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MINI REVIEW

Polymers and Functionalized Polymer Catalysts in Organic Synthesis: A Mini Review

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Polymeric materials, especially poly(ethylene glycol) (PEG)-based systems, play an essential role in biomedical applications due to their biocompatibility and tunable surface properties, which improve blood compatibility and reduce biofouling. Beyond biomedical uses, PEG and PEG-supported catalysts have become valuable in organic synthesis because of their hydrophilicity, broad solubility, low toxicity and ease of functionalization, aligning well with green chemistry principles. This review summarizes key PEG-assisted synthetic methodologies for organophosphorus compounds including dialkyl/aryl phosphonates, α-amino phosphonates and α-hydroxy phosphonates as well as PEG-catalyzed routes for diverse heterocyclic frameworks. These include coumarins, acylals, indazole-triones, dihydropyrimidinones, oxadiazoles, thiadiazoles, pyrroles, *bis*(indolyl)alkanes, *bis*(pyrazolyl)methanes and xanthenes. PEG-based protocols typically offer mild conditions, shorter reaction times, high yields and simplified purification. Overall, incorporating Brønsted or Lewis acidic moieties into PEG matrices provides efficient, recyclable and environmentally benign catalytic systems that support sustainable organic synthesis and combinatorial chemistry.

Keywords: Liquid polymers, Polymer supported catalyst, Phosphonates, Heterocyclic compounds.

INTRODUCTION

The development and advancement of biomedical systems and devices in medicine has been greatly aided by polymeric materials. In the field of biomaterials, surface properties of polymers have gained increasing significance due to their ability to come into contact with physiological components like blood and living tissue [1]. It has been demonstrated that surface modification with a variety of macromolecules, including heparin, albumin and poly(ethylene glycol) (PEG), enhances blood compatibility [2-4]. PEG is a well-known and frequently utilized part of practical biomaterials. Hydrophilic PEG exhibits kinetic chain mobility and a large thermodynamic steric volume in aqueous physiological conditions, which effectively rebuffs nearly all forms of foreign adherence and adsorption. Conversely, it was found that a negatively charged surface was more blood compatible than a positively charged one [5].

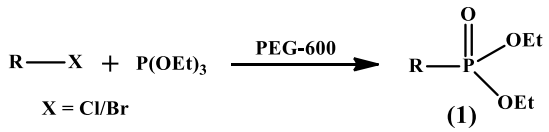
Functionalized polymers have been used as catalysts and stoichiometric reagents in organic synthesis for a long time [6,7]. Nonetheless, the unique demands of combinatorial and green chemistry are presumably driving a massive renaissance in their development and applications in organic synthesis at the moment. More and more researches are being done on poly(ethylene)s [8], both for combinatorial synthesis and as supports for organic reaction catalysis in solvent-free and solution-based media. Among these are PEGs, which are widely available commercially in a range of molecular weights, easily functionalized reagents and solvents with non-toxic properties (phase transfer catalysts, PTCs) [9], environmentally benign [10] and have a broad solubility profile [11]. PEGs are a significant class of polymers and catalysts in many areas of chemistry and industry, including organic synthesis, because of these outstanding qualities [12]. PEG is thought to be a fascinating solvent system when viewed through the perspective of green

chemistry. It is widely soluble, has a broad solubility profile, is inexpensive, thermally stable, non-volatile, non-toxic and readily degradable. It is also commercially available in various molecular weights. These outstanding qualities make PEGs a significant class of polymers and catalysts in many areas of chemistry, most notably organic synthesis. PEGs are good candidates for these uses based on recent efforts to use environmentally benign and eco-friendly reagents in chemistry [13].

Synthesis of phosphorus compounds

Synthesis of dialkyl, aryl/heteroaryl phosphonates: The Michaelis-Arbuzov rearrangement represents one of the most versatile methods for forming carbon-phosphorus bonds, owing to its adaptability and the wide range of biological activities exhibited by its products [14,15]. Relatively safe catalysts have recently been employed in this reaction, including $InCl_3$, Amberlyst-15, I_2, alkali-metal-iodide, trimethyl silyl halide (TMSX) and $BF_3{\cdot}OEt_2$. However, some of these methods' limited application is due to their laborious workup procedure, high temperature, longer reaction times and moderate product yields.

Alkyl/aryl halide and trialkyl phosphate were catalyzed by PEG-600 in a two-components reaction quickly produced corresponding alkyl/aryl phosphonates 1 (**Scheme-I**). This protocol is straightforward process, quick reaction time and use of an inexpensive, safe solvent for the phosphonate synthesis were its appealing aspects [16].

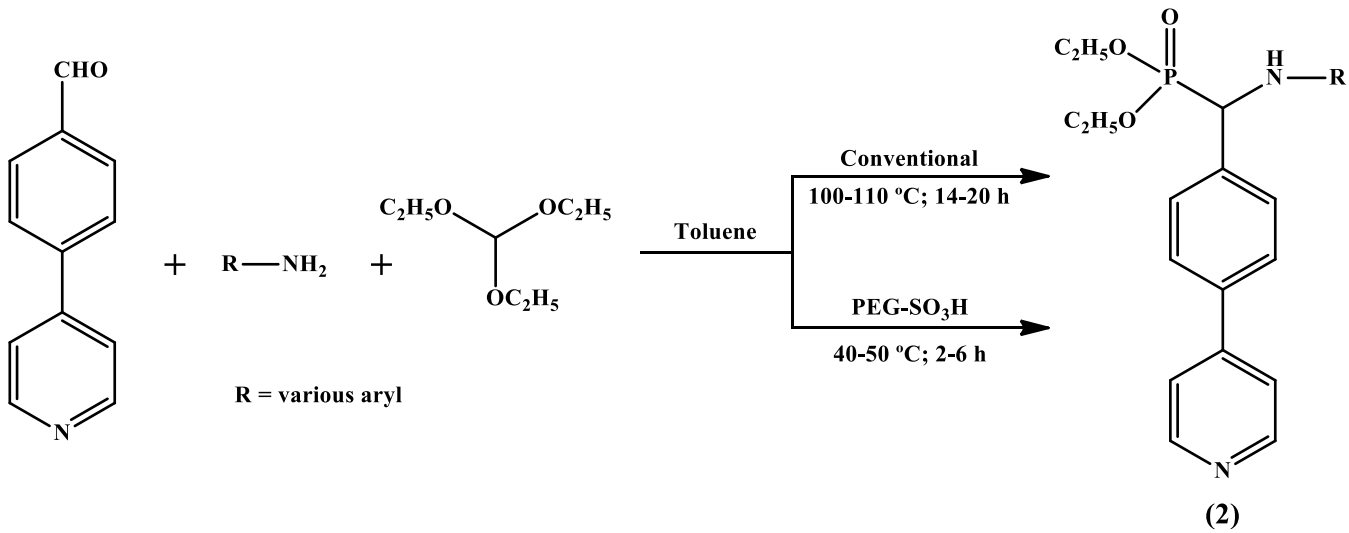


Scheme-I: Synthetic route of aryl/alkyl phosphonates

Synthesis of α-amino phosphonates: α-Aminophosphonates are an important class of bioactive substances. Their usage as peptide mimics [17], enzyme inhibitors [18], antibiotics [19], anti-HIV [20], anticancer [21] and thrombotic [22] medicines has proved their extensive potential as direct therapeutics as well as drug precursors for a range of diseases. These molecules often undergo advanced synthetic techniques like chemical synthesis, recombinant DNA technology or enzymatic synthesis, all of which help to understand their function or develop therapeutic applications. α-Aminophosphonates have been synthesized using a range of solvent-free synthesis methods, such as $CoCl_2{\cdot}6H_2O$ [23], $[Yb(PFO)_3]$ [24], nano Fe_3O_4 [25], $Mg(ClO_4)_2$ [26], metal triflate [27]. α-Aminophosphonates have also been synthesized in organic solvents using $In(OTf)_3$/$MgSO_4$ [28], GaI_3 [29], $BiCl_3$ [30], $Cu(OTf)_2$ [31] and $SbCl_3$/Al_2O_3 [32]. They have been explored using a variety of advanced synthetic strategies, including the application of Lewis acid–surfactant catalysts [33], as well as conditions in which the reactions proceed without any catalyst or solvent [34].

However, the catalysts discussed above have several limitations, including long reaction times, sensitivity to moisture, the need for stoichiometric amounts of hazardous materials, low product yields and excessive waste generation. In response, α-amino phosphonates have been synthesized using polyethylene glycol sulfonic acid (PEG-SO_3H) as a moderately effective and recyclable catalyst. The ongoing efforts to develop target-specific, safer and more efficient bioactive organophosphorus compounds have led to the establishment of this improved methodology. In order to synthesize **2** (**Scheme-II**), an aldehyde, an amine and diethylphosphite were reacted with PEG-SO_3H acting as an effective catalyst. This method proved both practical and attractive for the synthesis of these compounds, as it offered several advantages, including mild reaction conditions, high product yields, and a simple experimental procedure with straightforward work-up [35].

Synthesis of α-hydroxy phosphonates: α-Hydroxyphosphonates are the structural counterparts of α-hydroxy phosphonic acids [36,37] and serve as a wide spectrum of enzyme inhibitors for farnesyl protein transferase (FPT) [38], human renin [39,40], human protein tyrosine phosphatase (PTP) [41-43], purine nucleoside phosphorylase (PNP) [44] and 5-enolpyruvylshikimate-3-phosphate synthase (EPSP) [45]. They also showed anticancer action against a range of human cancer



Scheme-II: Synthetic route of novel α-aminophosphonates

cell lines [46,47] and suppression of the human immunodeficiency virus (HIV) [48]. Numerous methods have been employed to synthesize α-hydroxyphosphonates, but they are all dependent on the reaction between dialkyl phosphites and aldehydes or ketones in the presence of various bases [49,50] and acids [51]. There are few studies that discuss the reaction of trialkyl phosphites with aldehydes or ketones in the presence of acid catalysts such as TMSCl [52], $HCl·Et_2O$ [53], $LiClO_4·Et_2O$, ChOH [54] and guanidine hydrochloride [55]. The lack of success can be attributed to inherent mechanistic constraints associated with the acid catalysts examined. These systems fail to achieve the simultaneous nucleophilic activation of the trialkyl phosphite and electrophilic activation of the carbonyl substrate necessary for productive bond formation. Consequently, the reaction environment favours alternative, thermodynamically preferred pathways over the generation of the requisite reactive intermediate, leading to minimal or no formation of the targeted products.

Scheme-III illustrates the reaction of various substituted isatins with trialkyl phosphites in PEG-400 as reaction medium, affording the corresponding α^1-oxindole-α-hydroxyphosphonate (**3**) derivatives. Under these conditions, the products were obtained in high yields and the transformations proceeded with exceptional cleanliness throughout [56].

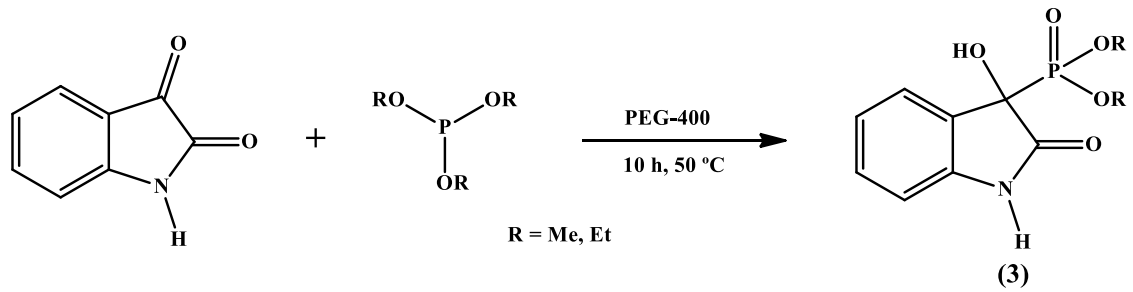
Synthesis of α-amino and α-hydroxy phosphonates: Reddy *et al.* [57] have developed a PEG/H_2O mixture as an environmentally benign solvent system for the synthesis of α-amino (**4**) and α-hydroxyphosphonate (**5**) derivatives (**Scheme-IV**). This solvent combination not only aligns with green chemistry principles but also provides an efficient medium that supports smooth transformations and good product yields.

Synthesis of heterocyclic compounds

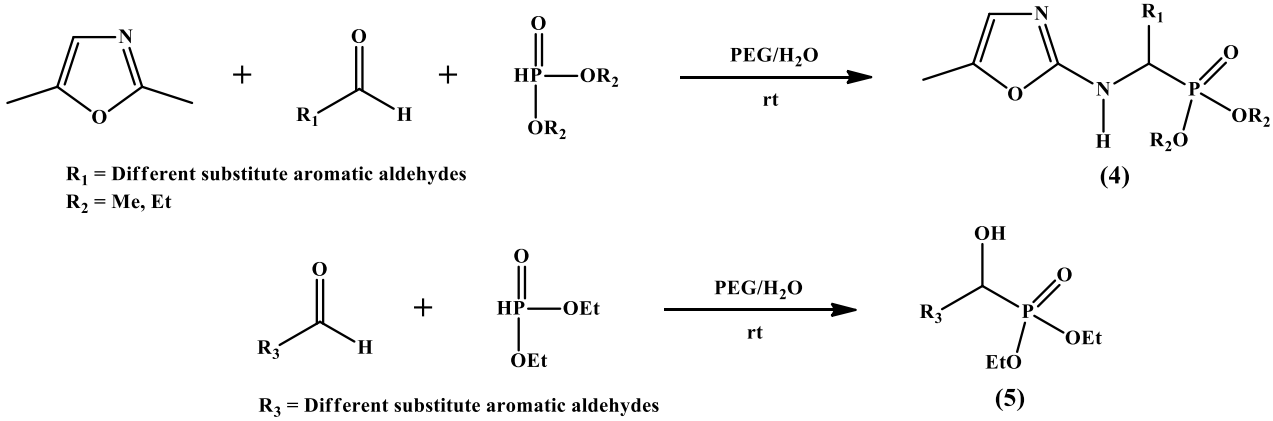
Synthesis of nitrogen heterocyclic compounds: Nitrogen containing heterocyclic compounds constitute a major class of chemical entities. They are widely represented in fine chemicals, natural products and numerous pharmacologically active agents that play essential roles in improving human health. Among these, urazole derivatives are particularly noteworthy, as they exhibit diverse biological and pharmacological activities, including reported anticancer and hypolipidemic properties [57]. In addition to their medicinal relevance, urazole based compounds are employed in the manufacture of polymeric materials with specialized performance characteristics such as heat-resistant coatings, high-traction tire rubbers, melamine resins and various agrochemical agents, including pesticides, herbicides and insecticides [58].

In combinatorial chemistry, the multi-component reactions (MCRs) are an essential tool that have various advantages over conventional synthetic techniques. These benefits consist of higher output, simple processes and simplicity of use [59]. Despite substantial research in combinatorial chemistry and its rapid expansion as a platform technology within the pharmaceutical industry, relatively few novel multicomponent reactions (MCRs) have been developed over the past two decades. As common instances, the Biginelli [60], Passerini [61], Ugi [62] and Mannich [63] reactions have been successfully used in a number of organic synthesis methods. These factors have led to considerable efforts being made and still being made, to find and develop new MCR catalysts.

The synthesis of the targeted compounds, including new [1,2-*a*]-indazole-trione and spiro-triazolo derivatives, has attarcted considerable attention due to their structural diversity and



Scheme-III: Synthetic route of novel α1-oxindole-α-hydroxyphosphonate



Scheme-IV: Synthetic route of novel α-amino (**4**) and α-hydroxy phosphonates (**5**)

potential functional relevance. In this context, PEG-SO_3H has been reported as a highly stable, reusable and environmentally benign polymer-supported catalyst for the preparation of indazole-tetraone derivatives (**6**) (**Scheme-V**) [64]. It robustness, ease of handling and biodegradability make it an appealing choice for promoting such transformations within a green chemistry framework.

Synthesis of Biginelli-type 3,4-dihydropyrimidine-2(1*H*)-ones: The Biginelli reaction involves the condensation of urea, aldehyde and β-ketoester in the presence of an acid catalyst [65]. However, its utility is often limited by the need for harsh reaction conditions, extended reaction times, and frequently low yields. The resulting 3,4-dihydropyrimidin-2(1*H*)-ones (DHPMs) are structurally intriguing compounds with significant applications in biological, synthetic and pharmaceutical chemistry [66]. Remarkably, by employing PEG-bound sulfonic acid both as a catalyst and reaction medium, libraries of Biginelli-type DHPMs (**7**) (**Scheme-VI**) can be efficiently synthesized under milder, more environmentally benign conditions [67].

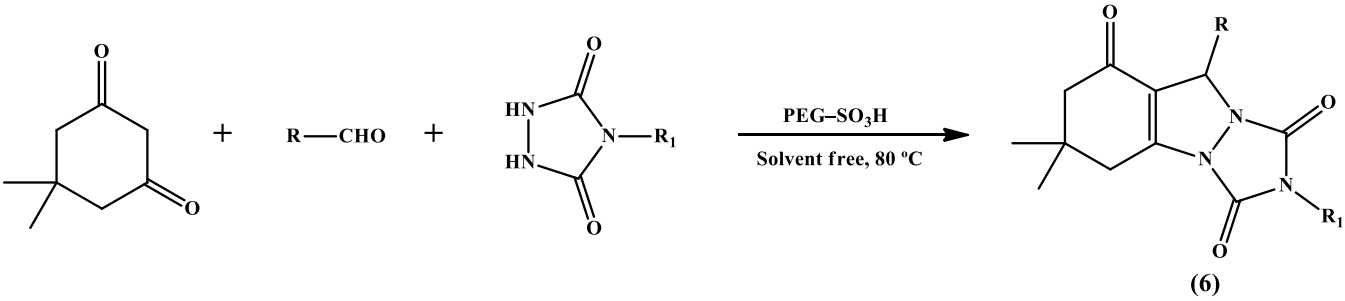
Synthesis of 1,3,4-oxadiazoles: Derivatives of 1,3,4-oxadiazole have long piqued the interest of medicinal chemists because of their antibacterial, antimitotic, anti-inflammatory, psychotropic, anti-aflatoxigenic and anticonvulsant qualities. Using conventional methodology, cyclization of diacyl hydrazides, cyclization of acyl thiosemicarbazides and oxidation of acyl hydrozones are three of the most popular processes for generating 1,3,4-oxadiazoles [68]. On the other hand, strong reagents as $SOCl_2$ [69], polyphosphoric acid [70] or silica-sulfuric acid [71], are commonly utilized, as are long reaction periods and high reaction temperatures. Nevertheless, these methods are insufficient for synthesizing 1,3,4-oxadiazole.

An efficient synthesis of 1,3,4-oxadiazole derivatives (**8**) has been reported *via* a two-component condensation of coumarin-3-carboxylic acid with various substituted benzoic acid hydrazides, employing a polymer-supported dehydrating reagent under microwave-assisted solvent-free conditions (**Scheme-VII**) [72].

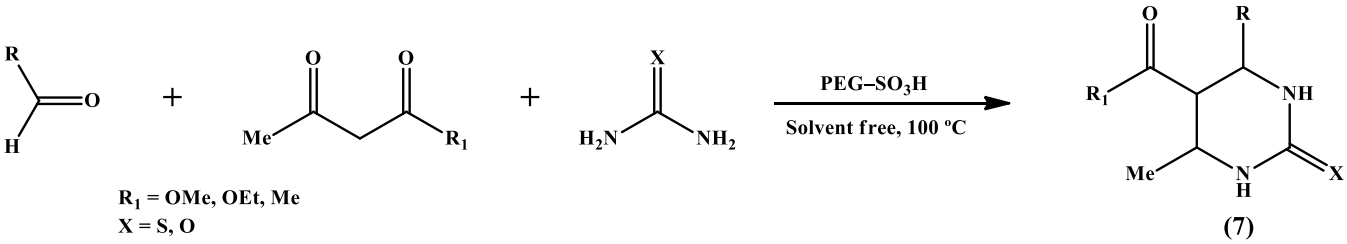
Synthesis of thiadiazoles: The broad biological activities of thiadiazole derivatives also attracted a lot of interest [73]. Li *et al.* [74] developed a successful solvent-free, microwave-assisted method for 2-amino-5 substituted 1,3,4-thiadiazoles (**9**) with poly (ethylene glycol)-supported dichlorophosphate ($PEGOP(O)Cl_2$) (**Scheme-VIII**) [74].

Synthesis of coumarins: Coumarins and their derivatives have attracted significant interest from organic and medicinal chemists due to the prevalence of this heterocyclic nucleus in numerous natural products. These compounds display a broad spectrum of biological activities, including anthelmintic, hypnotic, insecticidal, and anticoagulant effects [75]. Beyond their pharmacological relevance, coumarins are widely employed as optical brightening agents, fluorescent dyes, tunable laser dyes, food additives, cosmetics and agrochemicals [76]. They also serve as key intermediates in the synthesis of coumarins, chromenes, 2-acyl resorcinols and fluoro-coumarins [77].

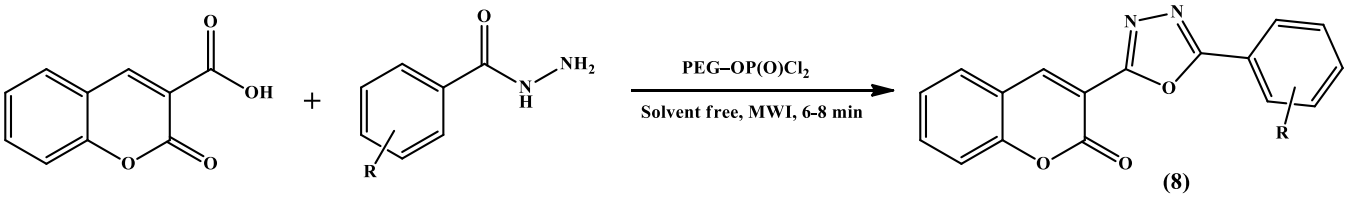
Various synthetic methodologies have been reported for coumarin derivatives, including the Perkin [78], Pechmann [79], Knoevenagel [80], Reformatsky [81] and Wittig [82]



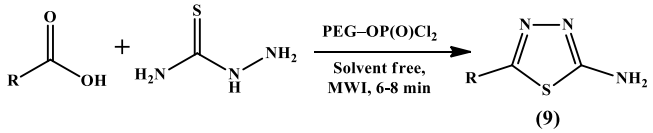
Scheme-V: Synthetic route of indazole-tetraones (**6**)



Scheme-VI: Synthetic route of DHPMs (**7**)



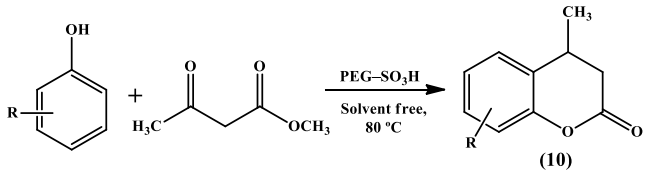
Scheme-VII: Synthetic route of 1,3,4-oxadiazoles (**8**)



Scheme-VIII: Synthetic route of 1,3,4-thiadiazoles (**9**)

reactions. In Pechmann reaction, a range of acidic condensing agents such as sulfuric acid, hydrochloric acid, trifluoroacetic acid, phosphoric acids, phosphorus pentoxide and Lewis acids, including $ZnCl_2$, $FeCl_3$, $AlCl_3$, as well as solid acid catalysts like ion-exchange resins, cellulose sulfuric acid, periodic mesoporous silica chloride (PMSCl) [83] and benzylsulfonic acid-functionalized mesoporous Zr-TMS have been employed. Other catalysts such as indium chloride [84] and dipyridine copper chloride [85] have also been reported.

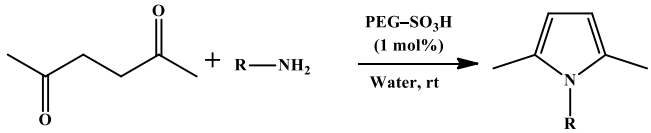
Microwave-assisted synthesis has been explored in recent years [86], offering faster reactions and improved efficiency. Nevertheless, many of these methods still suffer from drawbacks, including prolonged reaction times, harsh conditions, potential side reactions, limited reagent availability, and low product yields. To address these limitations, Nazeruddin *et al.* [87] reported the use of PEG-SO_3H as an effective, eco-friendly catalyst for the synthesis of coumarin derivatives (**10**) (**Scheme-IX**).



Scheme-IX: Preparation of coumarin derivatives (**10**)

Synthesis of Paal-Knorr pyrrole and bispyrrole: In medicinal chemistry, the pyrrole ring system is a helpful structural component has also been widely used in drug development as an antioxidant, antibacterial, anti-inflammatory, antiviral and antitumoral agent [88]. Moreover, pyrrole derivatives are a very useful class of intermediates that are used extensively in materials science and are used in the synthesis of heterocyclic compounds and natural products [89]. It follows that the development of numerous synthetic techniques for the synthesis of these compounds is not surprising [90]. Despite these advancements, the Paal-Knorr [91] reaction, which involves reacting 1,4-dicarbonyl compounds with primary amines in the presence of different promoting agents remains the most appealing technique for synthesizing pyrroles [92]. Some of these techniques do, however, have certain disadvantages, including the use of dangerous organic solvents, high costs, the use of a stoichiometric or relatively expensive catalyst, labour-intensive workup that results in the production of large amounts of wastes containing toxic metals and low product yields.

A green synthetic approach for the synthesis of bispyrrole and Paal-Knorr pyrrole derivatives (**11**) has been developed, as illustrated in **Scheme-X**. The reaction involves the condensation of a primary amine with 2,5-hexanedione and is carried out in water using a catalytic amount of PEG-SO_3H [93]. This environmentally benign protocol not only avoids the use of

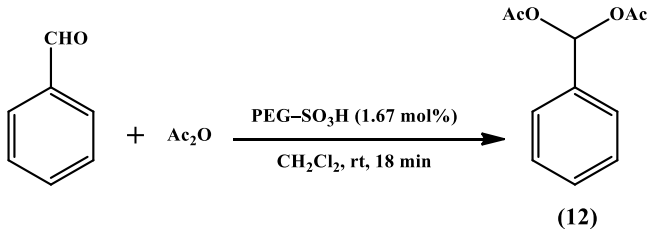


Scheme-X: Preparation of Paal-Knorr pyrrole derivatives (**11**)

hazardous organic solvents but also offers operational simplicity, mild reaction conditions and good efficiency, highlighting the advantages of polymer-supported acid catalysis in sustainable heterocyclic synthesis.

Synthesis of acylals: Acylals serve as important protecting groups in organic synthesis, offering enhanced stability under both strongly acidic and neutral conditions, making them valuable alternatives to acetals [94]. They find extensive applications in industrial and synthetic chemistry, including their use as stain-bleaching agents and crosslinking reagents in the cotton and cellulose industries [95]. In addition, acylals act as versatile synthons in various well-established organic reactions, such as the synthesis of nitriles [96], Grignard reactions [97], Prins reactions [98] and condensation reactions including Knoevenagel [80] and benzoin reactions [99], owing to their superior properties as protective groups and their significance in both industrial and synthetic contexts.

Recognizing the utility of acylals, Shiri *et al.* [100] developed an efficient method for their synthesis from various substituted aldehydes using acetic anhydride in the presence of PEG-SO_3H as a homogeneous polymer-supported catalyst. This approach offers several advantages, including a cleaner reaction profile, operational simplicity, rapid reaction times, high yields, and cost-effectiveness, with PEG-SO_3H exhibiting remarkable catalytic activity (**Scheme-XI**).

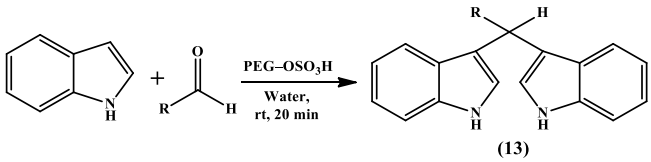


Scheme-XI: Protection of aldehydes with acetic anhydride (**12**)

Synthesis of *bis*(indolyl)alkanes: The chemistry of indole derivatives has been extensively reviewed in the literature [101], owing to their wide-ranging biological activities [102]. Among these, bis(indolyl)methanes (BIMs) represent an important class of compounds, typically synthesized *via* the reaction of indoles or indolyl Grignard reagents with amines, imines, alcohols, alkenes, alkynes, aldehydes, ketones, iminium salts or nitrones in the presence of protic or Lewis acid catalysts [102]. The synthetic importance of BIMs has prompted the development of numerous methodologies to improve efficiency, selectivity and yields.

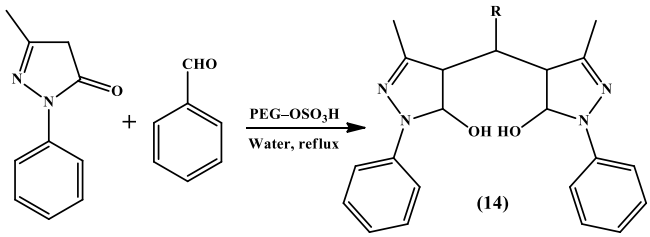
Recent studies have highlighted the use of PEG-SO_3H as a highly efficient, homogeneous polymer-supported catalyst for the synthesis of BIMs, offering environmentally benign reaction conditions and operational simplicity [103]. Similarly, Lasekan *et al.* [102] employed PEG-$OP(O)Cl_2$ as a catalyst

for the preparation of BIMs, achieving good yields and further exploring their anticancer properties (**Scheme-XII**). These developments underscore the growing interest in polymer-supported catalysts as versatile tools for the sustainable synthesis of biologically active indole derivatives.



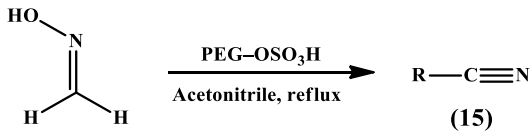
Scheme-XII: Preparation of *bis*(indolyl)methane's (**13**)

Synthesis of *bis*(pyrazolyl)methanes: Pyrazzolone derivatives, which have heterocyclic ring systems, exhibit a variety of special biological activities. Many commercially available medications for myocardial ischemia [104] and brain ischemia [105] now contain some derivatives of pyrazolones. Among them, *bis*(pyrazolyl)methanes (BPMs) (**14**) with a wide range of recognized biological activity are employed as antipyretic [106], anti-inflammatory [107], gastric secretion stimulatory [108], antidepressant [109], antibacterial [110] and antifilarial agents [111]. One example of a BPMs is 4,4′-(arylmethylene)-*bis*(3-methyl-1-phenyl-1*H*-pyrazol-5-ol)s. Furthermore, these substances have been used as dyestuffs, fungicides [112], pesticides [113], insecticides [114] and chelating and extracting reagents for various metal ions [115]. The importance of pyrazole derivatives has prompted the development of several synthetic methods; however, many of these approaches suffer from limitations such as low yields, harsh reaction conditions or long reaction times. Consequently, an effective catalytic strategy for the synthesis of pyrazoles has been reported, as illustrated in **Scheme-XIII**. This method offers improved efficiency and operational simplicity, addressing the shortcomings of previous protocols [103].



Scheme-XIII: Preparation of bis(pyrazolyl)methanes (BPMs) (**14**)

Synthesis of nitriles: Several catalysts, including chlorosulfonic acid [104], silica sulfate [105], sulfamic acid [106], cyanuric chloride [107], chloral [108], anhydrous oxalic acid and P_2O_5 [109], were used to carry out the Beckmann rearrangement. These techniques have several disadvantages, including the use of hazardous solvents, high reagent costs, significant byproduct formation, extended reaction times and low yields**.** Moreover, strong Brønsted or Lewis acids, such as conc. sulphuric acid, phosphorus pentachloride in diethyl ether and HCl in acetic anhydride, are typically required for the conventional Beckmann rearrangement, which results in significant amounts of waste and corrosion issues [110]. Thus, by Beckmann rearrangement in the presence of PEG-OSO_3H, Wang *et al.* [110] have described an effective method for converting a range of ketoximes and aldoximes into their corresponding amides and nitriles (**15**) with high yields (**Scheme-XIV**).



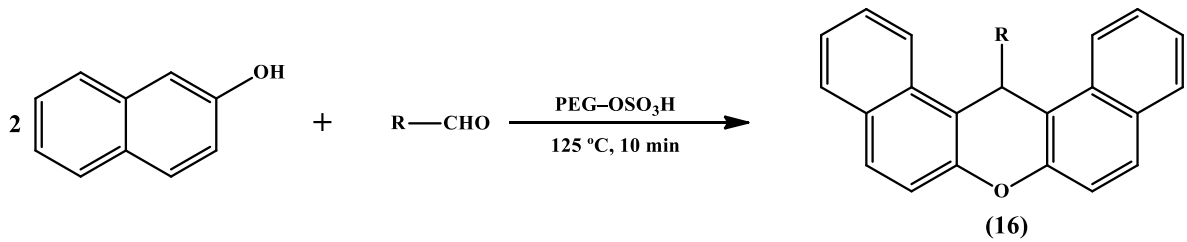
Scheme-XIV: Preparation of nitriles (**15**)

Synthesis of 14-aryl/heteroaryl-14*H*-dibenzo[*a,j*]xanthenes: Xanthenes represent an important class of oxygen containing heterocycles, exhibiting a wide range of pharmacological activities, including antiviral, anti-inflammatory, antibacterial and anti-proliferative effects [112]. Beyond their biological relevance, xanthene derivatives have found extensive applications as fluorescent dyes, pH-sensitive fluorescent probes for biomolecule visualization, photosensitizers in photodynamic therapy for tumor treatment, and in laser technologies [113]. The unique biological and photophysical properties of xanthenes have driven considerable interest in the development of efficient and sustainable synthetic methodologies.

A highly efficient and environmentally benign approach has been established for the synthesis of 14-aryl-14*H*-dibenzo-[*a,j*]xanthenes *via* a one-pot condensation of β-naphthol with various aromatic aldehydes. This protocol employs a catalytic amount of sulfonated polyethylene glycol 6000 (PEG–SO_3H), a stable, recyclable, and biodegradable polymer-supported acid, under solvent-free conditions. The method offers significant advantages, including operational simplicity, high yields and alignment with green chemistry principles (**Scheme-XV**) [114].

Conclusion

Incorporating catalytically active moieties, such as Lewis and Brønsted acid sites, into organic polymers represents one of the most effective and versatile strategies for developing novel, environmentally benign catalytic systems. Polymeric acids are easily separated from reaction mixtures and can often



Scheme-XV: Preparation of xanthenes (**16**)

be recycled without significant loss of activity, simplifying product isolation and streamlining workup procedures. Owing to the central role of acid-catalyzed transformations in organic chemistry, polymer-supported acid catalysts both Lewis and Brønsted have been instrumental in advancing green organic synthesis and the construction of combinatorial libraries. Research in polymer-supported acid catalysis generally focuses on three main areas: (i) the development of stable polymer supports with suitable hydrophobicity; (ii) the synthesis of new acidic species, including sulfonic acids, sulfonamides, and neutral Lewis acids based on metal complexes; and (iii) the exploration of effective immobilization techniques for attaching acid functionalities to polymeric matrices. While commercially available polystyrene resins remain widely used, advances in catalyst immobilization have extended far beyond simple sulfonation of polystyrene. Novel strategies, such as microencapsulation and polymer entrapment, have been developed, demonstrating remarkable efficiency and versatility for a wide range of applications.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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