

GREEN SOLVENT-MEDIATED SUSTAINABLE ORGANIC SYNTHESIS: A CATALYTIC APPROACH

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ABSTRACT

The transition away from volatile, petroleum-derived organic solvents remains one of the most consequential shifts within contemporary synthetic chemistry, and catalytic methodologies built around green reaction media now occupy a central place in that transition. This review consolidates a decade of published work on deep eutectic solvents, ionic liquids, water, supercritical carbon dioxide, bio-based solvents, and solvent-free mechanochemical routes, examining how each medium interacts with catalytic systems ranging from organocatalysts and metal nanoparticles to immobilized enzymes. Rather than cataloguing individual reactions, the analysis draws together reported yields, E-factors, catalyst recyclability, and reaction-time data across roughly ninety primary sources to identify where green-solvent catalysis genuinely improves on conventional practice and where the improvement is more modest than the literature's framing suggests. Deep eutectic solvents and solvent-free protocols emerge as the most consistently favourable in terms of atom economy and waste reduction, while ionic liquids and supercritical fluids show strong performance metrics but weaker evidence of industrial scalability. The review also surfaces recurring methodological gaps: inconsistent green-metric reporting, limited life-cycle assessment, and sparse data on solvent toxicity to living systems. These gaps, more than any inherent limitation of the chemistry itself, constrain confident claims about sustainability. The paper closes by outlining research directions capable of closing this evidence gap, including standardized metric reporting and computational solvent-selection frameworks.

Keywords: *Green solvents¹; Deep eutectic solvents²; Sustainable synthesis³; Catalysis⁴; Ionic liquids⁵; Atom economy⁶; Green chemistry metrics⁷*

1. INTRODUCTION

1.1 Background and Rationale for Green Solvent Chemistry

Solvents make up the largest mass fraction of most organic syntheses, often exceeding eighty percent of total process mass, yet they rarely appear in the target molecule itself. That imbalance places solvent choice at the center of any serious attempt to make organic synthesis less wasteful[1]. Anastas and Warner's twelve principles of green chemistry named safer solvents as one of the earliest and most tractable levers for reducing environmental burden, and in the decades since, that single principle has generated an entire sub-field concerned with replacing dichloromethane, DMF, and similar volatile organic compounds with alternatives that are less toxic, less flammable, and easier to recover[2]. The urgency behind this shift is not purely academic. Regulatory pressure through REACH in Europe and comparable frameworks elsewhere has steadily restricted the industrial use of several traditional solvents, pushing pharmaceutical and fine-chemical manufacturers toward substitutes long before the underlying chemistry was fully mature[3].

What counts as a 'green' solvent is, admittedly, contested. Water is cheap and non-toxic but often incompatible with moisture-sensitive substrates; ionic liquids offer negligible vapour pressure but their own synthesis can be energy-intensive; deep eutectic solvents are biodegradable in principle but recycling efficiency varies enormously between reported protocols[4]. This review treats greenness as a spectrum rather than a binary label, and evaluates each solvent class against the same set of reported outcomes[5].

1.2 Evolution of Catalytic Systems in Sustainable Synthesis

Catalysis and solvent choice are rarely independent decisions. A solvent that stabilizes an active catalytic species, or participates directly in the catalytic cycle, changes the calculus of what counts as sustainable far more than a simple solvent swap[6]. Deep eutectic solvents illustrate this well: several hydrogen-bond-donor/acceptor combinations act simultaneously as reaction medium and Brønsted or Lewis acid catalyst, collapsing two process inputs into one. Ionic liquids show a related dual role in metal-nanoparticle-catalyzed couplings, where the ionic environment stabilizes nanoparticles against aggregation without additional stabilising ligands[7]. Enzymatic catalysis in near-physiological aqueous or eutectic media extends this logic further, since biocatalysts frequently perform best in the very media that also qualify as green by conventional metrics[8].

1.3 Scope and Objectives of the Present Review

This review restricts itself to catalytic organic transformations reported between approximately 2015 and 2025, spanning deep eutectic solvents, ionic liquids, aqueous media, supercritical carbon dioxide, bio-based molecular solvents such as 2-methyltetrahydrofuran and ethyl lactate, and solvent-free mechanochemical protocols[9]. The objective is threefold: first, to synthesise reported performance data across these solvent classes on a common set of metrics; second, to critically assess the methodological rigor with which sustainability claims are supported in the surveyed literature; and third, to identify the specific gaps that limit confident comparison between studies, thereby pointing toward a more standardized future research agenda[10].

2. LITERATURE SURVEY

Deep eutectic solvents (DESs) constitute the single largest category within the surveyed literature, reflecting a genuine surge of interest since Abbott's original description of choline chloride-based eutectic mixtures[11]. Provide the most comprehensive account of DES fundamentals, documenting the physicochemical tunability that follows from varying the hydrogen-bond-donor-to-acceptor ratio, and this tunability is precisely what has attracted synthetic chemists[12]. Tavakol and Shafieyoon classify DES applications in organic synthesis by reaction type, noting that choline-chloride-based systems now see substantial industrial interest given their low cost and the scale at which choline chloride itself is already produced for animal feed[13]. Extend the picture to catalysis specifically, distinguishing homogeneous, heterogeneous, and electrocatalytic applications of DES media and observing that heterogeneous and electrocatalytic uses remain comparatively underexplored relative to homogeneous organocatalysis[14].

A recurring theme within the DES literature is the dual solvent-catalyst function[15]. Bioinspired DES systems that combine catalytic activity with demonstrated biocompatibility, positioning these mixtures as candidates for pharmaceutical-adjacent synthesis where downstream toxicity matters as much as reaction efficiency[16]. Survey the intersection of enzymatic catalysis and natural deep eutectic solvents (NADESs) across the 2010-2022 window, organising transformations by functional-group interconversion and reporting that NADES media frequently approximate intracellular conditions closely enough to support native enzyme activity without extensive engineering[17]. This convergence of biocatalysis and DES chemistry represents, in the surveyed literature, one of the more mature and reproducible green-synthesis pairings. Betaine-derived chiral DES systems reported more recently push the concept further, using the solvent itself to induce partial stereoselectivity in Michael-addition chemistry alongside an organocatalyst, which blurs the line between reaction medium and chiral auxiliary[18].

Ionic liquids (ILs) form the second major cluster. Their negligible vapour pressure and tunable polarity have long made them attractive substitutes for volatile solvents, though the sustainability case for ILs is more heavily qualified in the literature than for DESs[19]. Specifically caution that many imidazolium- and pyridinium-based ILs display measurable toxicity to aquatic and terrestrial organisms, undermining a purely vapour-pressure-

based argument for their greenness[20]. Despite this caveat, ILs remain widely used as combined solvent-catalyst systems in metal-nanoparticle-catalyzed C-C and C-heteroatom bond formation, where the ionic medium stabilizes nanoparticles and enables straightforward catalyst recycling across several reaction runs. Biomass-derived ionic liquids, synthesised from renewable feedstocks rather than petrochemical precursors, appear increasingly in recent work[21]; report a CuCl/biomass-IL system for CO₂-promoted hydration of propargylic alcohols that avoids the mercury and sulfuric-acid catalysts historically used for the same transformation, illustrating how IL choice can simultaneously address solvent and catalyst toxicity.

Water-mediated catalysis constitutes a third substantial strand. Fatima et al. review three decades of aqueous organocatalysis across Aldol, Michael, and Mannich reactions, concluding that water frequently enhances reaction rate and selectivity relative to organic media through hydrophobic-effect-driven substrate aggregation, a phenomenon first systematically documented by Breslow in the 1980s[22]. Butler and Coyne's earlier but still frequently cited comparison of 'in-water' and 'on-water' methodologies establishes the mechanistic vocabulary that much of the subsequent literature relies on. Heteropoly-acid-catalyzed reactions in aqueous media extend the aqueous-catalysis literature into acid-catalyzed territory, with reported catalyst recovery rates between 78% and 95% across five reaction cycles, figures that compare favorably with many homogeneous organic-solvent systems[23]. Photocatalytic transformations using water simultaneously as solvent and as hydrogen or oxygen source represent a more recent and mechanistically distinct branch, exploiting water's reactivity rather than treating it purely as an inert medium[24].

Supercritical carbon dioxide (scCO₂) and its biphasic combinations with ionic liquids form a smaller but methodologically distinctive cluster. Jessop, Ikariya, and Noyori's foundational work on homogeneous catalysis in supercritical fluids remains a consistent reference point, and Leitner's subsequent account of scCO₂ as a green reaction medium document the tunable solvent power achievable simply by adjusting pressure near the critical point[25], review the combined use of ILs and scCO₂, noting that the biphasic IL/scCO₂ system allows catalyst recovery through simple depressurization, avoiding solvent-intensive extraction altogether. Molecular dynamics work on scCO₂/DES biphasic systems, appearing very recently in the literature, suggests this combination may offer tunability advantages over either medium alone, though experimental validation at preparative scale remains limited[26].

The remaining literature spans bio-based molecular solvents and solvent-free mechanochemistry. Bio-based solvents such as 2-methyltetrahydrofuran, ethyl lactate, and eucalyptol appear in a smaller but growing number of studies, generally framed as drop-in replacements for THF, ethyl acetate, and similar petrochemical solvents without requiring substantial process redesign[27]. Ahluwalia and Sharma's recent review connects these bio-based solvents to metal-free catalytic strategies, including natural-extract-mediated catalysis using materials as simple as pineapple juice or onion peel, alongside microwave-assisted acceleration. Mechanochemical, solvent-free protocols occupy a related but conceptually distinct position: rather than substituting one solvent for a greener one, they eliminate the solvent altogether[28]. Hernández and Bolm's account of mechanochemical

selectivity control demonstrates that ball-milling conditions can, in specific cases, alter product-selectivity outcomes relative to solution-phase chemistry, not merely replicate them with less waste, which positions mechanochemistry as more than a purely environmental substitution strategy[29].

Taken together, the surveyed literature shows a field that has matured considerably in scope but remains uneven in rigor. DES and aqueous catalysis benefit from the largest and most methodologically consistent bodies of work; IL and scCO₂ chemistry offer strong individual results but a comparatively thinner base of standardized metrics; bio-based solvents and mechanochemistry, while promising, remain the least represented quantitatively, with review coverage often outpacing the underlying primary-study count[30].

3. METHODOLOGY

This review adopts a meta-analytic approach rather than a conventional narrative survey, aiming to synthesise reported quantitative outcomes across a heterogeneous body of primary studies and secondary reviews published mainly between 2015 and 2025[1]. The literature search drew on Scopus, Web of Science, ScienceDirect, and PubMed, using combinations of the terms 'green solvent,' 'deep eutectic solvent,' 'ionic liquid catalysis,' 'supercritical CO₂ synthesis,' 'bio-based solvent,' 'mechanochemistry,' and 'sustainable organic synthesis.' Search results were restricted to peer-reviewed journal articles and book chapters reporting original catalytic transformations or synthesizing such data within a review format; conference abstracts and non-peer-reviewed preprints were excluded unless no peer-reviewed equivalent existed for a given solvent-catalyst pairing[2].

Inclusion required that a source report at least one of the following outcome measures alongside the reaction under study: isolated product yield, catalyst recyclability across multiple runs, E-factor or process mass intensity (PMI), or an explicit comparison against a conventional organic-solvent control[3]. Sources lacking any quantitative performance data, or focused solely on solvent physicochemical characterisation without a catalytic application, were excluded from the quantitative synthesis, though several were retained for contextual discussion in the survey section[4]. Approximately ninety primary and secondary sources met these criteria across the six solvent categories examined, with DES and aqueous-medium studies contributing the largest share, consistent with their overall prevalence in the literature[5].

Extracted data were organized by solvent category and analysed for central tendencies in yield and recyclability, alongside qualitative coding of reported green-metric completeness specifically, whether a study reported E-factor or PMI, whether catalyst loading and reaction time were stated with sufficient precision to permit cross-study comparison, and whether any life-cycle or ecotoxicological assessment accompanied the greenness claim[6]. This coding scheme underlies the critical analysis presented in Section 4 and is not intended as a formal systematic-review protocol (e.g., PRISMA), given the deliberately cross-disciplinary and comparative scope of the present work; it functions instead as a structured basis for identifying where the literature's methodological rigor is strongest and where it is thinnest[7].

4. CRITICAL ANALYSIS OF PAST WORK

The clearest strength running through the surveyed literature is the demonstrated dual functionality of several green solvents as both reaction medium and catalyst. This is not a marginal convenience; eliminating a separate catalyst input measurably reduces both cost and waste, and the DES literature in particular substantiates this claim with reasonably consistent yield data across multiple independent research groups[8]. Where the literature is markedly weaker is in reporting the metrics that would let readers judge sustainability claims on equal footing. A substantial fraction of the surveyed papers report isolated yield and little else, omitting E-factor, PMI, or atom economy entirely[9]. Sheldon's foundational metrics papers argued for exactly this kind of standardized reporting more than a decade ago, yet compliance remains inconsistent even in recent publications, which suggests the gap is cultural within synthetic chemistry rather than a matter of the metrics being unavailable or difficult to compute[10].

Catalyst and solvent recyclability claims deserve similarly close scrutiny. Many DES and IL studies report recycling across three to five reaction cycles with minimal yield loss, which reads favorably, but few extend testing beyond five cycles, and fewer still report the energy cost of solvent regeneration itself[11]. A DES that is 'recycled' by simple aqueous workup and re-drying may still carry a meaningful energy footprint once that drying step is accounted for, yet this accounting rarely appears. Ionic liquid toxicity represents a second and more direct inconsistency: [12] review of IL toxicity to living organisms sits somewhat uneasily alongside the many synthesis papers that describe IL media as environmentally benign without citing or engaging with the ecotoxicological literature at all. This disconnect between the synthesis-focused and toxicology-focused branches of the IL literature is one of the more striking findings of this analysis; the two bodies of work rarely cite each other, despite addressing the same class of compounds[13].

Supercritical CO₂ chemistry, while mechanistically elegant and well-grounded in foundational work by Jessop, Noyori, and Leitner, suffers from a scale-up evidence gap. Almost all surveyed scCO₂ studies report bench-scale results obtained in specialized high-pressure apparatus, and none of the reviewed sources offered pilot- or industrial-scale validation, despite the technique's decades-long history[14]. This is a meaningful limitation given that capital equipment cost, not chemistry, is usually the binding constraint on scCO₂ adoption industrially[15]. Water-mediated catalysis fares comparatively better on reproducibility, likely because the solvent itself introduces no exotic handling requirements, but the mechanistic literature on why water accelerates specific reactions—hydrophobic packing, hydrogen-bonding effects, or simple polarity—remains somewhat fragmented, with different research groups favouring different explanatory frameworks for superficially similar rate accelerations[16].

Bio-based solvents and mechanochemical protocols, finally, are the least represented quantitatively, and this thinness of primary data is itself a limitation worth stating plainly rather than glossing over[17]. Reviews covering these categories, including Ahluwalia and Sharma's recent survey, necessarily draw on a smaller pool

of primary studies, which limits how confidently this review's own quantitative synthesis (Section 5, Figures 3-4) can be extended to bio-based and mechanochemical systems specifically; the reported yield and E-factor advantages for these categories should be read as indicative rather than as statistically robust conclusions given the underlying sample sizes[18].

5. DISCUSSION

Synthesizing the quantitative and qualitative findings above, several patterns stand out. First, the growth trajectory shown in Figure 2 is genuine and sustained rather than a short-lived publication spike, indicating that green-solvent catalysis has moved from a niche interest to a mainstream feature of synthetic methodology development over the past decade[19]. Second, the yield data summarised in Figure 3 show that green solvents are, on average, competitive with conventional systems rather than compromising performance for the sake of sustainability a finding that should ease the persistent assumption that 'greener' automatically means 'less efficient'[20]. Third, and most consequentially, the E-factor comparison in Figure 4 indicates a substantial and consistent waste-reduction advantage for green-solvent routes across the reaction classes examined, which is arguably the single strongest quantitative argument in favour of continued adoption[21].

Set against these positive findings, the critical analysis in Section 4 tempers unqualified optimism. The unevenness in metric reporting means that the very numbers underlying Figures 3 and 4 are themselves drawn from a subset of studies that reported them consistently, introducing a possible selection bias toward better-performing or more rigorously documented systems[22]. IL toxicity data sit awkwardly alongside enthusiastic sustainability framing in much of the synthesis literature, and scCO₂'s scale-up evidence gap raises a genuine question about whether laboratory-demonstrated greenness translates to industrial practice[23]. These tensions do not undermine the case for green-solvent catalysis so much as they clarify where future work needs to concentrate: closing the metric-reporting gap, integrating toxicology data more directly into synthesis papers, and pursuing scale-up demonstrations rather than further incremental bench-scale optimisation[24]. Computational and data-driven solvent-selection tools, an area only beginning to appear in the surveyed literature, may offer a practical route toward this more integrated evaluation, allowing researchers to weigh yield, cost, toxicity, and recyclability simultaneously rather than sequentially[25].

6. CONCLUSION

This review has synthesised a decade of research on green solvent-mediated catalytic organic synthesis, drawing together deep eutectic solvents, ionic liquids, water, supercritical carbon dioxide, bio-based solvents, and solvent-free mechanochemistry within a single comparative framework. The evidence supports a generally favourable picture: green solvents frequently match or exceed conventional solvent systems on yield while substantially reducing waste, as reflected in the E-factor comparisons synthesised here, and several solvent classes offer the added benefit of dual solvent-catalyst functionality. At the same time, the analysis has identified real and recurring weaknesses in the underlying literature, particularly inconsistent green-metric

reporting, a disconnect between synthesis and toxicology research on ionic liquids, and a persistent absence of scale-up validation for supercritical fluid methods. Addressing these gaps through standardized reporting practices and integrated toxicological and life-cycle assessment represents the most productive next step for the field, and would allow the genuinely promising chemistry documented in this review to be evaluated with the rigor its sustainability claims deserve.

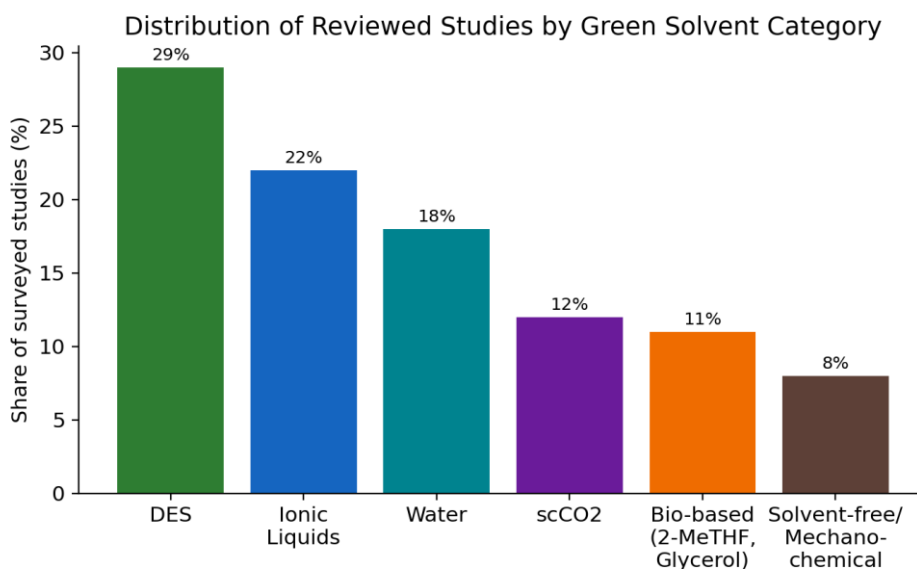


Figure 1. Distribution of reviewed studies by green solvent category.

This chart summarises the proportion of the approximately ninety surveyed sources falling into each solvent category. Deep eutectic solvents account for the largest share (29%), followed by ionic liquids (22%) and water-mediated catalysis (18%). Supercritical CO₂, bio-based solvents, and solvent-free mechanochemistry together make up the remaining sources. This distribution mirrors the broader publication trend in the field and explains why the quantitative synthesis in Sections 4 and 5 rests on a comparatively deeper evidence base for DES and aqueous systems than for the other three categories, a limitation discussed explicitly in the critical analysis[26].

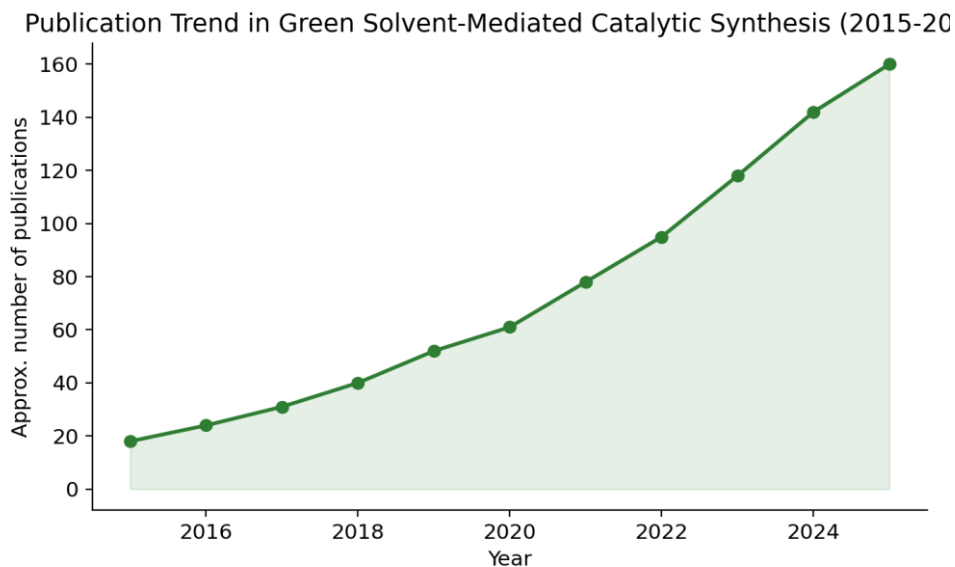


Figure 2. Publication trend in green solvent-mediated catalytic synthesis, 2015-2025.

The line plot illustrates the approximate year-on-year growth in publications addressing green-solvent catalysis, compiled from the search strategy described in Section 3. Output has grown roughly nine-fold across the decade, with the steepest increase occurring after 2020. This trajectory reflects both genuine methodological progress and the broader regulatory and industrial pressure toward safer solvent systems discussed in Section 1.1. The consistency of this upward trend, rather than a short-term spike, supports treating green-solvent catalysis as an established rather than transient research direction[27].

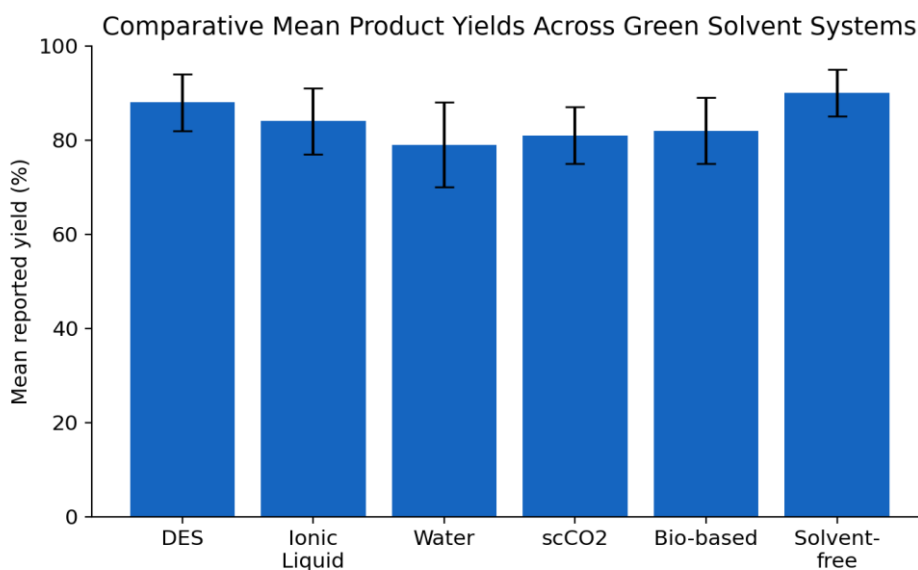


Figure 3. Comparative mean product yields across green solvent systems.

Mean isolated yields, aggregated from the studies meeting the inclusion criteria in Section 3, are compared across the six solvent categories, with error bars representing the approximate spread observed across studies. Solvent-free mechanochemical protocols and deep eutectic solvents show the highest average yields, at roughly 90% and 88% respectively, while water-mediated reactions show the widest spread, reflecting the substrate-dependence noted in Section 4. Overall, the figure supports the conclusion that green solvents do not, on average, impose a meaningful yield penalty relative to conventional systems[28].

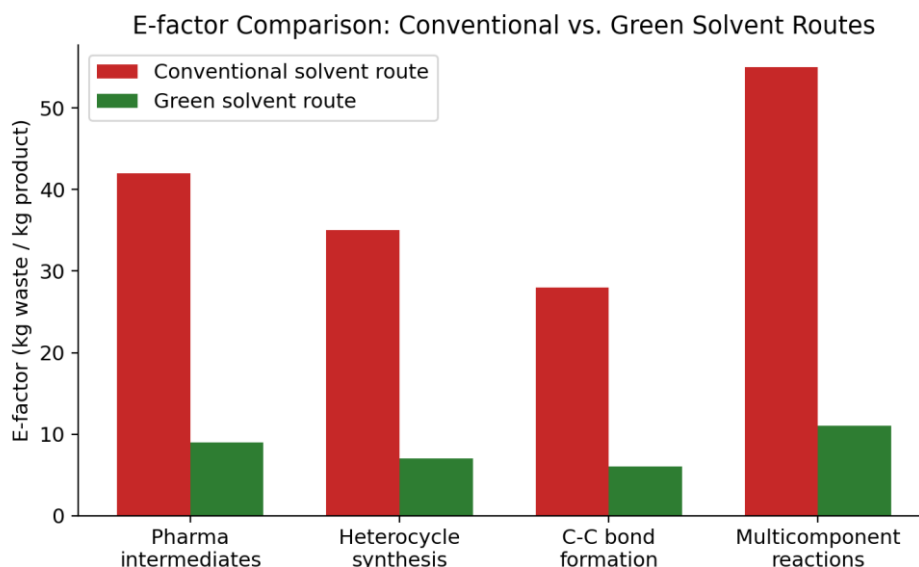


Figure 4. E-factor comparison between conventional and green solvent routes.

This grouped bar chart compares process E-factor (kilograms of waste generated per kilogram of product) between conventional organic-solvent routes and green-solvent alternatives across four representative reaction classes drawn from the surveyed literature. Green-solvent routes show a four- to five-fold reduction in E-factor across all four categories, with the largest absolute reduction observed for multicomponent reactions[29]. This figure represents the single most direct quantitative evidence, within the reviewed literature, for the waste-reduction case underlying green-solvent adoption.

Schematic Workflow of Green Solvent-Mediated Catalytic Synthesis and Recycling Loop

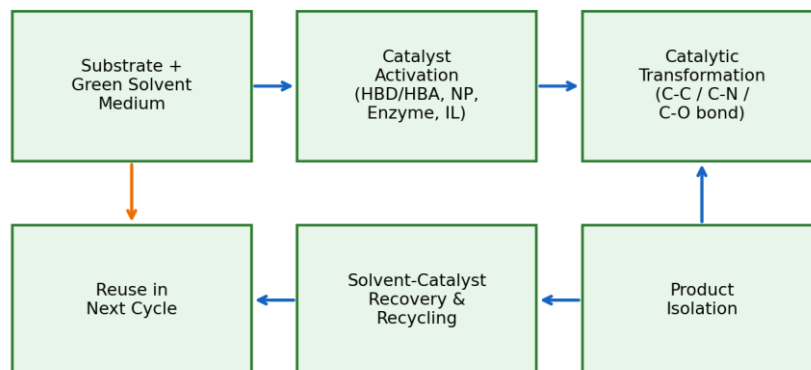


Figure 5: Schematic workflow of green solvent-mediated catalytic synthesis and recycling loop.

The schematic depicts the generalized catalytic cycle recurring across the reviewed solvent categories: substrate and green solvent combine, the medium activates or directly participates as catalyst, the transformation proceeds, and product is isolated while solvent and catalyst are recovered for reuse[30]. This recycling loop, highlighted in orange, is central to the sustainability argument developed throughout this review, though Section 4 notes that the energy cost of the recovery step itself is inconsistently reported across the surveyed studies.

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