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FULL PAPER

Chemical synthesis of various composites of chromen-2-one: A review

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Chromen-2-one composites are a type of heterocycles that has a wide range of uses in healthcare technology, biological research, and a variety of commercial fields. Many attempts are being made to create the innovative and more realistic ways of manufacturing such molecules in order to achieve this goal. Several chromen-2-one composite synthesis procedures are described in this study, including Von Pechmann condensation, Knoevenagel condensation, Kostanecki reaction, Baylis-Hillman retort, Michael addition, electrophilic reaction mediated by vinyl phosphonium salt, and the reaction of Heck-lactonization.

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KEYWORDS

Chromen-2-one composites; chemical synthesis; pharmacological properties.

Introduction

Chromen-2-one composites are of high significance due to their important biological activities [1]. These composites, as bioactive agents, can exhibit diverse biological activities which include antiviral [2], antibacterial [3], antimicrobial [4], anticoagulant [5], anti-inflammatory [6], anti-cancer [7], anticonvulsant [8], antioxidant [9], anti-fungal [10], and anti-HIV [11]. They also have features such as platelet aggregation inhibition [12] and steroid 5α-reductase inhibition. In addition, chemists are quite interested in them due to their huge range of applications along with photosensitizers [13] and optically fluorescent dyes and components in nutrition, fragrance, skincare, and medications.

A family of heterocyclic molecules known as benzopyrone is formed when the pyrone ring and the benzene nucleus fuse. As displayed in Figure 1, there are two varieties of benzopyrones. They are benzo-α-pyrone (A), also known as coumarin, and benzo-γ-pyrone (B), also known as chromones, and they merely differ in the location of the pyrone ring's carbonyl group [13].

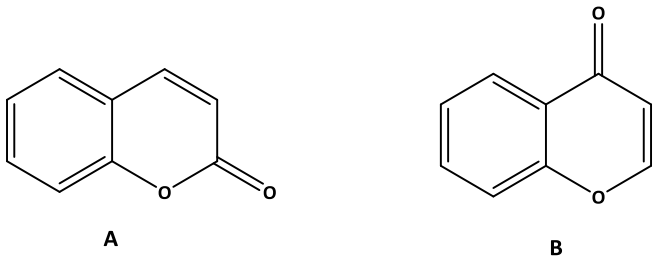


FIGURE 1 Two general types of benzopyrones

Chromen-2-ones, as depicted in Figure 2, are categorized according to their molecular constitution: the simple chromen-2-ones with the benzene ring hydroxylated, alkylated, or alkoxylated (e.g., Umbelliferone). The linear furanochromen-2-ones (e.g., psoralen) and the angular furanochromen-2-ones (e.g., Angelicin) are furanochromen-2-ones which have a five-membered furan ring attached to the chromen-2-one group [14].

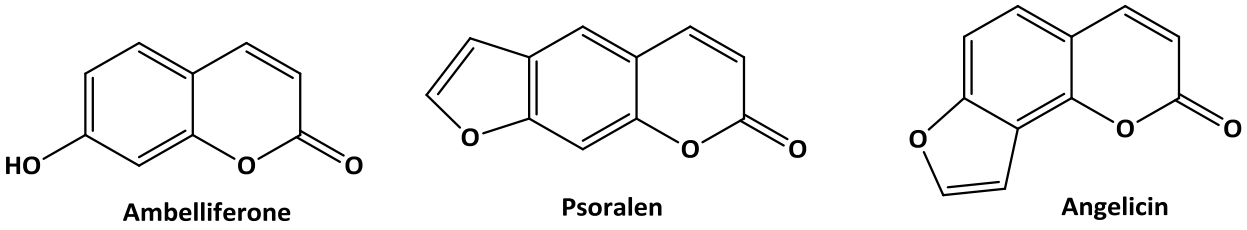


FIGURE 2 The classic examples of different types of chromen-2-ones

As demonstrated in Figure 3, a ring with six members linked to the chromen-2-one group in pyrano chromen-2-ones (e.g., Xanthyletin and Seselin). Chromen-2-ones have pyrone ring substitutes (e.g., Warfarin) [14].

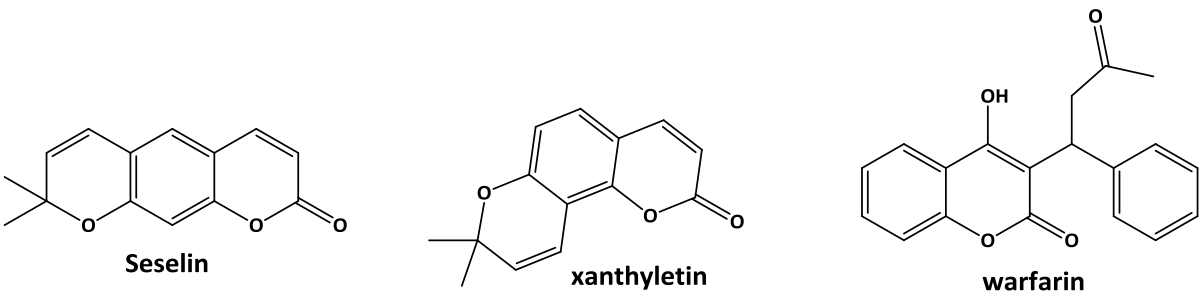


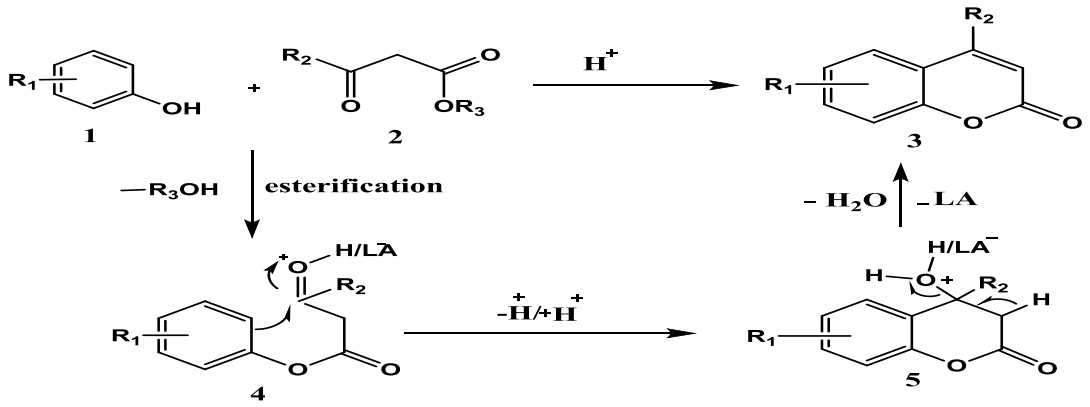
FIGURE 3 The classic examples of pyrano chromen-2-ones

Synthetic chemists are developing new strategies to isolate chromen-2-one composites in the light of these diverse activities of chromen-2-ones. The most extensively used technique for chromen-2-one synthesis is the Pechmann reaction [15], which includes the condensing of phenols with β-keto esters using acid as a catalyst. This approach involves both homogeneous catalysts, including concentrate sulphoric acid [15], F_3CCOOH [16], aluminum chloride [17], zinc chloride, titanium (IV) chloride [18], and many others. Nafion resin/silica composites [19], cation-exchange resins [20], and the other solid acids are some examples of heterogeneous catalysts.

Synthetic Methods for Chromen-2-One Composites

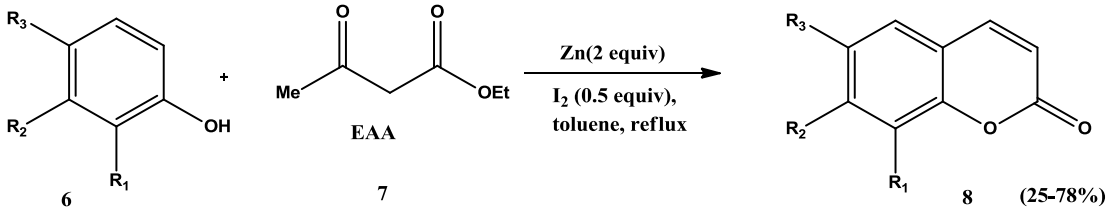
The reaction of pechmann condensation

In 1883, Pechmann & Duisburg were the first to mention the Pechmann condensation process. Because of its ease of preparation and low cost of starting material, it's been widely used in the production of chromen-2-ones. In the presence of an acid catalyst, as depicted in Scheme 1, the esterification/transesterification of phenol 1 & β-keto ester 2 forming species 4, then the activated carbonyl binds to the ortho position of the aromatic ring, giving species 5. Finally, chromen-2-one composites 3 are obtained by dehydrating species 5.



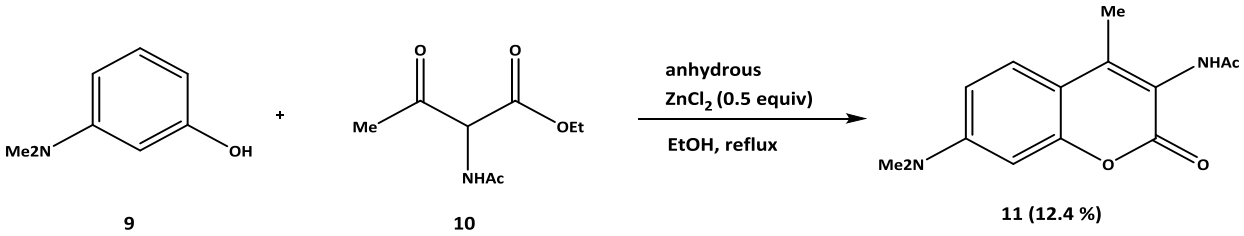
SCHEME 1 Pechmann condensation catalyzed by an acid

When substituted phenols 6 are combined with ethyl 3-oxobutanoate 7 in the presence of Zn-I_2 combination, a variety of the substituted chromen-2-ones 8 were generated in good yield, as depicted in Scheme 2 [21]. The non-substituted phenols and phenols with electron removing groups like NO_2 generate lower yields than phenols with electron giving groups like -CH_3.



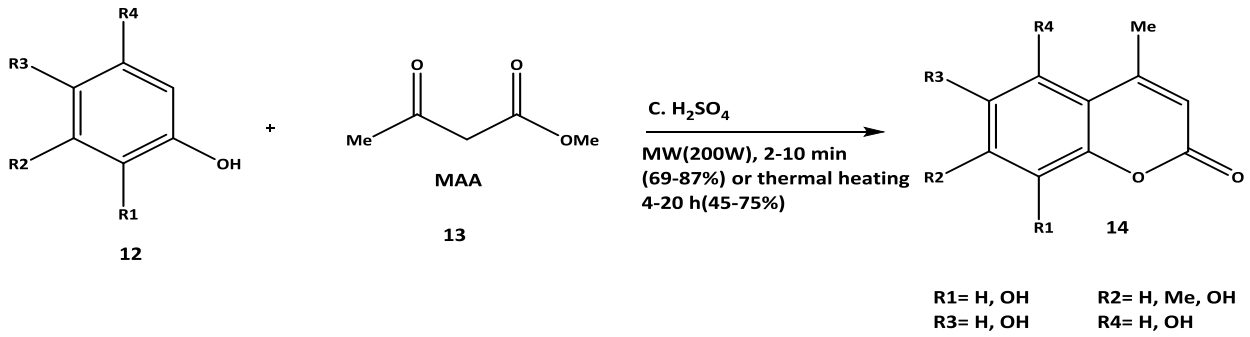
SCHEME 2 The reaction between various phenols and active methylene-containing group

As depicted in Scheme 3, only 12.4 % of the acetamido coumarin 11 is produced when 3-(dimethylamino) phenol 9 reacts with ethyl 2-acetamido-3-oxobutanoate 10 in absolute ethanol under reflux conditions using anhydrous zinc chloride as a catalyst (Scheme 3) [22].



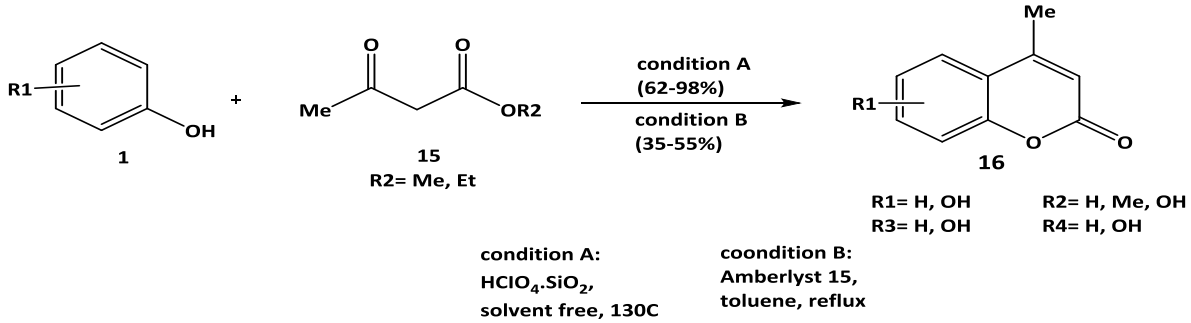
SCHEME 3 The synthesis of acetamido chromen-2-one composites using zinc chloride as promoter

Using concentrated sulphoric acid as a catalyst under microwave and conventional heating, the substituted chromen-2-ones 14 were produced in good yield from substituted phenols 12 and methyl 3-oxobutanoate 13, as depicted in Scheme 4 [23].



SCHEME 4 Synthesis of various substituted chromen-2-ones under different energy sources

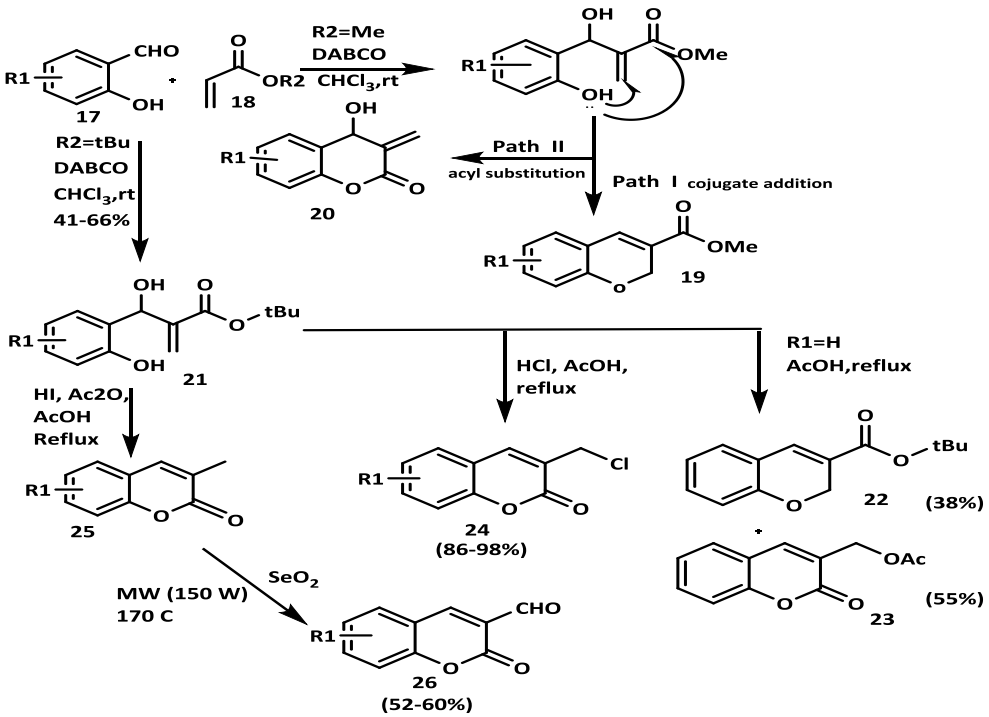
In a liquid free environment (Scheme 5, condition A), substituted chromen-2-ones 16 were generated in good yield by reacting β-keto esters 15 with substituted phenols 1 employing a heterogeneous catalyst, $HClO_4.SiO_2$ [24]. This approach uses a low-cost catalyst and produces good product yields in a shorter reaction time. However, employing a similar process catalyzed by acidified catalyst Amberlyst-15 [25] during refluxing conditions in toluene, less production of substituted chromen-2-ones 16 were obtained (Scheme 5, Condition B).



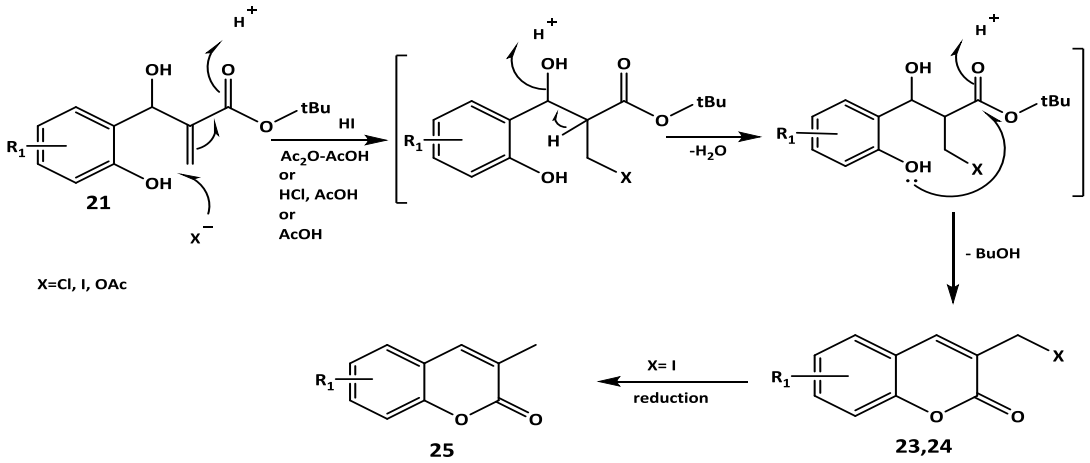
SCHEME 5 Synthesis of various substituted chromen-2-ones under a liquid free environment

Baylis-Hillman reaction

Scheme 6 reveals the Baylis-Hillman technique for synthesis of substituted chromen-2-one composites s. When 1,4-diazabicyclo [2.2.2] octane is presented, salicylaldehyde 17 reacts with methyl acrylate 18a to create a combination of chromen-2-one composites 19 and 20 [26]. The similar reactions of salicylaldehyde 17 with tertiary butyl acrylate 18b using the traditional approach [27] and/or irradiation via microwaves [28] generate analogous adducts of Baylis-Hillman 21, which are cyclized in acetic acid to yield a 3-substituted mixture of chromen-2-one composites 22 and 23. When adducts of Baylis-Hillman 21 are treated with strong hydrochloric acid in acetic acid reflux, the desired yields of 3-(chloromethyl)-2*H*-chromen-2-one 24 are obtained. Furthermore, the reaction of 21 with hydrogen iodide in a combination of acetic anhydride and acetic acid under reflux yields 3-methyl-2H-chromen-2-one 25, which is then converted into the equivalent 2-oxo-2*H*-chromen-3-carbaldehyde 26 by further reaction with selenium dioxide. Scheme 7 depicts the proposed pathway for chromen-2-one composites 23, 24, and 25 synthesis.



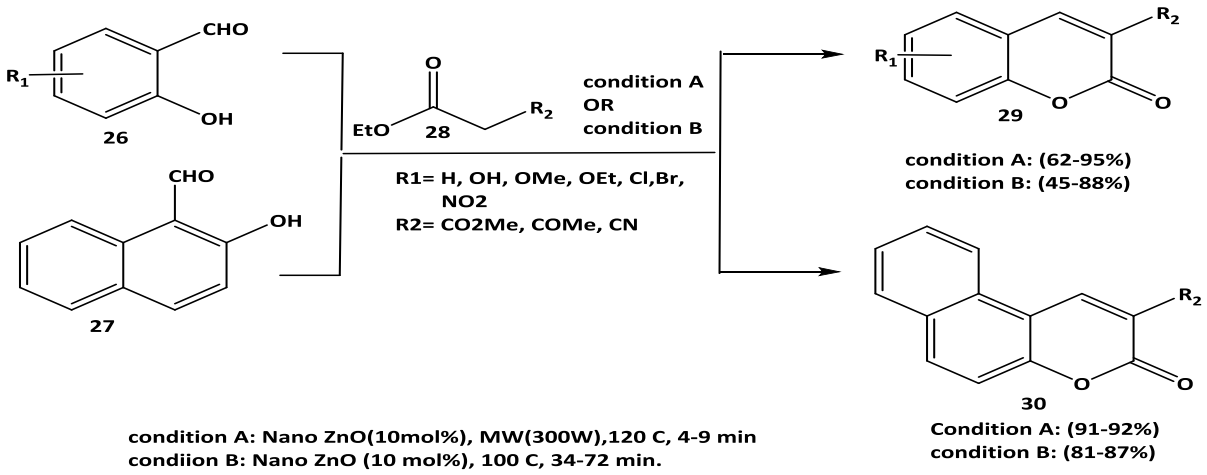
SCHEME 6 Synthesis of various 3-substituted chromen-2-ones by Baylis-Hillman technique



SCHEME 7 Synthetic mechanism of various 3-substituted chromen-2-ones utilizing a Baylis-Hillman reaction

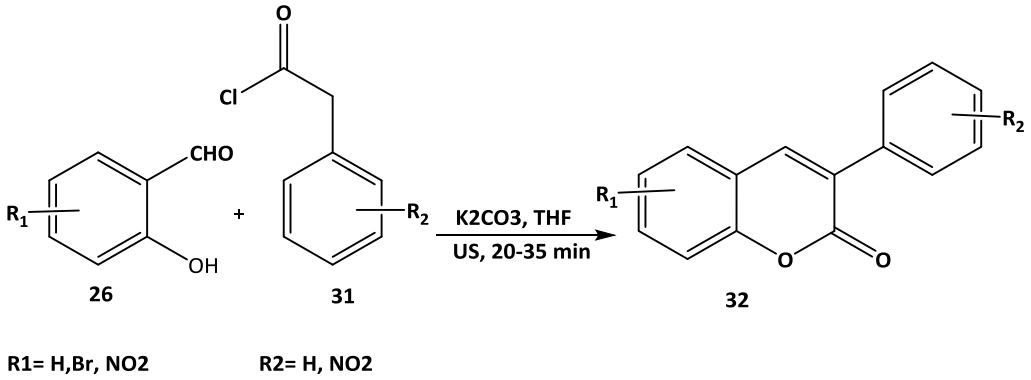
The condensation reaction of Knoevenagel

Knoevenagel condensation of different salicylaldehydes 26–27 with 1,3-dicarbonyl compounds 28 under microwave or heat conditions using zinc oxide nanoparticle catalyst has been discovered as an efficient way to synthesize 3-substituted chromen-2-one composites 29 and 30, as represented in Scheme 8 [29]. It has been proven that microwave-irradiated reactions are more efficient than thermal reactions.



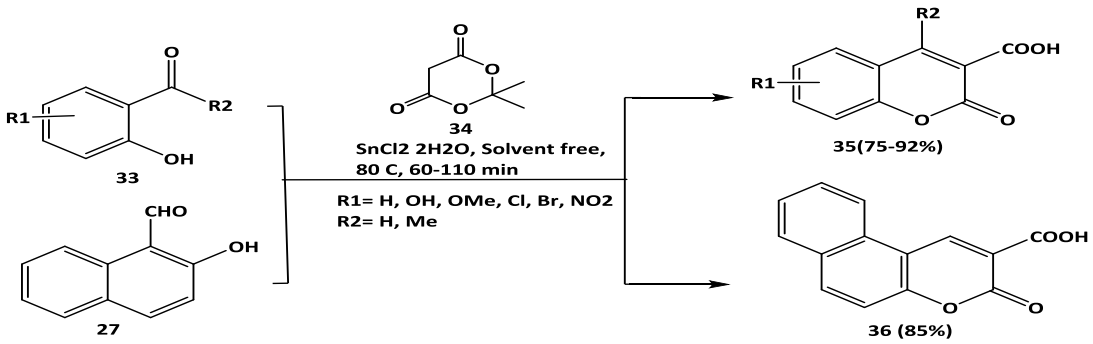
SCHEME 8 Synthesis of various 3-substituted chromen-2-ones utilizing zinc oxide nanoparticle as a promoter

The ultrasound irradiation technique was employed to make 3-aryl chromen-2-one composites. In tetrahydrofuran, the ultrasonic irradiation of salicylaldehyde 26 with acetyl chloride substituted with aryl group 31 using potassium carbonate as a catalyst results in the production of 3-aryl chromen-2-one composites 32 in good yields, as displayed in Scheme 9 [30]. This green approach looks to be a more handy and straightforward option than the traditional heating.



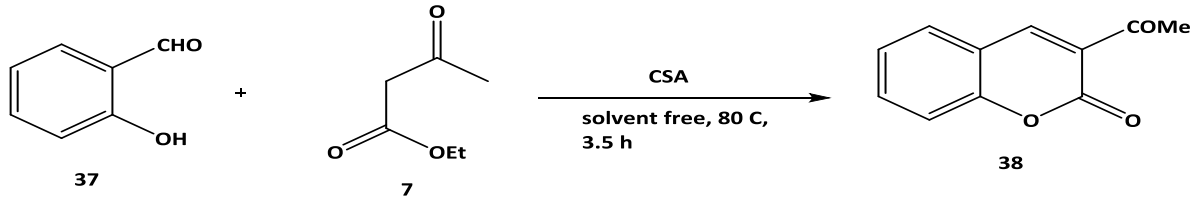
SCHEME 9 Synthesis of various 3-aryl chromen-2-ones utilizing potassium carbonate as a promoter

Under solvent-free conditions, several composites of chromen-2-one-3-carboxylic acid 35 and 36, as demonstrated in Scheme 10, have been produced in excellent yields employing catalytic quantities of stannous chloride dehydrate [31].



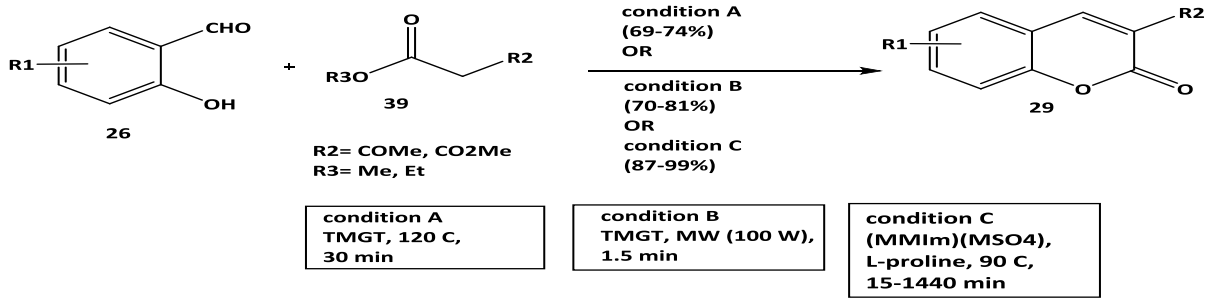
SCHEME 10 Synthesis of different chromen-2-one-3-carboxylic acid composites under solvent-free conditions

The Knoevenagel condensation process uses cellulose sulfonic acid as a promoter to produce chromen-2-one substituted at 3-position, as depicted in Scheme 11. In the presence of CSA, 3-acetyl chromen-2-one 38 is produced in a yield of 88 % under solvent-free conditions, in a reaction involving salicylaldehyde 37 and ethyl 3-oxobutanoate 7 [32].



SCHEME 11 Synthesis of chromen-2-one-3-substituted composite using cellulose sulfonic acid as a promoter

Through Knoevenagel condensation of salicylaldehyde 26 with dicarbonyl compounds 39 in the presence of 1,1,3,3-tetramethylguanidinium trifluoroacetate (TMGT) (33) under thermal heating (Scheme 12, condition A) and/or microwave irradiation conditions, 3-substituted chromen-2-ones 29 were produced in desired yields. Under heating conditions (Scheme 12, condition B), 3-substituted chromen-2-ones 29 are similarly generated from identical starting precursors using the 1,3-dimethyl-1*H*-imidazol-3-ium methyl sulfate ionic liquid in the presence of L-proline as an additional promoter (Scheme 12, condition C).

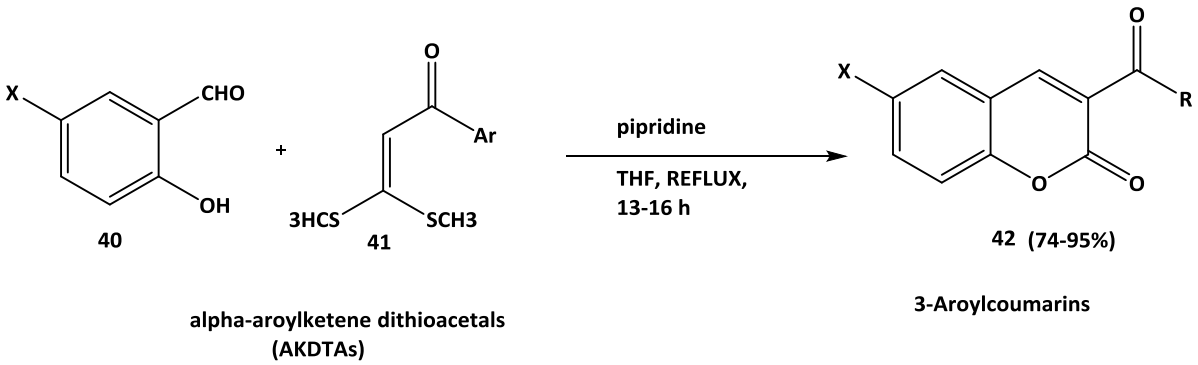


SCHEME 12 Synthesis of chromen-2-one-3-substituted composites using L-proline as a promoter in different reaction conditions

Michael addition reaction

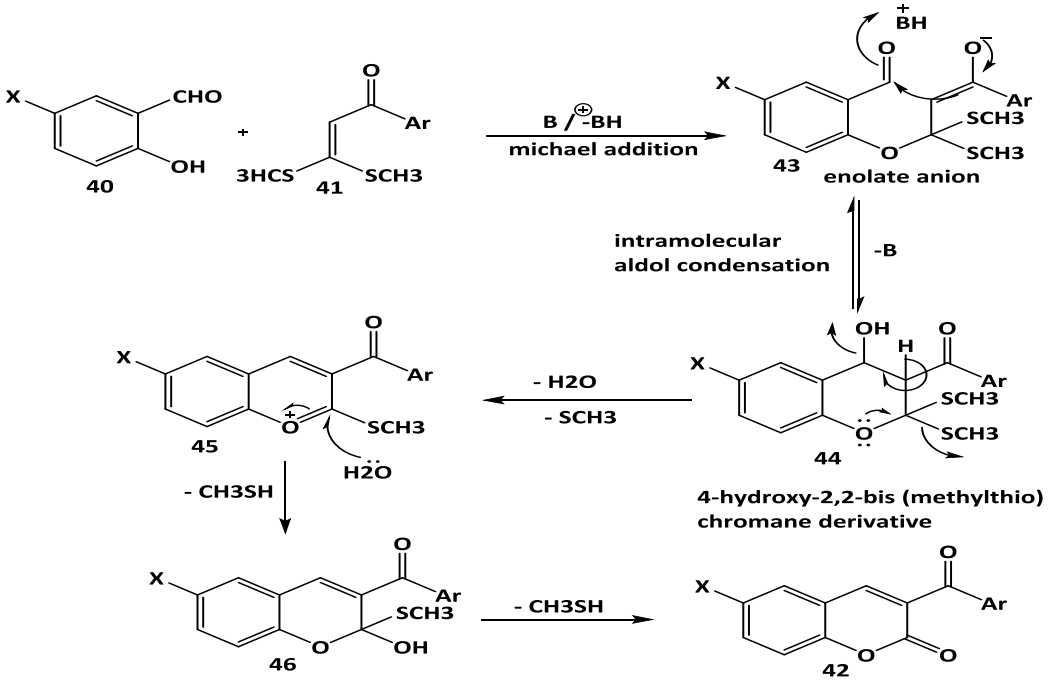
Michael addition, as depicted in Scheme 13, was used to synthesize 3-aroyl-chromen-2-

one composites 42 in excellent yields from salicylaldehyde 40 and dithioacetals of α-aroylketene 41 in a refluxing condition in the tetrahydrofuran solvent system using piperidine as a promoter [34].



SCHEME 13 Synthesis of 3-aroyl-chromen-2-one composites using a Michael addition reaction

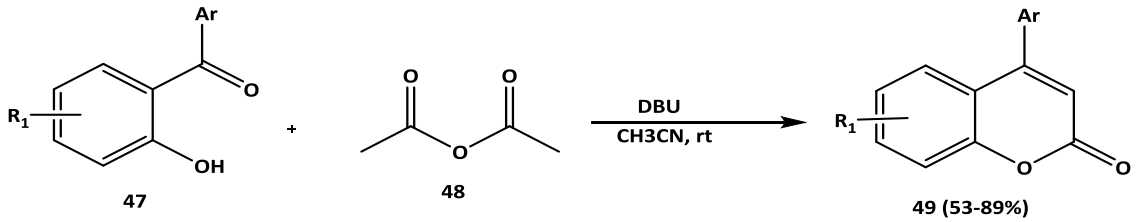
As presented in Scheme 14, the reaction mechanism of the aforementioned composites starts with a Michael addition, and then enhances to an intramolecular aldol condensation process [34].



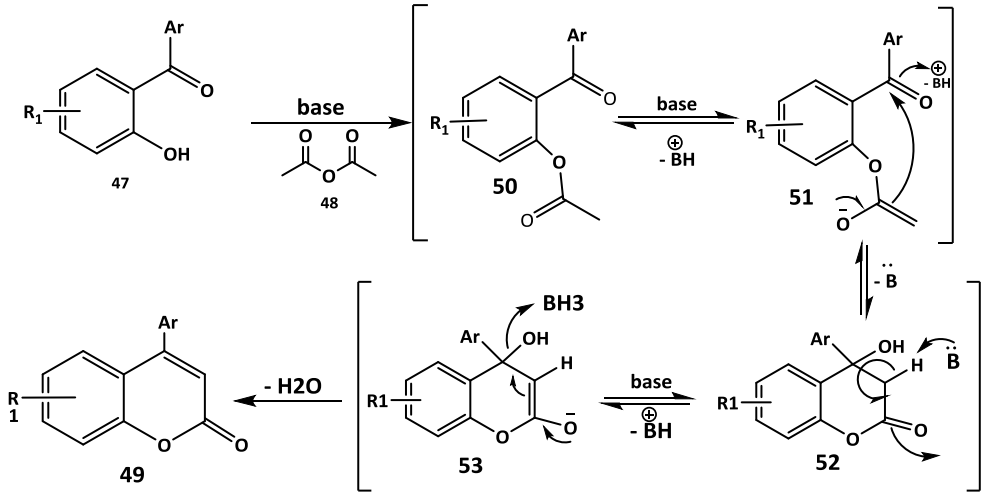
SCHEME 14 Synthetic mechanism of the 3-aroyl-chromen-2-one composites 42

Kostanecki reaction

Under relatively mild conditions, the Kostanecki reaction of 2-benzoylphenol 47 with acetic anhydride 48 under the catalytic influence of DBU yielded 4-arylchromen-2-one composites 49 in excellent yields, as depicted in Scheme 15. Also, the mechanistic steps concerning this reaction are illustrated in Scheme 16 [35].



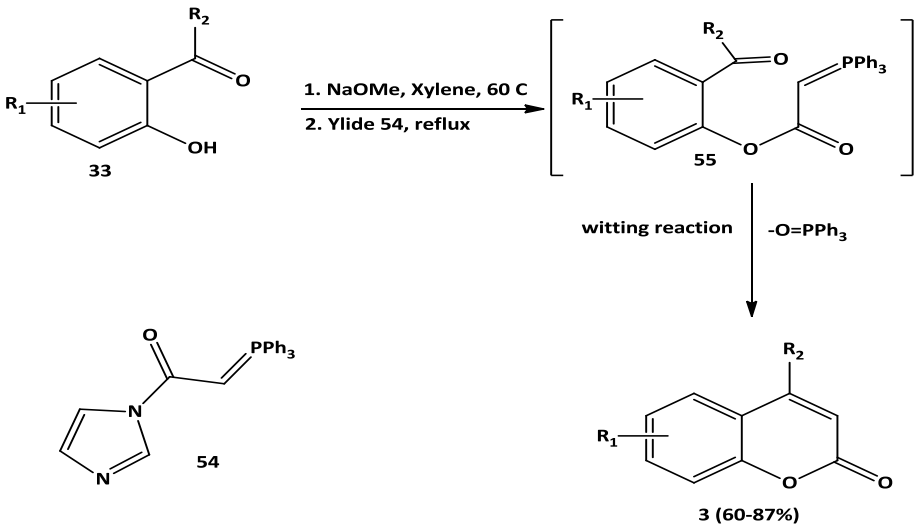
SCHEME 15 Synthesis of 4-arylchromen-2-one composites under Kostanecki reaction conditions



SCHEME 16 The kostanecki reaction mechanism

Wittig reaction

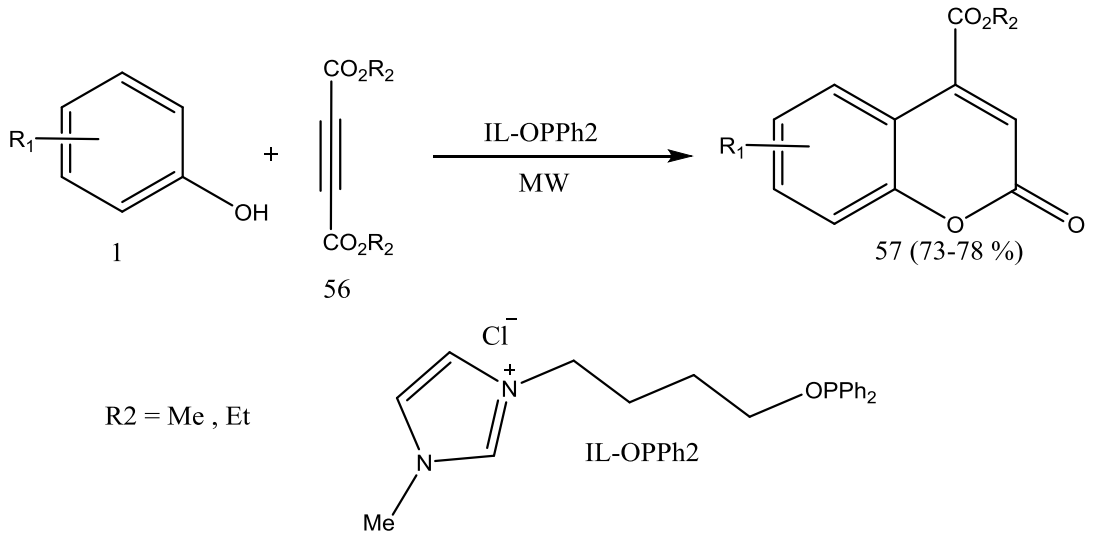
Kumar and colleagues used the intramolecular Wittig cyclization to effectively synthesize substituted chromen-2-one composites 3 from ortho carbonyl group substituted phenolic compounds 33 and imidazole triphenyl phosphorane ylide 54, as depicted in Scheme 17. As determined by spectroscopic data, all of the reactions take place via the production of phosphorane intermediates 55 [35].



SCHEME 17 Synthesis of substituted chromen-2-one composites via an intramolecular Wittig cyclization methodology

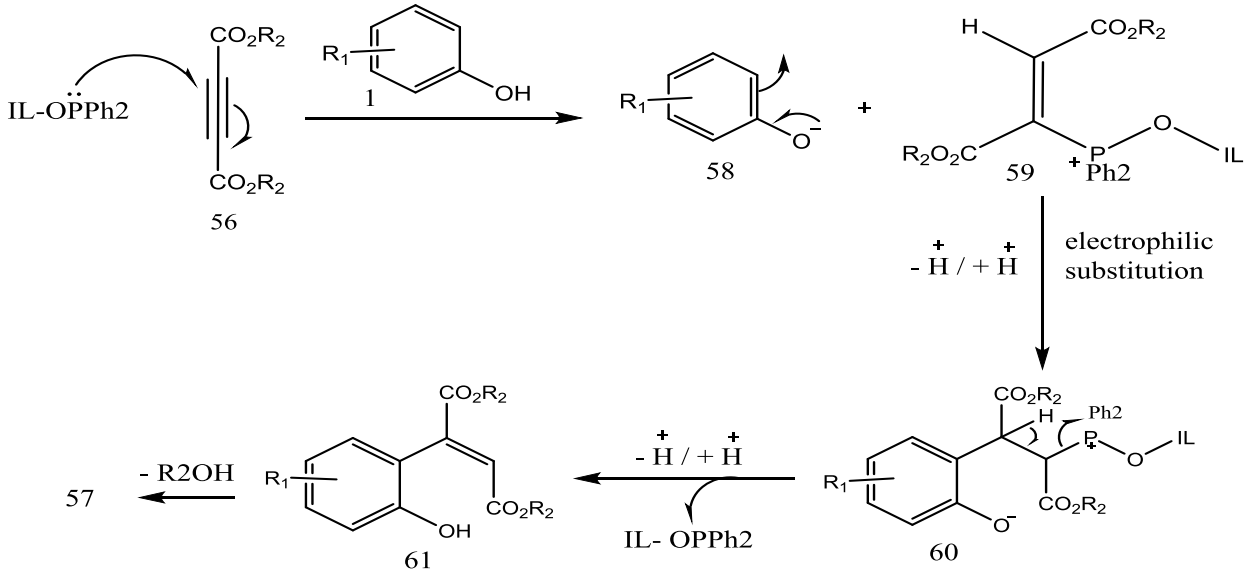
Electrophilic substitution process mediated by vinyl phosphonium salt

Under solvent-free microwave conditions, a number of 4-carboxy (methyl/ethyl) chromen-2-one composites 57 were produced in excellent yields from substituted phenols 1 and di(methyl/ethyl) 2-butynedioate 56 in the presence of phosphinite alkaline solution, as illustrated in Scheme 18. The process exceeded through the ionic liquid of diphenylphosphine group [37].



SCHEME 18 Synthesis of 4-functionalized chromen-2-one composites via an electrophilic substitution reaction

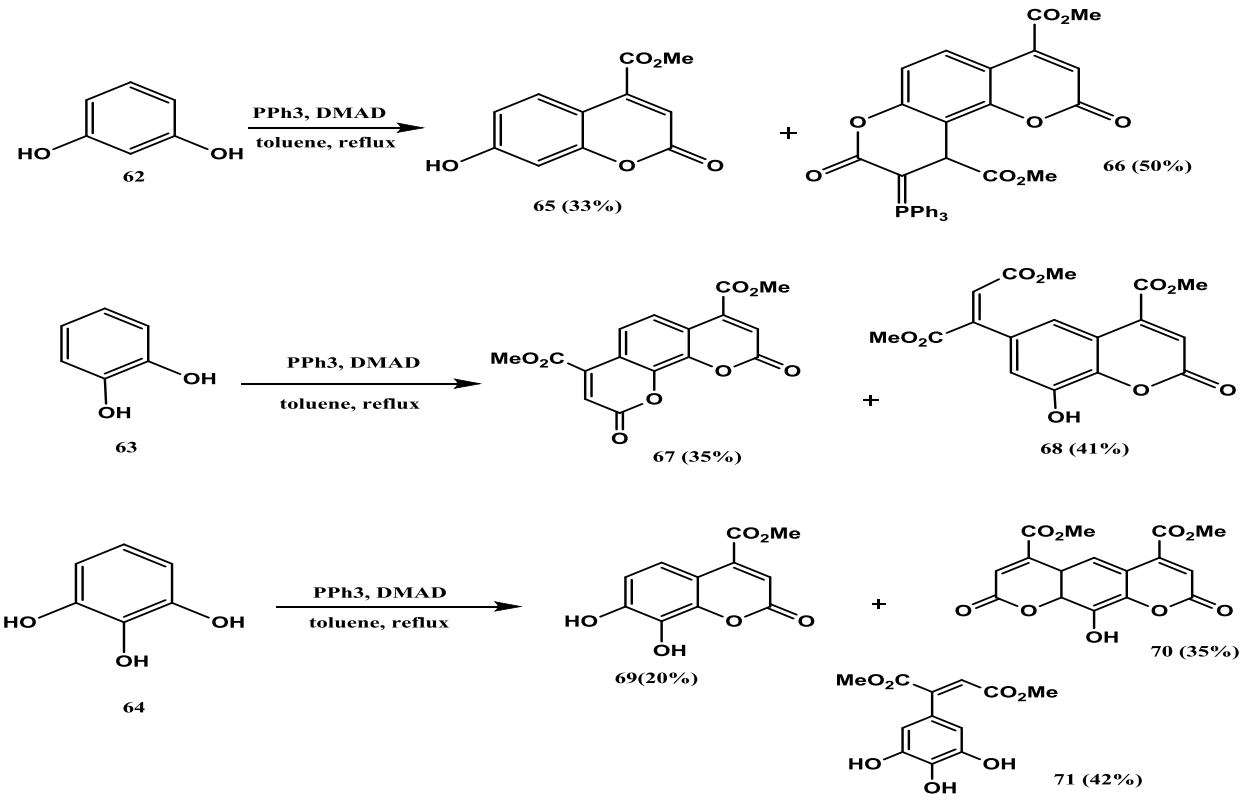
Scheme 19 depicts the suggested mechanistic steps involved in the generation of 4-functionalized chromen-2-one composites 57 by electrophilic substitution mediated by vinyl phosphonium salts.



SCHEME 19 Synthetic mechanism of 4-functionalized chromen-2-one composites via an electrophilic substitution reaction

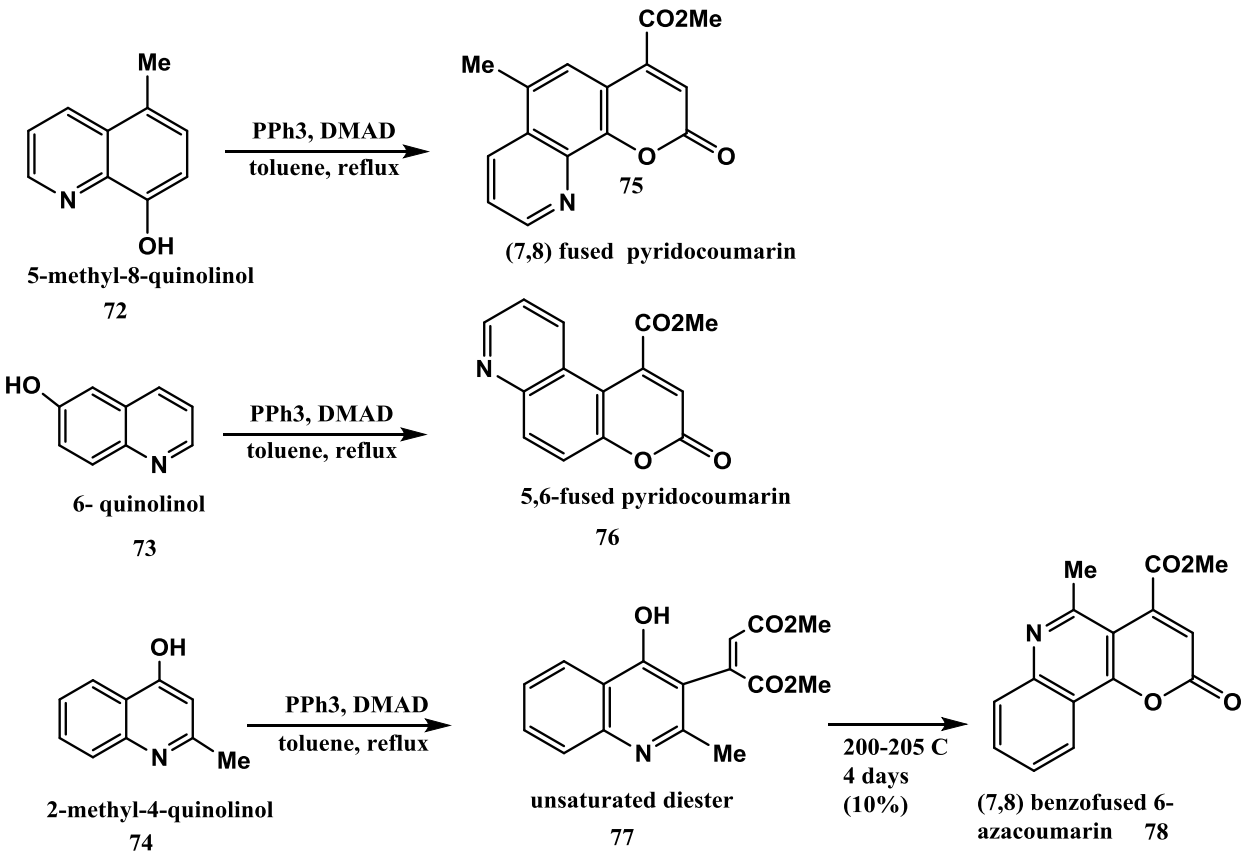
By reacting di-or tri-phenols with dimethyl 2-butynedioate under refluxing conditions in toluene as a solubilizing medium and with the presence of

triphenylphosphine, polyfunctionalized chromen-2-one composites are produced with a considerable number of the unwanted by-products, as displayed in Scheme 20 [38].



SCHEME 20 Synthesis of polyfunctionalized chromen-2-one composites employing triphenylphosphine as a promoter

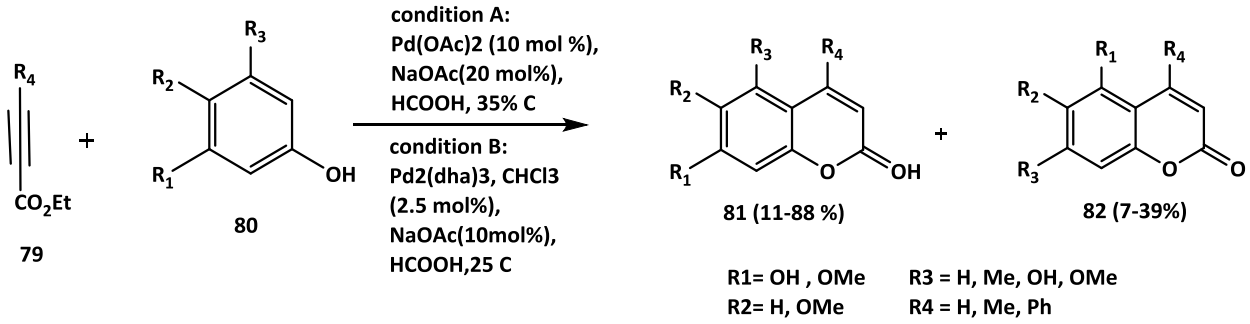
This method has also been utilized to make the angular pyridochromen-2-one composites 75, 76, and benzo-fused 6-azachromen-2-one 78, as demonstrated in Scheme 21 [38].



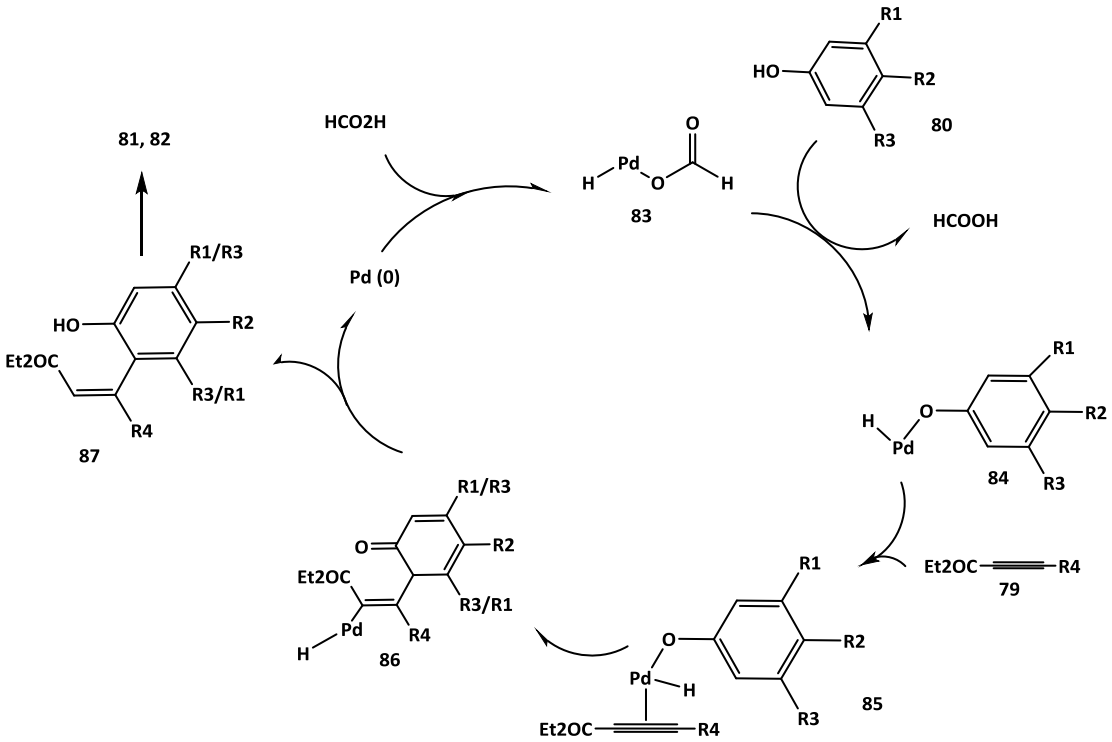
SCHEME 21 Synthetic of pyridochromen-2-one and benzo-fused azacohromen-2-one composites via an electrophilic substitution reaction

Palladium-catalyzed reactions

Ethyl propynoate 79 and substituted phenols 80 undergo a palladium-catalyzed reaction which produces polysubstituted chromen-2-one composites 81 and 82, as depicted in Scheme 22. In the same side, Scheme 23 depicts the hypothesized synthetic steps for this synthesis [39].

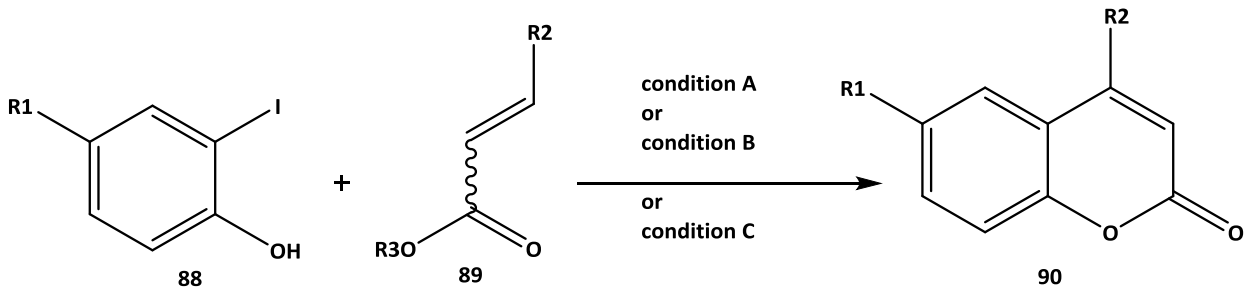


SCHEME 22 Synthesis of polysubstituted chromen-2-one composites under the palladium-catalyzed reaction conditions



SCHEME 23 Synthetic mechanism of polysubstituted chromen-2-one composites 81 and 82

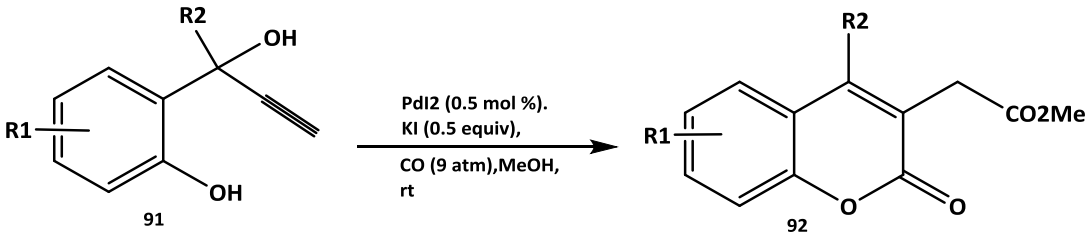
Heck-lactonization using palladium catalysis of *E*- or *Z*-enoates 89 with 2-iodophenols 88 yielded 4,6-disubstituted chromen-2-one composites s 90, as presented in Scheme 24 [39].



condition A : 10 mol% of Pd(OAc)2,3 equiv Et3N, H2O, 80 C, 40 h
condition B : 10 mol % of PdCl2, 3 equiv Et3N, H2O, 80 C, 40 h
condition C : 10 mol% of Pd(OAc)2, in the presence or absence of 20 mol % of pph3, 3 equiv AgCO3, acetone, reflux, 40 h

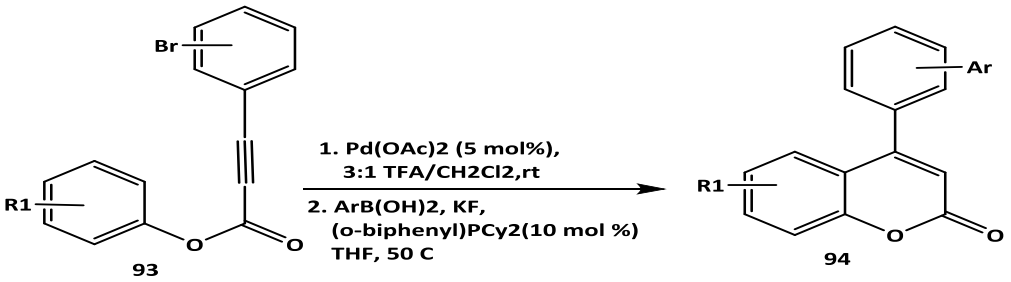
SCHEME 24 Synthesis of 4,6-disubstituted chromen-2-one composites via a palladium catalysis methodology

Furthermore, dicarbonylation of accessible 2-(1-hydroxyprop-2-yn-1-yl) phenols 91 catalyzed by palladium in methanol at room temperature gives polyfunctionalized chromen-2-ones 92 in good quantities, as displayed in Scheme 25 [39].



SCHEME 25 Synthesis of polyfunctionalized chromen-2-one composites via a palladium catalysis methodology

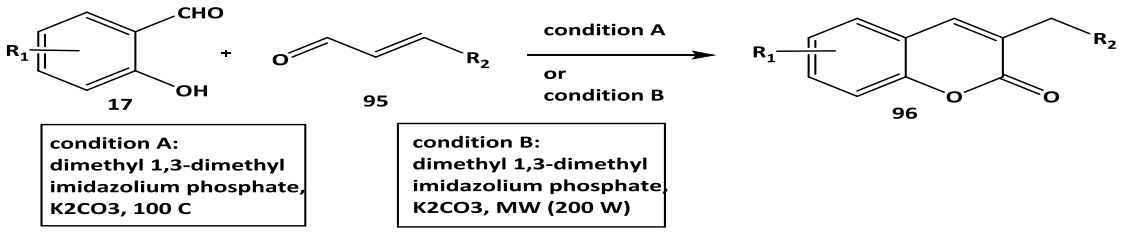
Moreover, the 4-arylchromen-2-one composites 94 are obtained in moderate to fair yields via cycloisomerization mediated by electrophilic palladium of brominated aryl propynoate 93 performed by Suzuki–Miyaura reaction using Ar-B(OH)$_2$, as depicted in Scheme 26 [39].



SCHEME 26 Synthesis of 4-arylchromen-2-one composites via a palladium catalysis methodology

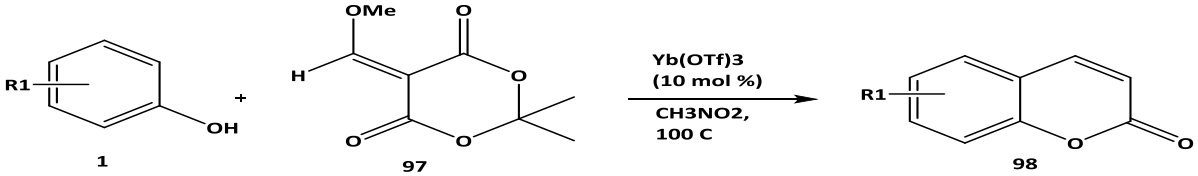
Other methods

By generating N-heterocyclic carbine intermediates under conventional heating (Scheme 27, condition A) or microwave irradiation (Scheme 27, condition B), a variety of 3-alkylchromen-2-one composites 96 are afforded in high quantities from salicylaldehyde 17 and α,β-unsaturated aldehydes 95 [40].



SCHEME 27 Synthesis of 3-alkylchromen-2-one composites under various energy sources

The substituted phenols 1 reaction with 5-(methoxymethylene)-isopropylidene malonate 97 in nitromethane at 100 °C promoted by Ytterbium (III) triflate create the substituted chromen-2-one composites s 98 in good yields, as illustrated in Scheme 28 [40].



SCHEME 28 Synthesis of monosubstituted chromen-2-one composites utilizing Ytterbium (III) triflate as a promoter

Conclusion

The benefits and/or drawbacks of chromen-2-one composite synthesis in one pot compared to the other techniques were explored in this review. The ionic liquids and/or solid acids catalyze reactions under microwave and/or the ultrasonic irradiation conditions. Both the Pechmann and Knoevenagel condensation reactions have several advantages, including high product yields, the simplicity of the product separation, short reaction times, and the environmental benefits by preventing hazardous precursors and solvents. Chemo- and regioselective synthesis of 3-substituted chromen-2-ones was accomplished using Baylis-Hillman reactions under the mild conditions. Under neutral circumstances, however, the electrophilic substitution reactions of phenols mediated by vinyl phosphonium salts give 4-carboxyalkyl chromen-2-one composites in high yields. For the production of chromen-2-ones with acid-sensitive functional groups, this approach has a lot of benefits. Furthermore, the regioselective synthesis of chromen-2-one composites from 2-iodophenols and enoates has been achieved using a palladium-catalyzed Heck lactonization technique. This reaction is demonstrated to be sensitive to steric hindrance in the enoates surrounding the double bond. The Kostanecki reaction conditions can improve the yields of chromen-2-one composites, particularly those which are highly functionalized 4-arylchromen-2-one composites with structural variety.

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Ethical issues[1]

The scientific committee of the Pharmaceutical Chemistry Department was approved this work

Competing interests

We have no conflicts of interest to disclose.

Authors' contributions

All authors contributed toward data analysis, drafting and revising the paper and agreed to responsible for all the aspects of this work.

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