistry and product formation to constrain global kinetic models, not just one of these techniques [33, 34].

The complete kinetic network is responsible for selectivity and not the first quenching step. After the formation of a radical ion, fragmentation, proton transfer, radical addition, cage escape and back electron transfer compete [3]. Thus, excited state quenching rates may be comparable and the product distribution may vary from one system to another due to changes in solvent polarity and ion pairing. With dual catalysis, the concentration and oxidation state of the transition metal partner is the key to capture the radical or background chemistry [29]. Mechanistic studies should be able to measure the productive quantum yield and determine the branching step that leads to product formation. This information is more applicable for designing catalysts than a list of thermodynamically feasible elementary reactions [3, 29].

Intensity dependence remains an underused diagnostic. For a simple catalytic cycle with one photon per reaction a linear relationship between rate and photon flux is expected if the concentration of catalyst and substrate are constant [10]. Sublinear behavior may be associated with ground state depletion, radical recombination, oxygen or mass transfer limitation, while superlinear behavior may be associated with the consecutive absorption of photons or the initiation of radical chains. The quantum yields >1 may represent chain propagation, but they need the rigorous measurement of the number of absorbed photons and cannot be understood exclusively based on the conversion [1]. Intensity dependence over the preparative range is important as a link

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Online ISSN 3007-3197
Print ISSN 3007-3189
http://amresearchreview.com/index.php/Journal/about

between mechanism and reactor design, and also to ensure that the preparative range does not cross into a new kinetic regime during the scale-up process [1, 10]. These photophysical and photochemical mechanisms are summarized in **Table 3.1.**

Table 3.1. Photophysical and photochemical mechanisms operative in bio-inspired photocatalysis.

Mechanism	Key features	Diagnostic evidence
Triplet formation (energy transfer)	Intersystem crossing populates the triplet state heavy atom (Ru, Ir) complexes use spin-orbit coupling, heavy atom free sensitizers rely on charge transfer, symmetry breaking or a small singlet-triplet gap	Triplet lifetime and phosphorescence, transient absorption
Single electron transfer	Oxidative or reductive quenching of the excited photocatalyst governed by redox potential, reorganization energy, diffusion, electronic coupling and back electron transfer	Stern-Volmer quenching, transient absorption, electrochemistry
Hydrogen atom transfer / PCET	Concerted or stepwise coupling of electron and proton motion; enables selective activation of O-H, N-H and C-H bonds, influenced by redox potential, acidity and hydrogen bonding	Kinetic isotope effect together with kinetic and spectroscopic evidence
Oxygen photosensitization (Type I / II)	Type I involves electron or hydrogen transfer forming superoxide, hydroperoxyl or substrate radicals Type II involves energy transfer to ground state oxygen forming singlet oxygen both can coexist	Product distribution, isotope effects, EPR and near infrared detection used together
Mechanistic and kinetic diagnostics	Global kinetic models built from spectroscopy, electrochemistry and product formation intensity dependence links mechanism to reactor design and preparative scale-up	Time correlated fluorescence, transient absorption, EPR, operando vibrational or X-ray methods

Light-Matter Interaction and Photon Management

Spectral matching is dependent upon the overlap between the source emission, catalyst absorption and useful excited state pathways. The absorption onset is too poor a measure since it is not representative of the fraction of incident photons absorbed in an operating reactor, or the quantum yield at each wavelength [35]. Smaller band gaps in semiconductors lengthen the visible absorption range but could decrease the redox potentials; defect states can be either useful stepping stones or recombination centers.

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http://amresearchreview.com/index.php/Journal/about
Volume 4, Issue 3 (2026)

Online ISSN 3007-3197
Print ISSN 3007-3189
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While plasmonic and photonic structures can enhance local fields, they also can modify the lifetime of photons, and their utility depends on the quality of the interfaces, as well as the geometry and scattering of the particles [35, 36]. **Fig. 4.1** shows light-matter interaction strategies used to increase photon capture and excited-state control.

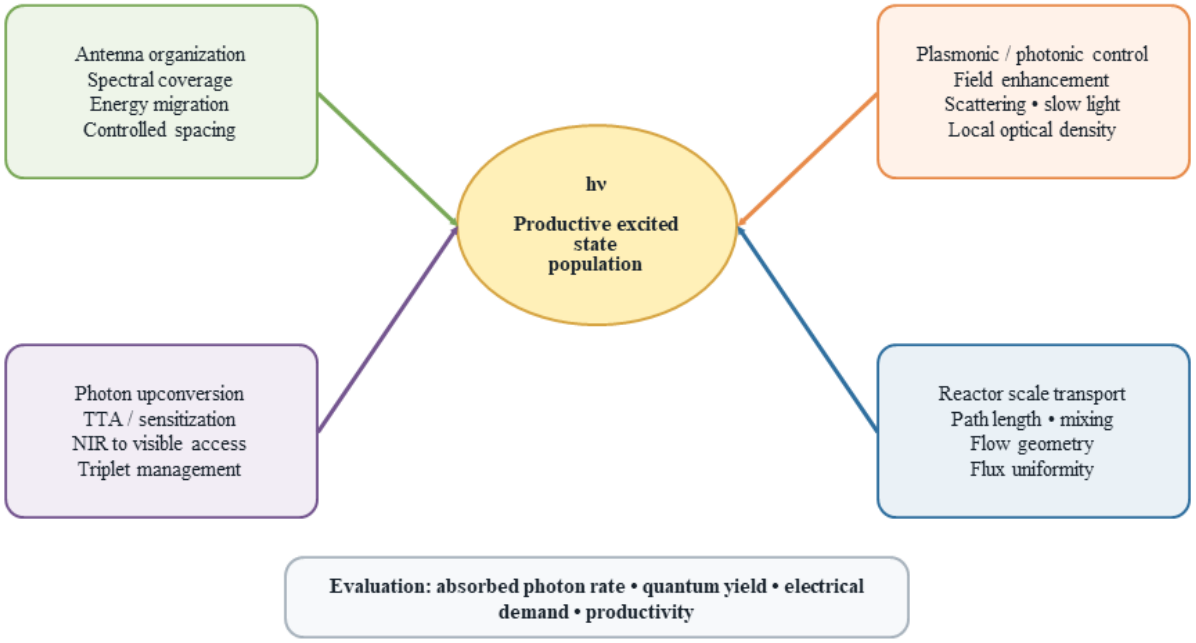


Figure 4.1. Light-matter interaction strategies used to increase photon capture and excited-state control.

In order for recombination to occur, excitons must travel to a dissociation interface or catalytic site. The diffusion length depends on the lifetime, energetic disorder, morphology and interchromophore coupling. In reported values for polymeric photocatalysts, the model assumptions and particle polydispersity can also lead to distorted values; therefore, revised measurements have been reported and increased attention is paid to separating out the optical and morphological effects from the intrinsic migration [21]. Hydrogen bond bridges and ordered donor–acceptor contacts can enhance dissociation and directional motion, but strong association can lead to traps or to back transfer [37].

The meaning of charge carrier lifetime is related only to catalytic rate constants. A long-lived trapped carrier may be chemically inactive, whereas a shorter-lived interfacial state may react efficiently with a pre-adsorbed substrate [38]. Incident photon yield and absorbed photon yield are different with respect to the questions they answer, while quantum yield relates to absorption and subsequent chemistry. When correlations between excited state rate constants and activity are obtained under similar conditions and are backed up by mechanistic assignments, they are useful [38]. Meaningful comparisons are not possible when only nominal lamp power is reported instead of calibrated photon flux [38].

Morphology controls light scattering, surface area, adsorption, diffusion and the density

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Online ISSN 3007-3197
Print ISSN 3007-3189
http://amresearchreview.com/index.php/Journal/about

of interfaces. Although nanostructuring can reduce carrier path length and increase the active facets, very small particles could either aggregate or disintegrate, or introduce a high density of recombination active defect [39]. Near the surface illuminated, dense catalyst beds absorb and deprive the interior of photons. Thus, slurries require a finite catalyst loading, which means an addition of more solid may reduce the utilization of photons (volumetric) [39]. In photofluidized beds or structured reactors, the particles are made to flow through the light rich regions; in the evaluation of the process, the energy needed for fluidization of the particles and attrition of the particles by the fluidization process must be taken into account [39].

Multiple scattering and Beer-Lambert attenuation result in spatially non-uniform reaction rates at scale. The increase in vessel diameter does not scale proportionally with the increase in the productive illuminated volume and the more powerful lamps can enhance side reactions close to the vessel wall [40]. The mixing and kinetics must be solved along with the radiative transfer models. A uniform irradiation and preservation of geometry is achieved with narrow path flow reactors and with numbering-up, but pumping, fouling and solids handling still have limitations [10]. Photon management is then not only a catalyst property, but a property of the reactor, too [10, 40].

There is an optical kinetic compromise in the case of the use of a catalyst by concentration. When the concentration is low, a large proportion of the photons pass through the reactor unabsorbed, and when the concentration is high, the photons are absorbed close to the wall leaving the inside dark [10]. Molecular catalysts can also undergo self-quenching and aggregation, and semiconductor slurries scatter light and protect the particles. Thus, for each geometry and wavelength, the optimum loading needs to be determined [39]. Reporting only the highest rate per catalyst mass can encourage artificially low loadings, and reporting only the volumetric productivity can mask inefficient photon use. Activity (catalyst normalized) as well as absorbed photon productivity (reactor level) is required [10, 39].

Photostability should be part of the spectral matching. A sensitizer can absorb intensely at the chosen wavelength, and it can also have an excited state that is destructive or absorbs via a degradation product [9]. But action spectra obtained at the beginning of a run may not be the same as later in the run after longer irradiation. Alternatively, these spectral drift measurements may be made online using UV-visible spectroscopy and periodic quantum yield measurements [24]. Under sunlight, it is the photocatalytic system rather than the source that must tolerate diurnal variation in ultraviolet content, temperature and intensity. A robust response to a spectral variation might be more relevant to annual productivity than a maximum quantum yield at a single monochromatic wavelength [9, 24].

Applications in Sustainable Organic Synthesis

As **Fig. 5.1** and **Table 5.1** summarize the most clearly sustainable way that photocatalysis has an impact on the synthesis of a compound is when it does not require a stoichiometric redox reagent, when it serves to activate a large-scale feedstock, when it makes it easier to shorten a synthesis route, and when it results in chemoselectivity. Selective aerobic oxidation is a process that includes oxygen as the terminal oxidant,

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http://amresearchreview.com/index.php/Journal/about

and it is possible to achieve the oxidation of C-H bonds, alcohols and sulfides under mild conditions [41]. The advantage is to control Type I and Type II pathways, to prevent overoxidation and to avoid using harmful solvents. The atom economy of a process is only apparent during scale-up, because oxygen mass transfer and flammability become significant and the use of sacrificial co-reductants may bring the apparent atom economy down [41, 42].

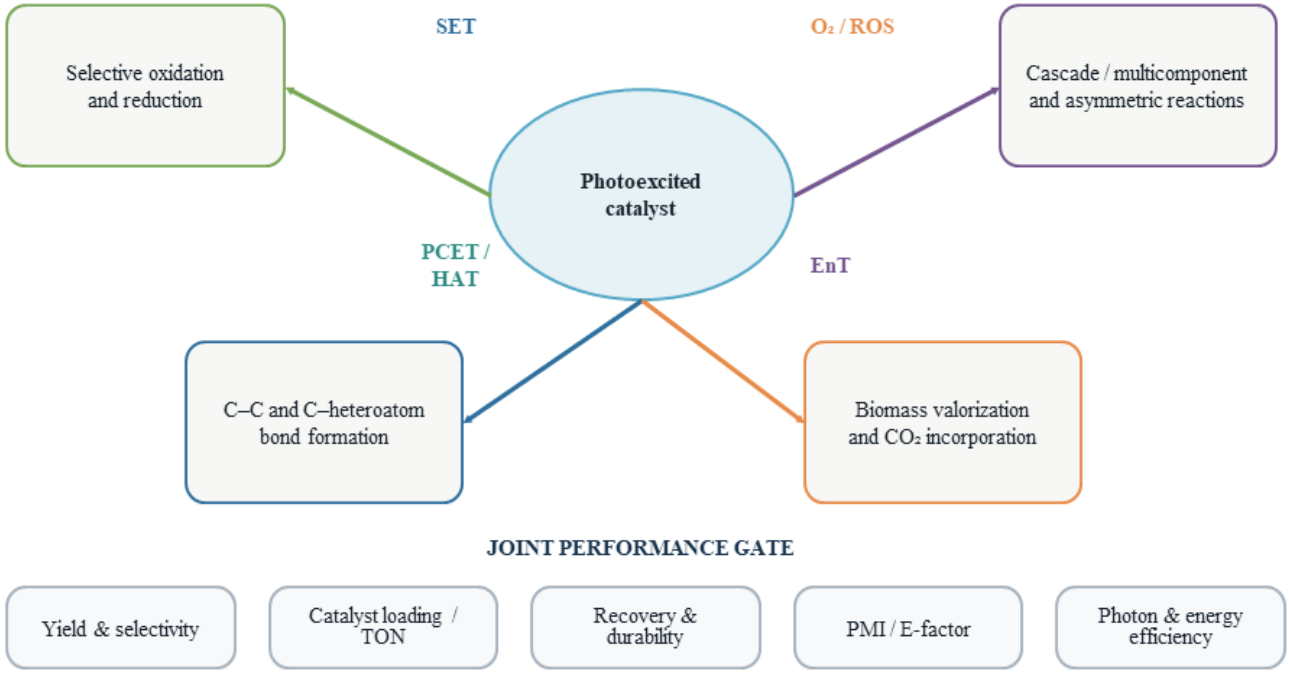


Figure 5.1. Mechanistic pathways linking bio-inspired photocatalysis to synthetic performance and sustainability.

Table 5.1. Synthetic applications of bio-inspired photocatalysis.

Reaction type	Mechanistic basis	Sustainability consideration
Selective aerobic oxidation	O_2 as terminal oxidant, type I / type II pathways	Avoid over oxidation and hazardous solvent, O_2 mass transfer and flammability at scale
Reduction & hydrogenation	Excited state electron transfer, semiconductor electrons, coupled dehydrogenation	Pair with biomass oxidation to avoid a sacrificial donor, confirm active catalyst by operando methods
C-C bond formation	Radical addition, Minisci reaction, cross electrophile coupling,	Weigh ligand complexity, metal handling and excess coupling partner against

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Online ISSN 3007-3197
Print ISSN 3007-3189

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Reaction type	Mechanistic basis	Sustainability consideration
	metallaphotoredox	route simplification
C-heteroatom coupling	Radical substitution and photoregulated transition metal cycles, for example red light metallaphotocatalysis	Longer wavelength does not automatically mean higher energy efficiency, measure loading and quantum yield
C-H functionalization / HAT	Proton-coupled electron transfer, polarity matching and bond dissociation energy	May require several equivalents of HAT donor; include reagent synthesis in the comparison
Biomass valorization	Selective cleavage of lignin, cellulose and hemicellulose domains	Model compound studies neglected real mass impurities; product recovery from water is limited
CO_2 incorporation	Photoredox-generated radical anions and organometallics add CO_2	Requires concentrated or purified CO_2; needs capture and energy accounting for net mitigation
Cascade and multicomponent reactions	One photogenerated intermediate drives successive bond formation steps	High yield can conceal difficult separation of closely related by-products
Asymmetric photocatalysis	Chiral confinement Brønsted acids, H-bonding catalysts, enzymes, supramolecular pockets	Strong binding can inhibit turnover report enantioselectivity together with conversion and background rate
Paired photochemistry	Coupling oxidative and reductive half reactions from one excitation	Requires kinetic and separation matching needs a real mass and energy balance not just product value

Photocatalytic reduction and hydrogenation provide reducing equivalents via excited state electron transfer, semiconductor electron or coupled substrate dehydrogenation. Oxidation and hydrogenation of biomass can be coupled to utilize both the redox equivalents, without the loss of an unproductive sacrificial donor [43]. Selectivity is still sensitive to proton source, hydrogen atom donor, adsorption, and catalyst transformation. The active catalyst can only be developed in the presence of light, in

Annual Methodological Archive Research Review
http://amresearchreview.com/index.php/Journal/about
Volume 4, Issue 3 (2026)

Online ISSN 3007-3197
Print ISSN 3007-3189
http://amresearchreview.com/index.php/Journal/about

which case, pre-reaction and post-reaction characterization will not provide operando evidence [24].

Radical addition, Minisci chemistry, cross electrophile coupling, cycloaddition and carboxylation are all involved in C-C bond formation. Photogenerated radicals allow for bond construction from partners that would not work otherwise via prefunctionalization or strong reagents [44]. Transition metals paved the way for this space with metallaphotoredox catalysis, which involves the control of the oxidation state of the metal and the capture of the transition metal radical. Its synthetic power should be balanced against the complexity of the ligands, the handling of nickel or precious metal, the recovery of the photosensitizer and the often use of excess coupling partner [44]. It is truly beneficial to use reductive coupling when it eliminates organometallic reagents and makes the pathway easier rather than just changing the stoichiometry to an external reductant [44].

The coupling of carbon with heteroatoms can achieve C-N, C-O, C-S and C-P bonds under visible light by radical substitution or photoregulated transition metals cycles. Red light metallaphotocatalysis helps penetration and limits the damage from photodestruction and may help recovery with semiheterogeneous catalyst format [45]. However, energy efficiency does not depend on longer wavelengths alone, and a more sophisticated sensitizer or longer time of irradiation might be required for lower photon energy. The method is better than the thermal coupling method if it has a loading of a catalyst, quantum yield and concentration and isolation.

The photoredox C-H functionalization and direct hydrogen atom transfer decreases the dependence on prefunctionalized substrates. Regioselectivity can be achieved by polarity matching and bond strength and catalysed association, whereas PCET can activate bonds which are not accessible to outer sphere electron transfer [46]. These reactions can lead to an improvement of step economy but may require more than one equivalent of trapping reagent or hydrogen atom donor. For a complete comparison, everything from the preparation of the functionalizing reagent, by-products and downstream separation must be included [46].

The biomass conversion aims to demonstrate the potential of photocatalysis to link renewable carbon with selective synthesis. Lignocellulose consists of chemically different domains of lignin, cellulose and hemicellulose, which need to be selectively cleavable under control of adsorption, radical formation, and oxygen chemistry [47]. In the case of photocatalysis, it is possible to obtain aromatic oxygenates, organic acids, hydrogen and platform molecules under mild conditions, but such studies on model compounds do not reflect the effect of mass transfer, impurities and product mixtures in real biomass [47, 48]. However, the practical performance can be determined by product recovery from the water and catalyst fouling by oligomers [48].

The carbon dioxide incorporation process converts a stable C1 feedstock into carboxylic acids or dicarboxylated products. Radical anions or organometallic intermediates can be formed via photoredox cycles, which can add CO_2 under less forcing conditions than classical carboxylation [49]. The alkyne 1, 2 dicarboxylation is a valuable bond construction that can be exemplified [49]. However, the process burden may be due to concentrated or purified CO_2, sacrificial reductants, and low substrate concentrations [49]. The carbon incorporation is not net carbon mitigation unless

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http://amresearchreview.com/index.php/Journal/about
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Online ISSN 3007-3197
Print ISSN 3007-3189

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capture, energy supply, and product lifetime are taken into account [49].

Cascade and multicomponent reactions use a single intermediate which is generated by a photochemical process and reacts in a series of bond-forming steps. They can enhance atom and step economy by minimizing the number of isolations and enabling multiple components to be activated in a single step [50, 51, 52]. Network complexity is their vulnerability; each component can quench the photocatalyst, each component can capture the radicals, and each component can inhibit the metal catalyst. High yield may mask problems with difficult separation of closely related by-products [50]. The best examples are those that are high in concentration, have a balanced stoichiometry, and are both broadly selective and direct to the isolated product [51].

Asymmetric photocatalysis is an example of how bio-inspired confinement can provide added value beyond rate. Chiral Brønsted acids, hydrogen bonding catalysts, enzymes and supramolecular pockets can direct the orientation of bond formation with a radical precursor or radical acceptor [12]. The problem is the need to align a fast-diffusing open-shell intermediate with a slower stereocontrolling association. Binding can enhance the selectivity of capture and ion pairing can raise the probability of selective capture, although high binding affinity may slow down catalyst turnover [51]. The interpretation should consider conversion and background rate since a highly selective minor pathway can be masked or diluted by nonselective photochemistry [12, 51].

Reaction concentration is an important parameter in all applications. Dilute operation improves light penetration and suppresses bimolecular radical termination, but it also increases solvent consumption, reactor volume and separation energy [10]. Raising the concentration generally lowers PMI and raises throughput, although its net effect depends on the solvent inventory across the full process boundary, and it simultaneously intensifies inner filter effects, aggregation, and oxygen or gas transfer limitations. A sustainable approach should seek the maximum concentration without loss of stability of selectivity and photon distribution [1]. Scaling up at the original dilute concentration is not process intensification, but just increasing the amount of solvent and equipment [1, 10].

Paired photochemistry increases redox economy since both the electron and hole generated during the excitation process will be utilized. For many semiconductor reactions, one half reaction results in the formation of the desired molecule and the counter reaction consume a sacrificial donor or acceptor [50]. Two convenient transformations, e.g., oxidation of biomass with proton reduction or dehydrogenation of substrate coupled with hydrogenation of substrate, can increase the atom utilization. The two reactions, however, need to be matched in terms of their separation and kinetics [43]. Low selectivity is not redeemed by the mere existence of a co-product, and the market value of that co-product is not a substitute for a physical mass and energy balance [43, 50].

Preparative relevance is established by isolated productivity rather than by analytical conversion alone. Analytical conversion in a closed vial may not be representative of the results when oxygen, CO_2 or hydrogen must be continuously supplied, when the catalyst particles settle out of the reaction mixture, or when the product soaks up the wavelength of the radiation to which it is exposed [10]. Isolated yield, reaction concentration, illuminated volume and irradiation time and space-time yield should be

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http://amresearchreview.com/index.php/Journal/about
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Online ISSN 3007-3197
Print ISSN 3007-3189
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reported. In continuous processes steady state conditions must be established for several residence times [53]. Reported on this basis, molecular, heterogeneous and biohybrid systems can be compared on equal terms, without privileging any single normalization.

Quantitative Green Chemistry Evaluation

As shown in **Fig. 6.1** an equivalent functional unit is the first step in making a defensible comparison, and is the same amount of product with the same purity at a similar size. Atom economy, reaction mass efficiency, E-factor, and process mass intensity are all used to measure waste [54]. These metrics are not interchangeable and show different weaknesses. In most cases, photocatalysis will lead to an improvement in step economy, but it can be outcompeted by dilution, sacrificial reagents, and chromatography [1, 54].

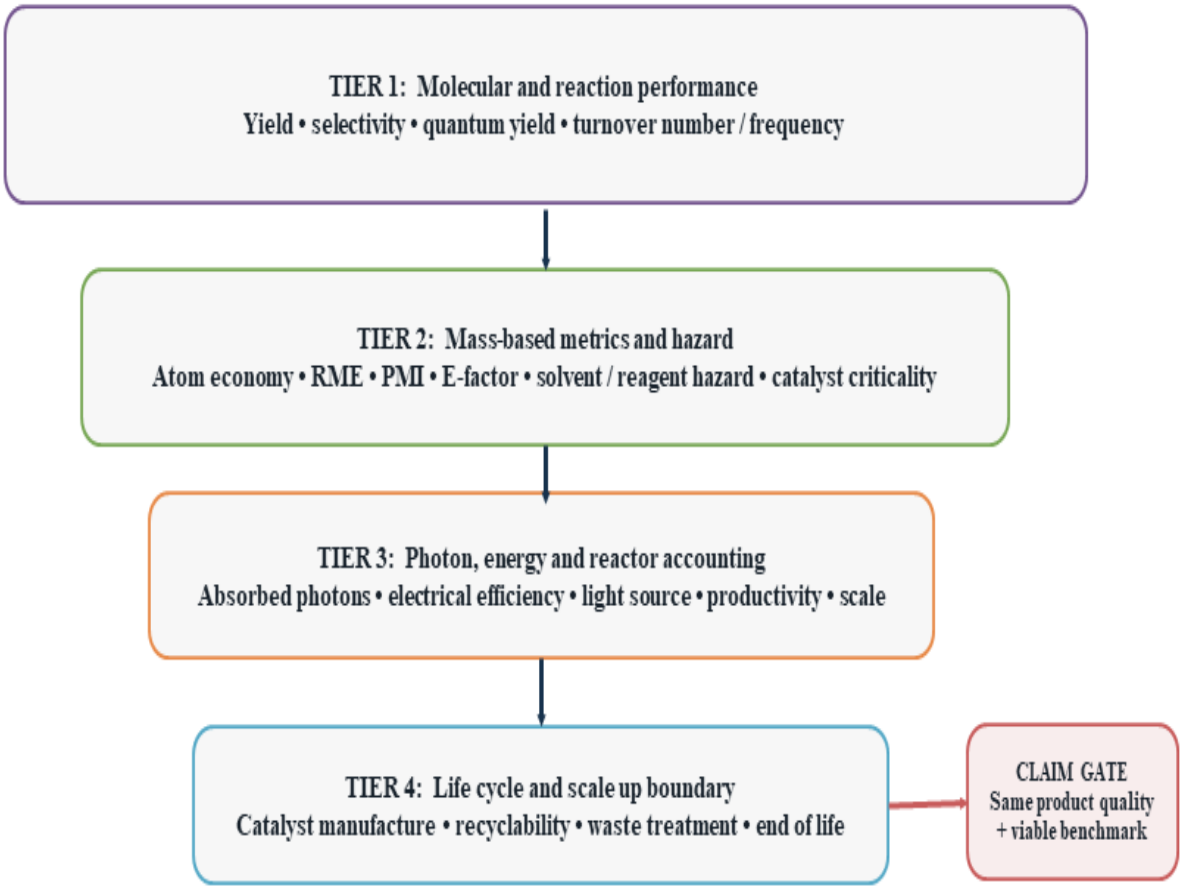


Figure 6.1. Tiered evaluation of green chemistry: molecular performance, life cycle and scale-up.

The energy assessment has to differentiate between the incident optical energy and the electrical energy consumed by the light source and any auxiliary equipment used. Photon flux, spectral overlap, quantum yield, irradiation time, temperature control, pumping and productivity are the relevant quantities [55]. Red light penetrates deeper and possibly lower-energy sensitisers, but blue LEDs can have a high electrical efficiency, with good catalyst absorption. Solar operation avoids the electrical cost of

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http://amresearchreview.com/index.php/Journal/about
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Online ISSN 3007-3197
Print ISSN 3007-3189
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lamps but introduces collector area, tracking, weather dependence and variable throughput, none of which apply to lamp-based systems. The preferred source is therefore system- and location-specific [55, 56].

Solvents are a common source of PMI and hazard. The use of acetonitrile, chlorinated media, water, alcohols, ethyl acetate and some bio-derived solvents can offer good photophysics, improved safety or recovery [57]. Replacement needs to be evaluated experimentally due to the variation of solubility, oxygen transfer, excited-state lifetime and quenching with medium. Even a nominally green solvent may have a higher overall impact when used to the extent of having to be diluted to extreme levels or extracted with difficulty [58]. Reaction performance and recovery should be used in combination with selection guides [57, 58].

There is a relationship between abundance, toxicity and durability of catalysts. However, the ruthenium and iridium sensitizers typically require rare elements as well as complex ligands, and generally require low loading [59]. The abundance can be enhanced by the use of copper, iron and organic photocatalysts, but may require higher loading or display limited stability windows [59]. With heterogenization, the recovery can only be achieved if the leaching is controlled and the leaching activity is measured within the time period that is relevant to the process [59]. Manufacture, support production and disposal of catalysts should be included within the inventory repeated conversion without mass balance does not indicate recyclability [59].

A graded assessment is feasible. The first level is reporting atom economy, yield, reaction mass efficiency, E-factor and PMI, including work-up [60]. The second introduces the concepts of solvent and reagent hazard, catalyst criticality, leaching and worker exposure. The third represents electrical energy, photon efficiency, reactor productivity and if available cradle-to-gate life-cycle impacts [61]. Laboratory inventories are not definitive but suitable assumptions and sensitivity analysis can determine the role of solvent recycling, catalyst lifetime or electricity mix in driving the outcome [60, 61]. Those parameters should be reasonably variable to ensure a sustainability claim withstands reasonable variation [60]. **Table 6.1** summarizes the tiered framework for green chemistry evaluation of bio-inspired photocatalytic routes.

Table 6.1. Tiered framework for green chemistry evaluation of bio-inspired photocatalytic routes.

Tier	Evaluation criteria
Tier 1 Molecular and reaction performance	Yield, selectivity, quantum yield, turnover number and frequency
Tier 2 Mass-based metrics and hazard	Atom economy, reaction mass efficiency, PMI, E-factor, solvent and reagent hazard, catalyst criticality
Tier 3 Photon, energy and reactor accounting	Absorbed photons, electrical efficiency, light source selection, reactor productivity, scale

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Online ISSN 3007-3197
Print ISSN 3007-3189
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Tier	Evaluation criteria
Tier 4 Life-cycle and scale-up boundary	Catalyst manufacture, recyclability, waste treatment, end of life, and where data permit cradle-to-gate life-cycle impacts

Work-up is a special case so it requires specific attention as it often accounts for most of the laboratory waste. The mass of the reaction inputs can be exceeded by quenching, extraction, washing, drying, adsorption onto silica and evaporation of solvents [54]. Even if the chemical selectivity is high, it is possible that the photocatalyst color or contamination by nanoparticles will require further purification. Process advantage can be obtained through direct crystallization, phase separation or membrane retention or telescoping into a subsequent step of the process from the photochemical selectivity [1]. Therefore, green metrics should be calculated for isolated product, and should account for all purification materials, not just the reaction mixture [1, 54].

Recyclability should be reported in terms of retained function per catalyst inventory. Catalyst activity may be masked by reporting identical conversion for five cycles, which can be due to longer reaction times, loss of the catalyst during handling or addition of additives [9]. Accompanied by metal or chromophore leaching, turnover number and cumulative product per initial catalyst mass are more informative and characterization by activity normalization. Both active component and support for supported catalysts need to be monitored [1]. The use of a catalyst which is easily filtered, but which retains a small amount of toxic metal per batch could result in more contamination at the downstream than a homogeneous catalyst which is recovered by closed loop separation [1, 9].

Life-cycle interpretation needs to differentiate between actual lab performance and the potential for optimized lab performance. High energy and solvent consumption are possible for a bench scale route, due to poor use of equipment, and scale-up may reduce energy and/or solvent consumption due to better heat integration and recovery [60]. On the other hand, the assessment of the technology for a prospective application can be slightly favorable without evidence. Information about the present should be reported in a scenario analysis, as should be a clear improvement case and break-even values [61]. If the photocatalytic approach is only feasible at an unproven quantum yield or lifetime, then that is a research objective, not a proven ability, and should be included in the requirement [60, 61].

Challenges, Knowledge Gaps and Research Priorities

As shown in **Fig. 7.1** one of the key challenges with limited visible light utilization is still present. Most semiconductors absorb primarily UV light, and some molecular catalysts absorb only small portions of the spectrum of an LED or solar light [62]. The possibility of absorption tails, dyes, intermediates and direct excitation of the substrate with the catalyst can be confused with catalyst activity. A broadband heterostructure, donor-acceptor frameworks and plasmonic architectures can be used to increase the absorption range, but can also reduce the redox power or result in recombination sites [36]. The missing measurement is a closed photon balance under reaction conditions,

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http://amresearchreview.com/index.php/Journal/about
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Online ISSN 3007-3197
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with separation into individual wavelengths or frequencies. The parameters which are not performance parameters are action spectra, incident and absorbed photon flux, and contribution of the catalyst, substrate and scattering should be reported together [36, 62].

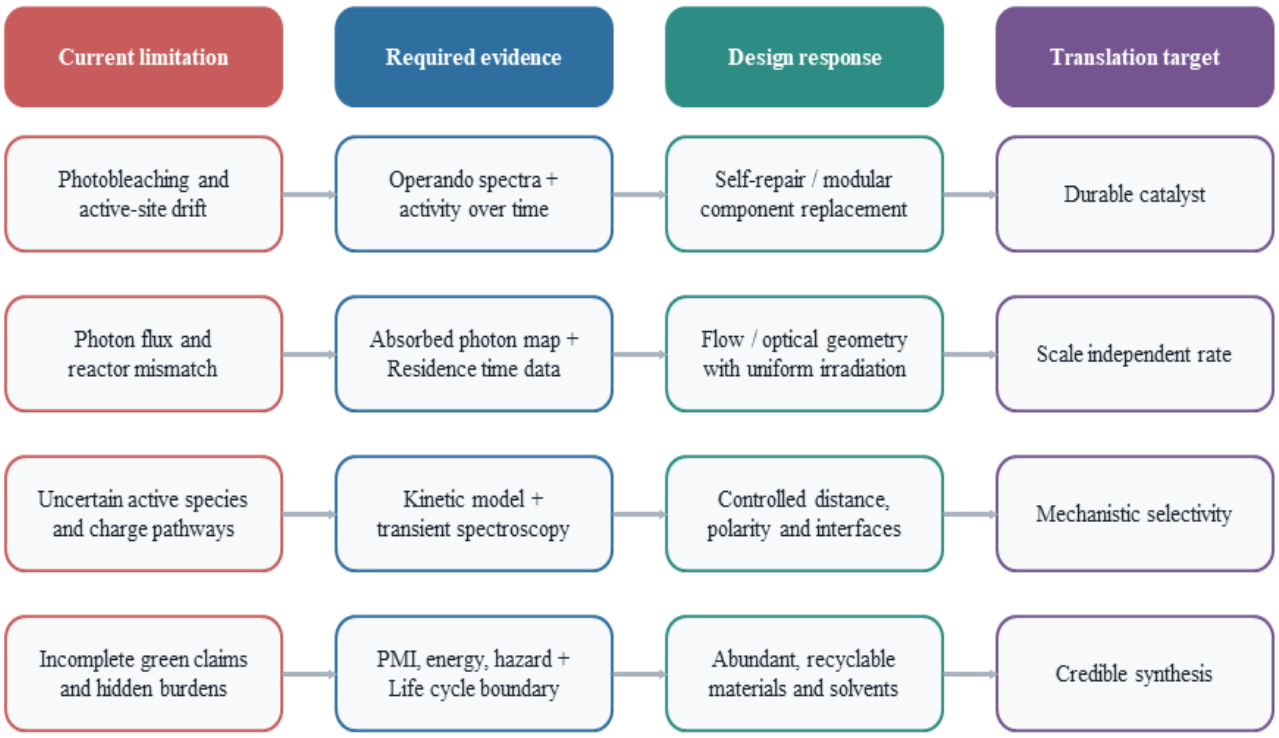


Figure 7.1. Challenges and priorities for scalable bio-inspired photocatalysis.

Charge recombination connects molecule invention to reactor productivity. From femtoseconds to milliseconds, there is competition among geminate recombination, trapping, bulk electron-hole annihilation, and back electron transfer [63]. Directional transfer can occur at heterojunctions and internal electric fields, but poor matching of interfaces introduces trap states. A longer spectroscopic lifetime may signal an inert trapped carrier rather than a reactive state [38]. The first goal is to link transient populations in a quantitative manner to the conversion of the substrates as modeled with global models based on spectroscopy, electrochemistry and product kinetics at catalytic photon flux [38, 63].

Most cases of catalyst degradation are not diagnosed. Ligand loss, ligand photobleaching, metal leaching, photocorrosion, aggregation, pore collapse and surface reconstruction are all processes that can be induced by light [24]. With a dual catalysis two changes of speciation occur, one with the photosensitizer and the other with the transition metal component. The active species that is responsible for the sustained activity may not be the precatalyst [24]. The reuse test using only the conversion may be confounded due to product carryover or leached homogeneous catalyst. Requirements for durability studies are mass balance, leaching, operando characterization, continuous irradiation, impurity challenges and start stop cycling [23].

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A self-healing claim must differentiate between true healing and consumption of an undetected precursor reservoir [23].

Mechanistic uncertainty exists due to the fact that electron transfer, energy transfer, HAT, PCET and direct excitation may generate similar products. Stern–Volmer quenching is a test for interaction, not productive turnover, while radical traps can alter the network that you are trying to diagnose [34]. Transient spectra may overlap, EPR spin trapping can steer the chemistry and high fluence laser measurements can enable the observation of non-catalytic states. Robust mechanistic assignment therefore requires convergent controls: intensity and wavelength dependence, isotope studies, electrochemical consistency, intermediate detection, and correctly predicted changes in selectivity. In porous and heterogeneous systems, methods that prevent the separation of intrinsic carrier dynamics from adsorption, diffusion and scattering are particularly needed [34].

The description of light and reactor conditions is incomplete, which limits reproducibility. Lamp name or nominal wattage does not define photons delivered to the sample [9]. The conversion is influenced by the emission spectrum, distance, illuminated area, optical path, stirring, vessel material, cooling, ageing of temperature and ageing of the lamp. At least the following parameters should be reported: calibrated photon flux at the reactor, geometry, spectrum, irradiated volume, temperature, mixing, catalyst particle properties, analysis calibration and replicate uncertainty [64]. The quantum yields should specify the incident or absorbed photons. Chemistry can be separated from apparatus-specific performance by using open-source reactor designs and benchmark reactions [9, 64, 65].

Radiative transfer remains the governing constraint on scale-up. In a large absorbing vessel, a bright boundary is surrounded by a photon-poor interior and if the lamp power is increased, then the side reactions can be increased without increasing the useful volume [53]. Mixing is used to ensure flow of material in both light and dark zones, which is compatible with both excited-state and reaction kinetics. Fouling, pumping and numbering up make flow reactors complicated, but they have short optical paths and defined residence times. Settling, abrasion and catalyst separation are added with heterogeneous systems. The following parameters should be reported absorbed photon efficiency, illuminated volume productivity, space-time yield, pressure, pumping and cooling energy, catalyst inventory and extended steady state operation [53, 66].

System-level economic and environmental trade-offs need to be considered. Operating concentration, photon efficiency, campaign duration and purification are more important than catalyst price [59]. A high-loading, effective photocatalyst could not be as desirable as a recoverable precious metal complex at extremely low loading. Water can lower the solvent hazard and energy of extraction, while immobilization can enhance the energy of extraction and add to support manufacture and mass-transfer limitations [1]. Comparisons should be made against the best realistic thermal, electrochemical or biocatalytic alternative at equal purity and throughput, rather than an outdated benchmark [1, 59].

The gaps show that the central challenge identified in this review is not a lack of active materials, but a lack of integration. The main trend in catalyst papers is to optimize for absorption or rate mechanistic papers focus on the ultrafast events synthetic papers

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Print ISSN 3007-3189
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focus on yield; and sustainability papers begin after route has been developed [9]. A reasonable design process should instead begin with a synthetic function and a sustainability boundary, choose overabundant and durable materials, and match excited state kinetics with turnover, design photon transport at the intended scale, and measure mass, hazard and energy metrics in the optimization process. Only this sequence can lead to a process advantage that is transferable to applications in the field of biological inspiration [1].

Inconsistent normalization makes comparison of catalysts even more difficult. The rates per gram for low surface area supports differ from those of molecular catalysts because the rates per surface area for the low surface area supports depend on the uncertain measurement of the active area, while turnover frequencies depend on a defensible number of active sites. At least absolute product amount, catalyst composition, catalyst loading, irradiated volume, time and photon flux together with the desired normalized rate should be reported in the studies. This more than one axis reporting will avoid an unearned sense of universal superiority [9, 65].

In oxidative reactions and biomass reactions, selectivity and carbon balance are always critical. Loss of substrate may be due to mineralisation or oligomerisation or adsorption [31] but not necessarily to useful conversion. Only knowing of the target product doesn't give you a complete picture of radical pathways and waste. Balance should be closed by total organic carbon, gas analysis and liquid product quantification via calibration [48]. This reasoning also holds true for CO_2 incorporation experiments, which may sometimes necessitate the use of isotopic labelling to differentiate external CO_2 from carbonate, solvent or substrate derived carbon [31, 48, 49].

More metadata are needed for biohybrid reproducibility biological strain and enzymatic preparation, activity assay, cell density, age, buffer composition, oxygenation, localization and viability of the sensitizer before and after the irradiation. A high chemical yield may also be associated with permanent biological damage, restricting reuse [15]. Proteins can bind to the photocatalyst nanoparticles or membranes can be disrupted, changing both the electron transfer and the toxicity. Normally, chemical productivity should be associated with the survival of the biocatalyst and it should indicate whether the biological part is catalytic, sacrificial or regenerated [15, 16].

The environmental release of nanoscale catalysts is not well studied. Dissolved ions and small fragments or colloids that pass through do not result in recovery in the filtration mass measurement [9]. Irradiation can induce transformation that can alter mobility and toxicity. In addition, activity measurements should be supplemented by analysis of leachate under reaction and cleaning conditions, particle size resolved recovery and realistic fate testing [24]. Designing larger recoverable architectures or covalently bound chromophores can minimize release but the impact on photon transport and mass transfer should be determined [9, 24].

These gaps indicate a research hierarchy that is practical in nature. First, determine the active species and close mass and photon balances [9]. Second, determine the kinetic loss which prevents the productive quantum yield. Third, enhance the architecture of the catalyst without compromising the redox driving force and transport. Fourth, check on the durability and reproducibility in the intended reactor. Lastly, compare the optimized process with an equally effective, but credible alternative with equal

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Online ISSN 3007-3197
Print ISSN 3007-3189
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functional units [9]. This hierarchy helps avoid unnecessary complexity in materials and forces focus on the most critical material that limits synthetic and environmental performance [65].

An additional gap in knowledge is due to publication bias. A vast amount of literature exists describing the successful catalysts and optimum conditions, while inactive structures, failed wavelength matches, and deactivation pathways are generally scarce [9]. This skews structure activity relationships and prompts re-exploration of unproductive design space. When coupled with full optical and mechanistic metadata, negative results can be particularly useful to reveal where the redox driving force, lifetime, transport and/or compatibility fails [67]. Raw spectra, calibration files and failed experiments should all be deposited in curated repositories, so that selectively reported successes do not create a misleading impression of causality when the literature is mined by meta-analysis or machine learning.

Language should be controlled. The term "solar-driven" must apply to a measured natural or simulated solar operation, not just the absorption in the visible [1]. The term "recyclable" should include the amount of catalyst recovered and amount of retained normalized activity. The "Metal free" should include contamination and synthesis, and the "Bio-derived" should include "renewable content" and "processing burden" [65]. For "Quantum efficiency" the following should be given: incident photons or absorbed photons, wavelength range. Qualitative labels can be avoided by using precise language to allow readers to compare claims on molecular, heterogeneous and biological platforms [1, 65].

The reporting requirements distributed across this section are consolidated in **Table 7.1,** which sets out the minimum evidence needed before a bio-inspired photocatalytic transformation can be compared meaningfully with an alternative route.

Table 7.1. Minimum evidence and reporting checklist for bio-inspired photocatalytic synthesis.

Domain	Minimum parameters to report	Preferred validation	Misleading practice to avoid
Photon input	Wavelength, emission spectrum, calibrated photon flux, geometry, irradiated volume	Actinometry or radiometry; action spectrum	Lamp wattage only
Mechanism	Excited-state lifetime, quenching constants, active species, quantum yield	Transient absorption, EPR, kinetic isotope effect, electrochemistry with kinetics	Stern–Volmer quenching alone
Catalyst	Loading, TON and TOF, lifetime, recovered mass, leaching	Operando analysis with a closed mass balance	"Recyclable" claimed from repeated conversion only
Preparative	Isolated yield, selectivity,	Scaled or steady-state	Analytical

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Domain	Minimum parameters to report	Preferred validation	Misleading practice to avoid
performance	reaction concentration, irradiation time, space–time yield	run	conversion only
Green metrics	Reaction mass efficiency, PMI, E-factor, solvent and reagent hazard, work-up inventory	Mass balance over the full isolated process	Reaction mixture considered in isolation
Scale-up	Absorbed photon efficiency, optical path, mixing, pumping and cooling energy	Continuous operation; break-even analysis	"Visible light therefore sustainable"
Biohybrid systems	Strain or enzyme identity and activity, viability, localization, regeneration	Biological activity measured before and after irradiation	Chemical yield without biocatalyst-health data

Future Perspectives

Future developments of artificial photosynthetic assemblies should move towards kinetically matched catalytic machines. The antenna absorption, directional charge separation, multielectron accumulation, proton management and substrate exchange should occur on compatible time scales [12]. These functions can be preorganized by supramolecular cages, membranes and porous frameworks, but future demonstrations should avoid sacrificial donors in favour of useful coupled oxidations, and should quantify the oxidative as well as the reductive products [12, 68].

Self-healing photocatalysts offer an alternative to static stability, using reversible coordination, component exchange, redeposition or dynamic covalent bonds. Operational definitions are required: the damaged state must be identified, a functionally equivalent active state must be demonstrably restored, recovery must be reproducible over repeated cycles, and any consumption of a repair agent must be reported [22]. The monitoring of PMI should be done in combination with activity, selectivity, leaching and repair agent. [22, 69]

Current datasets combine metrics on photons which are incompatible, and failures are underreported, but data-driven discovery can link composition, excited state descriptors, solvent, wavelength and reactor geometry. Multiple objectives for a physics-informed model should be optimized including selectivity, quantum yield, catalyst criticality, solvent hazard, space-time yield and life-cycle impact, while reporting uncertainty and domain of applicability [67, 70]. Interpretability is important since models with useful results should make mechanistic hypothesis, not just a ranking [67].

Natural light harvesting antennae can be integrated with modular covalent organic frameworks to enhance the properties of photon absorption, energy directionality, and selective photocatalytic CO_2 conversion on a future bio-inspired photocatalytic platform [71, 72]. These ultrafast studies of photosynthetic electron transfer pathways offer valuable mechanistic insights for artificial photosynthesis and triplet triplet

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annihilation provides an alternative method to converting near infrared photons to higher energy visible photons [73, 74].

When these optically sophisticated systems require available sensitizers, more complex preparation, or possibly critical materials like upconversion nanoparticles, however, the environmental benefit should be evaluated in terms of green metrics, in addition to life-cycle boundaries [75, 76]. The scale-up of COF-based photocatalytic processes, e.g., selective CO_2 reduction is one field of application where the continuous-flow version of photochemistry can contribute to enhance irradiation uniformity, residence-time control, heat and mass transfer, and reproducibility [77, 78].

Slurry Taylor flow microreactors could also be a potential scale-up option for the application of heterogeneous photocatalytic synthesis, and of course, their performance should be correlated with the generation, trapping, recombination, migration and interfacial transfer of charge carriers under the actual operating conditions [79, 80]. Photon up conversion materials that span the near infrared spectrum could extend the spectrum of solar energy that can be harnessed, and electron transport chain inspired coordination polymers could facilitate charge separation and transport over long space and time distances [81, 82].

While bio-inspired supramolecular columns offer controllable architectures for artificial light harvesting, triplet fusion upconversion can enable the use of low-energy infrared photons for synthetically useful photoredox transformations [83, 84]. In addition to the above, efficient triplet exciton formation for selective singlet oxygen-mediated oxidation can be optimized by donor-acceptor engineering of MOF photocatalysts, which also relies on sensitized triplet formation, triplet energy transfer, and triplet-triplet annihilation [85, 86].

Architectural changes of the excited state plasmon can be used to alter local electromagnetic fields and the dynamics of photochemical reactions precisely engineered cascade charge transfer chains can guide the flow of the photogenerated carriers across successive redox sites and minimize undesirable recombination [87, 88].

To translate these molecular and mesoscale developments to industry, controlled delivery of photons must be achieved, the reactor geometry needs to meet the requirements, continuous operation is necessary, the scaling up of the process can be achieved via a “numbering-up” approach, and the results must be reported in a standardized way and be evaluated on a preparative and industrial scale [89].

Catalyst ageing, feed variation and varying irradiance can be compensated by Continuous Photochemistry, Inline Spectroscopy and feedback control [89]. Direct solar synthesis is particularly appealing when high annual insolation is acceptable, or for efficient LEDs that produce predictable output and can control the spectrum. A hybrid solar LED operation could be a viable compromise. Comparisons should include collector or reactor area, annualized productivity, regional electricity, auxiliary energy and equipment lifetime [53, 55, 56].

It is important to include a standardized sustainability assessment as a design input. The core reporting should include photon metadata, mass balance, selectivity, catalyst lifetime, leaching and uncertainty, while system-specific modules should deal with scattering, biohybrid viability or adsorption of the porous material [1]. Interlaboratory validation and trustworthy learning would be possible if all benchmark reactions and

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Print ISSN 3007-3189

machine-readable raw data were shared. Finally, the most valuable advance will not be the catalyst that can be isolated and react at the highest rate but the system that can selectively produce, with a transparent material, energy and environmental performance [1, 65]. Artificial assemblies and self-repair are worth pursuing only where they reduce, rather than increase, the number of process components. Synthesis, separation and failure modes are caused by each antenna added, mediator, membrane and repair agent [22]. The design should allow for easy replacement of one function without the reconstruction of the entire catalyst, and the interfaces should be chemically characterized to a sufficient degree to allow mechanistic testing. Routine stress tests of oxygen exposure, water, salts and impurities, variable light and continuous turnover would show if biological sophistication has resulted in robustness, or merely laboratory complexity [22, 23].

The field must also contain prospective targets in process terms. Studies cannot just aim for higher absorption or longer lifetime, but can define the quantum yield, catalyst lifetime, concentration and space-time yield needed to outpace a conventional route [38]. These break-even targets link fundamental design to industrial relevance, and can be used to guide meaningful break-even goals for AI or high-throughput systems. Successful and failed experiments could be stored in a shared open dataset along with all of the information about the photons and the sustainability metrics of the experiment, which would enable such multi-objective discovery [38, 67].

These requirements can be organised as a sequential translation roadmap. **Fig. 8.1** links a bio-inspired design feature to mechanistic, preparative and reactor-level validation and, finally, to a sustainability gate, with an explicit redesign loop whenever a stage is not passed. Read in this way, the integrated framework advocated throughout this review becomes a decision procedure rather than a description.

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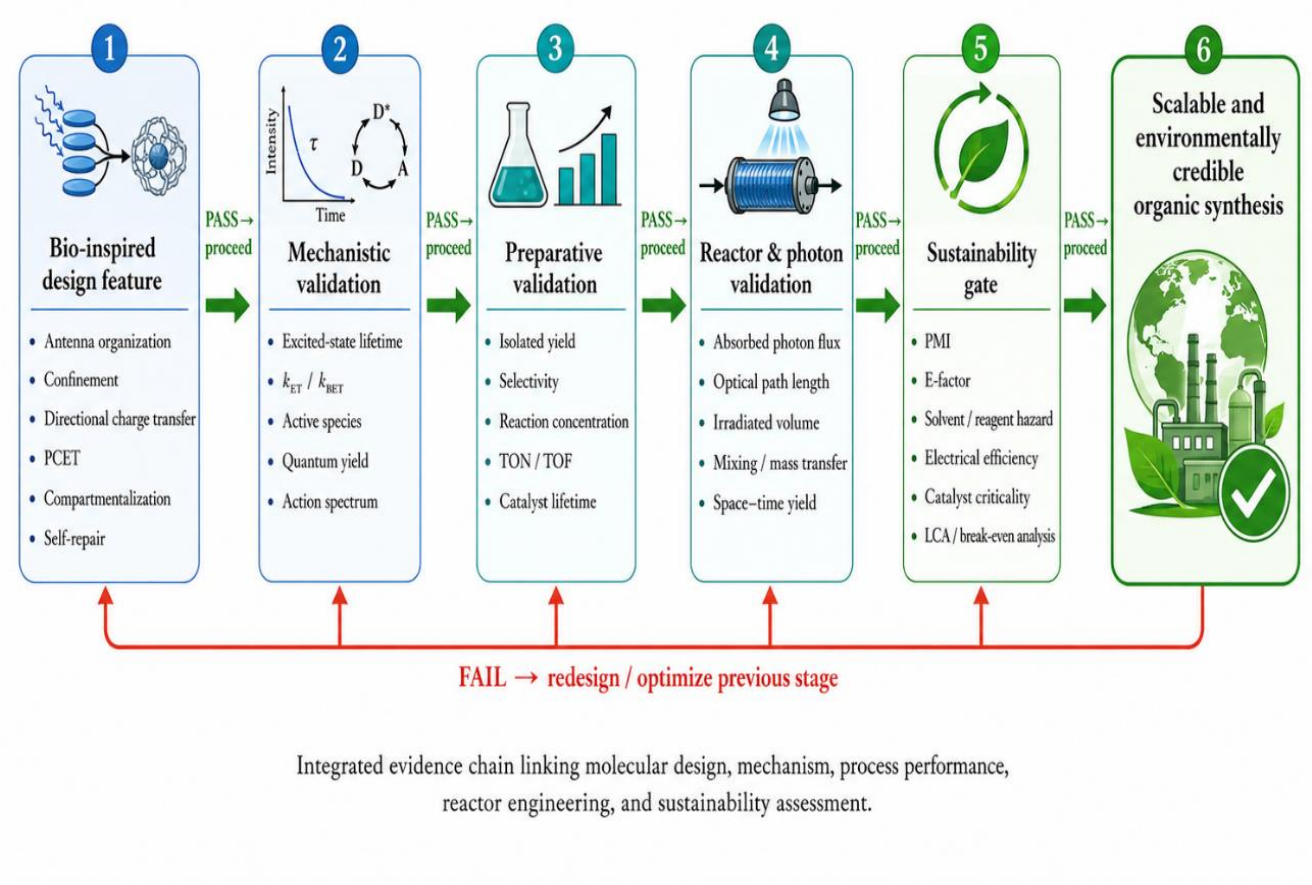


Figure 8.1. Translation roadmap from bio-inspired design to scalable, sustainable organic synthesis.

Conclusions

The ability of a photocatalyst to mimic biological systems is most effective when biological functions are integrated into engineering rather than biological shapes or compositions. The design of catalysts can be based on antenna-like absorption, directional energy migration, controlled donor-acceptor distance, coupled proton and electron transfer, compartmentalization and repair. Their synthetic value only comes about when they are in excited states, when charges arrive at selective catalytic sites and when photons are efficiently distributed throughout the reactor. This comprehensive analysis also alters the way sustainability is evaluated. There are desirable properties of visible light, renewable feedstocks and recyclable solids, but none of them singly is decisive. A credible route should be selective and concentrated, and have good and abundant catalysts, recyclable low-hazard media, minimal work-up, and favourable photon and electrical efficiency. Mass metrics, hazard and life-cycle boundaries must be reported at a similar functional outcome. Co-optimisation is therefore required at several levels: between molecular photophysics and mesoscale organisation, between catalytic mechanism and photon transport, and between catalytic mechanism and process sustainability. The closure of these interfaces will render bio-inspired photocatalysis beyond impressing positive laboratory proof and will turn it into predictable, scalable and environmentally defensible organic synthesis. A well-

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developed study should do four things. It should justify each bio-inspired feature in terms of a defined elementary process. It should demonstrate that this feature improves isolated synthetic performance. It should identify the active catalyst and establish its stability over the course of the reaction. Finally, it should quantify the photon and auxiliary energy input and report material and hazard metrics for the catalyst. This chain of evidence transforms novelty into a testable contribution. It also helps to provide grounding for the field when determining the merits of increasingly sophisticated architectures.

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