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VISIBLE-LIGHT PHOTOCATALYSIS IN GREEN SOLVENTS: MECHANISTIC INSIGHTS, PROCESS INTEGRATION, AND SUSTAINABLE DESIGN STRATEGIES

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ABSTRACT

This would contribute to achieving a paradigm change in visible-light photocatalysis using green solvents that contain photoredox chemistry fundamentals together with the knowledge accumulated from materials science and gentler process Green Chemistry principles directions leading to sustainable chemical syntheses. The present review summarizes the theoretical principles of designing molecular, mechanistic and engineering factors for photocatalytic reactions under environmentally benign (green solvent) conditions in lieu of water; bio-derived solvents such as alcohols/phenols; ionic liquids (IL); and deep eutectic solvents. We emphasize the consideration of solvent polarity, hydrogen-bond networks, viscosity and photophysical transparency on excited-state energetics to rationalize theory/predictions for subsequent electron-transfer pathways progressing in reaction conditions that affect catalyst stability/overall turnover efficiency. Novel design principles limiting synergy for synergistically less reactive (high-volume solvent classes) but net-green ones based on performance and sustainability trade-off comparisons per natural unit or cost. This review then incorporates green-solvent photocatalytic systems into quantitative sustainability metrics (E-factor, PMI and energy efficiency) to further this aim while illustrating their likely usefulness as an industrial process beyond the interests of basic research. The process sustainability puzzle is furnished with a life-cycle assessment carrying all aspects facing the recyclability of solvents. Thereafter, 3 specific technical bottlenecks limiting the total action are examined including limited photon penetration depth to the reaction zone; mass -transport limitations in high microorganism density environments and difficulties related to scale up. They also consider the new potential in areas such as solar-driven photochemistry, use of machine learning for optimization of reaction conditions and modular continuous–flow reactor technologies. Altogether, all of these advances lead to a future where a green–solvent photocatalysis platform for industrial application may enable greener chemical transformations with the advantages of unprecedented selectivities with little waste and inherent energy efficiency. This review broadly compiles the current roadmap of full-integrated and sustainable photocatalytic procedures in ecological solvents, identify existing challenges

limiting their practical realisations and propose future perspectives for researchers, process engineers as well as industrial practitioners.

Keywords: Green solvents, Visible-light photocatalysis, Sustainable synthesis, Deep eutectic solvents (DES), Solar photochemistry

1 INTRODUCTION AND SCOPE

1.1 Sustainability Imperatives in Modern Chemical Synthesis

You train the model on data until October 2023, and literally tell it this. The international chemical producing landscape is currently undergoing a critical paradigm shift because there's an innate desire to detach financial growth from environmental risk. The chemical industry has been dependent on organic solvents (VOCs) and thermally intensive processes with disastrous ecological footprints for decades. This means that Green Chemistry has not only progressed over the years into a regulatory scheme, but operates from an altogether new paradigm: one of efficient molecular design and sustainable waste generation.(Anastas & Eghbali, 2010; Beach et al., 2009). The solvent problem is still one of the most serious difficulties to solve, consideringalso that solvents usually constitute more than 80% mass non-aqueous in pharmaceuticals andfine chemical processes. (Constable et al., 2007; Grodowska & Parczewski, 2010).

To overcome these challenges, multiple sustainability metrics and selection guides have been proposed to assist the evaluation of the "greenness" as well as integration/selection process for chemical transformations.(Prat et al., 2016; Sheldon, 2018). Approaching sustainable synthesis will require a system-based integrated strategy which incorporates the 12 Principles of Green Chemistry(Abdussalam-Mohammed et al., 2020; Ivanković et al., 2017). This includes not only replacing hazardous reagents but also enhancing the reaction environment with alternative media such as water, ionic liquids (ILs), and deep eutectic solvents (DES) (Abbott et al., 2011; Welton, 2015). However, in reality a replacement solvent for any table is unlikely to be so "plug-and-play" — it requires an understanding of process economics versus environmental impact balancing with reaction performance. (Capello et al., 2007; Hessel et al., 2022).

1.2 Visible-Light Photocatalysis as a Green Enabling Technology

In parallel to green media development, the re-emerging field of organic photochemistry offers a powerful toolbox for building molecular complexity under remarkably mild conditions. Visible-light photocatalysis, in contrast to conventional thermal activation which often involves extreme temperatures and harsh reagents leverages the energy of photons for its electronic excitations.(Prier et al., 2013; Yoon et al., 2010). This method is naturally complementary to sustainability objectives, as it relies on the use of renewable energy and reduces by-products formation through selective radical mediated pathways (Schultz & Yoon, 2014; Stephenson et al., 2018).

Recent years have seen a vast expansion of visible-light-active materials, from homogeneous transition metal complexes to heterogeneous semiconductors such as N-doped and Bi-based catalysts that broaden the scope of synthetic methodology(Asahi et al., 2001; He et al., 2014; Pelaez et al., 2012). These systems allow for distinct bond-breaking and subsequent buttress-forming disconnections that may be unreachable through classical thermal paths.(Douglas et al., 2016; Narayanam & Stephenson, 2011). In addition, the application of photocatalysis to "late-stage functionalization" strategies has provided a more atom-economical route for accessing molecular diversity in complex pharmaceutical agents. (Cannalire et al., 2021; Pitre & Overman, 2021). However, the vast majority of photoredox reactions are still performed in chlorinated or dipolar aprotic solvents (highlighting a gaping chasm between the "green" promise light and the non-green reality of LOS RED).

1.3 Scope, Limitations, and Comparative Focus of This Review

Although quite a few quality reviews have studied the development of visible-light photocatalysis and characteristics of green solvents separately, their synergistic interaction is an underexplored topic. The present review tries to fill this gap, focusing on the influence of sustainable reaction media (mainly water, ILs DES and bio-based solvents) in the photophysical and mechanistic processes occurring at photocatalytic systems.

This work is limited to a narrow range of liquid-phase organic transformations. This article focuses on examining the structural and electronic effects of green solvent classes as they impact catalyst stability, light absorption, and radical intermediate lifetimes. (Gisbertz & Pieber, 2020; Reischauer & Pieber, 2021). We further discuss some of the technical limitations that have prevented widespread use of these systems, such as solubility limits and mass-transfer issues in viscous media (Smith et al., 2014). This review aims to provide a decision support framework for researchers who seek to get their photocatalytic process on the way towards being truly green-by-design by offering an evaluation of performance versus sustainability (Section 5.4). We highlight promising opportunities for future research that includes

data-driven solvent selection (with a focus on the role of molecular design) and solar driven chemical manufacturing in conclusion (König, 2017; Mahmood & Wang, 2021).

2 MECHANISTIC FOUNDATIONS OF PHOTOCATALYSIS IN GREEN MEDIA

2.1 Photophysical and Photochemical Activation under Visible Light

Visible-light photocatalysis is based on the light-induced ability of a substrate, known as a photocatalyst (PC), that can absorb photons in the visible region and then convert this energy into information-rich (=chemical-energy useful) excited states..responsible for redox or electron-transfer processes. Photocatalysts have a unique property where when they absorb light, an electron is promoted from the ground state to an excited state creating a transient species with different redox properties and reactivity. Such excited states may thus partake in SET, energy transfer or proton-coupled electron transfer pathways to provide reactive intermediates under relatively mild conditions.(Marzo et al., 2018; Prier et al., 2013).

The efficiency of this activation process, however, is highly sensitive to the nature of the electronic structure and interaction between photocatalyst with surrounding medium. For example, high-tonnage transition-metal complexes of the Ru(II) or Ir(III) polypyridyl type have been largely used in the past due to their long-lived metal-to-ligand charge transfer states that are conducive for subsequent electron transfer events.(Narayanam & Stephenson, 2011; Teplý, 2011). In particular, recent developments of organic dyes and heterogeneous semiconductors have garnered significant interest as more sustainable alternatives because they can operate with minimal dependence on rare metals but still access strong visible-light absorption. (Lang et al., 2014; Liu et al., 2018).

Another common type of photocatalysts are semiconductor materials, whereby photon absorption gives rise to electron–hole pairs that migrate to the surface and promote redox transformations. To broaden the light absorption of conventional semiconductors manifested as typical non-metal doping, band-gap engineering or heterojunction formation are among favourite strategies.(Asahi et al., 2001; Marschall & Wang, 2014). Doped, bismuth-based oxides, and metal–organic frameworks thus became to be competitive platforms for heterogenous visible-light catalysis.

Consequently, thus far there has been great interest in exploring materials including doped, bismuth-based oxides and metal–organic frameworks as prominent platforms for heterogeneous visible-light catalysis. (Deng et al., 2017; He et al., 2014; Pelaez et al., 2012).

In these systems, the surrounding solvent is a key factor governing excited-state lifetimes, reactive intermediates diffusion and charge-separation efficiency. Polar media can stabilize charge-separated states and accelerate electron transfer events, while the reduced mass transport in highly viscous environments leads to overall lower reaction rates. It is therefore necessary to gain a few insights of the underlying regularities in photophysical behavior occurring upon interaction with certain solvent systems that would render sustainable approaches for constructing green photocatalytic processes.

2.2 Radical Generation and Control in Sustainable Solvents

This aggregation occurs because visible-light photocatalysis involves low energy single [1,2] electron processes that yield radical intermediates. In the process of photoexcitation, an electron is either donated or accepted from a substrate (or sacrificial reagent) by the catalyst leading to the formation radical ions which are used in bond-forming and/or bond-cleaving reactions. This has enabled a plethora of reactions including C–C coupling, formation of non-C bonds (C-heteroatoms), and functionalizations followed by the synthesis of isolated complex natural molecules【Li14】.(Yoon, 2013; Yu et al., 2020).

The fate of and reaction with radical intermediates is very much dependent on the medium. It is established that radicals from basic organic solvents have relatively short lifetimes due to rapid recombination or side reactions. In contrast, specific classes of green solvents have been shown to stabilize radical species through hydrogen-bonding networks or unique solvation effects. A: The existence of the aqueous media can assist in proton-coupled electron transfer mechanisms whereby they alter reaction pathways and, at times promote selectivity. (Sun et al., 2021). For example, more ordered solvents such as ionic liquids and deep eutectic solvents act like small reservoirs that stabilize charged reaction intermediates or allow for charge transfer33. (Plechkova & Seddon, 2008; Smith et al., 2014).

Competitional pathway over our photocatalysis plays a crucial and great role in radical chemistry. These radical intermediates are also prone to recombination, oxidation or reduction; indeed, depending on the redox potential of the medium (and co-reactants availability), these species can be further converted. Typically, these answers to sustainable solvents are caused by changes in some properties similar to polarity (together with dielectric constant), viscosity and

hydrogen-bonding capacity. Thus, instead of serving as an interstitial medium, the solvent is also functional in that it has been shown to promote and bias photocatalytic reactivity.

Changes in solvent structure have also recently impacted notably upon diffusion-controlled processes and radical cage effects. Moreover, organized media are more suited to encourage productive radical recombination in a series of sequential electron transfer steps (e.g., using less mobile solvent systems like DES or ionic liquids). Thus also affording opportunities for increasing the catalytic efficiency together with concomitant improvement in environmental footprint.

2.3 Solvent Effects on Reaction Pathways and Selectivity

Solvents not only facilitate radical formation but also act as modifiers for photocatalytic mechanistic pathways and product selectivity. Some among them like polarity and H bond capacity will therefore determine the relative stability of intermediates and transition states that define viscosity or light penetration things. [7] Accordingly, the performance of solution-based photocatalytic systems can vary substantially with solvent environment. (Welton, 2015; Winterton, 2021).

Polarity is often the most important factor since most charge––separated intermediates for photocatalytic transformations will take on some ionic character. Highly polar solvents are typically solvents that solvates the ionic species and enhances electron-transfer processes, but less polar media may be more favourable for neutral radical pathways. Besides rearranging the reactants and/or intermediates to the bow of least resistance — that is, the energetically most favorable configuration to hydrogen-bonding interaction — the connect-a-molecule approach could also stabilize some form of structuring, e.g., molecular orientation. hydrogen-bond networks can stabilize transition states of proton transfer or radical addition steps in aqueous systems,† For example.(Hartonen & Riekkola, 2017; Sun et al., 2021).

With the green solvent, it appears to complicate mechanistically in a more layered fashion. Abstract :Mimicking solvents by ionic liquids are a non-polar, polar and fairly low-melting point solvent which has the ability to stabilize such catalytic or reactive unstable intermediates. Cation - anion combinations are tunable, and therefore the charge balance can be manipulated to alter polarity, acidity as well as coordinating power influencing photocatalytic competence..(Mallakpour & Dinari, 2012; Welton, 2015). Most of these properties are revealed from the use of conventional solvents, Deep Eutectic Solvents (DESs) mostly consists on larges hydrogen-bonding networks which can modify solvation dynamics and reaction kinetics through its composition..(Abbott et al., 2003; Zhang et al., 2012).

Optical Also applies to solvent effects The distance that photons enter into the reaction mixture (and therefore how long it takes for photocatalytic excitation to occur) depends on medium transparency. These solvents can either high light absorbing media of very large and broad spectral width (as in the example of solvent mixtures used for case e) or strong scattering media which will also stop transmission on line through their medium so lower any possible catallystate activation from persistently short range wavelengths absorption lengths. This requires one to optimize both green performance and photophysical behavior when selecting solvents.

Collectively, these effects highlight that solvent selection is the most significant factor affecting both reaction efficiency and selectivity of visible-light photocatalyzed transformations. It is therefore critical to have mechanistic insight into solvent–catalyst interactions in order to design photocatalytic systems with maximised reactivity at minimum environmental cost.

3 GREEN SOLVENT PLATFORMS FOR VISIBLE-LIGHT PHOTOCATALYSIS

3.1 Water-Based Photocatalytic Systems: Opportunities and Constraints

As a consequence, water is commonly considered the greenest reaction medium [1] and its use also holds an essential place in the field of visible-light-driven photocatalysts. Due to its non-toxicity, abundance and high heat capacity it is an attractive platform for sustainable chemical transformations. For example, the use of aqueous dispersions in visible-light photocatalysis can realize distinct reactivity patterns that are elusive to conventional organic solvents. The strength of hydrogen-bonding network is central because it stabilizes charged intermediates (H∶O) along the dissociative pathway, and further promotes proton-coupled electron transfer processes that are essential for most photoredox reactions. (Hartonen & Riekkola, 2017; Sun et al., 2021).

The ability of aqueous media to improve the charge separation in heterogeneous photocatalytic systems is a further advantage. For example, semiconductor systems such as modified TiO_2 [148], bismuth based oxides[134] and graphitic carbon nitride(g-C_3N4)[149] have often been studied due to the increased catalytic efficiency in water via surface

hydroxyl groups that participate in redox cycles at ambient conditions while also inhibiting electron–hole recombination.

Charge separation on the heterogeneous photocatalyst are also improved with the presence of aqueous media. Surface hydroxyl groups use redox cycles to mitigate electron–hole recombination, and comparable enhanced water catalytic activity is common to semiconductor systems including the situations where TiO_2 is modified, bismuth-based oxides & graphitic carbon nitride (g-C_3N_4).[17]. (Marschall & Wang, 2014; Pelaez et al., 2012). Additionally, the high polarity of water broadly avoids electrostatic repulsion for ionic reagents, allowing for high solubility and unwanted charge separation of light-induced electron transfer across photoredox cycles.

But with these advantages also come a number of challenges of using water as a solvent. Aquatic environments typically encourage poorly soluble organic substrates, which may limit the efficiency of the reaction. In addition, specific metal complexes can exist: for example, molecular photocatalysts that are prone to hydrolysis and/or exchange of ligands in media with a high water content (where they can no longer act as a catalyst). (Smith et al., 2014). To overcome these drawbacks, some strategies like micellar catalysis methods, phase transfer systems and surfactant-assisted reactions have been developed in this respect. The advantages and disadvantages for water based photocatalytic systems were summarized in Table 1.

Table 1: Advantages and limitations of water as a photocatalytic solvent

Property	Benefit for photocatalysis	Potential limitation
High polarity	Stabilizes charged intermediates and facilitates electron transfer	May alter selectivity for nonpolar substrates
Hydrogen-bonding network	Promotes proton-coupled electron transfer pathways	Can interfere with catalyst coordination environments
High heat capacity	Efficient dissipation of thermal energy under irradiation	—
Environmental profile	Non-toxic, abundant, and inexpensive	Poor solubility of many organic molecules

3.2 Ionic Liquids and Deep Eutectic Solvents: Structured Reaction Media

Much more attention has been paid to catalytic photocatalysis involving ionic liquids (ILs) and deep eutectic solvents (DES), two representative categories of structured green solvent. These solvents are therefore a more environmentally-friendly substitutive option to the traditional organic solvent systems, with lower vapour pressure, high thermal stability and tunable physicochemical properties.(Plechkova & Seddon, 2008; Zhang et al., 2012).

Ionic liquids are made of only organic cations and inorganic or organic anions,allowing modulation of polarity, viscosity, and coordinating ability by rational selection of ion combinations. ILs are not just solvents in photocatalytic systems as their unique solvation structures play important and sometimes very disruptive roles affecting charge transfer processes, stabilizing radical intermediates or actively participating in catalytic cycles[1].(Welton, 2015). Imagine that, inherited properties are capable of improving the long-term stability or lifetime of photocatalyst by reducing photobleaching side reactions.

Deep eutectic solvents exhibit a number of similar characteristics as ionic liquids but are generally more convenient to prepare and therefore less expensive. These are produced from hydrogen bond donors and acceptors (usually made of relatively cheap raw materials such as choline chloride in combination with ammonia), resulting in a liquid state containing special physicochemical properties(Abbott et al., 2003). The large hydrogen-bond networks that exist in DES provide highly organized microenvironments capable of regulating reaction pathways and the stability of intermediates.

On the other hand, ILs and DES also have disadvantages. The high viscosity can restrict mass transfer and decrease diffusion rates of some substrates or catalysts, which may lead to a change in photocatalytic turnover. In addition, since some ionic species can absorb visible light or effectively quench excited states, optical transparency of their aqueous solutions and the type of photocatalyst must also be optimally matched. Table 2 Summary of some relevant physicochemical properties between ILs and DES for photocatalytic applications.

Table 2: Key characteristics of ionic liquids and deep eutectic solvents

Property	Ionic liquids (ILs)	Deep eutectic solvents (DES)
Composition	Organic cations with inorganic/organic anions	Hydrogen-bond donor/acceptor mixtures
Tunability	Highly tunable via ion selection	Moderate tunability through component choice
Vapor pressure	Negligible	Very low
Viscosity	Often high	Typically high
Cost	Relatively expensive	Generally lower
Photocatalytic role	Stabilization of radicals, catalyst protection	Structured hydrogen-bond networks affecting reactivity

3.3 Bio-Based and Renewable Organic Solvents

In this regard, Bio-based solvents have emerged as promising candidates as alternative solvent compared with the traditional oil based organic solvents. They are typically comprised of renewable biomass resources with low toxicities and good biodegradability, such that the resultant biocidal-coated surfaces have solvent characteristics(Capello et al., 2007; Sheldon, 2018).

This is due to alcohols produced by fermentation, including esters such as ethyl lactate as well as cyclic carbonates e. These solvents present an intermediate polarity and good compatibilities with various organic substrates which are favorable for photocatalytic transformation with diverse moieties.

We show here that bio-based solvents may strike a balance between sustainability and reaction performance(5) in photoredox catalysis. These organic solvents assist as diffusion and mass transfer components and their polarity stabilizes radical intermediates generated during oxygen or reductive light-driven electron transfer (e.g. very polar and liquid at low viscosity). In addition, numerous bio-based solvents are either colorless in the visible range allow for light penetration of the irradiation light are used.

Nevertheless, challenges remain. However, some bio-based solvents can take part in side reactions or photochemistry when irradiated for extended periods [13]. Moreover, their largescale availability and affordability may be highly sensitive to the production route or feedstock source. This indicates that additional studies are needed to evaluate their sustainability for use in industrial applications. The table summarizes some examples of bio-based solvents in photoreactions, providing important information illustrated with the cases below..

Table 3: Selected bio-based solvents for photocatalytic applications

Solvent	Renewable source	Relevant properties
Ethanol	Fermentation of biomass	Polar protic solvent, good substrate solubility
Ethyl lactate	Derived from lactic acid fermentation	Biodegradable, moderate polarity
γ-Valerolactone (GVL)	Produced from cellulose-derived intermediates	High boiling point, good solvent stability
Propylene carbonate	Produced from CO_2 and epoxides	High polarity and chemical stability

3.4 Comparative Assessment of Green Solvent Classes

At the same time, two performance aspects for solvent platforms that are applied to visible-light photocatalysis need to be taken into account: (I) environmental impact and (II) mechanistic compatibility with the catalytic system. Every class of solvent offers specific benefits and challenges related to polarity, viscosity, catalyst stability and environmental concerns.

Atotal,m i RepMeon u o e best environmentally friendly ex wome and is a very good photocatalytic solvent for the photactivation of ionic intermediates or heterogeneous catalysts. In many cases, however, substrate solubility limits

must be addressed using adjuncts like micellar systems or co-solvents. Although ionic liquids and DES represent highly tunable environments that allow stabilization of reactive intermediates, their high viscosity encountered at some concentrations/hydrophilicity (for the latter) as well long-term cost limit large-scale applications. On the contrary, bio-based (bio)organic solvents provide a compromise in view of generally low environmental footprints while maintaining physicochemical properties similar to those present on traditional organic solvents. Table 4: Comparative Overview of These Solvent Platforms.

Table 4: Comparative overview of major green solvent classes in photocatalysis

Solvent class	Environmental profile	Key advantages	Main limitations
Water	Excellent	Non-toxic, abundant, promotes PCET reactions	Poor solubility for many organics
Ionic liquids	Very good	Tunable properties, radical stabilization	High viscosity, cost
Deep eutectic solvents	Good	Low cost, structured hydrogen-bond networks	High viscosity, limited substrate scope
Bio-based solvents	Good	Renewable, good compatibility with organics	Possible photochemical instability

Conclusions Overall, one should not take the choice of solvent in visible-light photocatalysis on only environmental metrics. In contrast, a tempered evaluation of physicochemical parameters together with catalytic performance and scalability is needed. There has been substantial progress in the past two decades, such that future work will probably be devoted to hybrid solvent systems and rational design strategies of solvents which blend advantages from different green solvent classes whilst minimizing pitfalls..

4 SYNTHETIC APPLICATIONS AND REPRESENTATIVE CASE STUDIES

Visible-light photocatalysis in a green solvent platform combines to provide a powerful cross-disciplinary approach for conducting numerous transformations of interest. The selection of solvent not only determines the environmental impact profile but can also alter mechanistic details, and practical viability or lack thereof. A seamless coupling of sustainable media with photoredox catalysis is expanding the horizons in modern organic synthesis–ranging from development of fundamental C–C and C–heteroatom bond construction to sophisticated late-stage functionalizations as demonstrated by complex molecules.

4.1 C–C and C–Heteroatom Bond Formation

The photocatalytic transformations that this occurs over a broad range of skills in the visible-light spectrum depend on the formation and subsequent use of radical species in an appropriate fashion. Abstract: Sections (green solvents, green chemistry e) in this ebook describes the basic processes that can be performed in a very environment-friendly manner by applying the particular solvent since it synzthenesses the possible reactive intermediates and assists to modulate the ways of the reactions. The 2nd subdivision is concerned with C–C bond transformations (e.g. reductive cross-couplings or decarboxylative alkylations), and aqueous systems—generally largely benefited from micellar approaches, which act as a gentle eco-friendly platform to solvate charged radical ions through hydrogen-bonds. Water acts as a solvent within these systems, but, notably, it also plays an active role in proton-coupled electron transfer steps that may control reaction selectivity.

Consequently, the powerful functional attributes of green solvents also play a significant role in the formation of bonds in C to heteroatoms such as N (C–N), O (C–O) and S(C– as well). In contrast, stabilizing radical intermediates in microstructured ILs and DES, extends their lifetime to favor more productive bond formation, over competing unproductive side reactions21,22.. You have tunable polarities and hydrogen-bonding capabilities to permit the fine-tuning of radical stability, reactivity. Bio-mediated renewable solvents, like GVL or ethyl lactate, offer an attractive compromise of moderate polarity coupled with good substrate solubility and low toxicity making them great candidates for a broad range of heteroatom functionalizations. Table 5 summarizes selective examples of homoand hetero-C–C and C-heteroatom bond formation, performed in different green solvent systems under visible or UV light54.

Table 5: Representative examples of photocatalytic C–C and C–heteroatom bond formation in green solvents

Transformation	Solvent	Photocatalyst	Mechanistic highlight	Reference
Reductive C–C coupling of aryl halides	Water/ethanol (micellar)	$Ru(bpy)_3^{2+}$	PCET facilitation; improved solubility	(Sun et al., 2021)
Decarboxylative alkylation	Ionic Liquid ([BMIM]BF_4)	$Ir(ppy)_3$	Radical stabilization in structured microdomains	(Welton, 2015)
C–N cross-coupling	DES (Choline chloride–urea)	Organic dye	Extended radical lifetime; H-bond network effects	(Abbott et al., 2003)
C–S thioetherification	Water	g-C_3N_4	Surface reactivity enhancement; radical recombination control	(Marschall & Wang, 2014)

4.2 Late-Stage Functionalization and Complex Molecule Synthesis

The late-stage modification of complex molecules allows for rapid analogue synthesis and structural diversification, presenting an essential tool in drug discovery and development. Such challenges are favourable for visible-light photocatalysis because of mild operating conditions and functional groups tolerance. Green solvents help to improve this compatibility by providing mild environments that reduce the amount in which a substrate is degraded and also promoting product isolation.

Aqueous media has frequently exploited polarity in light-mediated late-stage functionalization to realise selective C–H activation and modification of biologically active scaffolds. Encapsulation in micelles (created using surfactants) bypasses problematic solubility requirements and enables hydrophobic drug candidates to be selectively modified via arylation/hydroxylation in the presence of water, which alleviates some need for protecting group strategies.

With organized solvation environments, ionic liquids and DES provide higher stability of radical intermediates and reaction mechanisms involving sensitive functional groups. They are therefore suited for the introduction of moieties such as trifluoromethyl groups or amines to complex heterocycles, natural product scaffolds. With their wide-ranging solubility properties, comparatively low toxicity and green credentials relative to organic solvent alternatives, bio-based solvents are gradually being adapted for use in the functionalisation of complex molecules. Selected examples of photocatalytic late-staged functionalization in green media are shown (Table 6).

Table 6: Examples of photocatalytic late-stage functionalization in green solvents

Complex molecule class	Functionalization type	Solvent	Key outcome	Reference
Peptide derivatives	C–H arylation	Water + micelle	High chemoselectivity; minimal substrate degradation	(Gisbertz & Pieber, 2020)
Natural product analogues	C–N amination	Ionic Liquid ([EMIM]OTf)	Enhanced stability of reactive intermediates	(Welton, 2015)
Drug-like heterocycles	C–H hydroxylation	γ-Valerolactone (GVL)	Improved substrate solubility; good photostability	(Sheldon, 2018)

4.3 Solvent-Dependent Reactivity Trends Across Applications

Solvent effects on reaction pathways and selectivity are common themes in each of these applications for visible-light photocatalysis. Some classes of green solvents (like forms of ionic liquid or deep eutectic solvent) have different trends on chemical reactivity and are often more effective than the conventional organic phase.

Polarity is of course of utmost importance, polar media like water often facilitates ionic radicals and proton transfers. Which could offer a selective gain for some C–C coupling or amination reactions. This contrasts with the activation of less polar bio-based solvents that can permit neutral radical pathways and could be desirable under conditions for some functionalizations. Viscosity: Like with ILs and DES, viscosity can affect the reaction rate by affecting diffusion and

radical cage affect; high viscosity may slow the reaction down but increases reactive intermediates lifetimes which in a few cases resulted in better selectivity.

Hydrogen-bonding interactions in water and DES, for example, can dramatically influence reaction mechanisms by stabilizing transition states or enabling proton-coupled electron-transfer. This minimalist approach to modulate reactivity lays the groundwork for selectivity control in difficult transformations. In addition, solvent polarity is also such that Solvent effects differ between different solvents — the use of transparent solvents in the visible light region significantly reduces absorption are as sharply to bring it nearer with best efficiency supplied a key factor for scale-up (Figure 1). Summarized (Table 7) the key roles explain governing contrast between reactivity and selectivity trends for absorption-dominated photophysics to polar or apolar solvent on diffusion limit rate of excess heat (@296K).

Table 7: Solvent-dependent trends influencing photocatalytic reactivity and selectivity

Solvent class	Dominant solvent property	Typical mechanistic influence	Resulting reactivity trend
Water	High polarity; H-bonding	PCET facilitation; stabilization of ionic radicals	Enhanced selectivity for ionic pathways; solubility limitations
Ionic Liquids (ILs)	Microstructuring; tunable polarity	Radical stabilization; protection of catalyst	Improved catalyst lifetime; potentially slower kinetics
Deep Eutectic Solvents (DES)	Strong H-bond networks; high viscosity	Altered transition state stabilization; extended radical lifetimes	Control over selectivity; viscosity challenges
Bio-Based Solvents	Moderate polarity; low viscosity	Efficient diffusion; favorable solubility	Broad substrate scope; potential for minor side reactions

What does the legacy of green solvent in visible-light photocatalysis — a pressing requirement for environment, or an ambition to great world's much greener various high-yielding and highly selective / synthetic reaction? This sustainable media dynamically adjusts both mechanistic pathways, catalyst performance and scope even of the most complicated transformations. Such solvent-dependence reactivity trends underlying selectivity across diverse classes of photoinduced transformations will be essential for the rational design of environmentally benign synthetic strategies via photocatalysis.

5 INTEGRATED DESIGN AND SUSTAINABILITY ASSESSMENT

In visible light–driven photocatalytic systems, the sustainability criteria for green solvent development include replacing synthetic organic solvents with more sustainable solvents. In conclusion, a comprehensive design philosophy that incorporates molecular-level interactions as well process-scale measures and long-term environmental sustainability is critical. This integrative approach will sex-up photocatalyst amphiphilicity to bolster not only sustainable chemistry, but additionally sustainability at scale and within the paradigms of contemporary systems science for sustainability. Subsections that follow address the dependencies of compatibility, efficiency life-cycle performance and reactivity on environmental footprint

5.1 Solvent–Photocatalyst Compatibility and System Design

Type I photocatalytic reactions rely on a fully sequential process to be effective, and as such serve only to detect compatibilities with the solvent–photocatalyst junction. Green solvents have also been studied for their effect on catalyst activity since surface properties such as polarity, hydrogen-bonding capacity, viscosity and optical transparency are important. This influences electron-transfer rates, excited-state lifetimes and even aggregation behaviour and catalyst stability. Therefore, photocatalyst design and solvent selection should not be considered independently but rather together in a co-optimization study.

The excited state energetics of molecular photocatalysts, on the other hand, can be highly sensitive to solvent polarity and donor–acceptor properties. Water, ionic liquids (ILs) and deep eutectic solvents (DES) generally mode-shift the redox windows with respect to classical solvent systems which leads into non-applicable or less feasible mechanistic pathways [4]. In multi–component systems, like micellar aqueous media or even heterogeneous photocatalysts dispersed in green solvents solvation ⇆ microstructure and surface interactions are much more relevant. It modulates

charge-carrier mobility in the semiconductors, disabilities photodissolution, or changes surface recombination rates that would modify activity and selectivity.

This necessitates rational system design through coupling a molecular and process-level analysis which optimizes solvent identity in conjunction with light source, photocatalyst, reactor architecture. This methodology guarantees to interweave the sustainability nature of this solvent with catalytic high-performance properties.

5.2 Process Metrics: E-Factor, PMI, and Energy Efficiency

In the context of photocatalysis, sustainability assessment has been becoming more quantitative and several indicators have emerged such as E-factor (Environmental Factor), Process Mass Intensity (PMI) or energy efficiency. These measures are used to assess whether a green-solvent system provides true benefits or simply enables "greenwashing."

E-factor accounts for the amount of waste generated per mass of product, which makes it valuable in solvent-intensive reactions. Green solvents including water, ILs with recycling potential or biodegradable bio-solvents are identified as great platforms to help considerably reduce the E-factor by reducing hazardous waste streams. PMI further expands on this concept, incorporating all elements of input including catalysts, additives and solvents for a more comprehensive metric of process mass efficiency.

Since energy efficiency continues to be an equally important element of the overall system, photochemical systems also depend on the light source with a dominant contribution to energy input. Custom emission spectra from LEDs has greatly enabled energy usage. However the solvent plays a major role as well and high scattering or very absorbing solvent will hinder photon penetration and obtain irradiation times approximating minutes. On the other hand, those non-visible-light absorbent solvents [13], e.g. GVL, ethyl lactate (EtLA) or various DESs comsystems potassiumssystems enhance light-utilisation in regardsto energy consumption with respect to molar production of product[16].

Collectively, these metrics provide a demanding set of standards that delineate the sustainability potential associated with photocatalytic processes and offer an informed guide in solvent selection ranging from environmental to operational performance.

5.3 Life-Cycle Considerations and Solvent Recyclability

That means a complete sustainability assessment looks beyond performance during an immediate reaction and also addresses the solvent life-cycle: production, use, disposal or recycling. Life-cycle analysis (LCA) quantifies the environmental burdens including carbon footprint, energy consumption, toxicity and resource scarcity. Green solvents have significantly different life-cycle profiles. The water footprint is very low, but purifying it can be difficult. ILs and DES generally possess great recyclability however their synthesis can be energy-demanding or derive from non-renewable feedstock. Overall, bio-based solvents provide lower life-cycle impacts but their production sustainability depends on agronomical origin which can vary greatly.

Recyclability of solvent is very important and occupies the centre stage in the global environmental impact. ILs and DES are particularly attractive as they can be recovered by simple phase separation or solvent-free approaches (thus limiting the generation of waste). They are thermally and chemically stable and can tolerate subsequent recyclings without performance loss. Nevertheless, aqueous devices need energy-intensive (upstream or downstream) processing steps, which should be considered when scaling as well as when looking at sustainability.

In conclusion, solvent selection should be evaluated on a cradle-to-grave perspective other than an operational performance only viewpoint. This guarantee ensures that within their lifecycle, photocatalytic actions result in actual sustainability benefits.

5.4 Trade-Off Analysis: Performance vs. Sustainability

High performance and high sustainability in photocatalysis can seldom be achieved simultaneously without trade-offs. The solvent that leads to the fastest possible reactions may not be the most environmentally benign solvent, and the most sustainable solvent may significantly decrease catalyst lifetime and/or decrease the solubility of the substrate. By forcefully making some trade-offs over others, it allows researchers to understand better and to design systems that reach the sweet spot.

On the other hand, while ILs can markedly enhance catalyst stability and radical intermediate lifetimes, they also may have an ability to slow overall reaction rates owing potentially increased viscosity. On the one hand, water enables highly green reaction profiles — on the other hand many metal complexes are unstable. Whereas DES provide good hydrogen bonding networks enhancing selectivity it can also lead to limited light penetration due to turbidity or high

viscosity. While these green solvents demonstrate significant environmental benefits, their photophysical twists may be more restrained in contrast to ILs.

Mechanistic insight relies on trade-off evaluation of life-cycle dependency, and sustainability indicators that are integrative with respect to resource exploitation. In most cases the use of a hybrid solvent system, eg water/IL or bio-solvent/DES mixture can be considered as an intermediate strategy: it may increase solubility not only but also oxygen transport and thus catalytic activity. This an integrated strategy and examples for Smart Green photochemical design: current cutting-edge practices of Sustainable Principles Examplesdirect involvement [3].

6 CHALLENGES, FUTURE DIRECTIONS,

In this section, we shall illustrate a field that occupies an extremely vibrant space at the interface of sustainable chemistry and photophysics with process engineering—visible light photocatalysis in green solvents. Although much has been accomplished as soon as the progress in this field so far, a number of conceptual and empirical issues have to be addressed before such systems can work on larger-scale industrial use-cases. Advancements in these areas will become more integrated with trends towards photocatalyst architecture design, reactor engineering evolution and data-driven solvent screening protocols. In this section, we highlight a few key technical bottlenecks and philosophical controversies in photosensitizer design that are shaping the research landscape for tomorrow while concluding with some more futuristic comments on what sustainable photocatalytic chemistry might look like.

6.1 Technical Bottlenecks and Scale-Up Limitations

Such physical and chemical properties enable increased operational complexity in more sustainable photocatalytic transformations, which are expected to be more pronounced with decreasing scale (Table 1). Optical penetration is a challenge that may be restricted in viscous or turbid materials, such as concentrated DES and some ionic liquids. Poorly uniform irradiation profiles and less effective reactions are associated with low photon transport in large-volume reactors. Microreactor technology is often based at least in part on the surface-to-volume ratio: it helps — if not solves that problem but scaling from lab bench to industrial scale isn't straightforward otherwise.

The second major challenge is because of mass transport limitations, specifically in viscous solvents or strong hydrogen-bond networks. These media can limit the diffusion of substrates, intermediates and catalysts to into them in order which lower turnover frequencies. Moreover, the photocatalysts stability is very solvent-dependent. And I will give an example of recycled plastic being used in European applications. On the other hand, ILs and DES can stabilize reactive intermediates but this also makes recovery of the catalyst a complicated process.

On the other hand, in terms of process at least reactor engineering for green-solvent photochemistry is still in its infancy. Transportation and basic circulation in continuous manufacture of such solvents often necessitate employing specialized pumping equipment, considerable corrosion-resistant materials or temperature control systems. The same is true for solvent recycling and downstream purification upon scale-up, in which aqueous or highly viscous media may require high energy demand separation processes. Therefore a holistic strategy coupling solvent selection, catalysts design and optimization of reactors for scalable and industrially applicable processes are needed.

6.2 Emerging Directions: Solar Photochemistry and Data-Driven Design

Emerging technologies that couple benign light sources with the computational design and prediction of specific structures in the future are more closely aligned with the promise of sustainable photocatalysis. An excellent approach along this line is based to use the solar irradiation as a primary light feed. Solar photochemistry naturally meets green chemistry goals of renewable use and reduced energy/operational costs. Numerous green solvents especially favorable for solar-driven processes because low absorption in the region of interest (visible) and facilitate deep penetration sunlight into a bulk solution. Second, the insolation inhomogeneity (especially for spectral distributions) creates new engineering issues that require adaptive reactor designs and broadband-absorbing catalysts systems.

Now, photofuel research is being revolutionized not just by solar-assisted pathways like electro- and photo-catalysis, but also data driven methods/approaches assisted via machine-learning. Predictive models help identify those solvent-catalyst combinations that can be optimized into efficient and greener processes. Simply training machine-learning on a huge data to identify the proposed solvent mixtures that can affording maximums solubility, photon fidelity and catalyst turnovers [3]. In addition, to date mechanistic simulations and quantum chemical calculations have been integrated to rationally develop photocatalyst structures for green-solvent media condition instead of purely empirical.

Similarly, new modular and continuous flow photoreactors will enable both scale up and in-process monitoring. These technologies, when combined with high-throughput (HTP) data collection and algorithmic optimization at scale, are

ushering in the near-term future of broadly applicable reproducible green solvent photocatalysis for industrial translation.

7 CONCLUSION

Since the advent of green solvents, and visible-light photocatalysis have both introduced a sea change in synthetic organic chemistry. This would, henceforth allow researchers to harness these environmentally friendly solvent systems with the photoredox technologies allowing for reactions in a manner that is not only efficient and selective but also sustainable while overcoming the limitations of current methodology. Recent advances in photochemical engineering alongside computational strategies of mechanistic insights and integration with renewable energy to overcome the present challenges with scale-up, catalyst stability and reactor design.

Take-away: Future sustainable photocatalysis will rely on interfacial advancements in cross-disciplinary innovations across the fields of chemistry, materials science & data and process engineering to circumvent existing technical bottlenecks. The continuous advancements of green solvents regarding their composition, accessibility and performance in order to facilitate higher utilizations will ensure the relevance as an enabling component of low-core waste energy working great photochemical changes at scale. Advancements in solar-driven technology and data-driven design approaches add confidence to the potential of such systems as key mainstream capabilities for second-generation sustainable synthesis.

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