Bulletin of Environment, Pharmacology and Life Sciences
Bull. Env. Pharmacol. Life Sci., Vol 13 [2] January 2024 : 69-84

Online ISSN 2277-1808
Journal's URL:http://www.bepls.com
CODEN: BEPLAD

REVIEW ARTICLE **OPEN ACCESS**

Environmentally Benign Synthetic Approaches for the Synthesis of Thiazole Derivatives: A Review

Mir Mohammad Masood[a]* and Arshid Iqbal[b]
[a]Department of Chemistry, Government Degree College for Women Baramulla, Jammu and Kashmir 193101, India;
[b]Department of Botany, Government Degree College for Women Baramulla, Jammu and Kashmir 193101, India.
*Address for correspondence to this author at Medicinal Chemistry Lab., Department of Chemistry, Government Degree College for Women Baramulla, Jammu and Kashmir 193101, India. Mob. +919797392211; Fax: +91-1952234985; https://orcid.org/0000-0002-3788-7261, e-mail: i). mirmmasood@gmail.com; ii). mirmohammmas.771227@jk.gov.in

ABSTRACT
Thiazole is a well-known five membered heterocycle containing sulfur and nitrogen atom in the ring and is one of the most important heterocyclic compounds possessing diverse biological activities. The motivating molecular architecture of 1,3-thiazole makes them suitable moieties for drug discovery and development. In view of the biological significance, there has been extensive attention toward the development of synthetic routes for the synthesis of compounds containing thiazole ring. However, most of the synthetic routes suffer from drawbacks such as availability of starting materials, harsh reaction conditions, use of transition-metal catalysts, environmentally hazardous chemicals, long reaction times and difficult post-processing. Significant progress has been made in recent times in exploration of greener and environmentally benign synthetic approaches of thiazole synthesis. Several green chemistry-based synthetic methods like multi-component single pot reaction, recyclable green-catalyst, green solvent, reusable solvent, the solvent-free, ultrasonic-mediated synthesis and microwave-assisted technique have been developed for the synthesis of thiazole derivatives. In this review, we have summarized the various environmentally benign and ecofriendly synthetic approaches for the synthesis of thiazole derivatives as reported in the time window 2013 to 2023. The compiled data in the article will surely update the scientific community with the recent developments in this area and will help researchers to efficiently synthesize the thiazole ring using greener approach.
Keywords: *Thiazole, sulfur and nitrogen containing heterocycles, drug discovery, biological significance, green chemistry approach.*

Received 24.09.2023 Revised 19.10.2023 Accepted 28.12.2023

INTRODUCTION

Thiazole a five-membered heterocyclic ring with nitrogen and sulfur atoms, can adopt two isomeric forms (**Fig. 1**), i.e., 1,3-thiazole (thiazole) or 1,2-thiazole (isothiazole) in which the hetero atoms are in adjacent positions.

1,3-thiazole 1,2-thiazole

Fig. 1. Isomeric forms of thiazole.

Thiazole (1,3-thiazole) and its derivatives have inspired researchers since long. As a privileged heterocyclic nucleus, the thiazole ring is a fundamental and important structural component for an extensive range of biologically active compounds, including anticonvulsants [1], anticancer [2], antibacterial [3,4], antiviral [5], antifungal [6], analgesic [7], anti-inflammatory [8] and antidiabetic agents [9].

The thiazole moiety is also a key constituent of some naturally occurring compounds and many commercial antibiotics. Vitamin B_1 (thiamine), penicillin (broad spectrum antibiotic), tiazofurin (antineoplastic),

ritonavir (antiretroviral), thiabendazole (antihelminthic and antifungal) and abafungin (antifungal) are some of the examples of thiazole bearing compounds (**Fig. 2**).

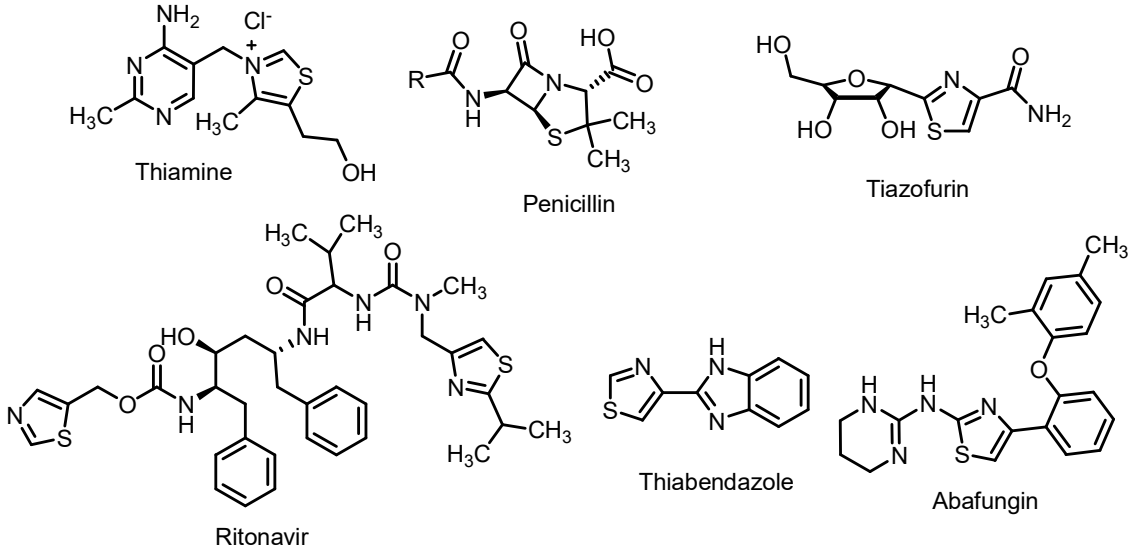
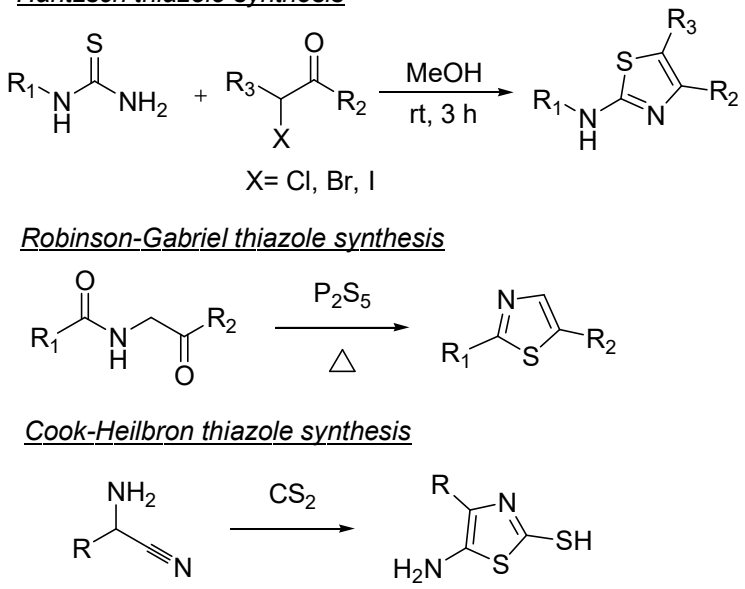


Fig. 2. Structure of vitamin B_1 (thiamine) and some 1,3-thiazole bearing drugs.

Synthetic methods toward thiazole derivatives have been summarized in many review articles documented recently [10–14]. Traditionally, the chosen method for the preparation of these compounds is via the Hantzsch thiazole synthesis, which involves the reaction of thioamides with α-haloketones [15]. However, various other earlier synthetic protocols, such as the Robinson–Gabriel [16,17] and the Cook–Heilborn [18] thiazole synthesis, among others are also documented in the literature (Scheme 1).



Scheme 1. Old methods of thiazole synthesis.

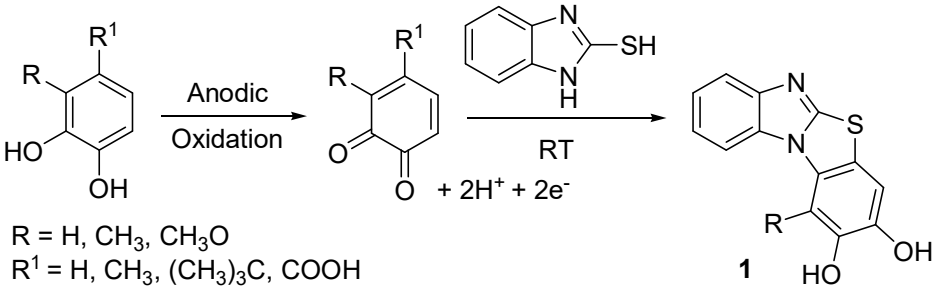
Many new methods of thiazole ring synthesis mostly involve the 'Hantzsch synthesis' method due to its simplicity, which in turn can introduce many functional groups and is used widely at present [19,20]. An inconvenient step of Hantzsch methodology is the use of toxic haloketones. Therefore, an alternative protocols based on the reactions of thioamides/ thioureas with ketones [21], α-diazoketones [22], α,β-unsaturated carbonyl compounds [23], alkynes [24], alkenes [25], β-keto esters [26,27], and nitroepoxides [28] or reactions of isothiocyanates with different substrates [29] have recently attracted attention as powerful strategies to prepare the thiazole nucleus. Although they are interesting approaches, most of them suffer from drawbacks, due to the availability of starting materials, harsh reaction conditions, use of environmentally hazardous chemicals, long reaction times and difficult post-processing or workup. Owing to shortcomings of these conventional methods, researchers have diverted their efforts towards the development of new environmentally benign synthetic routes for the synthesis of thiazole derivatives.

In the current review, focus is laid on the environmentally benign synthetic approaches adopted for the synthesis of thiazole derivatives, that have been published recently. A compiled data of all these recent articles will surely help researchers in providing a direction towards further research using greener approaches.

ENVIRONMENTALLY BENIGN SYNTHESIS OF 1,3-THIAZOLE DERIVATIVES

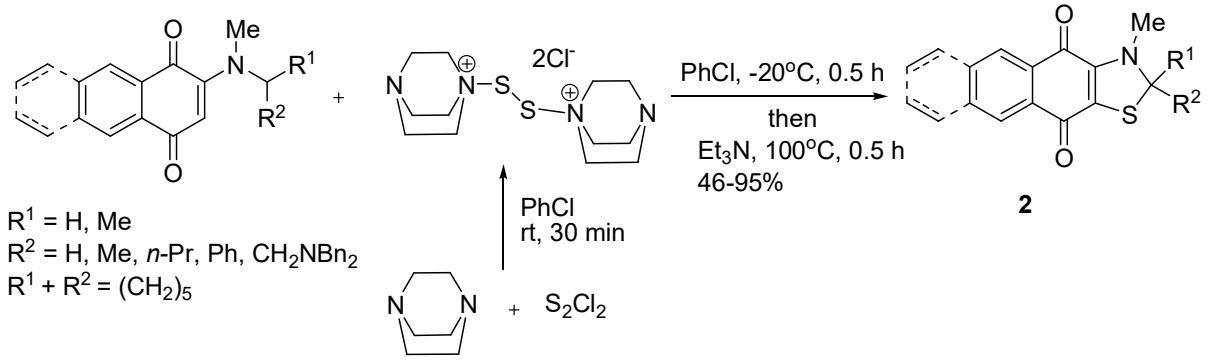
The article gives a brief review of the green and environmentally benign synthetic approaches of thiazole synthesis as reported from year 2013 to 2023.

Khodaei *et al.* successfully synthesized a novel series of catechol-fused tetracyclic compounds, with an imidazo[2,1-b]thiazole central core **1**, through the anodic oxidation of catechols in presence of 2-mercaptobenzimidazole in aqueous solution (Scheme 2) [30].



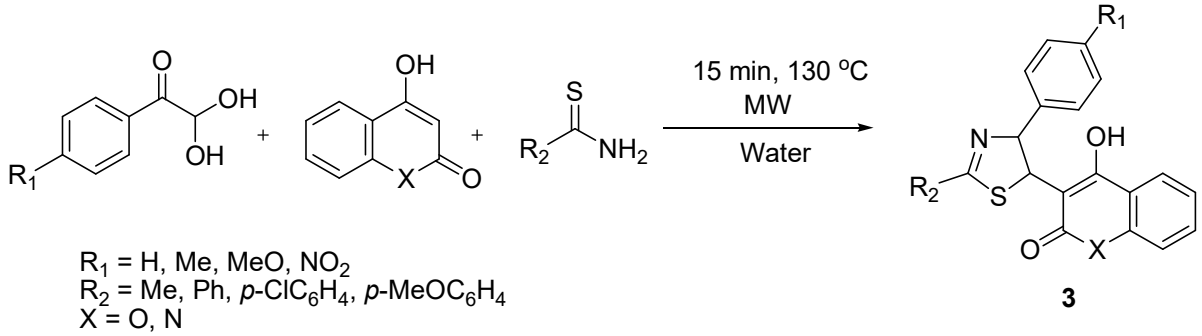
Scheme 2: Synthesis of thiazole derivatives **1.**

Konstantinova *et al.* reported reaction of N-substituted 2-(methylamino) nnaphthoquinones and 2-(methylamino)anthracene-1,4-diones with S_2Cl_2 and 1,4-diazabicyclo[2.2.2]octane (DABCO) leading to 2,3-dihydronaphtho[2,3-d][1,3]thiazole-4,9-diones and 2,3-dihydroanthra[2,3-*d*][1,3]thiazole-4,11-diones **2** (Scheme 3). DABCO being a small diazabicyclic molecule has many advantages, including commercial availability, safety, non-toxicity, affordability, etc., making this synthetic route green [31].



Scheme 3: Synthesis of thiazolediones **2.**

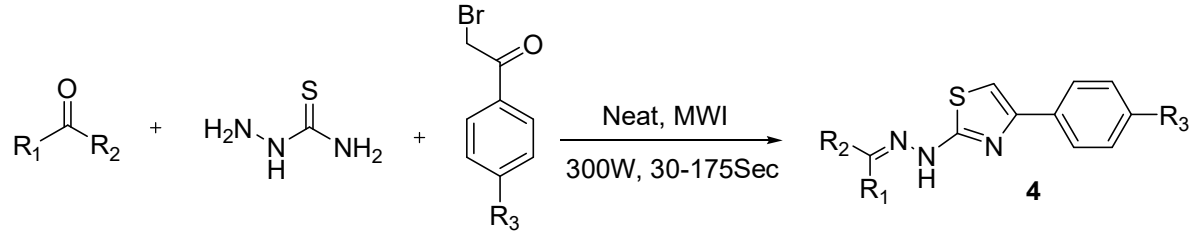
Karamthulla *et al.* reported a clean, efficient and catalyst-free multicomponent domino reaction of arylglyoxals, cyclic 1,3-dicarbonyls and thioamides in H_2O under microwave conditions. A wide variety of trisubstituted thiazoles **3** can be synthesized in good to very good yields using this green protocol (Scheme 4). This method provides advantages like use of green solvent, less reaction time, high yield, and use of microwave irradiation, and no harmful by-products [32].



Scheme 4: Synthesis of trisubstituted thiazole derivatives **3.**

Chinnaraja *et al.* reported an environmentally benign microwave assisted and efficient method for rapid synthesis of hydrazinyl thiazoles **4**. A rapid synthesis of hydrazinyl thiazoles under solvent and catalyst free

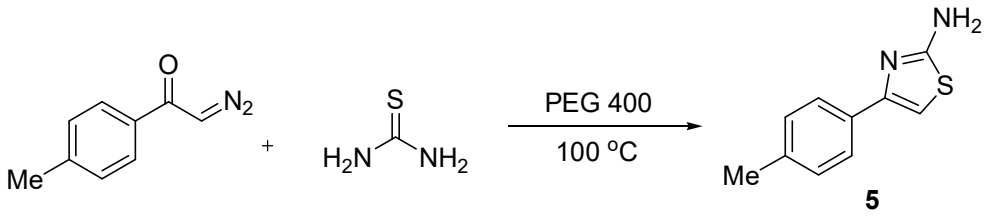
condition was carried by reacting a mixture of aryl ketones, thiosemicarbazide, and substituted phenacyl bromides in one pot under microwave irradiation at 300 W for 30–175 s (Scheme 5) [33].



R_1 = CH_3, 4-ClPh, 4-FPh, 4-NH_2Ph, 4-$NH_{2\text{-}5}$-NO_2Ph, 4-$NH_{2\text{-}5}$-NO_2Ph,4-$NH_{2\text{-}5}$-NO_2Ph
R_2 = Ph, 4-OCH_3Ph, 4-NO_2Ph, 4ClPh, 4-FPh, 4-OHPh, 3,4-$(OCH_3)_2$Ph, Thiophene, Furan
R_3 = OCH_3, Cl, NO_2

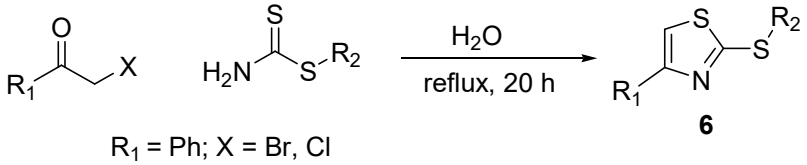
Scheme 5: Synthesis of thiazole derivatives **4.**

Babu *et al.* developed a simple, catalyst-free and efficient method for the synthesis of 2-aminothiazoles **5** from α-diazoketones and thiourea in the presence of PEG-400 for 2–3.5 h at 100 °C (Scheme 6). The desired products 25 were obtained in good yields (87–96%). The green protocol is simple, rapid and generates thiazole derivatives in excellent yields [34].



Scheme 6: Synthesis of thiazole derivative **5.**

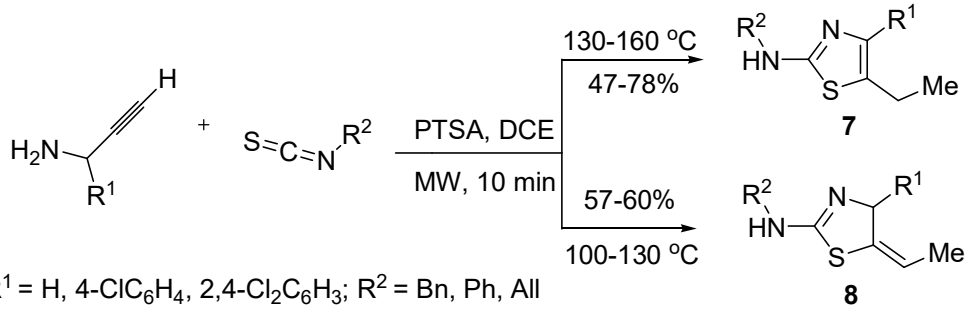
Halimehjani *et al.* reported a simple, green and high-yielding procedure for the synthesis of 4-substituted-2-(alkylsulfanyl)thiazoles **6** from the reaction of dithiocarbamates and α-halocarbonyl containing compounds using water as solvent (Scheme 7). The reactants were refluxed in absence of a catalyst for 20 h, substituted thiazoles were obtained in 75–90% yields. However, if phenacyl bromide was replaced by phenacyl chloride, the reaction got completed within 2 days. [35].



R_1 = Ph; X = Br, Cl
R_2 = Ph, Alkyl

Scheme 7: Catalyst-free **s**ynthesis of thiazole derivatives **6.**

Scalacci *et al.* synthesized 2-aminothiazoles **7** from propargylamines and substituted isothiocyanates under microwave conditions in the presence of catalytic amount of *p*-toluenesulfonic acid (PTSA) in 1,2-dichloroethane (DCE) at 130 °C for 10 min (Scheme 8). The desired 2-aminothiazoles **7** were obtained in 47–78% yields. However, when temperature was lowered to 100 °C, tautomeric 2-amino-4-methylenethiazolines **8** were obtained as major products [36].

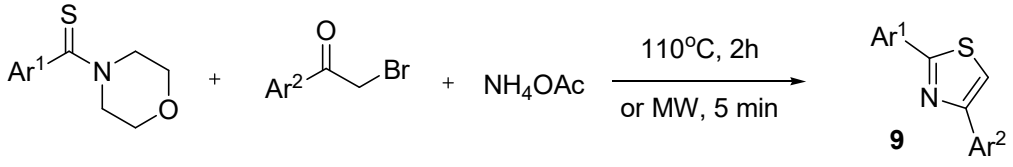


R^1 = H, 4-ClC_6H_4, 2,4-$Cl_2C_6H_3$; R^2 = Bn, Ph, All

Scheme 8: Synthesis of 2-aminothiazole derivatives **7** and **8.**

Zali-Boeini and Mansouri developed a novel one-pot three-component reaction for construction of thiazole derivatives **9** under solvent-free conditions. Tertiary thioamides, α-haloketones, and NH_4OAc were grinded

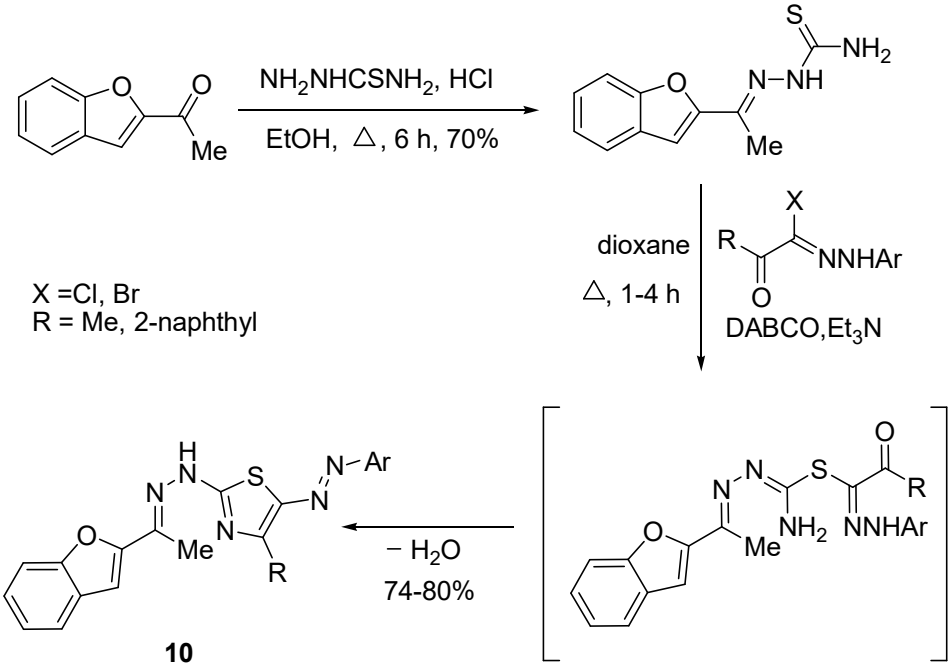
together and allowed to react thermally at 110 °C and/or under microwave irradiation to produce the corresponding thiazole derivatives in very good yields (Scheme 9) [37].



Ar^1 = Ph, 4-MeOPh, 4F-Ph Ar^2 = Ph, 4-MePh

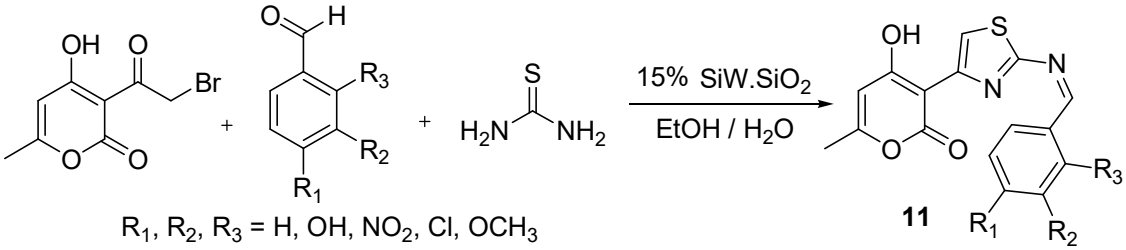
Scheme 9: Synthesis of thiazole derivatives **9.**

Gomha *et al*. reported a simple and convenient method for the synthesis of new thiazole derivatives containing benzofuran moiety **10** from 2-[1-(benzofuran-2-yl)ethylidene]-hydrazinecarbothioamide using 1,4-diazabicyclo[2.2.2]-octane (DABCO) as a catalyst. DABCO is a hindered base, and its advantage over other basic catalysts for that process is eco-friendliness, is safe to handle, recyclable, provides good to excellent yields, and reduces reaction time (Scheme 10) [38].



Scheme 10: Synthesis of new thiazole derivatives containing benzofuran moiety **10.**

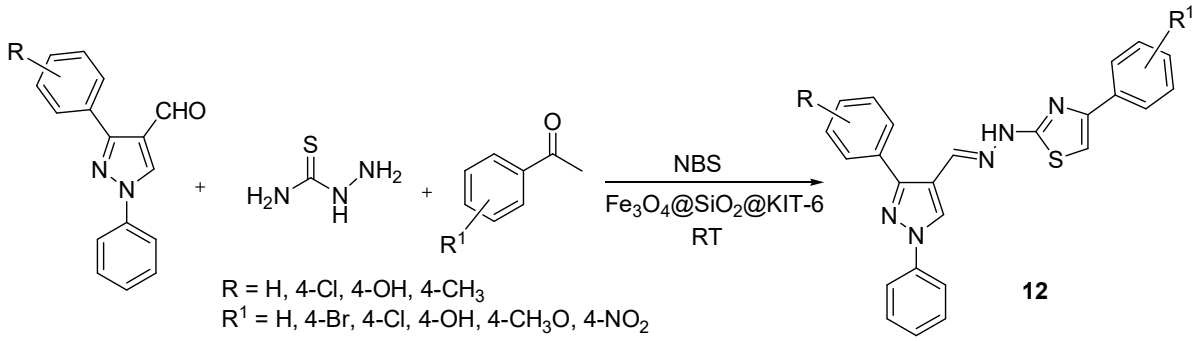
Bouherrou *et al*. developed an efficient and green method for the synthesis of new substituted Hantzsch thiazole derivatives **11** in 79%-90% yield, via the one-pot multi-component procedure, by the reaction of 3-(bromoacetyl)-4-hydroxy-6-methyl-2*H*-pyran-2-one, thiourea and substituted benzaldehydes in the presence of silica supported tungstosilisic acid, as a reusable catalyst, under conventional heating or under ultrasonic irradiation (Scheme 11) [39].



R_1, R_2, R_3 = H, OH, NO_2, Cl, OCH_3

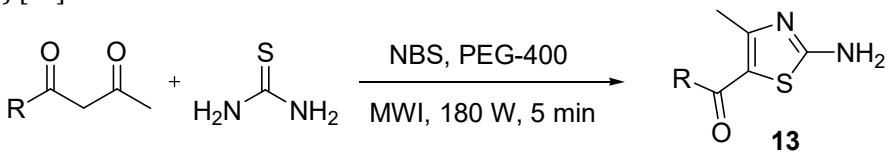
Scheme 11: $SiW.SiO_2$ catalysed synthesis of new Hantzsch thiazole derivatives **11.**

Nikpassand *et al.* reported a clean and environmentally benign route to synthesize a series of benzothiazole derivatives **12** using the reaction between various synthesized aldehydes, thiosemicarbazide, and different acetophenones and *N*-bromosuccinimide as a substrate instead of haloacetophenones in presence of a catalytic amount of silica coated magnetite nanoparticles (Fe_3O_4@SiO_2@KIT-6) (Scheme 12) [40].



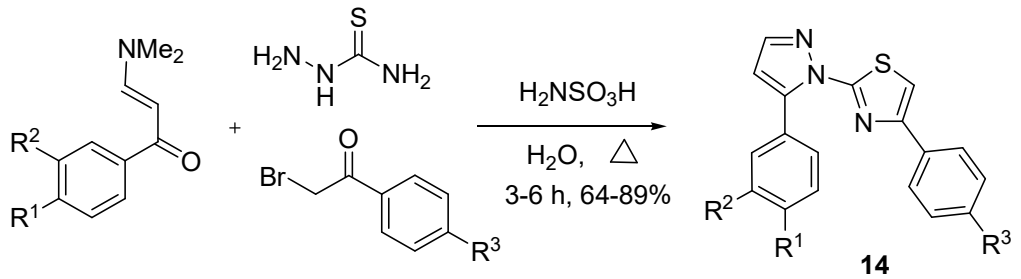
Scheme 12: Synthesis of thiazole derivatives **12.**

Vekariya *et al.* reported the green synthesis of 1-(2-amino-4-methylthiazol-5-yl)ethanone and ethyl 2-amino-4-methylthiazole-5-carboxylate **13** by one pot reaction of acetyl acetone or ethyl acetoacetate with *N*-bromosuccinimide (NBS) and thiourea in the presence of PEG-400 under microwave irradiation at 180 W (Scheme 13) [41].



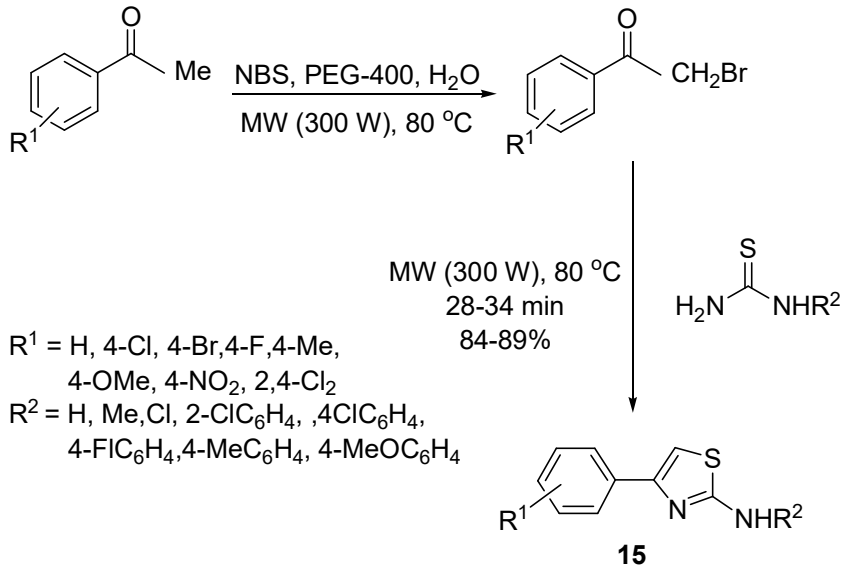
Scheme 13: Synthesis of thiazole derivatives **13.**

Sridevi *et al*. synthesized a series of pyrazolylthiazoles **14** from (*E*)-1-aryl-3-(dimethylamino)-2-propen-1-ones, hydrazinecarbothioamide and substituted 2-bromoacetophenones, catalysed by sulfamic acid using H_2O as green solvent (Scheme 14) [42].



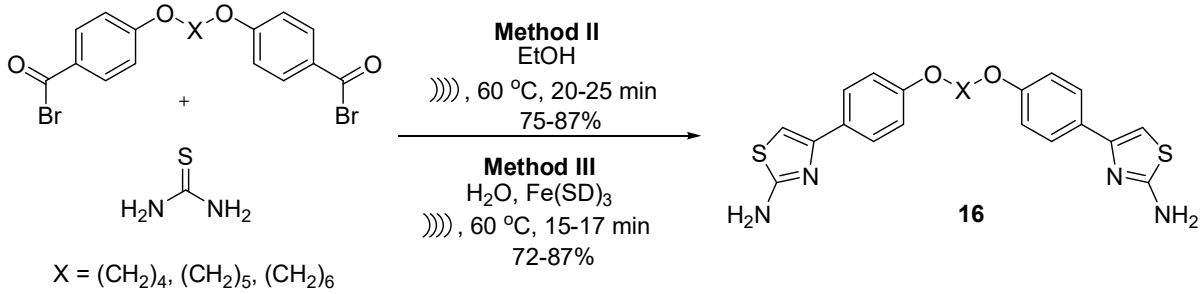
Scheme 14: Sulfamic acid catalysed synthesis of pyrazole-thiazole conjugates **14.**

Wagare *et al.* developed a one-pot protocol for the synthesis of 4-aryl-2-aminothiazoles **15** from the reaction of aromatic ketones, NBS (*N*-Bromosuccinimide) and thioureas under microwave irradiation (MWI) at 80–85 °C in PEG (polyethylene glycol)-400 with 84–89% yields in 28–32 min (Scheme 15). The method has several advantages such as use of water as green solvent, one-pot procedure, easy work-up, excellent yield and avoiding use of toxic α-haloketones [43].



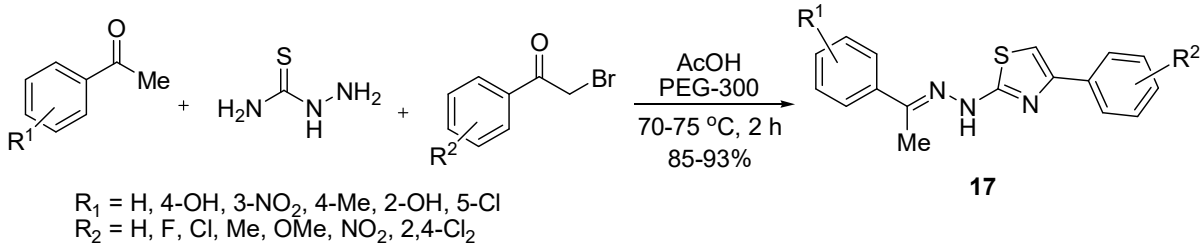
Scheme 15: Synthesis of 4-aryl-2-aminothiazoles **15.**

Parvizi *et al.* reported the ultrasound irradiated synthesis of bisthiazoles **16** using $Fe(SD)_3$ (iron(III) dodecyl sulfate) as a catalyst and H_2O as a solvent. α-Bromo derivatives of bisacetophenone were treated with thiourea under three different reaction conditions: method I – reflux, EtOH, 6 h; method II – ultrasound irradiation, EtOH; and method III – ultrasound irradiation, H_2O, $Fe(SD)_3$; at varied temperature (30–60°C) and time, to obtain bisthiazoles **16** (Scheme 16). The results showed that all three methods give product in significant yields 70–90%, 75–87%, and 72–87%, respectively, but ultrasound mediated methods II and III were considered to be best methods, taking less time for reaction completion, i.e., 20–25 min and 15–17 min at 60 °C, respectively, than method I which required 6 h reaction time. In addition, it is observed that $Fe(SD)_3$ helps in improvement of the yield and H_2O increases the rate of the reaction [44].



Scheme 16: Synthesis of bisthiazoles **16.**

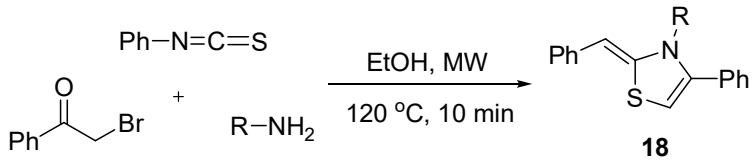
Raut *et al.* synthesized a series of 2-(2-hydrazinyl) thiazole derivatives **17** from substituted ketones, thiosemicarbazide & α-halo ketones using PEG-300 as a reaction medium via one-pot approach. The reaction mixture of substituted ketones and thiosemicarbazide with catalytic amount of glacial acetic acid was first heated for 2 h at 70–75 °C, followed by gradual addition of α-halo ketones and heating again for 15 min (Scheme 17). The desired products were obtained in good yield (85–93%) [45].



R_1 = H, 4-OH, 3-NO_2, 4-Me, 2-OH, 5-Cl
R_2 = H, F, Cl, Me, OMe, NO_2, 2,4-Cl_2

Scheme 17: Synthesis of a series of 2-(2-hydrazinyl) thiazole derivatives **17.**

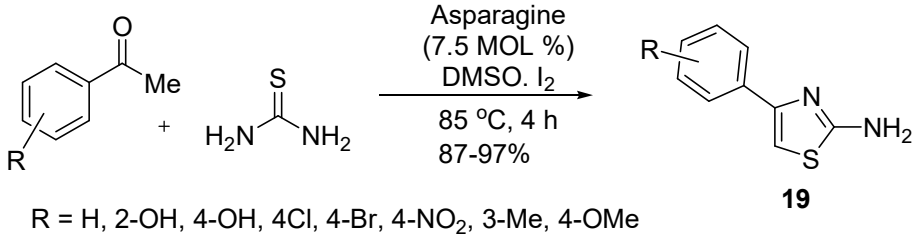
Shaikh *et al*. developed a microwave-assisted green approach for an efficient synthesis of thiazol-2-imines **18** under catalyst-free conditions – an improvised method for Hantzsch thiazole synthesis. The desired products were formed in excellent yields with high purity in 10–15 min by one-pot three-component reaction (Scheme 18) [46].



R = Ph, 2-ClPh, 3-ClPh, 4-ClPh, 4-BrPh, 4-FPh, 4-CH_3OPh, 4-CH_3Ph, 4-CF_3Ph,3-NO_2Ph, -NHPh, -$(CH_2)_2$

Scheme 18: One-pot three-component synthesis of thiazole-2-imines **18.**

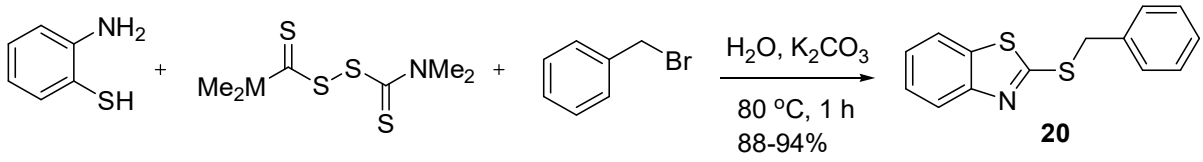
Safari *et al*. reported an efficient method for the one-pot synthesis of 2-aminothiazoles **19** via the reaction of thiourea with methylcarbonyls in the presence of I_2 as an oxidant reagent and asparagine as a green organocatalyst at 80 °C in DMSO solvent. Here, asparagine acts as bifunctional catalyst containing two groups, i. e., NH_2 group (Lewis base) and COOH group (Brønsted acid), which are involved in the mechanism of the reaction. Advantages of this efficient method include greener and cleaner conditions, easy isolation of products, higher purity and excellent yield of products, and convenient manipulation (Scheme 19) [47].



R = H, 2-OH, 4-OH, 4Cl, 4-Br, 4-NO_2, 3-Me, 4-OMe

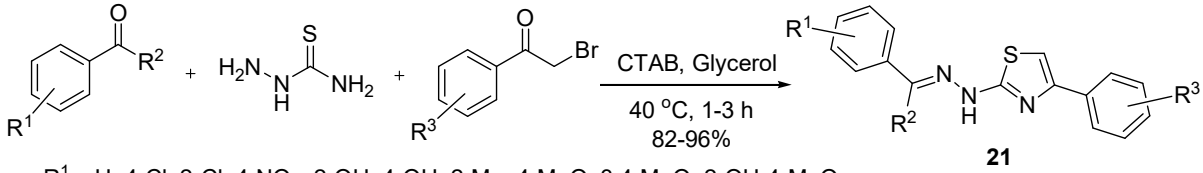
Scheme 19: One-pot synthesis of 2-aminothiazoles **19.**

Zhang *et al.* developed an efficient transition-metal-free protocol for the one-pot synthesis of 2-benzyl/2-allyl-substituted thiobenzothioazoles **20** in H_2O. The one-pot reaction of 2-aminobenzenethiol, tetramethylthiuram disulfide, and benzyl bromide was optimized using variety of bases and solvents at different temperatures (Scheme 20). The desired product **20** was obtained in excellent yield (88– 94%) in 1 h at 80°C temperature in presence of K_2CO_3 as a base and H_2O as a solvent [48].



Scheme 20: One-pot three-component synthesis of substituted thiobenzothioazole 20**.**

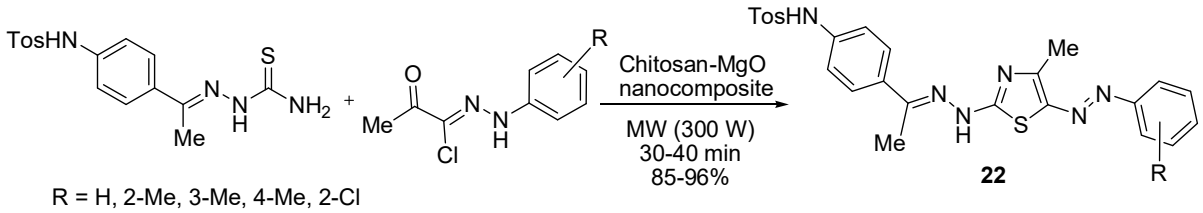
Tiwari *et al.* reported an efficient and green method for the synthesis of 2,4-disubstituted thiazole derivatives **21** through an efficient multicomponent reaction of substituted benzaldehydes, thiosemicarbazide, and phenacyl bromides. The reaction was first optimized using variety of solvents and catalysts. The products were obtained in short duration of time (1–3 h) with excellent yields (82–96%) in the presence of cetyl trimethyl ammonium bromide (CTAB) – a phase-transfer catalyst and glycerol as a solvent at 40°C (Scheme 21) [49].



R^1 = H, 4-Cl, 2-Cl, 4-NO_2, 2-OH, 4-OH, 2-Me, 4-MeO, 3,4-MeO, 2-OH-4-MeO
R^2 = H, Me; R^3 = H, 4-Cl,4-MeO

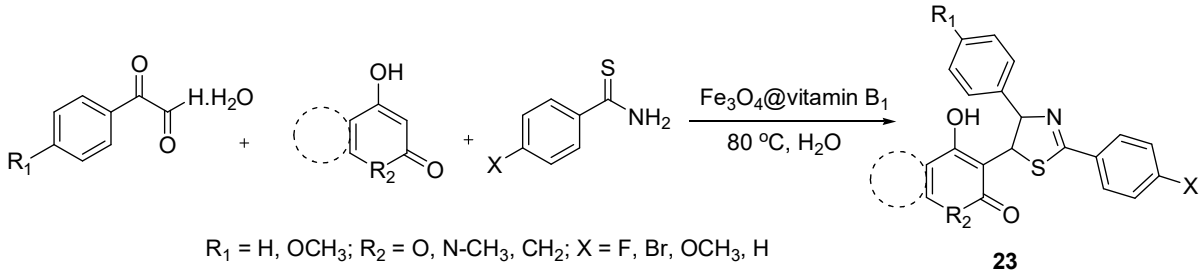
Scheme 21: Synthesis of 2,4-disubstituted thiazole derivatives **21.**

Riyadh *et al.* prepared a chitosan-MgO hybrid nanocomposite which served as a powerful ecofriendly basic catalyst under microwave irradiation in the synthesis of novel series of 5-arylazo-2-hydrazonothiazoles **22**. A comparative study in terms of yield between traditional catalyst Et_3N and chitosan–MgO nanocomposite has been done. Thiosemicarbazone and *N*-aryl-2-oxopropanehydrazonoyl chlorides dissolved in dioxane were treated with both catalysts (separately) under microwave heating (Scheme 22). The method using chitosan–MgO nanocomposite produced desired product (R = 3-Me) in higher yield (96%) than Et_3N (82%). The chitosan–MgO nanocomposite possessed more catalytic efficiency due to nano-size of MgO and synergistic effect produced by both basic MgO and chitosan. The catalyst can be recovered and reused many times in other synthetic reactions with the same catalytic efficacy [50].



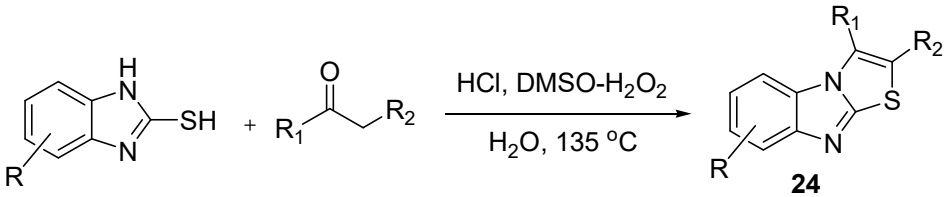
Scheme 22: Synthesis of 5-arylazo-2-hydrazonothiazoles **22**.

Shaterian *et al.* designed and prepared Fe_3O_4@vitamin B_1 as an inexpensive and efficient heterogeneous nano-catalyst for the synthesis of a new 1,3-thiazole derivative **23**. The three component, one-pot condensation of arylglyoxal monohydrate, cyclic 1,3-dicarbonyls, and thioamides in water as a green solvent was applied for the preparation of 1,3-thiazole derivatives (Scheme 23) [51].



Scheme 23: Synthesis of trisubstituted thiazoles **23.**

Zu *et al.* reported a green and regioselective synthesis for the construction of benzoimidazo[2,1-b]thiazole skeleton **24** from 2-Mercaptobenzimidazoles and Ketones using DMSO-H_2O_2 as an oxidant and water as solvent. This operationally simple and metal-free procedure could facilitates a diverse collection of benzoimidazo[2,1-b]thiazole derivatives (Scheme 24) [52].

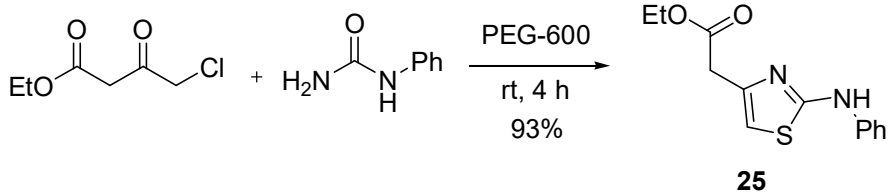


R = Cl, CH_3, CH_3O, NO_2

R_1, R_2 = $(CH_2)_{4\text{-}6}$, CH_3, CH_3CH_2, $CH_3CH_2CH_2$,C_5H_{11}

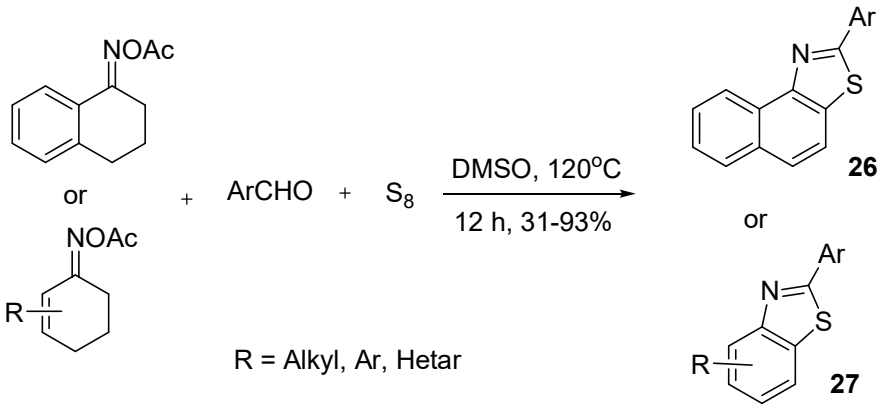
Scheme 24: Synthesis of thiazole derivatives **24.**

Kavitha *et al.* reported a facile pathway for the one-pot three-component synthesis of 3-(2-(phenylamino)thiazol-4-yl)-2*H*-chromen-2-ones **25** starting from ethyl-4-chloroacetoacetate and phenylthiourea in presence of PEG-600 (polyethylene glycol) at room temperature, in good yield (93%) and short period of time (Scheme 25). During optimization study, PEG-600 was found to be the best (green) reaction media in comparison to other solvents like EtOH, THF, glycerol, MeCN, etc. In addition, the reaction also completed at room temperature due to use of PEG-600. It also increases the rate of reaction by acting as phasetransfer catalyst, which initiates the nucleophilic substitution of ethyl 4-chloroacetoacetate with phenylthiourea, followed by cyclization to obtain thiazole 2**5** [53].



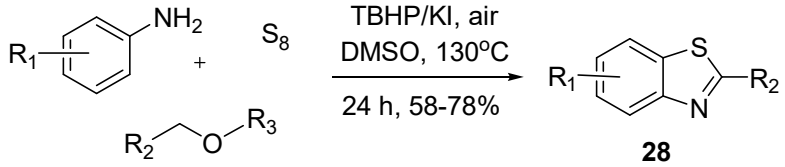
Scheme 25: Synthesis of thiazole derivative **25.**

Xu *et al.* reported a catalyst-free and additive-free synthetic route to naphthothiazoles **26** and benzothiazoles **27** using DMSO as a green solvent from 1-tetralone oxime acetate or cyclohex-2-en-1-one oxime acetates, aromatic aldehydes, and elemental sulfur (Scheme 26). The proposed method was efficient even on gram scale and broad range of functionalities were also well tolerated [54].



Scheme 26: Synthesis of naphthothiazoles **26** and benzothiazoles **27.**

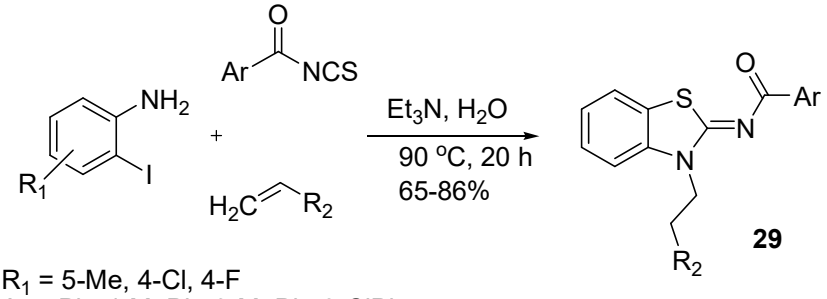
Zhang *et al*. reported a synthetic route for the synthesis of substituted benzothiazoles **28** via the annulation of substituted anilines with ethers and elemental sulfur using tert-Butyl hydroperoxide (TBHP)/KI as promoter through selective C–O bond cleavage of ethers under transition metal-free conditions. TBHP acts as a green oxidant, forming more stable radicals in comparison to other oxidants. A wide range of 2-aryl-, 2-heteroaryl-, and 2-alkyl-substituted benzothiazoles were easily synthesized with acceptable yields and good functional group compatibility (Scheme 27) [55].



R_1 = H, Me, Et, *t*-Bu, OMe, F, Br, Cl, CF_3, CN
R_2 = Alkyi, Hetar, R_3 = Me, Et, OMe, Cl. Br, NO_2

Scheme 27: Synthesis of substituted benzothiazoles **28.**

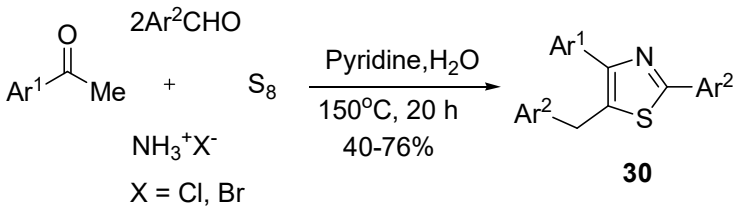
Saini *et al.* accomplished an environmentally benign, transition-metal-free organic base promoted one-pot cascade synthesis of highly functionalized benzo[d]thiazol-2(3*H*)-ylidene benzamide **29** in the presence of water by three-component reaction of *o*-iodoanilines, acrylates, and aroyl isothiocyanates. The protocol starts with in-situ generation of thiourea intermediate followed by Et_3N induced intramolecular SN_{Ar} displacement reaction and subsequent Michael addition onto acrylate leading to the formation of benzo[d]thiazol-2(3*H*)-ylidene benzamide (Scheme 28) [56].



R_1 = 5-Me, 4-Cl, 4-F
Ar = Ph, 4-MePh, 3-MePh, 3-ClPh
R_2 = CO_2Me, CO_2Et, CO_2n-Bu, CO_2t-Bu, $CON(Me)_2$. CN

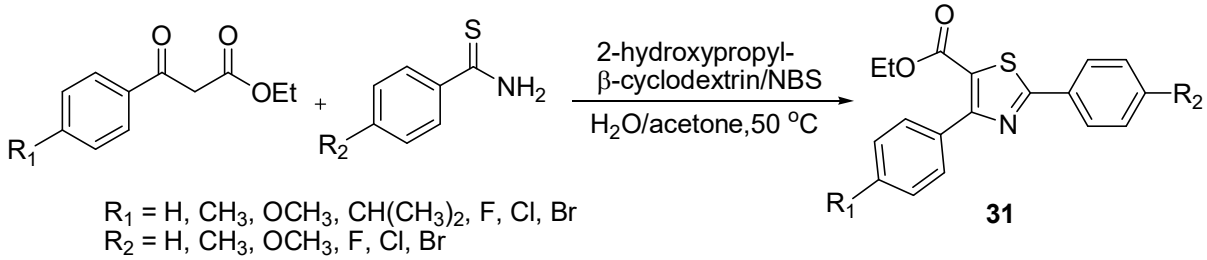
Scheme 28: One-pot cascade synthesis of highly functionalized benzo[d]thiazol-2(3*H*)-ylidene benzamide derivatives **29.**

Jiang *et al.* reported an environmentally benign, multicomponent reaction under metal-free conditions to synthesize polysubstituted thiazoles **30** from readily available chemicals utilizing ketones, aldehydes, ammonium salts, and elemental sulfur as substrates in pyridine. Pyridine as the reaction medium enhanced the yield. Besides, the solubilities of inorganic ammonium halide and elemental sulfur were improved by the addition of H_2O, which led to a further enhancement in yield with a range of functionalities tolerated (Scheme 29) [57].



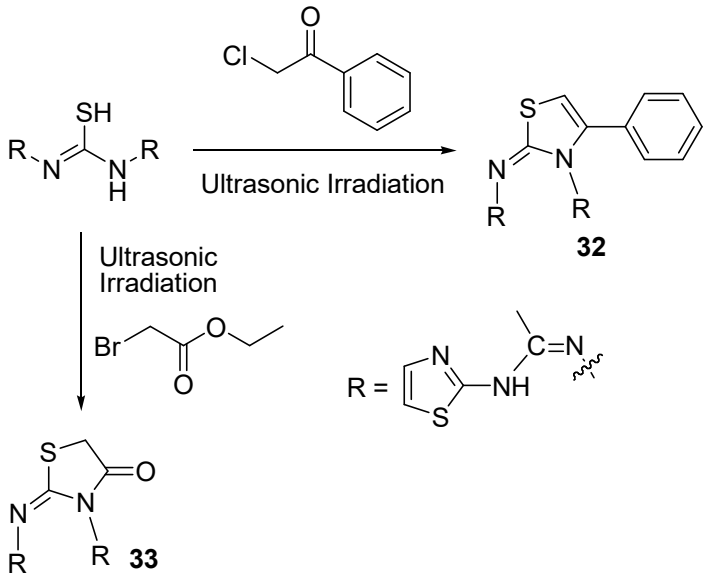
Scheme 29: Synthesis of polysubstituted thiazoles **30** by four-component reaction.

Zhang *et al*. reported a new protocol to synthesize 2,4-diphenyl thiazole analogs **31**, which involved the bromination of ethyl benzoylacetates with NBS in the presence of 2-hydroxypropyl-β-cyclodextrin, followed by a direct cyclization with thiobenzamides in water (Scheme 30) [58].



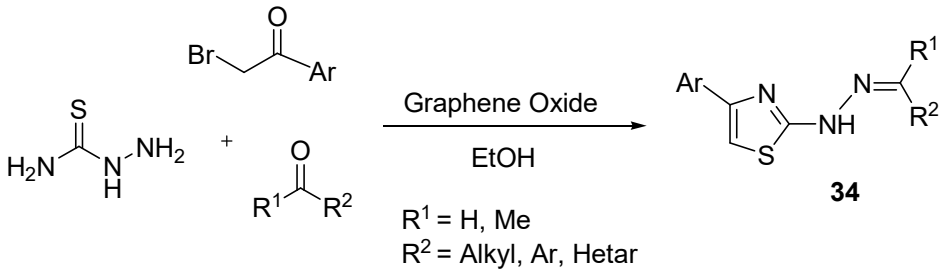
Scheme 30: Synthesis of substituted 2,4-diphenyl thiazoles **31.**

Shabaan *et al.* reported an eco-friendly, green, efficient approach for the synthesis of new 1,3-thiazoles using ultrasonic irradiation and solvent-free conditions. Thiocarbohydrazones were reacted with α-chloroketone or α-bromoester under ultrasonic conditions affording 1,3-thiazoles 3**2** and **33** (Scheme 31) [59].



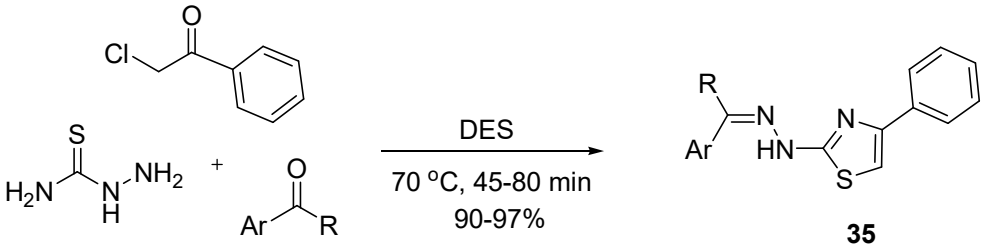
Scheme 31: Synthesis of thiazole derivatives **32** and **33.**

Das *et al.* developed an efficient, one-pot three-component reaction in the synthesis of a wide range of 2,4-disubstituted hydrazinyl thiazole scaffolds **34** in ethanol at room temperature by the reaction of carbonyl compounds, phenacyl bromides, and thiosemicarbazide, using graphene oxide (GO) as a catalyst. The GO catalyst has a high reusability rate and is simple to recover. This catalytic method provided a shorter reaction time, broad substrate scope, low catalyst loading, environmentally benign solvent media, easy handling, and operational simplicity at room temperature (Scheme 32) [60].



Scheme 32: Synthesis of 2,4-disubstituted hydrazinyl thiazole scaffolds **34.**

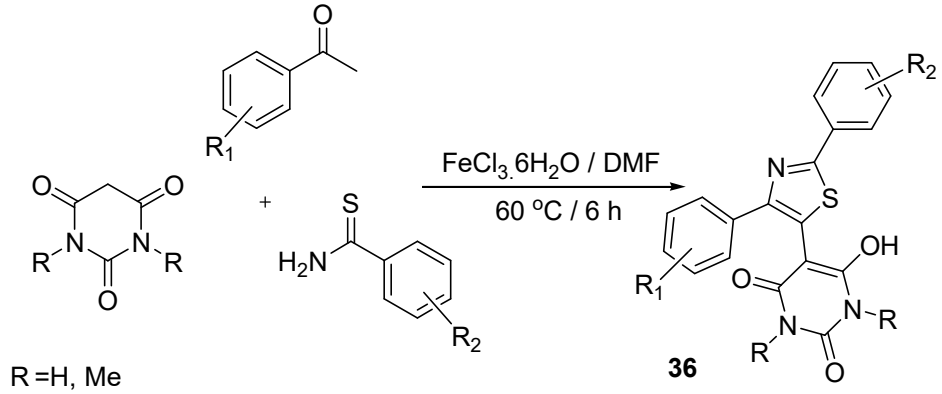
Kaldareh *et al*. developed a one-pot three-component synthesis of hydrazinyl-4-phenyl-1,3-thiazole derivatives **35** by reaction of various ketones or aldehydes, phenacyl chloride, and thiosemicarbazid in the presence of choline chloride/urea in 1:2 ratio as a deep eutectic solvent (DES) at 70 °C (Scheme 33). The products were formed in excellent yields over short reaction times under an environmentally friendly condition. Here, DES acts as a solvent as well as catalyst and afforded the desired products 35 in excellent yields (90–97%) [61].



Ar = Ph, 4-ClC_6H_4, 2-ClC_6H_4, 2,4-$Cl_2C_6H_3$, 4-$NO_2C_6H_4$, 3-$NO_2C_6H_4$
3-$MeOC_6H_4$, 4-$MeOC_6H_4$, 2-OHC_6H_4, 4-OHC_6H_4, 2,4-$(OH)_2C_6H_3$, 5-Br-2-OHC_6H_3,
1-naphthyl, 2-naphthyl; R = H, Me

Scheme 33: Synthesis of hydrazinyl-4-phenyl-1,3-thiazole derivatives **35.**

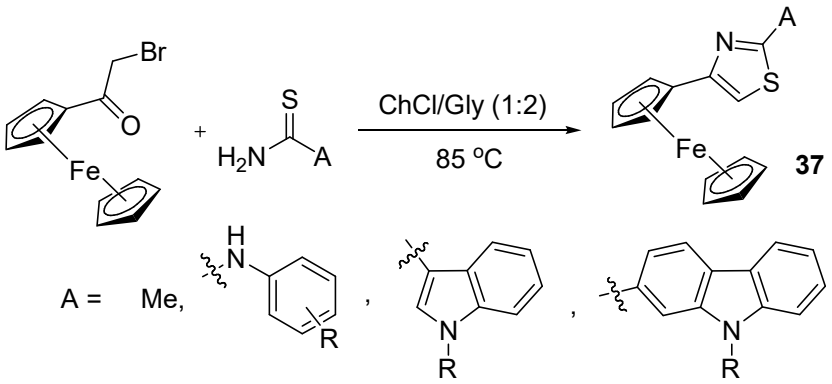
Singh *et al*. reported a clean and efficient, multi-component strategy for the synthesis of biologically important trisubstituted thiazole derivatives **36** through the reaction of readily available barbituric acid, acetophenone, and aryl thioamides in presence of $FeCl_3.6H_2O$ / O_2 (Air) in DMF solvent (Scheme 34). The advantages of the present strategy include a multi-component one-pot reaction, environment-friendly approach, cost-effectiveness, broad substrate scope, operational simplicity, short reaction time, easy workup procedure and high yields [62].



R =H, Me
R_1, R_2 = H, 4-CH_3, 2-CH_3, 4-OCH_3, 2-OCH_3, 4-Cl, 4-Br etc.

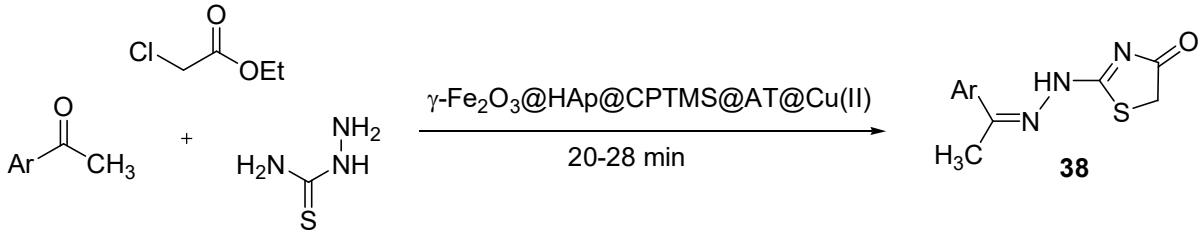
Scheme 34: Synthesis of trisubstituted thiazole derivatives **36.**

Zhao *et al*. accomplished a green Hantzsch synthesis of 4-ferrocenylthiazole derivatives **37** successfully by reacting bromoacetylferrocene with various aryl thioureas and 1-alkylindole-3- or 9-alkylcarbazole-3-carbothioamides efficiently in a deep eutectic solvent (DES) that is, choline chloride/glycerol (ChCl/Gly) (1:2 molar ratio) at 80 °C, avoiding the use of common volatile organic solvents (Scheme 35). This synthetic strategy has the attractive features such as mild and environmentally benign reaction conditions, experimental simplicity, easy work-up procedure and good yields [63].



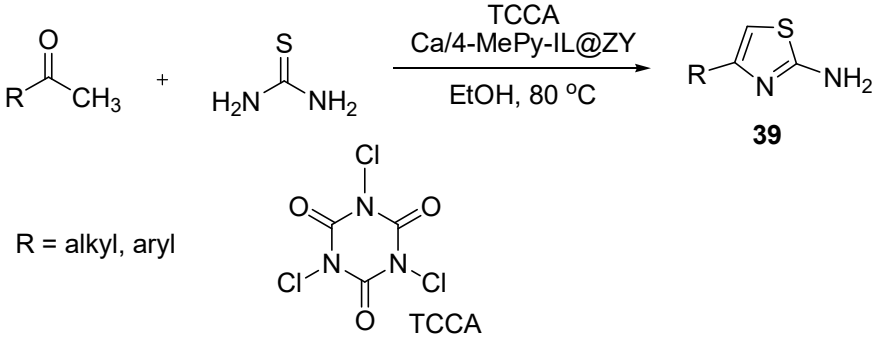
Scheme 35: Synthesis of ferrocene-based thiazole hybrids **37.**

Ghorbani Dehshal *et al.* devised a highly efficient one-pot three-component protocol for the synthesis of novel derivatives of 2-(2-(phenylethlidene)hydrazinyl)thiazol-4(5*H*)one **38** by the reaction of acetophenone derivatives, ethyl chloroacetate and thiosemicarbazide, in ethanol at room temperature in the presence of copper supported hydroxyapatite-encapsulated-γ-Fe_2O_3 (γ-Fe_2O_3@HAp@CPTMS@AT@Cu(II)) as the new magnetically recyclable heterogeneous nanocatalyst. The novel protocol furnished the desired products in excellent yields (88–95%) and short reaction times of 20–28 min (Scheme 36) [64].



Scheme 36: Synthesis of thiazole derivatives **38.**

Kalhor *et al.* synthesize a novel, multifunctional ionic liquid nanocatalyst based on zeolite-Y with 4-methylpyridinium chloride (4-MePy-Cl) and calcium ions (Ca/4-MePy-IL@ZY), then its catalytic activity in the one-pot synthesis of 2-aminothiazoles **39** using trichloroisocyanuric acid (TCCA) as a green supplier of halogen ions for intermediates was studied (Scheme 37). The use of non-toxic solvent and a cheap, safe, recyclable nano-catalyst, quick reaction times, high efficiency, and ease of nano-catalyst separation are additional benefits of this method [65].



Scheme 37: Synthesis of 2-aminothiazole derivatives **39.**

CONCLUSION

Heterocyclic compounds are continuously exploited for the development of libraries of biologically active compounds. Thiazole ring is one of the such frameworks possessing many biological activities, thus being explored by many researchers for the development of novel leads. Various conventional as well as green synthetic approaches have been reported for the synthesis of thiazole derivatives. However, the green synthetic approach involving use of environmentally benign chemicals, production of low waste, atom economy, excellent yields and short reaction time have overcome the problems generally associated with

the conventional methods of synthesis. In conclusion, we expect that the present review will assist many researchers toward the development of new thiazole derivatives employing greener approach.

ABBREVIATIONS

DABCO: 1,4-diazabicyclo[2.2.2]octane; PEG: Polyethylene glycol; PTSA: *p*-toluenesulfonic acid; DCE: 1,2-dichloroethane; NH_4OAc: Ammonium acetate; NBS: *N*-bromosuccinimide; MWI: Microwave Irradiation; $Fe(SD)_3$: Iron(III) dodecyl sulfate; EtOH: Ethanol; DMSO: Dimethyl sulfoxide; Et_3N: Triethyl amine; THF: Tetrahydrofuran; MeCN: Methyl cyanide; TBHP: tert-Butyl hydroperoxide; SN_{Ar}: Aromatic Nucleophilic substitution; GO: Graphene oxide; DES: Deep eutectic solvent; ChCl: Choline chloride; Gly: Glycerol.

ACKNOWLEDGEMENTS

M. M. Masood acknowledges the Department of Chemistry, Government Degree College for Women Baramulla, J&K India, for providing the logistical support throughout the review work.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR'S CONTRIBUTION

MMM is the main author and he has collected, analysed, conceived and designed the study and wrote the first draft. AI reviewed the draft and gave additional inputs to improve the scientific accuracy required and corrected the manuscript wherever required. Both authors read and approved the final draft of manuscript.

FUNDING

Not applicable

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CITATION OF THIS ARTICLE

Mir Mohammad Masood and Arshid Iqbal. Environmentally Benign Synthetic Approaches for the Synthesis of Thiazole Derivatives: A Review.. Bull. Env. Pharmacol. Life Sci., Vol 13[2] January 2024: 69-84