



Synthesis and processing of π -conjugated derivatives in aqueous media: Introducing sustainability in printed electronics

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Sustainability in the synthesis and processing of organic semiconductors is becoming increasingly important as the field progresses from fundamental research to technological deployment. Volatile organic solvents dominate the environmental footprint of both laboratory-scale and industrial production, significantly increasing hazardous waste generation. Substituting these solvents with water dramatically reduces environmental impact while enabling new reactivity modes and formulation strategies. Recent developments demonstrate that water is not only a benign solvent for synthetic chemistry but also a robust medium for preparing stable, printable, and device-compatible inks of conjugated materials. This review highlights advances in aqueous-phase synthesis as well as modern aqueous processing routes such as surfactant-stabilized dispersions, hydrotropic systems, and self-emulsifying conjugated polymers. These approaches illustrate how water can serve as a unifying platform for high-performance and environmentally responsible printed electronics.

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Intro

The development of green-chemistry-compliant processes has become a cornerstone of modern synthetic and process chemistry. Although its principles are well

established in pharmaceutical and natural product manufacturing, their adoption in emerging fine-chemical sectors—particularly in the synthesis and processing of organic semiconductors—remains limited. Indeed, sustainability metrics such as the E-factor (weight ratio between waste and product), process mass intensity (PMI), and solvent environmental impact are still only sporadically discussed in the organic electronics literature. This gap is striking given the growing industrial interest in flexible and printed optoelectronic devices.

Recent work by Sheldon, underscores the overwhelming contribution of volatile organic solvents (VOCs) to the carbon footprint of multistep syntheses and materials manufacturing [1]. Although initially framed in the context of bulk synthesis, these considerations are equally relevant to printed optoelectronics, where most fabrication routes rely on solution processing. Established device protocols still depend heavily on halogenated or aromatic solvents which pose toxicity, environmental, and regulatory challenges [2].

Efforts to mitigate solvent impact have led to the exploration of alternative media with reduced toxicity or derived from renewable feedstocks [3,4]. While these approaches provide benefits, they do not fundamentally address the high solvent volumes and associated waste intrinsic to organic processing. Water, by contrast, offers a compelling route to sustainability: it is inexpensive, non-toxic, non-flammable, and widely available. With a high heat capacity and unique polarity, water can also promote reaction pathways inaccessible in organic solvents.

Most high-performing conjugated materials are intrinsically hydrophobic, presenting challenges for aqueous-based workflows. Nevertheless, two complementary strategies have significantly expanded the role of water in organic semiconductor chemistry (Figure 1). The first involves molecular design, wherein ionic or hydrophilic side chains (e.g., ammonium groups, sulfonates, carboxylates, polyethylene glycol moieties) impart intrinsic water

solubility [5]. While effective, this approach demands substantial synthetic effort, particularly during purification, and may compromise electronic properties.

The second strategy relies on formulation chemistry, enabling aqueous dispersibility without altering the underlying conjugated backbone. Surfactant-assisted dispersion, hydrotropic solvents, and self-emulsifying materials allow fully hydrophobic semiconducting polymers to be synthesized and processed in water [6]. Although surfactants may affect device performances if not properly removed, this approach offers the advantage of leveraging widely used, high-performance materials with validated device metrics and commercial availability.

Synthesis – intrinsically water-soluble polymers

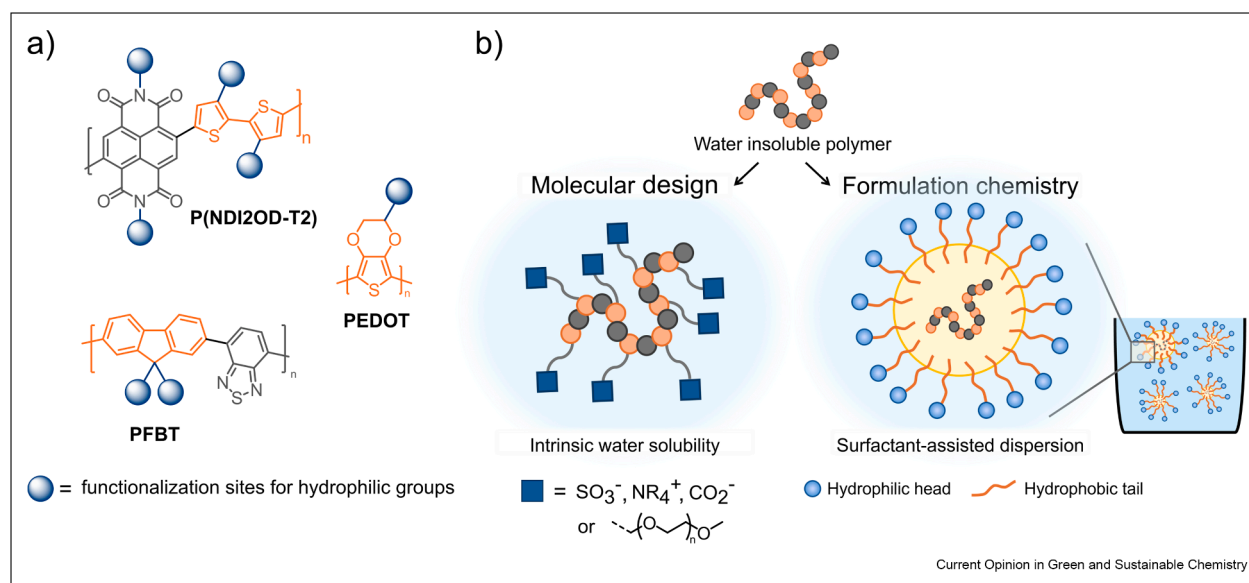
Processing of conjugated polymers from water was the main original driver for the synthesis of polymers featuring intrinsic water solubility. Lately, the development of electrochemical and electrochemically gated transistors – a major research area for applications in biosensing and bioelectronics – fostered a renewed interest for the development of such materials. The most common approach in the field is the functionalization of the conjugated backbone with side chains bearing ammonium salts or sulfonate group, as well as the partial or complete substitution of the alkyl solubilizing chains with polyethoxylated ones (Figure 1a). Zhang and co-workers reviewed advances in CPE design, demonstrating how backbone planarity, side-chain density and

counterion selection tune film morphology and device performance in organic electrochemical transistors and solar cells [5]. Li et al. recently reported a highly conductive n-type conjugated polymer complementing the performances of PEDOT:PSS and synthesized directly in water thanks to the introduction of suitable water solubilizing units within its structure [6]. Meanwhile, Liu et al. reported that ground-state electron transfer between donor-acceptor polymer blends enables aqueous processing of water-insoluble conjugated polymers—demonstrating that intrinsic water solubility is no longer a physical limit [7]. Their work demonstrates that waterborne conductive films derived from such blends had conductivities $\sim 10,000\times$ higher than pristine polymers, underscoring the potential of aqueous routes for high-performance device applications. The direct solubilization in water has advantages, like direct compatibility with aqueous environment (e.g., bioelectronics), reduced toxicity and easier handling, potential for high ionic conductivity. At the same time synthetic complexity, cumbersome purification, and long-term hygroscopicity and stability concerns motivate interest in a complementary formulative approach enabling dispersion in water of established hydrophobic polymers.

PEDOT:PSS as the foundational aqueous system

PEDOT:PSS (poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)) is the most successful material developed for printed electronic applications and remains the only conjugated polymer scaled to industrial

Figure 1



Examples of common semiconducting polymers. Functionalization sites suitable for the introduction of hydrophilic groups are highlighted with blue spheres (a); two approaches for enabling water processability of π -conjugated materials: (left) side chain engineering to impart intrinsic water solubility, and (right) surfactant-based chemistry to disperse the hydrophobic backbone in aqueous media (b).

quantities. This interpenetrated network of PEDOT (per se an insoluble material) and PSS (a polyelectrolyte) is commercially available as a wide range of aqueous formulations whose properties are optimized for applications ranging from capacitors to bioelectronics [8]. PEDOT:PSS dispersions are prepared directly in water by emulsion polymerization of the EDOT monomer in the presence of PSS and a suitable oxidizer [9]. The success of PEDOT:PSS has inspired the broader field to explore water-mediated routes for other conjugated materials.

Overcoming hydrophobicity: role of surfactants

The intrinsic water solubility of precursors and/or products is not a rigid limitation: hydrophobic conjugated backbones can be synthesized and processed in water environments, when supported by appropriate self-assembling amphiphilic derivatives like surfactants [10]. In this context, one can distinguish between reactions that occur “in water”, where substrates are homogeneously dispersed as various form of association colloids (micelles and microemulsions) or dispersed systems (emulsions and dispersions) within the aqueous bulk (Figure 2a), and those that occur “on water”, where reactivity is modulated by specific interactions at the water surface.

On water surface approaches

Prasoon and co-workers provided a compelling demonstration of interfacial control by showing site-selective imide formation between porphyrin and perylene units at a surfactant-coated air–water interface [11]. A sodium oleyl sulphate monolayer preorganized the protonated amine near the interface, enabling a reaction pathway entirely inaccessible in homogeneous solution. Their subsequent work in micellar interfaces further illustrated how hydration shells and interfacial ordering can mimic “on-water surface” environments even under fully dispersed conditions [12]. These studies collectively reveal that reactant preorganization, the features of interfacial electric fields, the hydration-layer structure and microenvironment polarity gradients are crucial variables that determine reactivity and selectivity in aqueous-phase organic chemistry.

In water approaches

The in-water approach is by far the most common and relies heavily on surfactants features. Industrial surfactants are chemical commodities offering the advantage of ready availability, low cost and robustness. They can be used to prepare conjugated materials, both molecular [13] and polymeric [14], sometimes also offering selectivity alongside with efficiency mostly due to turning of the reagents vs product relative affinity at the reaction

site [15,16]. On following up the pioneering contribution of Lipshutz, the field shifted from generic amphiphiles to designer surfactants, specifically engineered to promote organic reactions rather than merely provide homogeneous formulations (Figure 2b). TPGS-750-M, a provitamin-derived surfactant, remains the most iconic example, exhibiting remarkable performance across a wide range of transformations [17,18], including those most relevant for conjugated materials [18].

More recently, Hauk and co-workers introduced a next-generation functional surfactant optimized for mild C–H arylation [19], a reaction of increasing relevance in the field of organic semiconductors [20].

The same system mediated Suzuki–Miyaura couplings of heteroaromatics in water, demonstrating that surfactant structure can fine-tune the metal coordination environment, accelerating catalysis while maintaining colloidal stability.

Sengoden *et al.* reported a metallo-surfactant in which a Pd complex is covalently bound to a CO₂-based triblock polycarbonate [21]. In water, this material self-assembles into micelles that localize both the metal centre and the organic substrates, leading to efficient cross-couplings with facile catalyst recovery. This “catalyst-in-surfactant” design directly addresses concerns about metal leaching in electronic materials (residual metals can impair device performance) and offers an attractive route toward recyclable reaction media for conjugated-polymer synthesis.

Sanzone *et al.* described the first example of surfactant specifically designed for the synthesis of conjugated materials, demonstrating particularly fast reactions under remarkably mild conditions and selectivity directly associated to the reactant/products vs surfactant π – π interactions [22]. Finally, the polysarcosine endowed Savie deserves a special mention, being the only non-PEG derived designer surfactant to date, connecting excellent performances and biodegradability [23].

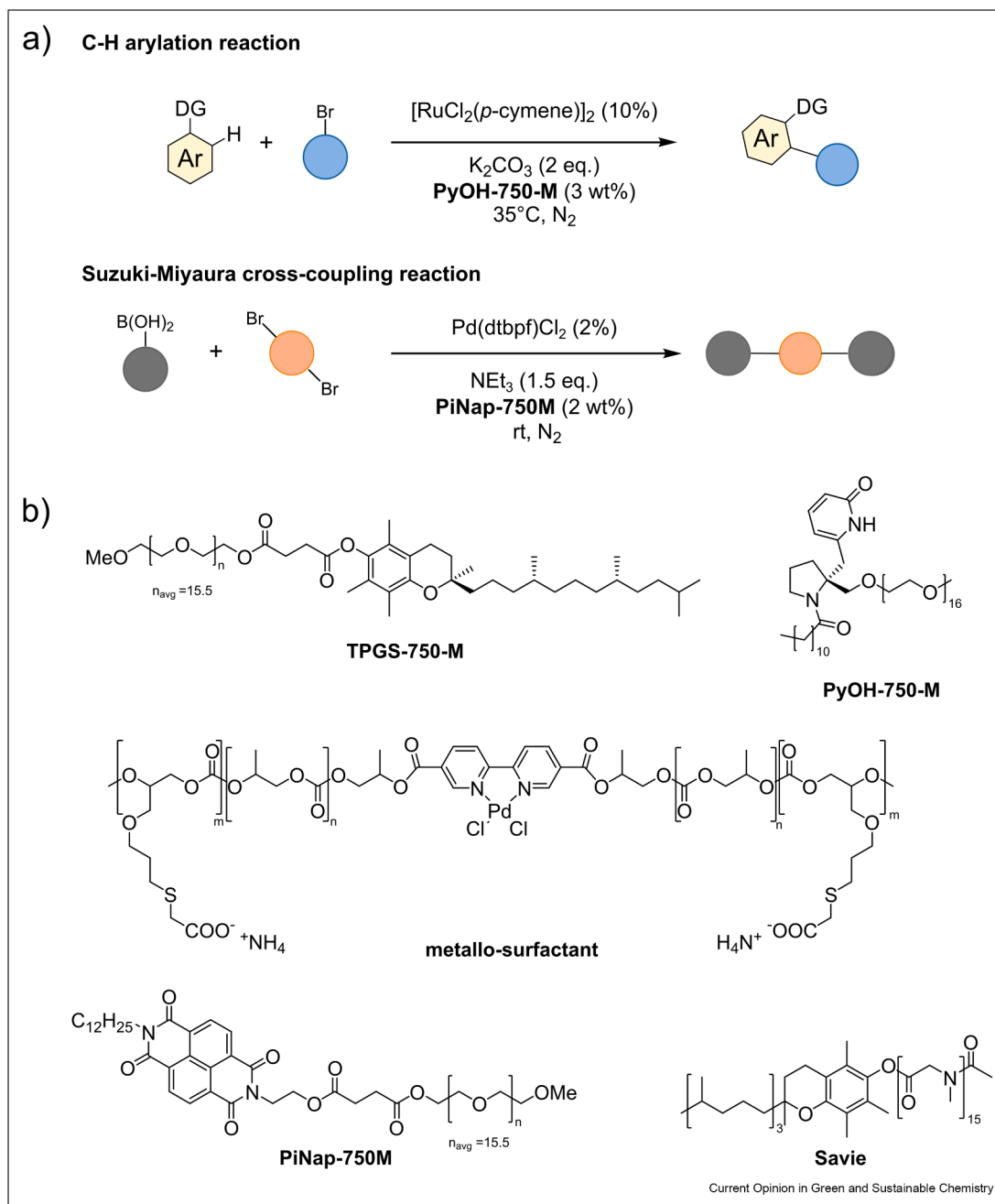
Polymerization carried out in the presence of surfactants and for the specific purpose of growing conjugated polymers has only relatively recently gained traction. The early work of Turner on microemulsion polymerization [24,25] is seminal and recently evolved towards a mostly solvent free synthesis and processing integrated protocol which is still State of the art in the field [26]. Sanzone *et al.* demonstrated the first example of in water polymerization featuring the industrial surfactant Kolliphor EL [14]. More recently, Ceriani *et al.* extended the approach to improve on the previous report by Turner

[26], through the development of an integrally organic solvent-free protocol going from synthesis to processing of field effect transistor devices (Figure 3a) [27]. Together, these developments confirm that aqueous environments can support the entire synthetic pathway of conjugated polymers—including monomer activation, bond formation, chain growth, and dispersion stabilization.

Preparation and use of stable aqueous formulations of conjugated polymers

As the field of organic electronics increasingly embraces sustainability principles, the development of stable aqueous formulations of conjugated polymers has become a central objective. Green chemistry metrics—including E-factor, PMI, and solvent hazard assessments—provide quantitative justification for replacing organic solvents

Figure 2



Selected examples of in water reactions relevant for the preparation of conjugated materials enabled by functional surfactants (a) [19,22] Examples of designer surfactants engineered to promote specific reactivity (b).

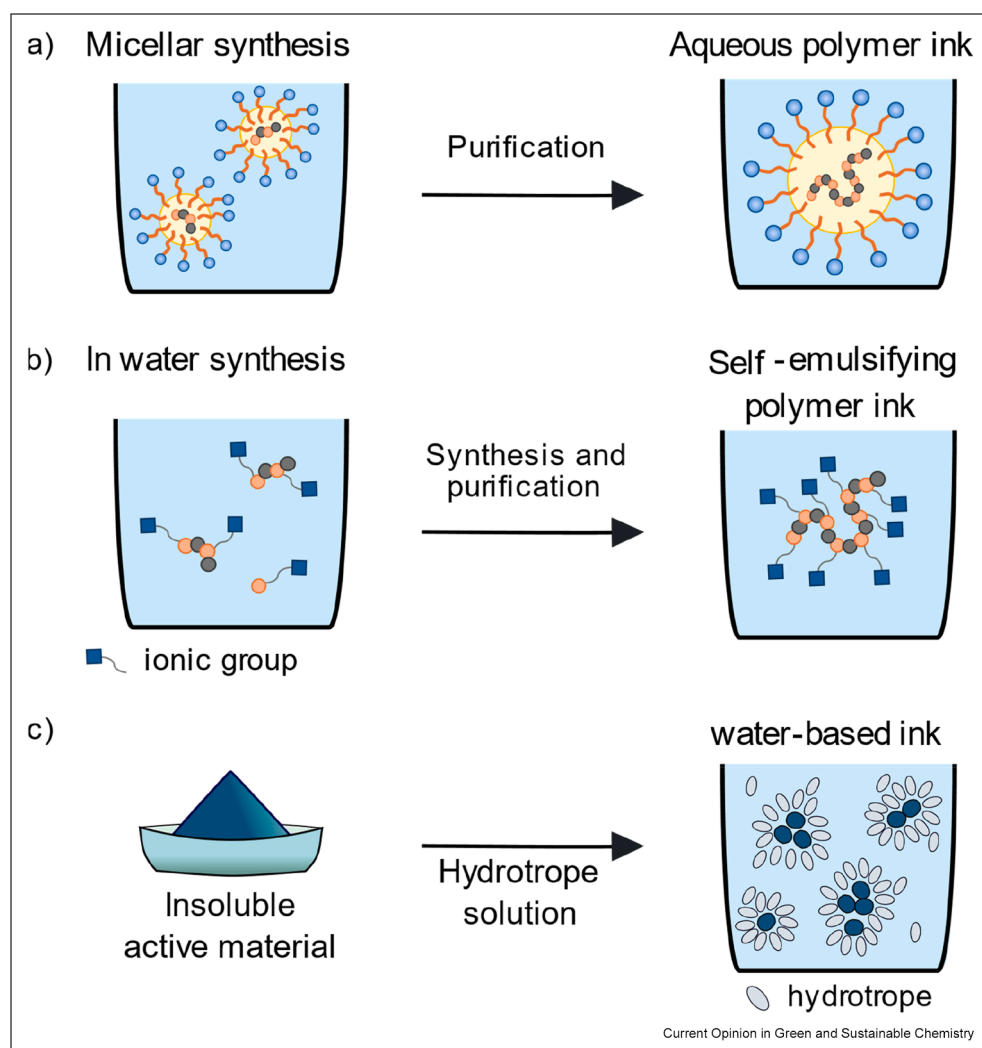
with water [28–30]. In printed electronics, however, these metrics must be extended beyond synthesis: the majority of solvent usage occurs during ink formulation, deposition, and post-processing [31].

For printed electronics, the advantages of using water as the processing medium extend beyond reaction metrics. Aqueous inks mitigate flammability, reduce worker exposure to hazardous vapours (particularly in roll-to-roll processes) [32] and simplify compliance with solvent emission regulations.

Surfactants and stabilizers enable the formulation of stable aqueous dispersions of otherwise hydrophobic

materials — enabling long-term colloidal stability, printability and compatibility with aqueous manufacturing lines. Unfortunately, the dispersing additive is insulating and non-volatile, thus requiring removal after deposition by washing with organic solvents [26,27]. As recently demonstrated by Lee *et al.*, the use of an hydrotrope (benzyl alcohol) offers an alternative solution. They demonstrated the formulation as water stable dispersion of the lipophilic n-type high performance polymer DPP-TT. The dispersing additive is volatile and can be removed directly by suitable thermal treatment (Figure 3c) [31]. Hydrotropes thus represent an interesting bridge between formulation chemistry and device-grade film purity.

Figure 3



Different approaches for the preparation of aqueous conjugated materials inks. Micellar synthesis of the active material, followed by dialysis (a); synthesis of a self-emulsifying material bearing ionic groups to grant dispersibility in water (b); aqueous ink formulation of a water-insoluble active material by mean of an hydrotrope (c).

Finally, the conjugated polymer itself can act as its own emulsifier. Gwiazda et al. showed that fluorene-based monomers bearing sulphate groups can polymerize into self-emulsifying conjugated polymers, producing aqueous dispersions without surfactants (Figure 3b) [33]. Benefits include removal of insulating residues, simplified formulation and improved batch-to-batch reproducibility. Although this strategy requires synthetic modification of monomers, it avoids the performance penalties associated with classical surfactants.

Outlook for aqueous synthesis and processing of conjugated derivatives

Water is consolidating its position as the solvent of choice for both the sustainable synthesis and processing of organic semiconductors. The work discussed throughout this review demonstrates that most of the limitations historically associated with aqueous media—poor solubility, incompatibility with organometallic catalysts, and uncontrolled aggregation—can be overcome through innovations in surfactant design, formulation chemistry, and molecular engineering of conjugated systems. From a processing perspective, water-based dispersions, hydrotropic formulations, and surfactant-free strategies provide a promising solution for sustainable ink formulation and film deposition. These developments collectively reduce VOC emissions, PMI, flammability risk and the generation of hazardous waste. Nevertheless, challenges remain. On the synthetic side, extensive collaboration between organic chemists and materials engineers is required to establish a trade-off between sustainability of synthetic procedures and performance of the obtained materials. On the processing side, the removal of insulating surfactants, control of water-induced microstructure, long-term stability under humidity, and compatibility with flexible substrates are still active areas of investigation. A key frontier is establishing fully aqueous workflows, where materials are synthesized, formulated in water and deposited exclusively from water.

Such a unified approach promises unprecedented sustainability and scalability for printed electronics. Continued innovation in biodegradable surfactants, recyclable aqueous media, water-compatible catalysts, and water-stable device architectures will be essential to achieving this vision.

Ultimately, water is no longer a compromise medium—it is a functional, tunable, and sustainable platform capable of supporting the next generation of organic semiconductors and printed electronic technologies.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as

potential competing interests: Luca Beverina reports financial support was provided by European Commission. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

No data was used for the research described in the article.

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